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Towards a circular management of nitrogen and phosphorus in human excreta: current state, global agricultural potential, and spatial constraint in France

Thomas Starck

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Towards a circular management of nitrogen and phosphorus in human excreta: current state, global agricultural potential, and spatial constraint in France.

École doctorale N°531, Sciences, Ingénierie et Environnement (SIE)
Sciences et Techniques de l'Environnement

Thèse préparée au Laboratoire Eau Environnement et Systèmes Urbains (LEESU)

Thèse soutenue le 22 octobre 2024, par
Thomas STARCK

Composition du jury:

Anne, TREMIER *Présidente de jury*
Directrice de Recherche, INRAE

Sylvie, GILLOT *Rapporteuse*
Directrice de Recherche, INRAE

Anne, TREMIER *Présidente de jury*
Directrice de Recherche, INRAE

Jeremy, GUEST *Examineur*
Associate Professor, University of Illinois

Bruno, TASSIN *Directeur de thèse*
Directeur de recherche et Professeur, ENPC

Fabien, ESCULIER *Encadrant de thèse*
Chercheur et Ingénieur IPEF, ENPC

Julia, LE NOE *Invitée*
Chargée de recherche, IRD

Mathilde, BESSON *Invitée*
Post-doctorante, INSA Toulouse

Abstract and Research Question

Short Abstract

Nitrogen (N) and Phosphorus (P) in human excreta can serve as fertilizers, but are seen as a pollution. In this thesis, I first focus on the disturbed N and P flows and how excreta are handled. I then study the French sanitation system: about 10% of the N and 50% of the P in French excreta is reused in agriculture, while national wastewater treatment plant (WWTP) removal efficiency are 70% and 80%. I then estimate the global agronomic potential of N in human excreta, which represents 15-20% of N inputs to croplands, with a lot of variation between countries; in a less wasteful and more vegetarian food system, this could be around 30-40%. The global quantity of N in human excreta is similar to the N in manure spread on croplands. Finally, I estimate that 60-70% of French excretions could be spread on croplands within less than 10 km. For this work, I produced 2 detailed datasets: one of the 20,000 French WWTPs over 2000-2020; and one of fertilization at fine spatial resolution in France for 2015-2020.

Detailed Abstract

Nutrients in human excreta could be used as fertilizers for croplands, but are mostly seen as a polluting waste. In this thesis, I assess the current fate of nitrogen (N) and phosphorus (P) in human excreta and their potential contribution to fertilization, at the global scale and with a specific focus on France. I first describe the current situation of the French sanitation systems, based on a dataset of its more than 20,000 wastewater treatment plants (WWTP) over 2000-2020. About 10% of the N in French excreta is reused in agriculture, through wastewater sludge; 50% is lost as inert atmospheric N_2 following denitrification; and the remaining 40% is lost as reactive N in waters and the atmosphere. The indicator currently followed by policy makers, the wastewater removal efficiency (70% for N in France), is blind to most of these losses, as it only looks at the share of N not discharged in waters. For P, I estimate the national reuse rate to be around 50%. The main difference with N is that P removal relies on precipitation in sludge, allowing better recycling if

removal efficiency is high (80% in France) and if sewage sludge are mostly spread on agricultural land (75% in France); but losses still happen outside WWTPs, in sewers and individual autonomous systems. I also show the influence of the European legislation on removal efficiency performance, namely for large WWTP in areas classified as N or P sensitive to eutrophication.

The current low reuse rate of N in human excreta testifies of an unfulfilled agronomic potential, were it to be reused as fertilizer. I assess this potential for the current food system. N in human excreta expressed as a percentage of N inputs to croplands usually spans from 5% in high inputs agricultural exporters to 50% in low inputs agricultural importers, and most countries are between 10 to 30%. At the global scale, with no imports nor exports, the rate is 15-20%, and corresponds to the global food system N use efficiency. I also give tentative estimates for a more frugal food systems (higher N use efficiency, less carnivorous diets, reduced food waste) and find upper plausible values of 30%, maybe 40% with considerable changes and mostly vegetarian diets. Also, in most countries and at the global scale, quantities of N in humans and non ruminant excretions are roughly similar. The global quantity of N in human excreta is also roughly equivalent to the N in all livestock manure collected and spread on croplands.

Actual recovery rate of nutrients in human excreta will depend on socio-technical factors, and I focus on one particular feature related to the logistics of handling human excreta, in France. Based on a fine resolution dataset of fertilization and excretions over 2015-2020, I assess the distance from excretions to crops, were all the human excretions to be recovered and spread in an optimized manner. With the current N fertilization rates, the mean distance would be 14 km, quite similar to the 17 km I find with lower inputs fertilization practices for an agro-ecological scenario. In both cases, 60-70% of N in excretions could be spread within a short distance of 10 km; 20-25% of excretions could be spread within 10-50 km, mostly corresponding to medium-size cities (0.1-1 million inhabitants). Finally, 5-15%, all located in the populated regions of Paris and the Mediterranean coast, have to be spread further than 50 km. Based on existing life-cycle analyses, my results suggest that in France, in most of cases, concentration of human excreta before transport would not be needed on environmental grounds, but would rather be a matter of logistical convenience (i.e. in densely populated areas) or of farmers preferences to handle smaller quantities.

To conclude, reusing nutrients in excreta can contribute to decrease pollution and lessen the need for new inputs in agricultural systems, and contribute to build less wasteful food/excreta systems.

Research Question

Nitrogen and Phosphorus are nutrients essential to life and agricultural systems, but in excess they result in pollution and health impacts: eutrophication, contamination of groundwaters, atmospheric pollution, acidification of soils and contribution to climate change... These excess mainly originate from N and P fertilizers used as new inputs in a wasteful food system. Reusing N and P in human excreta could reduce the food system needs for new inputs, and its losses and pollution; but this options is often barely considered. All this general background is detailed in Chapters 1, 2, and 3. From this I formulate the following research question:

What is the current state regarding the fate of nitrogen and phosphorus in human excretions used as fertilizers, and what could be their agronomic potential ?

To answer this question, I first undertake a nutrient mass balance of the current French sanitation system, based on operational data of the more than 20,000 wastewater treatment plants. This is the focus of Chapter 4. Besides the recycling rate of nitrogen and phosphorus, this assessment allows to take a deep dive into the sanitation system features. The main conclusion is that in industrialized countries more than 90% of excreted nitrogen is lost. For that reason, I will focus more particularly on that nutrient in the following.

I address in Chapter 5 the question of the agronomic potential of nitrogen in human excretions. I purposely do not focus on how much could be realistically recovered; this is studied in details by other sections of the OCAP program, through field studies. I hypothesize a 100% recovery rate, so that my results could easily be adapted to more realistic rates, and look at how much of the current agricultural nitrogen inputs could be covered with human excretions. I also focus on a wide range of countries, to study a more diverse set of situations. I also focus on the potential in more frugal and less wasteful agricultural systems with lower nutrient inputs, as described in existing agro-ecological scenarios.

In Chapter 6, I go beyond this unconstrained potential and assess one particular constraint, the geographical one, as human excretions and fertilized crops are not necessarily close to each other. I study the case of France, where I can produce detailed geospatial data for both human excretions and croplands fertilization. Under certain hypotheses, this allows to assess typical distances to transport human excretions to the surrounding fields. I also look at how these results are changed in a low nitrogen input scenario.

To summarize, my method to answer the research question is to proceed in progressive steps: description of the current situation; maximum theoretical potential,

without any constraint; study of one particular constraint, the geospatial matching, which affects logistics and transport.

Foreword

This thesis is divided into two main parts.

Part I focuses on the concepts I mobilize during my dissertation: nitrogen and phosphorus roles in life and ecosystem, and the pollutions resulting from their excessive quantity (Chapter 1); the cause of these pollutions, namely new anthropogenic N and P inputs disturbing the global biogeochemical flows (Chapter 2); and finally how humans handle these nutrients after their excretions, and how they could be better recovered and used as fertilizers (Chapter 3). This Part I can be seen as a general detailed introduction, based on a large literature review, but in which I bring very few original and personal results, even though I sometimes include personal reflection. Yet I hope that I contribute to some originality by bringing together subjects that are often treated separately.

Part II presents original results that I produced, some of which have been published in scientific journals at the time of writing (Starck et al. 2024a; Starck et al. 2024b), or as a repository database (Starck 2023). This Part features a detailed N and P assessment of the French sanitation system (Chapter 4); a first-order estimate of the potential of human excretions used as N fertilizers in most countries in the world and in more frugal food systems (Chapter 5); and finally a fine geospatial analysis in France of the matching of human excretions to crops fertilization (Chapter 6).

Abbreviations

- **WWTP**: WasteWater Treatment Plant
- **N**: Nitrogen
- **P**: Phosphorus
- **IAS**: Individual Autonomous System
- **NUE**: Nitrogen Use Efficiency
- **RPG**: Registre Parcellaire Graphique
- **NPP**:Net Primary Production

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Part I

General Background: Nitrogen and Phosphorus disrupted flows; Handling Human Excreta

Chapter 1

Nitrogen and Phosphorus in life and ecosystems

This chapter focuses on the role of nitrogen and phosphorus in life and ecosystem. First I review their indispensable roles in living organisms, and more particularly what are considered the basic nutritional needs in humans (Section 1.1). Then I review in Section 1.2 nitrogen and phosphorus compounds and their interactions in ecosystems (soil and freshwaters). I finally describe the many pollutions resulting from too much nitrogen and phosphorus (Section 1.3).

1.1 The role of N and P in life

In this section I detail how N and P elements appeared in the universe (Section 1.1.1) and the essential role they play in living organisms (Section 1.1.2). Finally, in Section 1.1.3, I focus more particularly on humans and their N and P intakes requirement in order to have a properly functioning metabolism.

1.1.1 How atomic elements appeared

This section is inspired from Elser et al. (2020a) and Brownlee (1992); the reader can also refer to DOE (2020) for a popularized explanation of nucleosynthesis or Burbidge et al. (1957) and Alpher et al. (1948) for seminal and more detailed papers.

According to the current understanding of the universe story, the first atomic elements (H, He and Li) emerged in the first minutes of the universe, during the Big Bang nucleosynthesis. But the universe temperature was too hot to allow electrons to be associated to protons. These elements stayed in a plasma state, in the form of a nucleus of protons and neutrons. Once the universe temperature was sufficiently low, these nuclei bound to electrons, around 380,000 years after the Big Bang, forming the first neutral atoms, and allowed the photons to escape from matter. This event is now detected as the oldest electromagnetic signal in the universe, the cosmic microwave background.

The next atoms required more energy to be synthesized, and were mostly formed in gravitational collapse of local heterogeneity of matter: the first stars¹. The gravitational forces at stake in them allowed to reach critical thresholds for new nucleosynthesis. This concerns elements with atomic numbers from 6 (C) and higher, and especially the nutrients I focused on during my thesis: nitrogen and phosphorus. A general trend is that heavier elements require higher energy to be synthesized, and are thus formed in more dense stars (supernovae, white dwarf, neutron stars), but there are important overlaps.

All these elements escaped stars when these latter exploded, forming interstellar clouds, of which new stars can emerge and sometimes other particular local heterogeneity: the planets. Figure 1.1 presents the abundance of the atomic elements in our solar system. Generally, large elements with high atomic numbers are less stable, because of the electromagnetic repulsion of the positively charged protons in the nucleus, while the strong interaction maintaining the nucleus only occurs at very short distances. In addition to this trend, even atomic numbers are more stable than uneven numbers: even numbers allow opposite spins coupling, without leaving

¹Actually, this distinction is very simplified. For instance, Li was both formed with the first Big Bang fusion and in dying stars. See the periodic table of elements in Johnson (2022)

one spin unpaired, which decreases the atom global energy and stabilizes it.

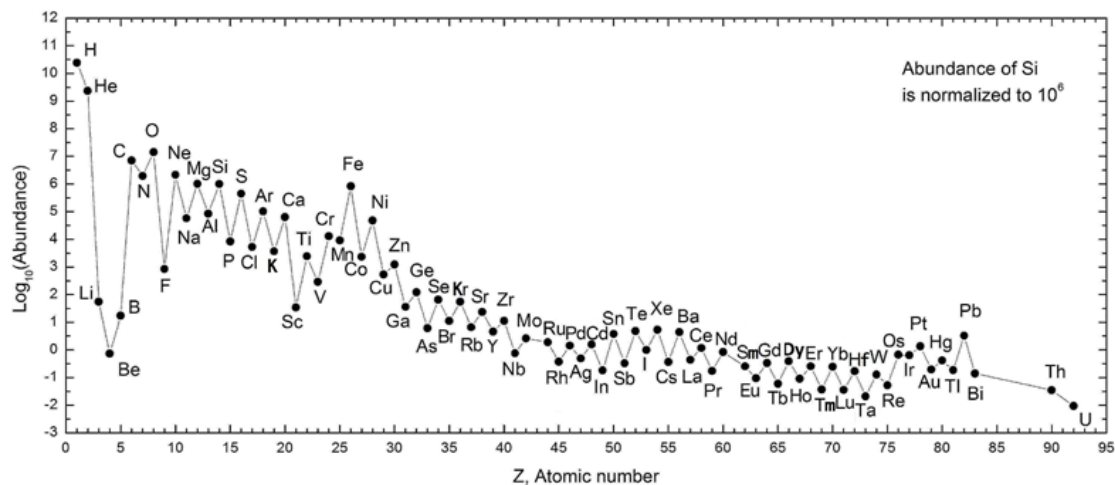


Figure 1.1: Relative molar abundance of the different atomic elements in the solar system, normalized by Si abundance, in log scale. Source: Wikimedia Commons (2022b)

In the next section, we will see that N and P are constituents of molecules essential to life on Earth. But Figure 1.1 shows one particularity of phosphorus: its relative rarity. It is hundreds to thousands times less abundant than other elements essential to life, like carbon, oxygen or even nitrogen. The only other essential elements to human body that are less abundant than phosphorus are K and Cl, but they are 6 to 7 less abundant in human constitution than phosphorus (Table 1.1).

1.1.2 Backbones of molecules essential to life

The reader interested in N and P role in life can refer to Elser et al. (2020b) and Butusov et al. (2013c) for phosphorus, and Leigh (2004a) and Gorman (2013a) for nitrogen.

Phosphorus and Nitrogen were constituents of RNA precursors, and were thus involved in the origin of life. These precursors assumed 2 main roles in life emergence: storing information for future reproduction, and catalysis of chemical reactions. In virtually all modern complex organisms, phosphorus and nitrogen are a constituent of molecules storing and transferring energy (ATP/ADP, Figure 1.3), storing genetic information (DNA, Figure 1.2, but also RNA for some viruses), and, for eukaryotes, transporting genetic information outside the cell nucleus (RNA).

In DNA and RNA, phosphorus ensures the link between the different nucleotides. Westheimer (1987) argues that phosphorus is particularly suited for this function because of its properties: even with 2 hydroxy groups OH, phosphorus keeps a negative charge, preventing OH^- ions in water to hydrolyze the carbon chain, and conferring a very stable structure to DNA based on phosphorus. Nitrogen in DNA

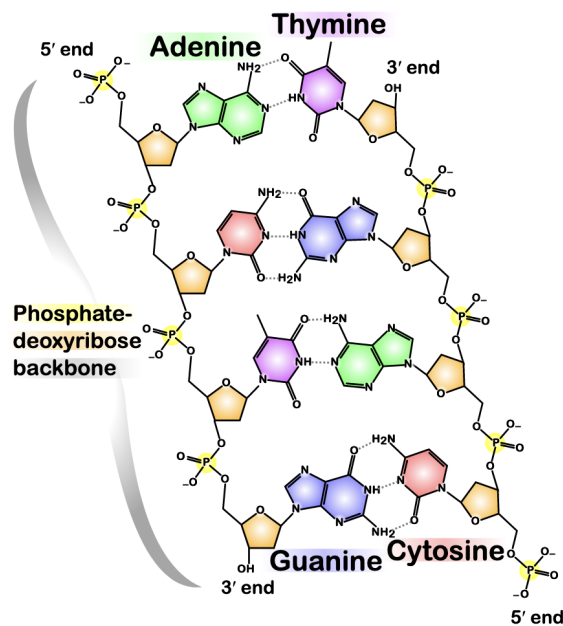


Figure 1.2: Molecular representation of a DNA sequence. Nitrogen allows the binding of the base pairs (A/T and C/G) of the double-stranded DNA. The backbone of DNA is made of chains of phosphates and sugars (phosphate-deoxyribose). The principle is the same for RNA, except that it is single stranded that folds on itself. Source: Wikimedia Commons (2024b)

allows the binding of the Adenine/Thymine and Guanine/Cytosine pairs of bases, between the secondary amine NH_2 and amide $\text{C}(=\text{O})\text{-NH}$ groups, and another N-HN chemical bond between the 2 cycles (Figure 1.2).

While nitrogen is a constituent of the adenosine diphosphate (ADP) and adenosine triphosphate (ATP) molecules (Figure 1.3), it is a phosphate ion PO_4^{3-} that is at the core of the reaction allowing to store energy. The reverse reaction releases energy in cells, for instance for chemical transport between different cell compartments.

Phosphorus also distinguishes itself from nitrogen because it is part of the phospholipids constituting cells membranes. Despite these essential functions, actually most of P in the human organism (85%) is stored in bones, teeth and cartilage in the form of a Ca-P mineral named apatite; 14% in tissues (muscles, liver, heart, kidney); and 1% in extracellular fluid (EFSA 2015b). While P is relatively uncommon in Earth's crust and seawater, and virtually nonexistent in the atmosphere, it is one of the most common element in vertebrates as it is central to bones formation, and makes up about half of a percent of the human's total body weight (Table 1.1).

But phosphorus is absent from the amino acids forming proteins, contrary to nitrogen which is an essential constituent of these elements. Amino acids are made of a carbon chain surrounded by a carboxylic acid ($\text{-C}(=\text{O})\text{-OH}$) group and one

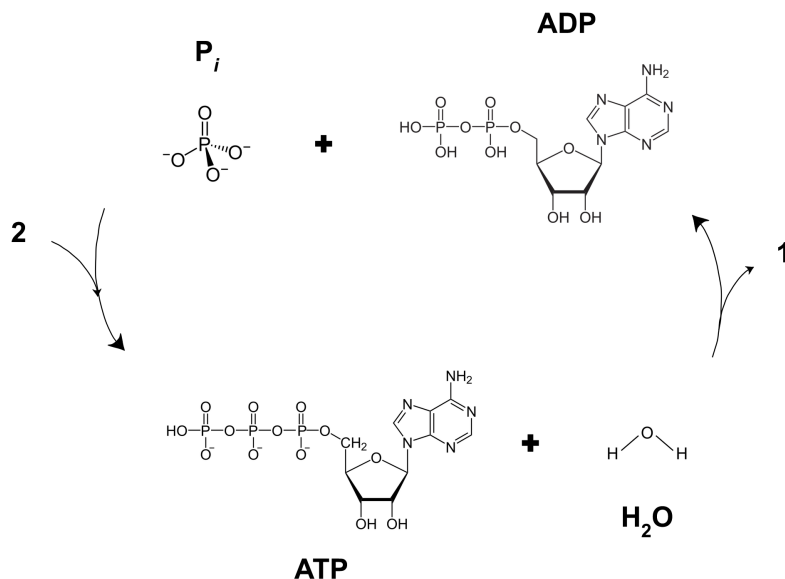


Figure 1.3: Chemical reaction releasing energy by breaking adenosine triphosphate (ATP) with water into adenosine diphosphate (ADP) and phosphate ion PO_4^{3-} ; or storing energy from ADP and PO_4^{3-} , into ATP. On the diagram, 1 and 2 respectively represent output and input of energy. Source: Wikimedia Commons (2023b)

secondary amine ($-NH_2$) group. Two amino acids can merge these 2 functional groups by releasing H_2O and creating a $R-CO-NH_2-R$ bond (Figure 1.4). Repeating this operation creates proteins.

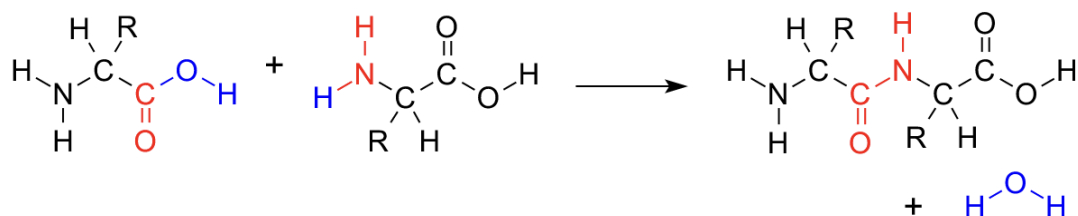


Figure 1.4: Synthesis of a peptide bond by merging the carboxylic acid ($-C(=O)-OH$) group and secondary amine ($-NH_2$) group of 2 amino acids. Source: Wikimedia Commons (2023a)

Some of the amino acids cannot be synthesized by animals, and are called essential amino acids. In humans there are 9 of them, out of the 22 used by human metabolism. These essential amino acids have to be eaten from plants proteins that can synthesize them, or animals that have eaten these plants.

According to EFSA (2012), in a 70 kg adult man, proteins represent 10-12 kg (14-17% of body mass). As a 6.25 factor (Jones' factor) is commonly used to convert proteins mass to nitrogen, the weight body is made of 2-3% of N (Table 1.1). The main pools of proteins are: muscles (42%), skin (15%), blood (15%), and viscera (10%). Half of proteins are in the form of collagen, myosin, actin or hemoglobin.

So in the human body, nitrogen is about 4 times more abundant than phosphorus in mass terms, and 5 times more abundant in molar terms. Like phosphorus, it is relatively very rare in sea water and Earth crust; but contrary to phosphorus, nitrogen is present in virtually unlimited quantities in the atmosphere (Table 1.1). But we will see that this abundance is only apparent, as most organisms cannot directly extract nitrogen from the inert N_2 gas. So, contrary to O and H that are abundantly available from air and water, the human metabolism will have to concentrate N and P (and carbon) from its environment to properly operate.

Table 1.1: Chemical composition of the Human Body, Earth Crust, Sea Water and Atmosphere. Note that topsoil composition can be very different from Earth crust, and enriched in life elements, due to biotic processes. Sources: Sea water and Earth crust: CRC Handbook from Lide (2004), reported in Wikipedia (2024); Atmosphere: NASA (2024)

Human Body				Earth crust		
	Elements	% mass	% molar		Elements	% mass
O	Oxygen	65	24	O	Oxygen	46
C	Carbon	18.5	12	Si	Silicon	28
H	Hydrogen	9.5	62	Al	Aluminum	8.2
N	Nitrogen	2.6	1.1	Fe	Iron	5.6
Ca	Calcium	1.3	0.22	Ca	Calcium	4.2
P	Phosphorus	0.6	0.22	Na	Sodium	2.4
S	Sulfur	0.3	0.038	Mg	Magnesium	2.3
K	Potassium	0.2	0.03	K	Potassium	2.1
Na	Sodium	0.2	0.037	H	Hydrogen	0.0014
Cl	Chlorine	0.2	0.024	P	Phosphorus	0.0011
Mg	Magnesium	0.1	0.015	C	Carbon	$2 \cdot 10^{-4}$
	Others	<0.1	<0.3	N	Nitrogen	$2 \cdot 10^{-5}$

Sea Water			Atmosphere		
	Elements	kg/L		Gas	% molar
O	Oxygen	86	N_2	Dinitrogen	78
H	Hydrogen	11	O_2	Dioxygen	21
Cl	Chlorine	1.9	Ar	Argon	0.9
Na	Sodium	1.1	CO_2	Carbon Dioxide	0.04
Mg	Magnesium	0.13	Ne	Neon	0.002
S	Sulfur	0.09	He	Helium	$5 \cdot 10^{-4}$
K	Potassium	$4 \cdot 10^{-4}$	CH_4	Methane	$2 \cdot 10^{-4}$
C	Carbon	$3 \cdot 10^{-5}$	Kr	Krypton	$1 \cdot 10^{-4}$
N	Nitrogen	$5 \cdot 10^{-7}$	N_2O	Nitrous Oxide	$3 \cdot 10^{-5}$
P	Phosphorus	$6 \cdot 10^{-8}$	H_2O	Water vapor	0 to 3

1.1.3 Nutritional aspects for humans

Humans have to ingest N and P to compensate for the inevitable losses happening mainly through excretions. Besides the compensation of these losses, during growth at young ages, additional needs are required to acquire the N and P that will constitute their body. Here I review recommendations regarding N and P intakes in humans.

1.1.3.1 Definitions

There are multiple notions regarding dietary recommendation, summarized by the European Food Safety Agency (EFSA 2017)²:

- **Average Requirement (AR)**: the mean adequate level of intake for a population. In practice, this is the median level of intake that is adequate for half of people in a population, assuming that intakes follow normal distribution. Average Requirement assumes that everyone gets exactly the phosphorus they need; if some people get more, others will get less than their requirements. For that reason, agencies usually take a security margin, with the *Population Reference Intake*.
- **Population Reference Intake (PRI)**: level of intakes that is adequate for virtually (97.5%) all people in a population. Consequently, for most people, intakes will be higher than what is required.
- **Adequate Intake (AI)**: when data is insufficient to rigorously set AR and PRI. In practice, Adequate Intake is similar to PRI, but established with less "rigorous" methodological standards. For a concrete example, see the case of phosphorus below.
- **Tolerable Upper Intake Levels (UL)**: the maximum level of total chronic daily intake of a nutrient (from all sources) which is not expected to pose a risk of adverse health effects to humans (EFSA Panel on Nutrition et al. 2022).

1.1.3.2 Phosphorus and nutrition

Most of the elements in this subsection come from the European Food Safety Authority EFSA (2015b), but it is also inspired from Butusov et al. (2013d), Scholz et al. (2014), and Elser et al. (2020c).

Recommended Intakes: Once aliments are ingested, EFSA (2015b) estimates that 55-80% of P intakes are actually absorbed by the human metabolism. Factors affecting this absorption are the phosphorus form in food (organic or inorganic), the ratio to other nutrients, namely calcium with which it forms bones, and the food type (P in animal products are better absorbed than in plant-based products). This

²Other agencies use other denominations for very similar concept. For instance, for the US National Academy of Medicine framework, the Estimated Average Requirement (EAR) is the intake that would be adapted to at least 50% of a given age category and the Recommended Dietary Allowance (RDA) is the threshold that would meet 97.5% of the needs of an age category (NIH 2023).

absorbed P enters the blood, regulated by vitamin D among others, before being excreted through urine. The unabsorbed part is directly excreted in feces.

Humans absorb P rather efficiently compared to monogastrics animals like poultry or swine, which cannot absorb P when it is in the form of phytate. Yet phytate is a common form of P in cereal grains fed to farm animals, which have particularly high needs since their breeding were selected to grow rapidly. This results in P deficiency with consequences such as less P mineralization and more sickness. This unabsorbed phytate is also why monogastrics manure is rich in P, mostly in its inorganic form. For that reason, farmers often give phytase enzymes to monogastrics fed on cereal grains, to help them process phytates (Jacela et al. 2010). There are even attempts to produce genetically modified pigs (EnviroPig™) that can absorb phytate and thus remove the need for P-supplements.

The EFSA did not identified an upper intake level for P for humans, above which harmful effects appear under realistic consumption values. While there can harms with too much P intakes, the quantities involved appear uncommon in normal Western diets (Schnee et al. 2014). The U.S. National Institutes of Health establish Tolerable Upper Intake Level of $4 \text{ gP}\cdot\text{day}^{-1}$ for adults (NIH 2023), or 2 to 3 times higher than actual intakes in European countries (Table 1.3).

Adverse effects of P deficiencies are rare, in the form of hypophosphatemia or bones malformations. But generally this does not occur because of insufficient P intakes, but rather due to low amounts of vitamin D, which regulates phosphorus and calcium concentrations.

The EFSA judged the data insufficient to establish Average Requirements (AR) and Population Reference Intakes (PRI) for phosphorus, and rather established an Adequate Intakes (AI) estimate based on the following reasoning. Since phosphorus is mainly used for bones, in association with Ca, they used the Ca:P molar typical ratio in human body (1.4:1 to 1.9:1). As Ca is usually the limiting factor of bones formation, they used the Ca Population Reference Intakes (EFSA 2015a) and applied the lower Ca:P ratio (resulting in higher P intakes, for security) to determine P Adequate Intakes. The Ca intakes requirements are based on Ca balances to compensate inevitable metabolism losses.

This results in an Adequate Intakes of $0.55 \text{ gP}\cdot\text{day}^{-1}$ in adults. The value is higher for adolescents, accounting for bones formation (Table 1.2).

Actual Ingestion: The review undertaken by EFSA (2015b) found that actual intakes across 9 European countries are on the order of $1.3\text{-}1.6 \text{ gP}\cdot\text{day}^{-1}$ (or $0.45\text{-}0.55 \text{ kgP}\cdot\text{year}^{-1}$) in adults 18-65 years. Differences between males and females are reported in Table 1.3. Consequently, actual intakes are about double what the EFSA and other agencies estimate as adequate in Table 1.2.

Table 1.2: Estimated Phosphorus Adequate Intakes for adults of both sexes by EFSA (2015b), and comparison with other agencies. Adults refers to people around 18-65 years old (differs between agencies). See EFSA (2015b) for agencies sources. EFSA also sets different recommendations for other ages, and pregnant lactating women (not shown here). DH: Department of Health; NL: Netherlands Food and Nutrition Council; SCF: Scientific Committee for Food; IOM: Institute Of Medicine; Affsa: Agence française de sécurité sanitaire des aliments; NCM: Nordic Council of Ministers; DACH: Deutschland, Austria, Confoederatio Helvetica.

		$\text{gP}\cdot\text{day}^{-1}$	$\text{kgP}\cdot\text{year}^{-1}$
EFSA (2015) for:		Adequate Intakes	
adults		0.55	0.20
11-17 years		0.64	0.24
4-10 years		0.44	0.16
1-3 years		0.25	0.09
7-11 months		0.16	0.06
Other agencies	Countries	Population Reference Intakes	
DH (1991)	United Kingdom	0.55	0.20
NL (1992)	Netherlands	0.7-1.4	0.25-0.50
SCF (1993)	European countries	0.55	0.20
IOM (1997)	United States	0.7	0.25
Affsa (2001)	France	0.75	0.27
NCM (2014)	Nordic countries	0.6	0.22
DACH (2015)	German countries	0.7	0.25

But it is difficult to deduce from this that people eat "too much" P. First, the 1.3-1.6 $\text{gP}\cdot\text{day}^{-1}$ is still way below the estimated tolerable upper intake of 4 $\text{gP}\cdot\text{day}^{-1}$. Second, the amount of the different eaten nutrients is not independent from each other, and adequate intakes of one nutrient, e.g. protein, can lead to intakes in higher amounts than necessitated for another, e.g. P, just because of the range of protein:P ratios in food.

P-rich food includes meat (especially if processed with additives), fish, cereals, legumes, sodas with P-additives (to regulate acidity and thus dissolved CO_2). On the contrary, some foods do not contain any P: sugar, fats or oil. Processed foods tend to have higher P contents than home-made meals, owing to the numerous P-additives and preservatives.

Indeed, the market of P-additives is not restricted to P supplementation for farm animals, and is also pervasive in human food, mostly under the form of phosphorus inorganic salts. Disodium phosphate is used to get nice coloration and texture (coagulation and emulsifiant), for instance in cheese or meats. Pyrophosphates efficiently absorb water, and are used to dry and conserve meat, making processed

Table 1.3: Actual P intakes in European countries, circa 2000-2010, for adults around 18-65 years old (different between studies). They also report intakes across other age classes (data not shown here). See EFSA (2015b) for sources.

	actual P intake			
	gP·day ⁻¹			kgP·year ⁻¹
	males	females	mean	mean
Finland	1.6	1.3	1.5	0.55
France	1.4	1.1	1.3	0.47
Ireland	1.8	1.3	1.6	0.59
Italy	1.4	1.2	1.3	0.47
Netherlands	1.7	1.3	1.5	0.55
Sweden	1.7	1.3	1.5	0.55
United Kingdom	1.5	1.1	1.3	0.47

meat rich in P. In soft drinks, phosphoric acid is used to lower pH and keep the drink acidic. Sodium aluminum phosphates are added to baking powder to help their leavening. Finally, polyphosphates salts act as emulsifiers, pH buffer or humectants (Scholz et al. 2014)

1.1.3.3 Nitrogen and nutrition

The concepts presented in this subsection are detailed in EFSA (2012).

A close link to protein: Nitrogen in food is closely linked to protein intakes. A common conversion factor is that N makes 16% of protein weight, transcribed in the Jones' factor of 6.25 to convert N to proteins on a mass basis, even though there is room for more precision (Mariotti et al. 2008); for instance, EFSA (2012) reports conversion factors of 6.38 for milk, 5.83 for wheat and 5.71 for soy proteins.

Ingested proteins are digested so that the body can reuse the amino acids lost with metabolic processes, and synthesize new proteins. For humans, there are 9 amino acids labeled "essential", meaning the human metabolism cannot synthesize them from other amino acids and needs to acquire them from food intakes: histidine, isoleucine, leucine, lysine, methionine, phenylalanine, threonine, tryptophan, valine.

The human metabolism also needs a particular stoichiometry between the different amino acids (WHO/FAO/UNU 2007). Generally, animal products fit humans' amino acid stoichiometry, owing to animals' metabolism which is close to humans. Plant-based products generally have a deficiency in at least one amino acid: lysine for cereals, methionine and cystine for legumes. As a result, an adequate vegetarian diet should rely on meals mixing the complementary foodstuffs, for instance cereals and legumes, to compensate their respective deficiencies.

For that reason, amino acid scores have been developed, based on the limiting

amino-acid in a given foodstuff. A score below one indicates that at least one amino-acid is limiting. Besides amino acids composition, one needs to also adjust for actual digestibility of proteins, as some of the proteins are not absorbed by the organism. The Protein Digestibility-Corrected Amino Acid Score (PD-CAAS) accounts for both of these factors (Table 1.4) (FAO/WHO 1991; FAO 2011a). A score of 70 out of 100 for beans means that one has to eat $1/0.7 =$ about 40% more to get the same amount as a reference protein with the adequate stoichiometry. Note that these scores relate to isolated foodstuff, and limitations can be overcome by mixing different products.

Table 1.4: Protein Digestibility- Corrected Amino Acid Score (PD-CAAS) for different foods. Source: EFSA (2012)

	PD-CAAS	limiting amino-acids
animal food		
egg	1	-
milk, cheese	1	-
meat, fish	1	-
plant source		
soy	0.95	methionine, cysteine
beans	0.7-0.75	methionine, cysteine
rice	0.65	lysine
wheat	0.5	lysine
maize	0.5	lysine

Protein / N dietary guidelines: EFSA (2012) based its recommendation of protein intakes on estimates of balance for obligatory losses in human metabolism. They established a protein Average Requirement of $0.66 \text{ g}\cdot\text{kg}^{-1}$ body weight per day ($105 \text{ mgN}\cdot\text{kg}^{-1}$ body weight per day) and a Population Reference Intake of $0.83 \text{ g}\cdot\text{kg}^{-1}$ body weight ($133 \text{ mgN}\cdot\text{kg}^{-1}$). This is very similar to recommendations made by other health agencies (Table 1.5). All these values refer to a PD-CAAS protein value of 1.0. However, the EFSA states that it is unlikely that indispensable amino acids are limiting considering European omnivorous diets, and in approximation no further PD-CAAS adjusting is required.

The recommendation for Population Reference Intake translates, for a person of 70 kg^3 , into about 3-3.5 kgN per year, or 50-60 g of protein per day (Table 1.5). But if we hypothesize that everybody could get exactly its needs with the Average Requirement, a 70 kg would only need 2.5-2.7 kgN per year, or 42-46 g of protein

³EFSA (2012) reports weighted median body weights of 75 kg for adult men and 62 kg for women

per day.

Table 1.5: Estimated protein and nitrogen Adequate Intakes and Population Reference Intakes for adults (mean of men and women) by EFSA (2012), and comparison with other agencies. Adults refers to people around 18-65 years old (differs between agencies). EFSA also sets different recommendations for other ages, and pregnant lactating women (not shown here). and Nitrogen obtained from protein with Jones' factor of 6.25. See EFSA (2012) for agencies sources. DoH: Department of Health; SCF: Scientific Committee for Food; IoM: Institute Of Medicine; AFFSA: Agence française de sécurité sanitaire des aliments; D-AC-H: Deutschland, Austria, Confoederatio Helvetica.

Agency	Average Requirement		Population Reference Intake	
	per kg body weight	for a 70 kg adult	per kg body weight	for a 70 kg adult
	g protein per day			
EFSA (2012)	0.66	46	0.83	58
FAO/WHO/UNU (1985)	0.6	42	0.75	53
DoH (1991)	0.6	42	0.75	53
SCF (1993)	0.6	42	0.75	53
Health Council Netherlands (2001)	0.6	42	0.8	56
IoM (2005)	0.66	46	0.8	56
WHO/FAO/UNU (2007)	0.66	46	0.83	58
AFSSA (2007)	0.66	46	0.83	58
D-A-CH (2008)	0.6	42	0.8	56
	kgN per year			
EFSA (2012)	0.039	2.7	0.048	3.4
FAO/WHO/UNU (1985)	0.035	2.5	0.044	3.1
DoH (1991)	0.035	2.5	0.044	3.1
SCF (1993)	0.035	2.5	0.044	3.1
Health Council Netherlands (2001)	0.035	2.5	0.047	3.3
IoM (2005)	0.039	2.7	0.047	3.3
WHO/FAO/UNU (2007)	0.039	2.7	0.048	3.4
AFSSA (2007)	0.39	2.7	0.048	3.4
D-A-CH (2008)	0.035	2.5	0.047	3.3

Actual supply and intakes: According to the review made by EFSA (2012) for European countries, daily protein intakes are usually around 90-100 g/day for adults males and 65-80 g/day for women, resulting in a population mean around 75-90 g/day for both sex. Using Jones' factor to convert to nitrogen, European intakes are around 4.5-5.5 kgN/year (Table 1.6), and are thus about 50% higher than the recommended 3-3.5 kgN/year that I calculated from the different agencies Population Reference Intake, on the basis of a 70 kg body weight.

Data relative to protein intakes on a body weight basis are scarcer, but when reported they vary between 0.8 and 1.25 g·kg⁻¹ body weight per day for adults in Occidental countries, higher than the 0.6-0.66 Population Reference Intake across agencies (Table 1.5). Relative to energy intakes in adults, proteins contribute 12-20% of total energy intake in the surveyed European countries.

Table 1.6: Actual N and protein intakes in European countries, circa 2000-2010, for adults around 18-65 years old (different between studies). They also report intakes across other age classes (data not shown here). See EFSA EFSA (2012) for sources.

	protein intake			N intake
	g/day			kgN/year
	males	females	mean	mean
Denmark	87	67	77	4.5
Finland	89	67	78	4.6
Hungary	102	80	91	5.3
Ireland	100	70	85	5
Italy	93	76	85	5
Portugal	103	98	101	5.9
Sweden	90	73	82	4.8
United Kingdom	88	65	77	4.5

Table 1.7: Range of protein content in typical foodstuff. Source: EFSA (2012). N content was computed using the 6.25 Jones' factor. Includes moisture content.

		% protein content	% N content
		(g/100g)	(g/100g)
animal food	red meat	20-33	3.2-5.3
	poultry	22-37	3.5-5.9
	fish	15-25	2.4-4
	eggs	11-13	1.8-2.1
	cheese, hard	27-34	4.3-5.4
	cheese, soft	12-28	1.9-4.5
	milk products	2-6	0.3-1
plant food	vegetables	1-5	0.2-0.8
	legumes	4-14	0.6-2.2
	fruits	0.3-2	0.05-0.3
	nuts and seeds	8-29	1.3-4.6
	pasta, rice	2-6	0.3-1
	breads and rolls	6-13	1-2.1
	breakfast cereals	5-13	0.8-2.1

1.2 N and P in ecosystems

In this section I review the processes regulating P (Section 1.2.1) and N (Section 1.2.2) exchanges between their respective compounds in terrestrial ecosystems.

1.2.1 Phosphorus in the soil

The reader interested in this subsection subject can refer to *Phosphorus cycle* in the *Encyclopedia of Soil Science* (Blake et al. 2008); Elser et al. (2020d); and Pierzynski et al. (2005)

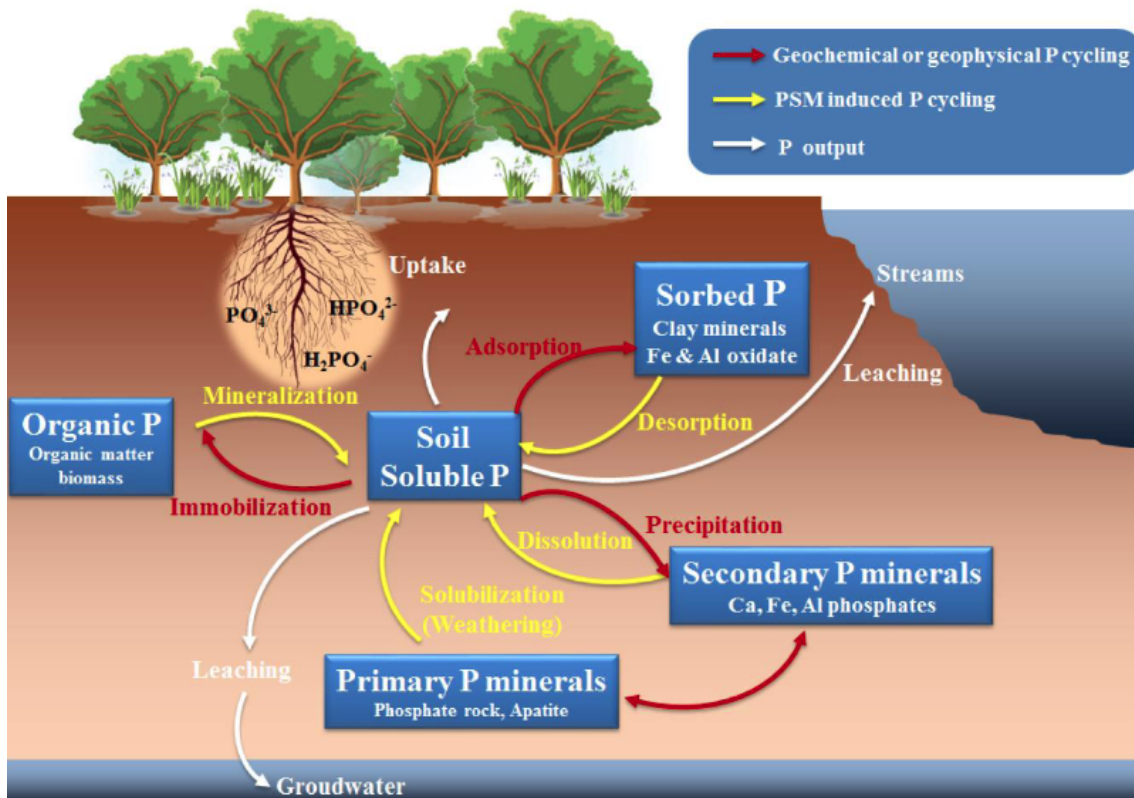


Figure 1.5: P soil cycle. PSM: Phosphate solubilizing microorganisms. Source: Tian et al. (2021).

1.2.1.1 Organic P

In soils, organic matter is one way phosphorus is stored, into nucleic acids, phospholipids, ATP/ADP, and other molecules (see Section 1.1.2). This organic matter is constitutive of living organisms (microbes, fungi...) but also dead organisms, plant residues, and humus. Organic P is not readily available to plants, and first has to be released into simpler ionic phosphate PO_4^{3-} , HPO_4^{2-} , and H_2PO_4^- , following digestion with enzymes by detritivore organisms. For plants to uptake these phosphates, they have to be dissolved in the soil water. The exchanges between the organic

P and the soil soluble phosphates are called mineralization (when organic matter is digested and metabolized as mineral P) and immobilization (when mineral P is ingested and incorporated into organic matter).

The absorption of phosphate ions by plants is not passive. First, symbiotic association between fungi and plants (called a mycorrhiza), can enhance the ions transfer from soil water to plants roots within vesicles; in exchange of energy in the form of carbohydrates provided by plants, they build a bridge from the soil to the interior of plants roots cells, and bring them phosphates. Second, plants roots and microorganisms can release enzymes making P more readily available, such as phosphatase and phytase, which release the P in organic compounds (Elser et al. 2020d).

1.2.1.2 Inorganic P pools

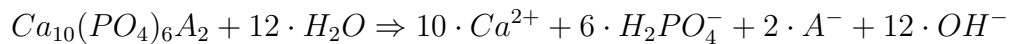
Besides these biological exchanges, the soil soluble P interacts through physico-chemical reactions with other less available P pools. A concept of the four P pools in soil have been developed to describe these exchanges (see Figure 1.5, and also Elser et al. (2020d)):

- **Soil solution**, with dissolved soluble P anions available for plant uptake (PO_4^{3-} , HPO_4^{2-} , H_2PO_4^-).
- **Sorbed P**, readily available for plant uptake after desorption, where the element is loosely bound to soil particles, like clay, Fe and Al oxides, or organic matter. **Adsorption/Desorption** regulate the exchange between the soil solution and the soil particles.
- **Secondary P minerals**, less available for plant uptake. P is more strongly bound to soil particles and so less available for plant uptake. This includes minerals bounding P with Ca, Fe, Mn or Al. Some of these minerals can come from dead organisms, like Calcium/Phosphorus minerals which constitutes bones. With these minerals, P is released through **Dissolution/Precipitation** reactions.
- **Primary P minerals**, where P is almost imprisoned in phosphate rocks or apatite. It is released to the soil solution or secondary minerals by **solubilization** through **weathering**.

1.2.1.3 P exchanges and movements in the soil

Many factors regulate the exchange between these different pools, among which: soil acidity; presence of Ca ions; soil organic content; amount of soil water (Butusov et al. 2013e).

Phosphates anions PO_4^{3-} are negatively charged, and will thus preferentially bind to positive charges. Organic matter can have both negative and positive charges, so it is possible for phosphate to be adsorbed on its surface. Clay is negatively charged, however ions Fe^{2+} , Fe^{3+} and Ca^{2+} can bind to phosphates and still keep a positive site, that can be adsorbed on clay negative sites, or on other anions present in the soil, and form precipitate salts. Here is a general equation for the dissolution of precipitate salts, with A symbolizing a soil anion like Cl^- , F^- or OH^- (Butusov et al. 2013e):



One widespread test to estimate available P for plant uptake in soils is for instance the Olsen test (Olsen 1954), but there are many more, with sometimes high variability between methods (Pierzynski 2000).

Compared to nitrogen (discussed in the following subsection 1.2.2), the particularity of the P cycle is that phosphates are easily adsorbed to minerals, and are thus way less mobile in water than nitrates. This has several consequences. First, leaching to groundwaters, when dissolved ions are dragged along by the soil water flow, is uncommon for phosphorus, contrary to nitrogen in its nitrate NO_3^- form. Losses of P mainly occur through erosion, when soil particles on which phosphate are adsorbed are torn away by heavy water flow and runoffs (and, to a lesser extent, wind erosion). Losses of P with erosion require strong rainfalls to carry soil particles. The majority (50-98%, see Hart et al. (2004)) of P losses during runoffs happen during particular heavy rains or storms.

This is actually the only main way to lose P from the soil, as there is no gaseous form of P, while nitrogen can also be lost to the air as N_2 or N_2O following denitrification, and NH_3 following volatilization. Also, P can stay bounded in the soil during years or decades before being finally released to the soil solution, contrary to nitrogen where losses are more immediate. In the case of the stock of P accumulated in cropland soils during decades of fertilization, this has been labeled as a "P legacy" (Sharpley et al. 2013), which could jeopardize water quality with its future release.

Once erosion finally brings P to the ocean, it can be considered definitively lost, as it takes tens to hundreds of millions of year before tectonic uplifting can bring it up to the continental surfaces⁴. In natural ecosystems, the only way to add more or less readily available P (i.e in organic P, soluble P, sorbed P and secondary P minerals) is through the weathering of apatite and phosphate rocks. Animals can spatially transfer ingested P to another location through their excretions, as can

⁴There are nonetheless small P flows from oceans to terrestrial ecosystems, when animals eat fish and excrete on the land.

wind erosion (Yu et al. 2015), but this is not a real new input when considering the whole ecosystem. It is assumed that in ecosystems with limited human impact, P losses are sufficiently low to be balanced by new inputs, but human activities have intensified both processes, through enhanced erosion on croplands and P inputs with fertilizers.

1.2.2 Multiple N compounds and flows

Elements in this subsection are mainly issued from Leigh (2004a), Gorman (2013a), and Leigh (2004b).

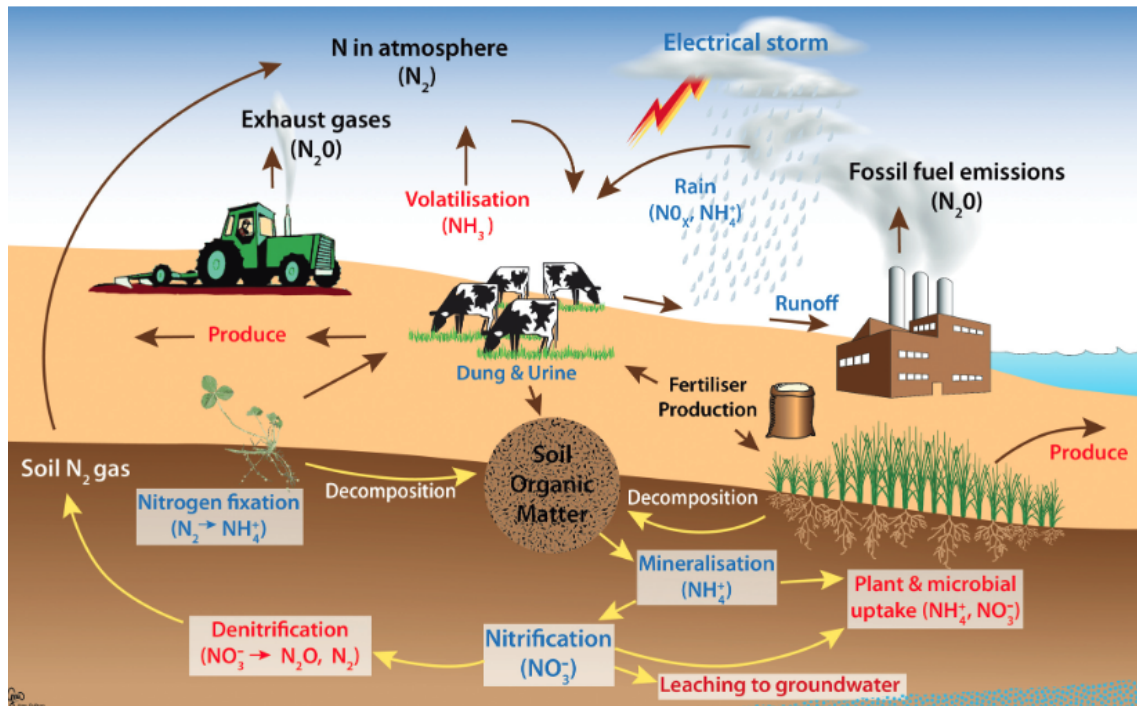


Figure 1.6: The N cycle in terrestrial ecosystems. Here is an anthropic ecosystem (farmland), but the principle remains the same in a natural ecosystem. Source: (Schipper et al. 2010)

1.2.2.1 Forms of N

Nitrogen in nucleotides and amino acids is trapped in complex carbon chains, and constitutes what is called the **organic N**. **Mineral N** includes much simpler molecules of nitrogen. They constitute the **reactive nitrogen**, noted N_r , by opposition to **inert dinitrogen gas** N_2 , due to its triple bound between the 2 N atoms ensuring a very stable molecule. Besides the $N_{organic}$, forms of reactive nitrogen include:

- **nitrate** (NO_3^-), **nitrite** (NO_2^-) and **ammonium** (NH_4^+) aqueous ions that

can be found in water. Ammonium also easily binds to negatively charged soil particles like clay.

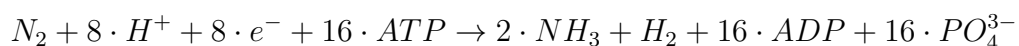
- **ammonia** (NH_3), a gas found in the air but that can also be dissolved in water, where it is in physical equilibrium with NH_4^+ .
- **nitrous acid** (HNO_2) and **nitric acid** (HNO_3), for instance found dissolved in rain, and involved in acidic rains. They are also found in the soil solution, following nitrification.
- **nitrogen oxides** (NO_x) gases, comprising **nitrogen dioxide** (NO_2) and **nitric oxide** (NO), released by combustion of fuel containing N, be it biomass or fossil fuels. They can also be the result of N_2 fixation following electric lightning. NO can also be emitted nitrifying and denitrifying bacteria.
- **nitrous oxide** (N_2O), a potent greenhouse gas, emitted by nitrifying and denitrifying bacteria.

1.2.2.2 Transfers between the different N forms, modulated by biogeochemical processes

Table 1.8: The different forms of Nitrogen

Total or Global N					
Kjeldahl N (reduced N, noted TKN)					
Inorganic				Organic	
Oxidized		Ammoniacal			
Nitrites	Nitrates	Ammonia	Ammonium	Urea	Other organic
NO_2^-	NO_3^-	NH_4^+	NH_3	$\text{CO}(\text{NH}_2)_2$	Amino Acids, Proteins

From nonreactive N_2 to reactive N_r : The major path in which inert N_2 is transformed in reactive N_r is through biological fixation, by bacteria that manage to transform N_2 into NH_3 , at the expense of considerable energy consumption of 8 ATP for 1 atom of N fixed. Here is the simplified fixation equation:



In reality this involves numerous intermediate steps, mediated by catalytic enzymes like nitrogenase. This can occur with free-living bacteria in the soil (*Azotobacter*) but also with bacteria (*Rhizobae*) that benefit from their symbiotic association with leguminous plants (*Fabaceae*) (Stewart 1969). Free-living non-symbiotic fixation can occur in anthropic systems like sugar cane fields or paddy rice, but the

amount of fixed N are then less important than with symbiotic legumes. Finally, some organisms are able to perform photosynthesis and also fix N from the air, like cyanobacteria (blue-green algae).

In the case of legumes, the symbiosis takes place in nodules in their roots (Figure 1.7). The plant provides energy in the form of carbohydrates to bacteria to perform the fixation, and in exchange bacteria provide nitrogen. Symbiotic fixation can happen in aerobic or anaerobic conditions, but some key enzymes involved in the process are sensitive to oxygen, so biofixating organisms must establish internal protection mechanism against aerobic conditions. Finally, the symbiosis with N-fixing bacteria does not only concern plants, but also insects as termites or fungi.



Figure 1.7: Nodules on the roots of *Vigna unguiculata*. Source: Wikimedia Commons (2022a)

The second minor way to transform inert atmospheric N_2 to reactive N_r is with energy released during lightnings. N_2 is oxidized by atmospheric O_2 into oxidized NO_x forms. Atmospheric N aerosols can then be deposited on land through dry deposition (in the forms of NO_2 , HNO_3 , NH_4^+ , NO_3^- , NH_3) or dissolved in acidic rains as wet deposition (NH_4^+ and NO_3^-). A minor introduction of N_r also happens with volcanic eruptions.

Changing forms in the soil and losses through leaching Biofixating organisms fix atmospheric N_2 in the form of NH_3 , which is then in equilibrium in the soil solution as NH_3/NH_4^+ . Nitrifying bacteria can extract energy from the reduced NH_3/NH_4^+ couple by breathing O_2 , and transform it into nitrites NO_2^- or nitrates NO_3^- , and H_2O . This nitrification process emits small amounts of N_2O , which is a potent greenhouse gas, as well as small amounts of NO .

The NO_2^- and NO_3^- oxidized forms are less attracted by negatively charged soil particles. They can dissolve more easily in soil water and reach groundwaters through leaching (but NO_2^- quantities are way smaller than NO_3^-). Contrary to phosphorus, for which phosphate is not easily soluble, N leaching is a major pathway of N losses in soils.

Organic N and mineralization/immobilization cycling Nitrogen also plays a role in the carbon biogeochemical cycle. C is the only element plants find directly in the air, through CO_2 . Other nutrients, among which is nitrogen, have to be taken up by plants from the soil. Plants absorb nitrogen under its nitrate (NO_3^-) or ammonium (NH_4^+) form, and incorporate it into their organic matter. Living organisms need to respect a C:N ratio range suitable with their nutrients composition.

In dead organisms, the decaying organic matter is generally broken down by detritivores, which extract energy from N_{organic} and transform it into digested ammonia NH_3 and ammonium NH_4^+ , bringing mineral N back to the soil. There is theoretically no limit to the mineralization/immobilization cycle $N_{\text{organic}} \rightarrow \text{NH}_4^+ \rightarrow \text{NO}_3^- \rightarrow N_{\text{organic}}$, as long as solar energy and water are present and no losses occur; but in practice the cycle is broken by losses to waters or the atmosphere.

In some particular conditions, organic matter is not broken down and ends up buried, and after millions of years can produce fossil fuels, still containing some amounts of nitrogen.

Bringing N back to the atmosphere: denitrification, volatilization, combustion Combustion of biomass or fossil fuels containing nitrogen emits NO_x gases to the atmosphere. Other emissions of reactive N can occur in the form of NH_3 gas. Indeed, dissolved NH_3 in the soils solution is in equilibrium with its gaseous form, and particular conditions such as windy weather can pull the equilibrium towards NH_3 volatilization. NO_x and NH_3 are locally lost but stay in a reactive form that can be further remobilized after atmospheric deposition. The only way to effectively remove nitrogen from the reactive pool is through denitrification.

Denitrification happens in absence of oxygen, under anoxic conditions, typically when soils are saturated with water. In this situation, denitrifying bacteria can transform nitrates into dinitrogen $\text{NO}_3^- \rightarrow \text{N}_2$. As during nitrification, this process also emits small amounts of N_2O and NO . In pre-anthropoc ecosystems, it is generally considered that there was global an equilibrium between fixation inputs and denitrification outputs; the situation has changed ever since (see Section 2.3)⁵.

I do not expand on the N cycle in oceans, but the main fact to keep in mind is that nitrogen compounds, especially NH_4^+ , are much more mobile as they do not bind to fixed soil particles and clay and can be driven by currents. There is little coupling between oceanic and terrestrial N cycles (Gruber et al. 2008a), except locally where rivers flow into the ocean or through fishes traveling back to freshwaters.

⁵This is not true on geological timescales, when the quantity of slowly increased as a result of increased life activity.

1.3 Too many nutrients: N and P pollutions

While N and P are essential to life and ecosystems functioning, their excess, mainly driven by human activities, result in pollutions impacts on biodiversity and human health.

1.3.1 Eutrophication: a problem common to N and P

The manifestations of eutrophication are toxic blooming of mostly cyanobacteria in lakes and water courses, and the proliferation of green macroalgae in coastal zones, caused by an excess of nutrients. This blooming eventually results in anoxia (i.e., not enough dissolved oxygen in water), leading to a loss of biodiversity, especially for organisms which need oxygen to breathe.

A comprehensive collective scientific expertise has been undertaken by Pinay et al. (2017). Eutrophication can be a natural phenomenon, but the term is usually used when triggered by human activities. For this reason they give 2 distinct definition:

- **anthropic eutrophication:** clearly caused by N and P excesses which induce overproduction of organic matter, within a short time frame of about a decade. N and P excesses are mainly due to intensive agriculture (fertilizers losses) and urbanization (discharge of municipal wastewater).
- **natural eutrophication:** in that case, N and P do not specially play a particular role compared to other nutrients. The blooming occurs because of accumulation of organic matter on geological times, not observable during human lifetime.

In the following, by default when I will use eutrophication, this will refer to human-caused eutrophication.

Following large introduction of nutrients in water, the primary producers (i.e., organisms able to photosynthesize) are the first to react, before the cascading process impacts all of the ecosystem. Through photosynthesis, they generate the *Net Primary Production (NPP)*, i.e. organic matter, out of CO₂ and light. This organic matter can then be eaten and breathed backed to CO₂ by heterotrophs. Globally, about 100 Gt of carbon are produced from NPP (Pinay et al. 2017), half in oceans and half in terrestrial ecosystems (this includes water ecosystems on terrestrial land).

Net Primary Production is limited by the first lacking element given the stoichiometry of organisms. For instance, Redfield in 1934 observed that the molar C:N:P ratio tended to be 106:16:1 in oceans (Redfield 1934). Extensive measurements in a variety of contexts show that there can be much more variability than

this single ratio (Martiny et al. 2014), but the concept remains similar. Subsequent works have added other nutrients, namely silicon. In freshwaters, the stoichiometry appears to be less stringently bounded (Pinay et al. 2017).

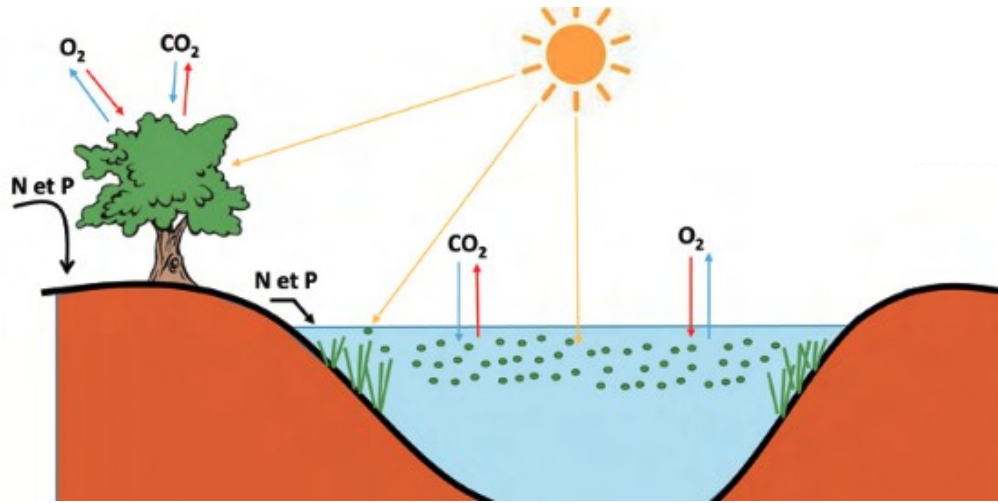


Figure 1.8: Flows at stake during eutrophication. Increased N and P flows (black arrow); light (yellow); primary production through photosynthesis (blue); organic matter degradation through respiration (red). Source: Pinay et al. (2017)

Following anthropogenic inputs of N and P, the increase in primary producers number on the water surface is so important that it prevents the light from crossing the water column and reaching the bottom. Except on the surface, the new limiting factor becomes light. Primary producers cannot replenish the oxygen consumed by heterotrophs, and dissolved oxygen begins to be depleted. In lakes, this is amplified by strong thermal stratification preventing oxygen diffusion from the surface to the bottom.

Some organisms can change their oxygen source when oxygen levels are too depleted. Instead of using O₂ to oxidize organic matter into CO₂ and produce energy, they instead can use other electron acceptors such as (in that order) nitrates NO₃⁻, manganese oxide MnO₂, and iron oxide FeO₂. A small feedback loop occurs with iron oxide FeO₂, because this element can fix phosphates, so its oxidization further releases phosphorus. The next oxidizing agent is sulfate SO₄⁻, reduced not into CO₂ but to malodorous H₂S. When all these oxidized forms are reduced, a shift occurs towards anaerobic organisms, which transform organic matter into methane CH₄. H₂S and CH₄ are toxic to herbivores, further increasing primary production, which is not regulated anymore by heterotrophs. The resulting eutrophic state is characterized by an excess of primary producers, such as cyanobacteria or algae, and the emission of toxic compounds.

The transition from an oligotrophic state to an eutrophic state is abrupt and non linear below a certain threshold of dissolved oxygen concentration. Each of

these 2 states is resilient to changes in dissolved oxygen, and it is difficult to come back to an oligotrophic state once the eutrophic one has been reached. While we understand the broad mechanisms, Pinay et al. (2017) insist that the complexity of the processes makes it difficult, if not impossible, to predict how the system will evolve. However some factors are known to influence eutrophication risks:

- **water residence time:** the longer it is, the more primary producers can absorb nutrients, enhancing the eutrophication risk.
- **vertical stratification (by temperature or salinity):** it isolates the deeper layers, prevents their reoxygenation, and can also increase residence time. It's especially common during spring when the air starts to warm and water is not yet homogenized, or along coastlines when fresh water meets the ocean.
- **temperature:** increased temperature decreases the capacity of water to carry dissolved oxygen, favoring anoxic conditions and thus eutrophication.
- **light:** for instance light is less accessible in troubled waters such as swamps, preventing the photosynthesis below the surface.

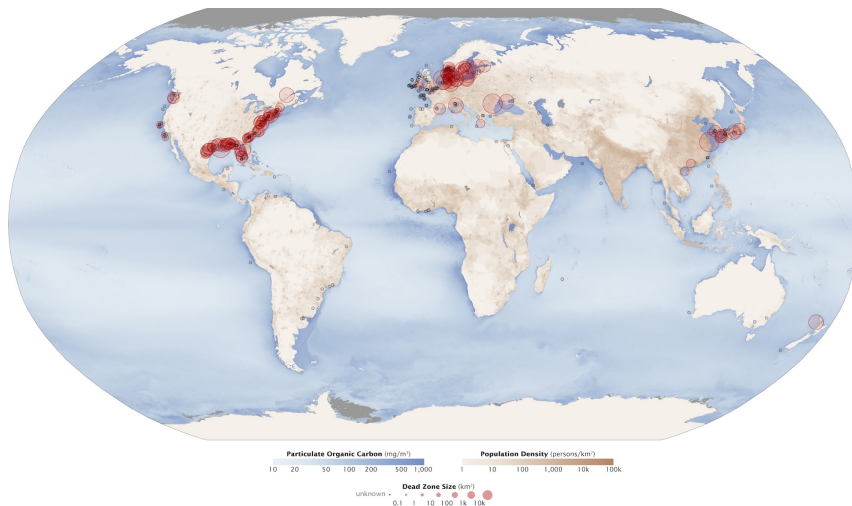


Figure 1.9: Coastal dead zones induced by eutrophication in the 2000s. Source: NASA Earth Observatory (2008) based on Diaz et al. (2008)

One concrete manifestation of large-scale eutrophication are the coastal "dead zones". During the 2000s, more than 400 coastal hypoxic dead zones were identified (Diaz et al. 2008). Some of these large zones have become emblematic of eutrophication. The Baltic Sea is a confined water body, with high resident time, favoring eutrophication surrounded by densely populated countries. The Gulf of Mexico drains water from Mississippi huge watershed, with high intensive farming

activities... We can also name Seto Inland Sea (off the coast of Japan) and Chesapeake Bay on the Eastern side of the USA (Figure 1.9). This count does not include eutrophication occurring in freshwaters.

1.3.2 A multitude of N impacts

The European Nitrogen Assessment (Sutton et al. 2011b) summarizes the numerous impacts associated with too much reactive nitrogen into 5 threats (Figure 1.10):

- **Climate:** mainly due to N_2O emissions, a potent greenhouse gas; but other N_r forms have opposite cooling effect (fertilization increasing carbon sinks, cooling aerosols...).
- **Air quality:** associated with NO_x emissions from mainly from transport and to a lesser extent from industry and NH_3 emissions from agriculture
- **Water quality:** N enrichment of aquatic system leads to eutrophication, already described in the previous section. Another impact particular to nitrogen is the increased nitrates NO_3^- concentration in drinking water, which can have health impacts.
- **Soil quality:** a main impact of N_r on soil quality is soil acidification because of N deposition. This results in a decrease in crops and forest production.
- **Terrestrial biodiversity:** terrestrial biodiversity is impacted by N_r deposition and acidification; N enrichment of the ecosystem also modifies the species relative abundance, by favoring species adapted to N-rich environments

Below I particularly focus on climate, air pollution and water quality.

1.3.2.1 Climate

One main contribution of reactive nitrogen to climate change is the emissions of nitrous oxide N_2O , from soils, particularly in agriculture, and from livestock effluents. At the global scale in 2020, 80% of N_2O emissions came from agriculture, 10% from industry, and the rest mainly from waste (which includes wastewater treatment) and land use change (Ritchie et al. 2020). Consequently, its atmospheric concentration have been increasing during the past century (Figure 1.11).

While N_2O is much more potent than CO_2 as a greenhouse gas (about 250-300 greenhouse warming potential over 100 year), it is emitted in much smaller quantity. Consequently, it represents about 5% of the total CO_2 equivalent emissions in the world, while the value is 75% for CO_2 , and 20% for methane.

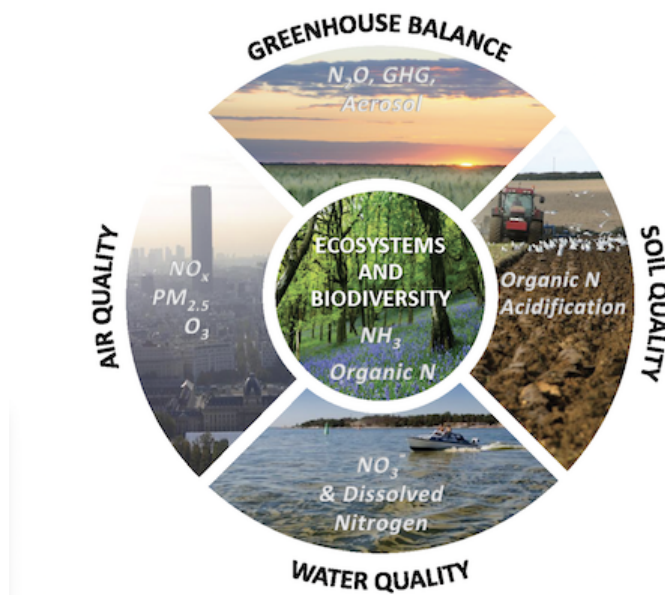


Figure 1.10: The five threats of excessive reactive nitrogen. Source: Sutton et al. (2011a), from Shutterstock.com and garysmithphotography.co.uk.

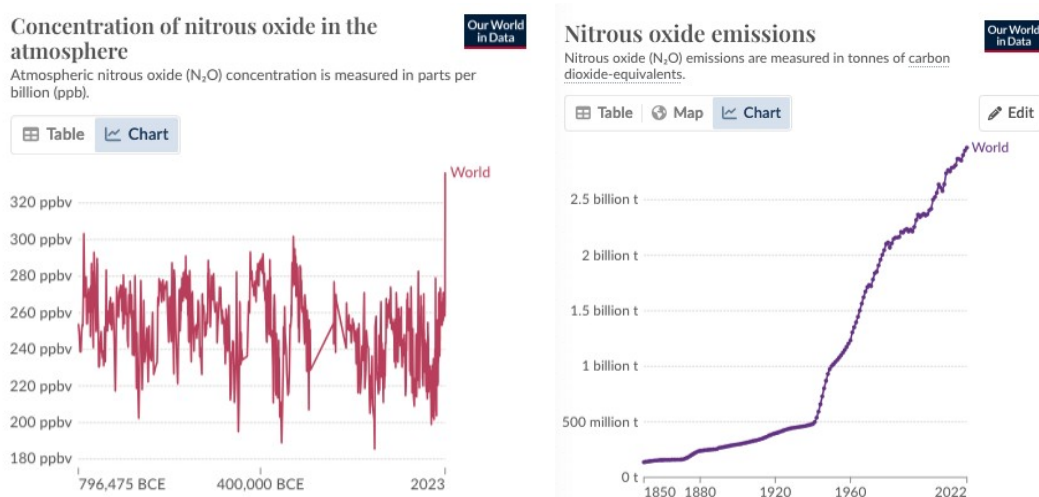


Figure 1.11: Global concentration (left) and anthropic emissions (right) of N_2O . Sources: Ritchie et al. (2023) and Ritchie et al. (2024)

But the total net effect of all reactive N forms is less straightforward (Sutton et al. 2011a). First, NO_x and NH_3 aerosols have a cooling effect, as they promote aerosols formation. Second, other indirect effects are involved like increasing CO_2 sinks with N deposition and fertilization, or alteration of the CH_4 flows. Consequently, in Europe, the net effect of European emitted N_r is a net cooling, but with uncertainty bounds including a net small warming. Large uncertainties come from the soil N_2O emission factors.

Also note that besides its warming potential, N_2O emissions also play a major role in stratospheric ozone depletion (Ravishankara et al. 2009).

1.3.2.2 Air pollution

Several N_r forms contribute to air pollution: reduced ammonia NH_3 and oxidized NO_x , of which nitrogen dioxide NO_2 . They can directly impact human and ecosystems, but also indirectly through ground level ozone O_3 and particulate matters. Ozone results from the photochemical alteration of NO_2 in the presence of volatile organic compounds, while particulate matters are formed when NO_2 is oxidized and combined with NH_3 , resulting in ammonium nitrates particles (Vries et al. 2011).

NO_x are mainly emitted during combustion, particularly from fossil fuel engines and industry, while NH_3 emissions are virtually all issued from agriculture, particularly from livestock effluents and fertilizers containing nitrogen as ammonium (there is less volatilization with nitrate-based fertilizers). While emissions of NO_x have quite successfully decreased due to regulations on transport emission (two-thirds reduction in 30 years in France, Citepa (2022b)), NH_3 emissions are much more stagnating (-20% in 30 years in France, Citepa (2022a)). While some measures like manure management can limit volatilization, more drastically reducing NH_3 emissions would necessitate to reduce the agricultural N flow, and thus total inputs, which is still not the case.

The impacts on human health include asthma and cardiovascular events. NO_2 and ozone result in foliar damage, diminishing vegetation growth.

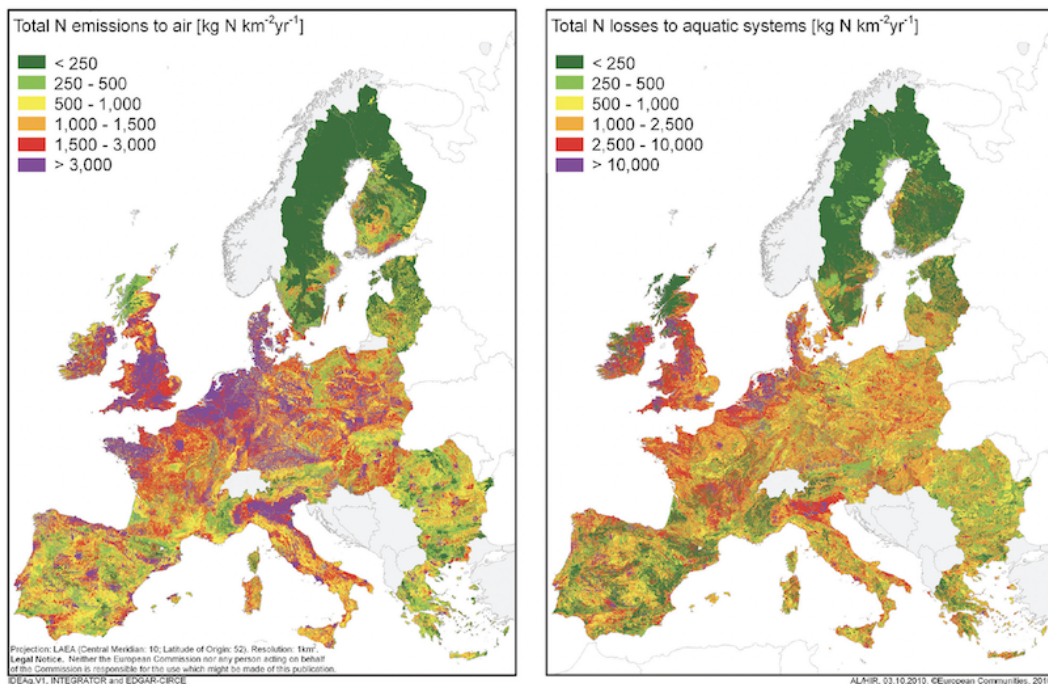


Figure 1.12: Reactive Nitrogen emissions to the atmosphere (left, includes NO_x , NH_3 and N_2O) and to aquatic systems (right, mainly nitrates but also other leaching and wastewater discharge), in 2000. Source: Sutton et al. (2011a)

It is possible to correlate the hotspots of N emissions on Figure 1.12, in purple, to human activities. Brittany and the Netherlands correspond to regions with high livestock densities, whose manure emits NH_3 . The Paris area or Northern Italy are examples of densely populated areas with NO_x emissions from vehicles or industrial combustion.

1.3.2.3 Water quality

I will not expand on the effect of excess nitrogen in water bodies resulting in eutrophication, which has already been treated in Section 1.3.1. Within the European Nitrogen Assessment, Grizzetti et al. (2011) estimated that above a concentration of $1.5 \text{ mgN}\cdot\text{L}^{-1}$, the potential risk of surface water eutrophication is high, and low below when $0.5 \text{ mgN}\cdot\text{L}^{-1}$. With these values, the vast majority of Europe territory was in high risk of eutrophication in the early 2000s.

Nitrates in drinking water are related to two possible health issues, i.e. "blue baby syndrome" (methaemoglobinaemia) and stomach cancers. Methaemoglobinaemia is a quite rare condition happening only with the most extreme concentrations of nitrates. Leigh (2004c) reports that the last known cases due to nitrates in the US and UK were in the 1950s, and Grizzetti et al. (2011) states that in the Europe this condition is mostly due to pathogens in the water rather than nitrates.

Concerning the risks of cancer, Grizzetti et al. (2011) note that it is very difficult to establish adverse effects from nitrates in drinking water with epidemiological studies, because of multiple sources of nitrates in diets: less than 20% come from drinking water, the rest mainly from meat and vegetables. A compromise value for drinking water of $50 \text{ mgNO}_3^- \cdot \text{L}^{-1}$ (or $11.3 \text{ mgN}\cdot\text{L}^{-1}$), established by the WHO has been adopted by the European Union Drinking Water Directive, while the United States adopted $44 \text{ mgNO}_3^- \cdot \text{L}^{-1}$ (or $10 \text{ mgN}\cdot\text{L}^{-1}$). The European Union also applied this threshold for groundwater and surface waters in the European Nitrates Directive.

In Western Europe, during the 2000s, most of the surface waters⁶ were below the $50 \text{ mgNO}_3^- \cdot \text{L}^{-1}$ threshold (Figure 1.13); but due to the population density distribution, about one fifth of the European population lived in areas where it was higher than $50 \text{ mgNO}_3^- \cdot \text{L}^{-1}$.

⁶but note that most (2/3) of the European population got its drinking water from groundwaters

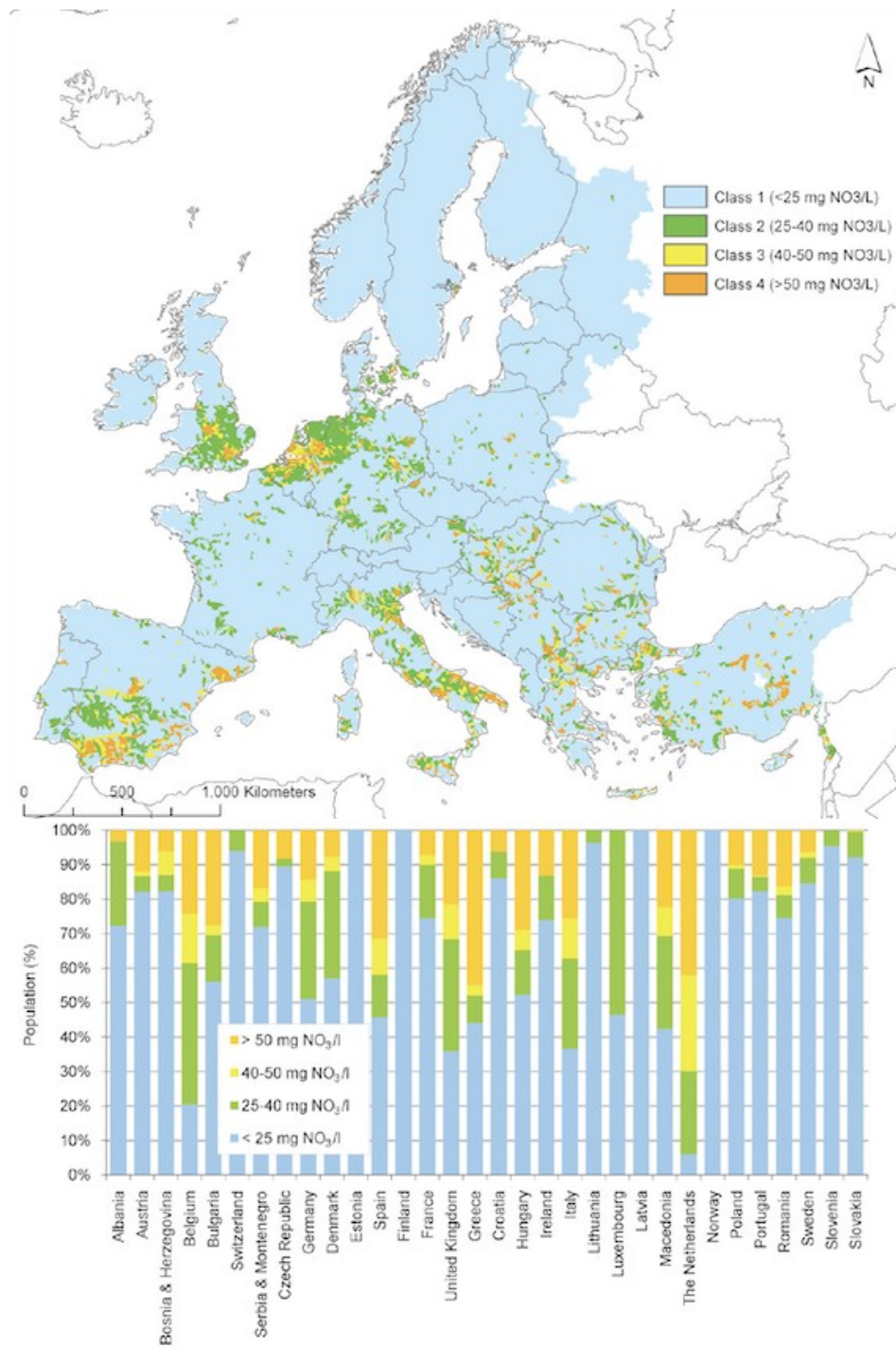


Figure 1.13: Nitrates levels in surface waters in Europe in the 2000s. Source: Grizzetti et al. (2011)

Chapter 2

Disruption of global biogeochemical flows

N and P are often limiting biomass productivity, and some humans have searched for increasing nitrogen and phosphorus availability for agricultural production, through P rocks exploitation (Section 2.1) and atmospheric N fixation (Section 2.2). But these huge new inputs significantly disrupted the planet biochemical flows, described Section 2.3.

2.1 Exploitation of a finite resource: Phosphate Rocks

In this section, I begin with a quick history of the first inorganic and mineral P fertilizers used during the XIXth century, such as guano, bone meals and superphosphates (Section 2.1.1). Despite considerable attention given to them, their actual role in the global food production has been minimal. As Barles (2005) puts it:

” The few works dealing in any detail with the history of soil fertilization give, in our view, excessive importance to guano, sodium nitrate and superphosphates in the XIXth century, even though their role was initially quite secondary”

This is easily visible in Figure 2.1, where guanos are barely visible, as phosphate rocks during much of the XIXth century. However, they had local influence on soil fertility, namely for some farmers in the UK or the US. These farmers got familiarized with the concept of mineral fertilizers, and that they could skip a fallow rotation, laying the ground for the XXth century industrial farming. The influence of these fertilizers was also felt geopolitically in countries where they were extracted, with conflicts and wars waged for these precious minerals. Finally, most of the processes used to transform insoluble phosphate minerals to soluble phosphates were developed during the XIXth century.

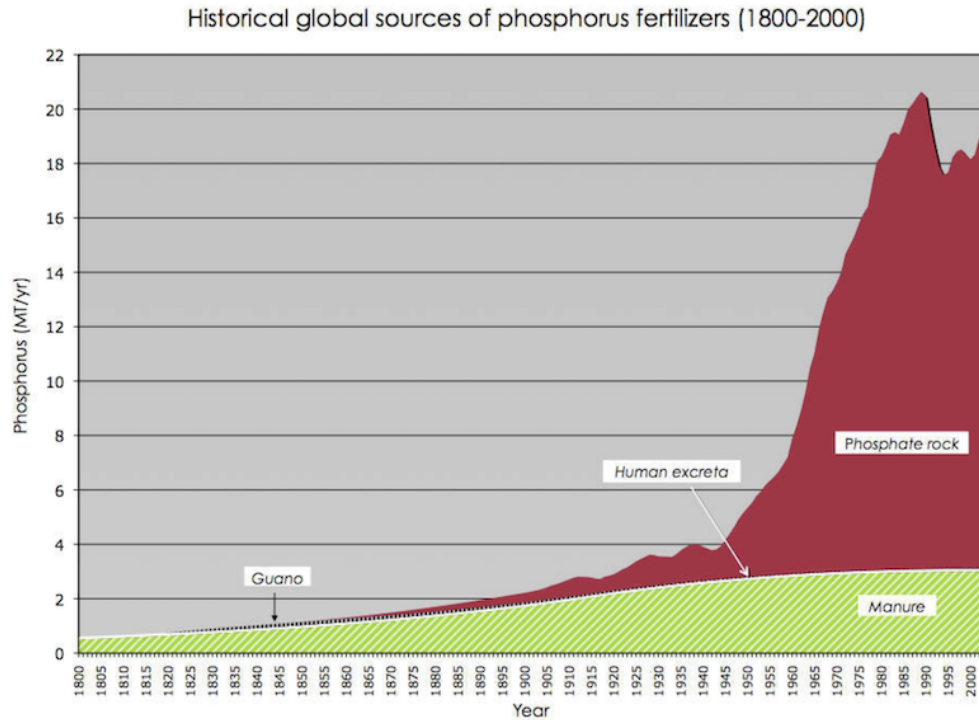


Figure 2.1: Global P fertilization sources during the XIXth and XXth centuries. Source: Cordell (2010)

I then describe the modern P fertilizer industry based on the exploitation of P rock (Section 2.1.2). As this is a finite resource, there are important stakes concerning its total reserves quantities and their distribution, that I address in Section 2.1.3. Finally, linked to these reserves issue, I weigh in the peak P debate that has known some momentum in the late 2000s, and try to go beyond this framework and address P vulnerability (Section 2.1.4).

2.1.1 Emergence of a P fertilizer industry: XIXth century

The reader eager to get a summary of the historical development of the P industry can refer to Mikkelsen (2019).

2.1.1.1 Identification of an element¹

The discovery of the elemental phosphorus is reported to have occurred in 1669, when Hennig Brand attempted to produce the philosophers' stone by boiling 5,000L urine from dozens of German soldiers, producing in the end a glowing mixture. Brand named it phosphorus ("light bearing" in Latin). This elemental phosphorus is very reactive and must be kept away from air, as it exothermically reacts with

¹The elements detailed in this subsection are reported from Butusov et al. (2013d), Scholz et al. (2014), and Elser et al. (2020a).

oxygen at ambient temperature, starting a fire.

The procedure was subsequently documented and reproduced, namely by Leibniz in 1710, and in 1776 Lavoisier officially identified the element, the 13th in the chronological list of discovered atoms.



Figure 2.2: Painting by Joseph Wright, 1771. *The Alchemist, In Search of the Philosopher's Stone, Discovers Phosphorus, and prays for the successful Conclusion of his operation, as was the custom of the Ancient Chymical Astrologers.* Source Wikimedia Commons (2024e)

2.1.1.2 Non soluble crushed bones²

Bones and teeth are rich in phosphorus owing to their composition in phosphate calcium, and the human skeleton typically contains about 80-90% of the body P. The demand for pounded bone began at the beginning of the XIXth century, particularly in England, where demand became higher than domestic supply. The imported bones peaked around 30 ktons·year⁻¹, and quickly England turned to Europe to expand its sourcing, and battlefields of the Napoleonic wars were a privileged location to collect it on the bodies of dead soldiers, as noted by Justus von Liebig, reported in Mikkelsen (2019):

"England is robbing all other countries for their fertility. Already in her eagerness for bones, she has turned up the great battlefields of Leipzig, and Waterloo, and of

²The elements detailed in this subsection are issued from Elser et al. (2020d), Butusov et al. (2013f), Scholz (2018), and Mikkelsen (2019)

Crimea: already from the catacombs of Sicily she has carried away the skeletons of many successive generations. Annually she removes from the shores of other countries to her own the manuerial equivalent of three million and a half men.... Like a vampire she hangs from the neck of Europe”

But phosphorus in crushed bones was still rather insoluble and unavailable to plants. Soon other sources of mineral P, more readily soluble, gained favor over untransformed bone meal.

2.1.1.3 Guanos: 1840s-1850s³

A particularity of the Pacific Ocean West of South America is its upwelling zone. Winds along the coast draw from the deep ocean cold and nutrient-rich waters. These nutrients support a rich ecosystem, from phytoplankton to coastal birds feeding on an abundant fish resource. The nutrients eaten by these birds are finally excreted on the coast shore or islands they use as reproductive sites. Here, they accumulated for millennia, sometimes up to 50m like on the Chincha island, as they are deposited in arid zones and are not washed away by rainfalls, contrary to European climates. These deposits of birds excretions are called guano.

Guanos were already identified as a precious resource by the pre-Colombian Amerindians. A logistic system was in place to transport them from the coast to the mountains; it was forbidden to disturb the birds during nesting, and considered a crime to steal guano from other villages. Garcilaso de la Vega, born in Cuzco in 1539 of an Inca mother and Spanish father, reported this in his *Comentarios reales de los incas*, describing Incas' life (Leigh 2004d). However conquistadors did not pay much attention, as they were more interested in gold (Gorman 2013b).

Centuries later, around 1800, the explorer Alexander von Humboldt, who gave his name to the Peruvian current involved in this upwelling zone, did took notice of the guano boats : *”You can smell them a kilometer away. The people sailing them were accustomed to the smell, but we could not help sneezing when they approached”* (Butusov et al. 2013f). He brought some guano back to Europe, and influenced Justus von Liebig, who advocated for the use of guano instead of crushed bones. Analyses revealed guano from the Chincha island contained about 10% of phosphoric acid. This became a source of commercial mineral fertilizers during the 1840s-1870s. The growth in British imports of Peruvian guano were particularly impressive: from only 2,000 tons in 1841, to 4,000 tons in 1843 and more than 200,000 tons in 1845, a 100-fold increase.

In 1842 the Peruvian government nationalized the Chincha Islands. Prisoners and slaves were used to mine the guano, and even after the official Peruvian abolition

³The elements detailed in this subsection are issued from Elser et al. (2020d), Butusov et al. (2013f), and Scholz et al. (2014)

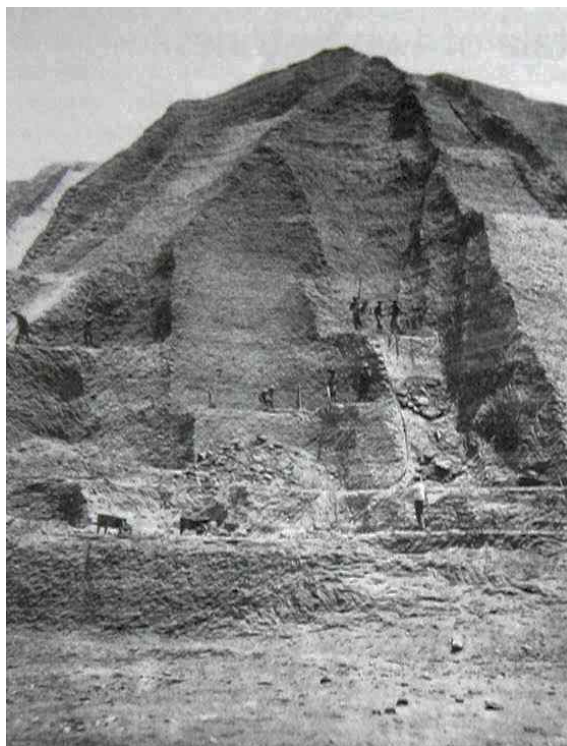


Figure 2.3: Guano mining by Chinese workers on the Chincha Islands, Peru, around in 1860. Source Wikimedia Commons (2024c)

of slavery in 1854, workers' conditions did not change that much (Figure 2.3). This attracted the colonial appetite from other nations, most exemplified by the guano Island Act from US Congress, enacted in 1856, stating that US citizens could take ownership of any guano island (Gorman 2013c). The very next year, the possession of Navassa Island was claimed. The act was suspended in 1863, but meanwhile more than 60 islands were appropriated. Despite Haiti protests concerning Navassa Islands, the US supreme court unsurprisingly judged in favor of its own citizens in 1890. Nine Islands are still the property of the US. The claimed islands were often abandoned after complete exploitation and devastation.

US farmers draw some of the demand for guano in the 1840s-1850s, yet most of the demand still came from British farmers. But guano exploitation and exports quickly declined by the end the XIXth century. First, because it was a limited resource that took millennia to form. Second, because of the development of competitive soluble superphosphates made from much more abundant P rocks.

2.1.1.4 First acidulation of P rocks; second half of the XIXth century⁴

Superphosphates are chemical compounds containing more or less important quantities of phosphates ions PO_4^{3-} . The businessman John Bennet Lawes (1803–1873),

⁴The elements detailed in this subsection are issued from Scholz (2018), Elser et al. (2020d), and Butusov et al. (2013g)

landowner of the Rothamsted Experimental Station, along with the chemist Henry Gilbert, induced a turning point with an agronomic experiment: compared to simple ground bones fertilizers, they found that treating them with sulfuric acid resulted in higher yields. In 1842, Lawes registered a patent on phosphorus acidulation with sulfuric acid to produce a soluble single superphosphates, mono-calcium phosphate, from bones. The production started the very next year in Deptford, London, and during the 1870s, Lawes business was producing about 40,000 t superphosphate per year. The process was not limited to England. In the United States, when the federal governments incited to slaughter buffalo during the XIXth century, in part to drive indigenous Amerindians off their land, a market opportunity was to transform all the buffalo skeleton to bones treated with acid (Figure 2.4).



Figure 2.4: Buffalo skulls in the US, 1892, ready to be treated with sulfuric acid to produce superphosphates. Source Wikimedia Commons (2011)

While acidulation was first used on crushed bones, the process was soon applied to a much more abundant resource, P rocks. Mineral phosphate fertilizers were widely used in Lawes' Rothamsted Experimental Station in the following decades, delivering 169 scientific papers. The classic experiments of Broadbalk Winter Wheat, started in 1843-1856, and studying the response of wheat yields to nutrient, is currently still undergoing (See Section 2.2.2 for data on that matter).

The first large P mine was located in England in 1847; in 1867 a first mine opened across the Atlantic in South Carolina. During the XXth century, P fertilizers production was pursued with the development of triple superphosphates, made from phosphate rocks treated with phosphoric acid, resulting in a P product 90% soluble. Moreover, triple superphosphate contains 46% phosphate (or 20% P) whereas

simple superphosphate contains 16-20% phosphate (or 7-9% P). This concentrated P allowed to reduce transport costs and the labor needed to fertilize land compared to animal manure and organic fertilizers (Table 2.1).

Table 2.1: P content of different fertilizers. Sources: for guanos, Guanos: Smil (2004a), appendix E; Superphosphates: Mikkelsen (2019); Manure: Li et al. (2019).

	PO_4^{3-} content	P content	Moisture Content
manure - ruminants	-	0.2-2%	60-95%
guanos	-	3-6%	10-15%
single superphosphate	46%	7-9%	0-5%
triple superphosphate	90%	20%	0-5%

2.1.2 The modern Phosphate Production Industry

2.1.2.1 A growing production - for now

The rise of phosphate rocks exploitation can be seen as the end of guano, but certainly not the end of extractivism. Maybe one place mostly exemplifying the desolation associated with the exploitation of P rocks is Nauru island, a small state of 20 km², North East of Australia, successively colonized by Germany, Australia, and Japan. P rock mining began at the dawn of the XXth century, and really took off during the 70s, after independence. The intense exploitation increased the GDP per capita close to the highest in the world. But in the 90s the deposits were depleted, leaving the island devastated, and the population poor.

In 1927 the world produced 10 Mtons phosphate rocks (Figure 2.5); after 3 decades of exponential growth starting the 1940s, the 100 Mtons threshold was crossed in 1972, just before a price spike during the first oil crisis, followed by a 3 decade stagnation around 120-150 Mtons. Following a second price spike after the 2008 financial crisis, production almost doubled and is now around 220-250 Mtons.

The vast majority of mined P rocks are used to produce fertilizers (more than 80%). The rest is used as P additives or supplements, or in some niche industrial applications such as chemical industry or for strengthening of steel (Butusov et al. 2013f). In the following I present the processes to produce mineral P compounds, and their applications.

2.1.2.2 Production processes⁵

There are two main mineral P compounds extracted from P rocks: a diversity of phosphoric acids (the majority); and white phosphorus.

⁵Elements in this subsection are mainly from De Boer et al. (2019) and Scholz et al. (2014)

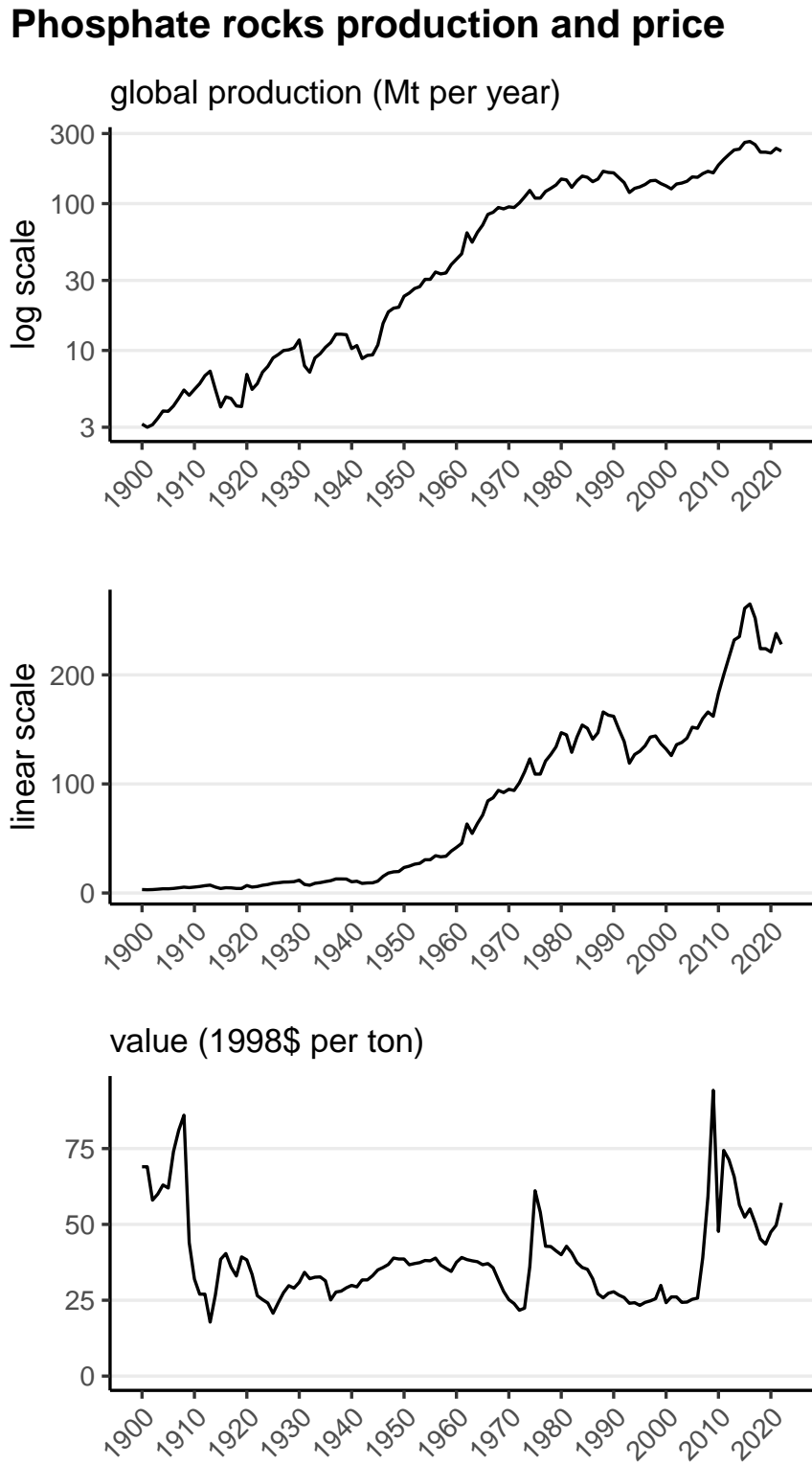


Figure 2.5: Phosphate rocks production and price (in constant 1998 dollars), 1900-2024, (USGS 2024c)

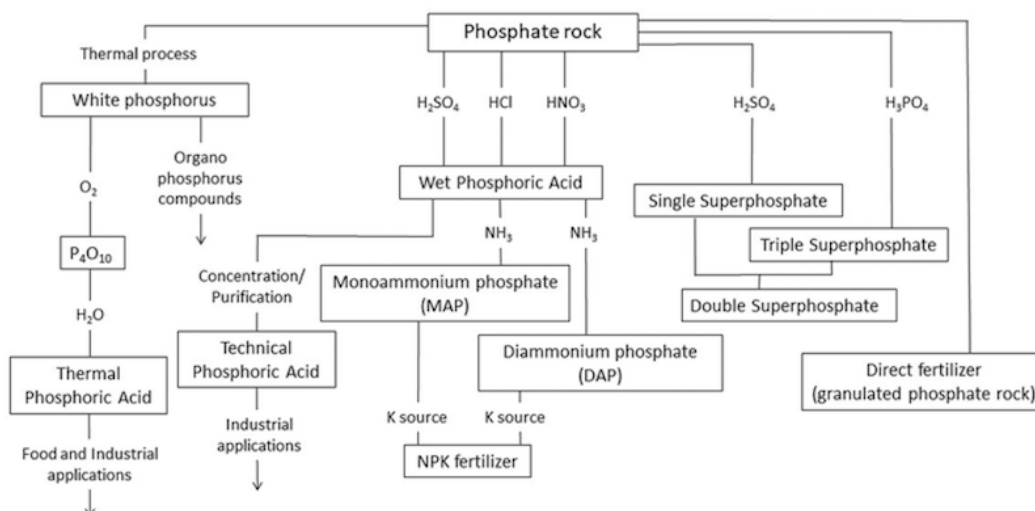


Figure 2.6: Production processes for P chemicals and fertilizers. Source: De Boer et al. (2019)

Sources of P rocks: Most (>80%) of phosphate rocks used to produce phosphorus P_4 come from sedimentary rocks deposited on continental marine shelf across millions of years, the rest from igneous magmatic rocks and metamorphic rocks (Butusov et al. 2013a).

Sedimentary rocks are usually made of fossilized deposits of shells and aquatic animals, in a location where there was once an upwelling zone rich in nutrients. Exploited deposits contain about 20-40% of phosphate (9-18% P) (Butusov et al. (2013g), and table 5.1 in De Boer et al. (2019)). A common retained order of magnitude is that P rocks contain about 30% P_2O_5 , or 13% P (Vaccari et al. 2014). Sedimentary rocks are made of apatite, i.e. calcium phosphate combined with a OH, F or Cl ion.

Igneous rocks are issued from magma cooling. They are much less concentrated in P_2O_5 , from 0.005 to 0.4%, sometimes up to 2%. They can be exploited after beneficiation, which increases their P_2O_5 up to 35-40%.

Metamorphic rocks are sedimentary or igneous that have been buried deep by tectonic movements and encountered high pressure and heat constraints, crystallizing their minerals. They are currently not widely exploited.

Acidulation to produce phosphoric acid (see Butusov et al. (2013g), Vaccari et al. (2014), and Scholz et al. (2014)): The main route to produce mineral P is similar to the one developed 100 years ago by John Lawes to produce superphosphates. Sulfuric acid H_2SO_4 is applied to the P rock, to produce an intermediary wet phosphoric acid H_3PO_4 (Figure 2.6). The main by-product waste is phosphogypsum ($CaSO_4$ mixed with phosphate rock), about 5 tons for every ton of phosphoric acid.

Processes producing less waste exist, namely the nitrophosphate one with nitric acid HNO_3 , or with HCl , but they are more expensive and thus not commonly used.

From then, the wet phosphoric acid can be simply purified and concentrated, to produce P for technical and industrial applications. But for fertilizers, it is usually combined with nitrogen (NH_4^+ , NH_3) and potassium compound to produce NP (monoammonium phosphate MAP, diammonium phosphate DAP) or NPK fertilizers. Another option is to remix the extracted phosphoric acid with P rocks, to obtain triple superphosphates.

White phosphorus: White phosphorus is produced by heating phosphate rocks with electrodes in presence of coke, a very energy intensive process. Most of the production occurs in China. The reader interested in more details about white (and red) phosphorus production can refer to De Boer et al. (2019).

White phosphorus used to be the constituent to light matches during the XIXth century (at that time white phosphorus was extracted from bone meal), but was highly toxic and this application was banned by the 1906 Berne Convention, and now red phosphorus is used in matches.

Virtually all the industrial demand for elemental phosphorus is met via white phosphorus (P4). Elemental phosphorus is used among other things to produce organo phosphorus compounds and chemicals, such as PH_3 , PCl_3 , P_4S_{10} , P_4S_3 ... Industrial products made from white phosphorus include feed additives, detergents, flame retardants, pesticides, medicines, incendiary derivatives (napalm, explosives, fireworks)... It is an extremely reactive and inflammable compound, that must be stored with great precautions.

Non fertilizer uses of phosphoric acid: While most (80-90%) of phosphate rocks are transformed to fertilizers (Scholz et al. 2014), there are numerous other industrial uses, summarized in Table 2.2.

Of particular interest for the following of this manuscript is the industrial use of P to produce detergents additives, namely sodium tripolyphosphate, widely used in the second half of the XXth century for washing machines and dishwasher (Scholz et al. 2014). Sodium tripolyphosphate can bind to other cations, increasing the power of detergents. Widely used in industrialized countries before there progressive ban (this will be discussed in the results of Chapter 4), these detergents, first in greywater, then in sewers, and finally discharged to the environment after partial P treatment in wastewater treatment plants, were the major cause of the important increase of P content in rivers. For instance, in US effluents, P content increased from $3 \text{ mg}\cdot\text{L}^{-1}$ in the 1940s to $11 \text{ mg}\cdot\text{L}^{-1}$ in the 1970s (Litke 1999).

Table 2.2: Industrial uses of phosphoric acid in 2011. Source: De Boer et al. (2019), based on Gantner (2015).

Industrial Application	share of industrial P_2O_5 use
Detergents and cleaners	38%
Food and beverages	23%
Metal treatment	10%
Water treatment	10%
Tooth paste	3%
Special fertilizers	2%
Miscellaneous	14%

Contaminants An issue of sedimentary phosphate rocks is their enrichment in some heavy metals, namely cadmium, and radionuclides, namely uranium. For igneous rocks, the heavy metal contamination mostly concerns arsenic and selenium (Vaccari et al. 2014).

Table 2.3: Content of phosphate rocks in cadmium and uranium. Source: De Boer et al. (2019)

		Cadmium (mg/kg P_2O_5)	Uranium (mg/kg)
Igneous rocks	Russia	0.25	20
	South Africa	0.38	-
Sedimentary rocks	USA	24-210	80-150
	Morocco	55	100-160
	Jordan	18	80-110
	Togo	147	100-110

European countries set limits on Cd content during the 2010s, concerning fertilizers (21.5 to 90 mg·kg⁻¹ of P_2O_5), inputs in agriculture (0.15–150 g·ha⁻¹·year⁻¹), and content in soils (0.5–3.0 mg·kg⁻¹ of dry soil) (Vaccari et al. 2014). For that reason, phosphate rocks exports to European markets usually require a cadmium removal step (Butusov et al. 2013g).

Waste and environmental impact: The environmental impact of phosphorus production begins with mining, when several meters of topsoils are removed. Then, large amounts of sulfuric acids are used for the majoritarian wet process acidulation producing phosphoric acid. For every ton of phosphoric acid, about 5 tons of phosphogypsum waste is produced. The vast majority of this phosphogypsum is just deposited in landfills, without further treatment (Tayibi et al. 2009). It could be theoretically used in constructions, but is in practice too contaminated and considered

not economically interesting (Butusov et al. 2013g). Pollutants in phosphogypsum include fluorine, cadmium, radium, sulfur, uranium, and rare earth (De Boer et al. 2019).



Figure 2.7: Top: Phosphate mine in Utah, USA; Bottom: Phosphogypsum stack in Florida, USA. Source: Wikimedia Commons (2021) and Wikimedia Commons (2024d).

In the coastal Togo mine, mining sludge were simply piled up and 40% went to the sea in the 2000s (Gnandi et al. 2006). These waste contaminate the water because of their heavy metals content, but also induce eutrophication because they still contain non negligible amounts of bioavailable P. Even in an industrialized country like the USA, the Environmental Protection Agency of Florida assessed the site comprising 3/4 of the national reserves and concluded that the company exploiting it "failed to elaborate a sustainable strategy" (Butusov et al. 2013g).

Finally, the impacts can be also visible on the very body of surrounding populations. Dental fluorosis is a condition, whose symptoms manifest with opaque areas on the teeth enamel, sometimes up to brown stains or teeth distortion; it is caused by ingestion of fluoride in excess quantities during the enamel formation. Phosphate-rich soil areas are naturally rich in in fluorine, resulting in dental fluorosis for animals bred in these regions (El Amiri et al. 2021). But besides this naturally occurring dental fluorosis, industrial production of phosphoric acid can exacerbate this by emitting fluorine-rich dusts, like in the Moroccan city of Khouribga (nick-

named *Phosphate city*). A study (Møller et al. 1975) found that in the 70s, dental mounting in children was related to the proximity to the Khouribga phosphate rocks factory, and that the incidence decreased in children growing after the installation of dust air filters removing.

2.1.3 Moving reserves and unequal distribution between countries

The reserves are defined as the known deposits that "could be economically extracted or produced at the time of determination" (USGS 2022). By construction, reserves are thus linked to economic and technological conditions, and are based on moving grounds. In 2012 the USGS estimated global reserve of phosphate rocks to be around 74 Gt (Figure 2.10)⁶.

From this definition of reserve, it appears that their quantity can vary if: new deposits are discovered; economic conditions change; new technologies are developed. This is most exemplified with the reassessment of Moroccan reserves in 2010. In 2007, the USGS estimated the global phosphate rocks reserves to be around 16 Gt, of which 6 were in Morocco. This corresponded to around 100 years of the then 150 Mtons/year consumption. But in 2010, following a report from the International Fertilizer Development Center, Moroccan reserves were upgraded to 50 Gtons, bringing the global reserves to about 60 Gtons, or 400 years of the global consumption (Figure 2.9). This huge increase has probably been in part triggered by the price spike that occurred a few years earlier (Figure 2.5).

The economic reserves for the principle mineral fertilizers have been investigated by Jönsson (2019) (Figure 2.8), expressed in terms of years left at the annual production rate. Most exemplifying the concept of moving reserves is zinc, whose reserves have stayed below 20 years of exploitation for more than 20 years. Similarly, for almost 50 years, from 1965 to 2010, Scholz et al. (2013) report that the resource/consumption ratio of uranium has remained roughly stable at around 100 years, despite increasing consumption.

The distribution of reserves is extremely unequal, with 2/3 in Morocco and the contested territory of Western Sahara; China, Russia, Tunisia and Algeria have each between 2 to 4 Gtons reserves, making 15% of the global total; the rest of the world shares the remaining 20% (Figure 2.10). On that list, the only European country is Finland, with 1 Gton.

⁶Even more uncertain are the resources, whose definition is "A concentration of naturally occurring solid, liquid, or gaseous material in or on the Earth's crust in such form and amount that economic extraction of a commodity from the concentration is currently or potentially feasible" (USGS 2022). All is in the "potential": estimating resources lays on assessment of future technological and economic developments. Resources were estimated to be 200 Gtons of phosphate rocks (Vaccari et al. 2014).

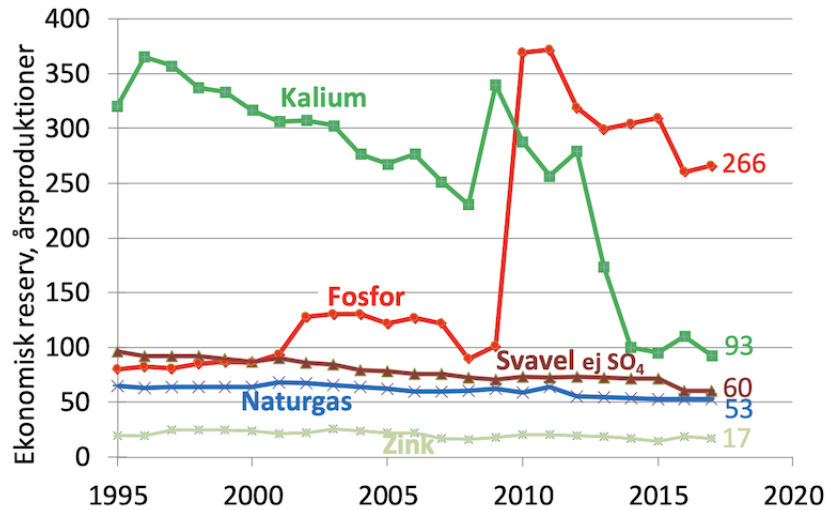


Figure 2.8: Years left to exploit economic reserves at the annual consumption rate, for potassium (green), phosphorus (red), sulfur (brown), natural gas (blue, used to produce N fertilizers with the Haber-Bosch process) and zinc (light green). Source: Jönsson (2019), based on USGS and BP.

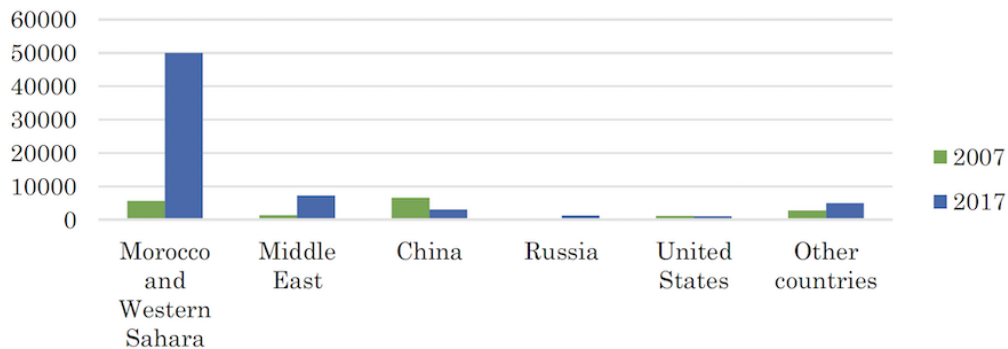


Figure 2.9: Estimation of phosphate rocks reserves in 2007 and 2017. Source: De Boer et al. (2019)

But reserves distribution is not reflected in the production. By far the largest producer is China, with 40% of the global production (Figure 2.10); adding Morocco, the United States and Russia brings the total to 75%. This concentration of production is also reflected in the corporations exploiting the mines: 10 companies are responsible for more than half of the mined P (Butusov et al. 2013b).

2.1.4 Peak Phosphorus: bogeyman or Cassandra ?

“Whether or not there will be a phosphorus (P) peak within decades, centuries or millennia [...], one thing is for sure – phosphorus is a limited and, in its function as a nutrient, an essential and irreplaceable resource” (Kabbe 2019).

I agree with that statement, with one small nuance. It is not “phosphorus”

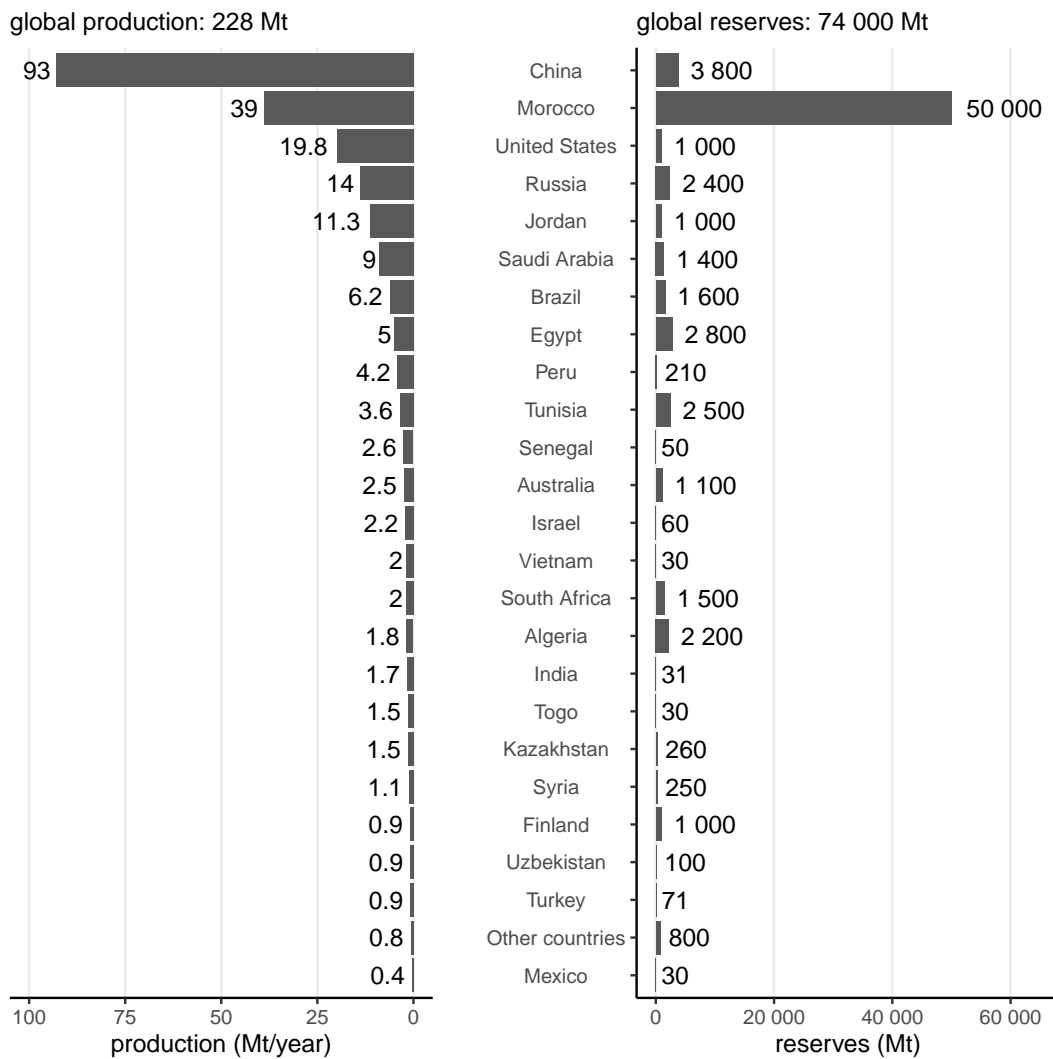
Phosphate rocks production and reserves in 2022

Figure 2.10: Phosphate rocks production and reserves in 2022, (USGS 2024a)

that is limited, but concentrated phosphorus, particularly in phosphate rocks. For instance, P in oceans constitutes an important reservoir⁷ replenished from the P lost from terrestrial ecosystems through erosion, but at too low a concentration to be of economic interest.

As it makes no doubt that concentrated P rocks are a limited resource, the key question is whether the time horizon until depletion is sufficiently close to be of concern for present and future generations. Back in 2009, Cordell et al. (2009) most influential paper argued for a peak within 2 decades, around 2030; on the contrary, Scholz et al. (2013) argued that peak demand will probably happen way before any peak production, and that economically mineable phosphorus can last for centuries.

⁷Considering oceans P concentrations of $6 \cdot 10^{-8}$ kgP/L (Table 1.1), and that oceans are 1.370 Gkm^3 , this yields a total P in oceans of about 80 GtP, an order of magnitude confirmed by Smil (2000). By comparison, the 74 Gtons of phosphate rocks reserves (USGS 2024a), hypothesizing a 30% PO_4^{3-} content, yields a total reserve of about 10 GtP.

In the following I review the issues surrounding this peak P debate. Since peak P projections are relatively recent, starting from the late 2000s, I try to offer an insight on their predictive power by preliminary focusing on peak oil predictions made in the last 50 years, as the quantitative methods applied in both cases are identical.

2.1.4.1 A concept coming from oil: Hubbert model

The concept of "peak" for a finite commodity is most certainly associated to oil. I find it important to introduce peak P with the past developments about peak oil, because seminal papers about peak phosphorus clearly follow the framework laid out by the peak oil models and refer to them (see e.g. *"Peak Phosphorus: the sequel to Peak Oil"* by White et al. (2011), namely the Hubbert model. Other peak P advocates make no mystery of its influence: *"M. King Hubbert [...] correctly predicted that oil production in the lower 48 states would peak in 1970."*; *"The depletion analysis pioneered by Hubbert can be applied to other non-renewable resources"* (Déry et al. 2007).

Indeed, the geophysicist Marion King Hubbert is widely credited for having forecasted the US peak oil production ten years in advance (Hubbert 1962). Retrospectively, in 1976, he boasted on the accuracy of his method (Grenon 1976), and, reflecting on the fact that his prediction was for 1968 vs an actual peak in 1970, he stated "I know of no other estimate that comes anywhere near this accuracy. Some have over-estimated the date of peak production by as much as 30 years". Questioned about the fact that his approach does not consider prices, and whether he finds it reasonable, he simply replied "the use of such data has given results of remarkable accuracy".

Many followed and expanded his projections to the world. A summary can be found in the 1977 Workshop on Alternative Energy Strategies (WAES), cited in Basile (1977): *"The supply of oil will fail to meet increasing demand before the year 2000, and most probably between 1985 and 1995. [...] Numerous other studies support the WAES conclusion, such as reports by the OECD (1977) and by Exxon (1977). Some others were even more pessimistic: the CIA report (1977)."* Actual production data, featured in red on Figure 2.11, clearly shows that such a production peak did not occur in the 80s nor the 90s.

Simple updates of past predictions sometimes gave widely different outcomes, as shown on Figure 2.11 (top left and top right): Hubbert's 1969 model for global oil production is about right until its predicted peak in the 90s, but then real production kept increasing, and currently reality is 3 to 4 times higher than the projection; another of Hubbert's projections, just 3 years later in 1972, gives on the contrary a much higher production peak, not even reached today. Even Hubbert's exact

Peak oil predictions

vertical line: date of projection

gray line: prediction

red line: actual oil production (dotted: only conventional oil)

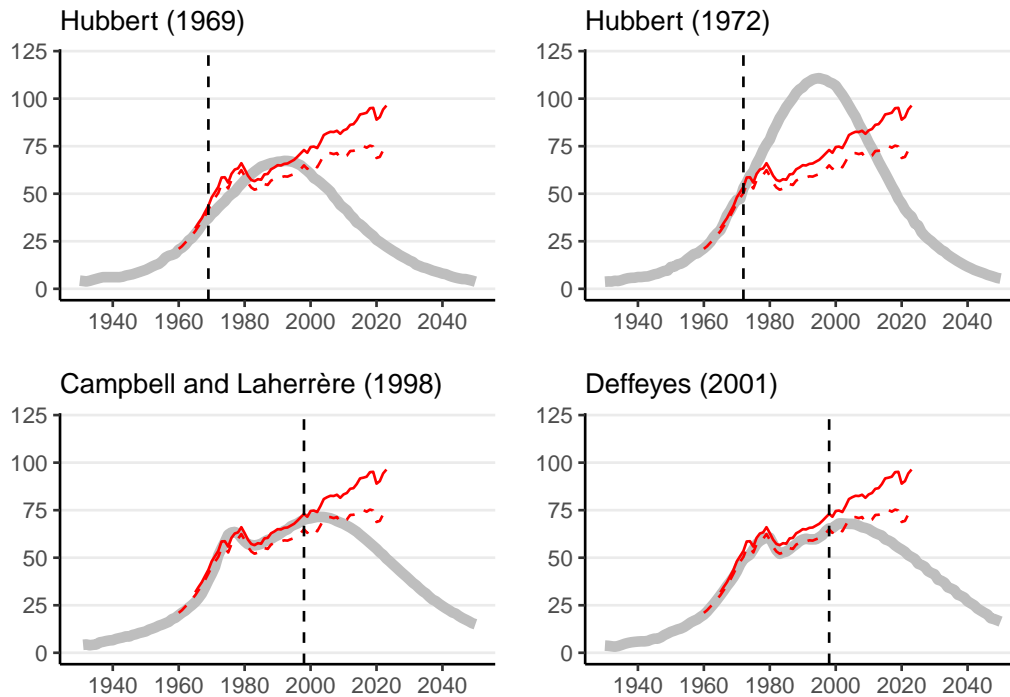


Figure 2.11: Hubbert’s projections in 1969 and 1972 (top left and top right) reflect that slightly changing the model’s parameters can drastically change the peak level. In both cases they are way off reality. Campbell and Laherrère (1998) and Deffeyes (2001) similar projections (bottom) show the vagueness of the criteria to define “true” oil production. In both cases, the original sources simply refer to “peak oil”. But in the first case, the scenario seems to be calibrated up to 2000 to the total oil production, i.e. the plain red curve (bottom left); while in the second case, it seems calibrated to conventional oil production, i.e. the dashed red curve (bottom right). See text for models sources, whose curves have been manually extracted with the software WebPlotDigitizer (<https://apps.automeris.io/wpd4/>). Total oil production (plain red curve): Energy Institute (2023). Conventional oil production (dashed red curve): OPEC (2023).

prediction of US production peak actually neglects the fact that the US shale gas boom increased the domestic production during the 2010s at levels higher than the 1970s, and that the production has not yet decreased.

Some argue that the projections only concerned conventional crude oil, but at the time of the predictions the distinction is usually much blurrier. Deffeyes published in 2001 an influential book about peak oil (Deffeyes 2001), with peak occurring in 2005. But global oil production continued to increase and by 2022 actual production is double his projection for that year (Figure 2.11, bottom right). Even when generously only considering conventional crude oil, reality is 50% higher. Yet in his

strong assertions he did not discriminate between conventional or unconventional oil: *"Around 1995, several analysts began applying Hubbert's method to world oil production, and most of them estimate that the peak year for world oil will be between 2004 and 2008."* *"There is nothing plausible that could postpone the peak until 2009. Get used to it."* (Deffeyes 2009).

The vagueness regarding whether projections are applied to all oil or simply conventional is also exemplified with a projection very similar to Deffeyes', made by Campbell et al. (1998) (Figure 2.11, bottom left). *"Predicting when oil production will stop rising is relatively straightforward once one has a good estimate of how much oil there is left to produce. We simply apply a refinement of a technique first published in 1956 by M. King Hubbert"*. Their main figure in their original article clearly refers to both conventional and unconventional, even though the text is less clear.

To conclude on Hubbert's method applied to oil, multiple projections with widely different estimates (see Figure 2.11 or also Figure 9.3 in Butusov et al. (2013h) and flexibility about what is defined as "real" oil as opposed to unconventional oil guarantees to retrospectively obtain at least one "correct" projection among many incorrect ones. It seems that, as Adelman (1992) puts it: *"Finite resources is an empty slogan; only marginal cost matters"*. Owing to changing situations in technology, economic perspectives, and explorations, reserves will keep evolving.

Half a century of retrospective hindsight about peak oil projections should at least make suspicious when the same methods are applied to another finite resource, in our case phosphorus. If anything, 50 years of peak oil projections have learned us some lessons that can be applied to phosphorus: conventional resources can expand over time; "unconventional" resources can add up to the previous ones.

2.1.4.2 Moving P peaks

Scholz et al. (2013) argue that the Hubbert model cannot be applied to phosphorus reserves; that the scarcity regarding phosphorus is much more economic than physical; and that phosphorus reserves and resources are dynamic and deeply uncertain. This latter point is particularly highlighted in the following with my quick review of past peak phosphorus predictions. For a more in depth analysis of the papers I review below, the reader can refer to Vaccari et al. (2014).

The first occurrence of a supply-driven peak P that I encountered is in Déry et al. (2007). Testing the Hubbert model on the Nauru island, where the P rocks have been exploited until depletion and where exploitation has stopped (see Section 2.5), they found a very good match, by applying the model to the data just before and just after the peak. An epistemological shortcoming is that the time frame on which the model is applied is chosen with hindsight of the historical peak. They then

applied their method at the global scale to estimate a future peak P. But they chose an ultimately recoverable resource way lower than what was reported at the time: only 35% of the USGS reserve official figures (Vaccari et al. 2014). Their approach, based on data up to 2005, predicted that the global peak P already occurred in 1989, and a near complete depletion by 2050: ” *We are probably on a world decline of rock phosphate production*”. As can be seen on Figure 2.12 (left), in the following 5 years production actually exceeded the 200 Mtons threshold and has stayed beyond ever since. In 2022, production was about 230 Mtons, 6 times higher than their predicted 38.5 Mtons for that year.

Two years later, Cordell et al. (2009) followed the same approach, this time applied to the full 16 Gtons reserves reported by USGS at that moment (Figure 2.12, middle). A widely cited quote from this paper is that ” *while phosphorus demand is projected to increase, the expected global peak in phosphorus production is predicted to occur around 2030*”. Yet they subsequently updated their work with the new estimates of USGS reserves of 60 Gtons a few years later (White et al. 2011), postponing the peak to somewhere around 2070, at a 480 Mtons value double their original prediction (Figure 2.12, right). Whether the demand will reach such a high amount remains an open question. Since these articles, global reserves have been further increased to 74 Gtons.

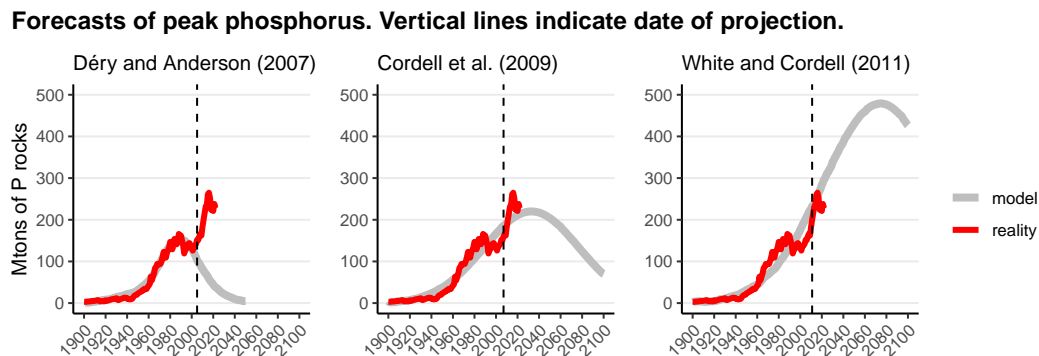


Figure 2.12: Comparison of subsequent peak phosphorus predictions to actual production data from USGS (2024c). See text for models sources, whose curves have been manually extracted with the software WebPlotDigitizer (<https://apps.automeris.io/wpd4/>)

This rapid review exemplifies that every estimate of a phosphorus peak is based on a blurry estimate: the amount of ultimately recoverable resource. This estimate can substantially change, sometimes by an order of magnitude, following technological and economic developments. Until now it always went in the direction of expanding reserves and postponing the peak; but one can speculate a future where economic conjuncture makes phosphate rocks exploitation less profitable, and decreases the reserves. Probably, we will know the date of phosphorus peak production

only decades after it has already been passed, like in Nauru island.

I hope that this detailed retrospective regarding oil and phosphorus convinced the reader that projecting a peak date is vain. At best and on the short-term, it can mobilize people, but on the long term it probably undermines scientists' credibility. Moreover, there is no need of an hypothetical short-term peak to foster the need for less consumption. Resources are concentrated in an handful of countries, making importers sensible to geopolitical shocks; high consumption of P fertilizers only mirrors the losses along the food chain, which are a major driver of environmental pollution (Section 1.3); and P fertilizers are based on mining with widespread environmental and societal consequences.

2.1.4.3 Beyond peak: P vulnerability

Even without environmental or societal concerns, there are still good reasons to pursue the aim of reducing the reliance on P rocks for phosphate fertilization.

Based on the arguments developed in the previous section, I think there is little basis for concerns regarding a physical scarcity of P rocks on the medium run. Much more concerning should be vulnerability to factors limiting access to these fertilizers. Figure 2.5 illustrates how volatile P rocks prices can be. While it features a spike by a factor 3 to 4 during the years 2007-2008, this hides higher increases on a monthly basis. This happened simultaneously to food riots in Africa, Asia, the Middle East, the Americas and the Caribbean, caused by a spike in food prices: +130% for wheat and +100% for rice in the first 3 months of 2008 compared to 2005 (Bush 2010).

Besides the simple vulnerability to price instabilities, the sheer concentration of P rocks reserves and production in a handful of countries and companies makes the importing countries vulnerable to geopolitical tensions (Childers et al. 2011). In 2008, China, by far the number one phosphate rocks producer (Figure 2.10), implemented an export quota on its phosphate rocks production, contributing to the price spike on that period by diminishing offer (De Boer et al. 2019). More stringent export bans in the future could result in more restricted access to P rocks.

Particularly concerned is Europe. While being an important consumer of phosphate rocks, almost all of its consumption is imported. In 2010, it imported 7.5 Mtons of phosphate rocks, but the only mine on European territory, the Siilinjärvi mine in Finland, produced only 10% of that amount (Van Dijk et al. 2016; HCSS 2012). Previous mines in Belgium, Italy or the UK have all been closed out of environmental concerns; in France the Terres Rouges mine was closed in 1998 (De Boer et al. 2019). For that reason, in 2014 the European Commission added phosphorus on its list of 20 critical raw materials, based on its economic importance and supply risk (JRC 2014). Several other indicators are being developed, such as the Worldwide Governance Index focusing on political stability, or the Herfindahl–Hirschmann

Index, assessing spatial concentration of supply (Scholz et al. 2014).

Europe will have to decide whether it wants to keep outsourcing the social and environmental damage of its P rock production, and stay vulnerable to economic and geopolitical instabilities; decrease vulnerability by relocating production and social and environmental damage; or decrease vulnerability by decreasing its needs through decreased P fertilizers consumption and increased recycling; or whatever other option...

2.2 The emergence of the N fixation industry

Here I describe the first exploitation of mineral N in the form of saltpeter during the Middle Ages and the Renaissance for explosives (Section 2.2.1), whose quest triggered the understanding of the different N reactive forms during the Industrial Revolution (Section 1.2.2). Like phosphorus, nitrogen exploitation began with the mining of different finite deposits in the late XVIIIth century: guanos, Chilean nitrates, and nitrogen imprisoned in coal.

But contrary to phosphorus, some industrial processes were developed in the early XXth century to fix reactive N from the virtually unlimited inert N₂ atmospheric pool. First with the cyanamide and the Norwegian electric arc (Section 2.2.4). But the Haber-Bosch process finally prevailed (Section 2.2.5), triggering a massive disturbance of the global biogeochemical flows.

All these processes are based on physico-chemical reactions, but during the second half of the XXth century, progress in genetics held out the prospect for an unfulfilled promise: enabling biofixation in major cereal crops (Section 2.2.6).

2.2.1 Saltpeter: a substance of pow(d)er⁸

Besides the obvious important role of N in plant nutrition since the beginning of agriculture, it also started to emerge as an important geopolitical factor during the first millennia of our era, when the recipe for gunpowder was developed in China. Its key ingredient to enable explosions is saltpeter (potassium nitrate, KNO₃), mixed with charcoal and sulfur.

Saltpeter (or niter) can naturally crystallize on decomposing matter when water evaporates. As long as water is present, nitrifying bacteria transform NH₃ into NO₃⁻. Once water evaporates, NO₃⁻ will crystallize with other cations (Na, Ca or K), forming nitrate salts. To replace NaNO₃ or CaNO₃ to niter, K-rich wood ash were usually added. In India, the climate allows the water to naturally evaporate, while in Europe saltpeter was usually found on porous rocks, for instance in underground cellars.

But relying on the naturally formed saltpeter was not enough to keep up with the demand, and so niter beds were constituted (Figure 2.13). Urine, manure and decaying organic matter were concentrated in holes where water was filtrated, allowing nitrate salts to form on the top of the pile. The Chinese Song dynasty (around XIth-XIIIth century) already used saltpeter as an incendiary weapon. Once the recipe was adapted in the XVth century by Europeans to make firearms and

⁸The elements detailed in this subsection are reported from Gorman (2013d), Gorman (2013b), Multhauf (1971), and Leigh (2004d)

guns, securing its supply was a key concern, and during the XVth century European states supported the transformation of urine, manure and ash to saltpeter. This usage obviously competed with local agricultural use.



Figure 2.13: German nitrary circa 1580. Leaching deposits (C), where saltpeter crystallizes, are filled with manure and vegetal materials. Source: Wikimedia Commons (2024f)

In 1631, Charles I of England gave order to save urine to be collected for saltpeter production. In Sweden during the XVIIth century, each household was supposed to provide materials for saltpeter production (soil sheep dung, ashes, wood, straw...). Facing the increasing demand for gunpowder, the United Kingdom, chose to out-source the production to its colonial empire. In 1800, it relied almost completely on India, from which 3,000 tons of saltpeter were imported each year via the East Indian Company (Gorman 2013d). In France around 1750, the consumption was 2,000 tons per year, first collected by private companies and then by the *Compagnie Royale*, considering the importance of this strategic resource.

2.2.2 Discovering N minerals and plant nutrition⁹

Johann Rudolf Glauber, through experiments, established during the XVIIth century a link between ammonia, saltpeter and plants, and he identified niter (i.e. potassium nitrate) as common substance in these three cases. Identifications followed during the XVIIth century, with scientists / alchemists such as John Evelyn or Edme Mariotte. Finally, during the XVIIIth century, Lavoisier, using the concept of mass conversion, officially identified nitrogen as an atomic element. He also identified that nitric acid contained oxygen in 1776, but its precise formula will be established in 1816 by Gay-Lussac and Berthollet. In 1785, Henry Cavendish, investigating the

⁹The elements detailed in this subsection are reported from Leigh (2004b), Gorman (2013e), and Smil (2004b)

composition of air, established that it was made of 1 part of oxygen with 4 parts of inert nitrogen.

With all these works, in the early XIXth century, there was a true understanding of the different N compounds. Namely, it was established that nitrogen in the air was inert, and that it could be transformed in a reactive oxidized compound with an electric arc. The fact that some plants like legumes might be able to fix it was barely considered. Transformations from one N form to another were also barely understood, as many of these process involve bacteria (nitrification, denitrification, mineralization), and microorganisms were unknown. The XIXth century was a century focused on the role of mineral nutrients in plant nutrition. In the UK, this scientific study of nitrogen and fertilization was investigated by businessmen like John Bennet Lawes, while in Germany it was supervised by the state through academia.

The work of Liebig during the XIXth century allowed to understand that plants do not use directly manure, but rather the NO_3^- and NH_4^+ (and other minerals) once organic matter decayed. In 1840 he published his seminal treaty "*Organic chemistry in its application to agriculture and physiology*". Chemical testing was being developed in that particular context to characterize and standardize the guano imported from South America, which contained from 1% to 15% N (Smil (2004a) in appendix E). This allowed to quickly establish that N in manure was insufficient on its own to restore cropland fertility. At first, Liebig advocated for mineral N fertilization, but quickly flip-flopped and vehemently argued that N deposition from atmosphere was sufficient. But this was based on unrealistic NH_3 deposition rates, in excess of $100 \text{ kgN}\cdot\text{ha}^{-1}$, two orders of magnitude higher than reality. (Smil 2004b)

The predominance of the mineral fertilization, and the absence of consideration for organic matter and nitrogen can be exemplified by one of Liebig's quotations: "*A time will come, when fields will be manured with a solution of glass (silicate of potash), with the ashes of burnt straw, and with the salts of phosphoric acid, prepared in chemical manufactories*" (Agricultural Chemistry (1847), 4th edn.). Except the role of nitrogen, this is a pretty accurate description of what would become industrial farming nearly a century later.

Experiments and measures by other scientists demonstrated that atmospheric N deposition was much less important than assumed by Liebig. More importantly, Rothamsted experiments disproved Liebig disdain for nitrogen. While it was already known that manure application greatly improved yield (more than x2 particular in that case), mineral fertilization experiments enabled to disentangle the effect of the different nutrients (Figure 2.14). Fertilization with all nutrient except N was barely different than no fertilization at all, while mineral fertilization with N only was significantly higher, but still way less than manure. Only the combination of all

nutrients could reach the yields obtained with manure, illustrating Liebig's *law of the minimum* (actually first stated by Carl Philipp Sprengel), stating that growth is limited by the scarcest nutrient. But contrary to Liebig's belief, nitrogen was the limiting nutrient, only then followed by phosphorus.

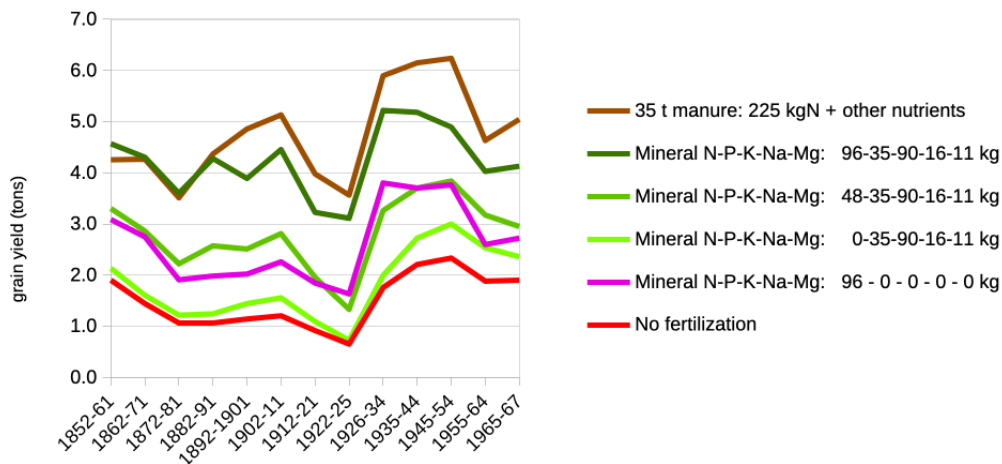


Figure 2.14: Effect of N and P mineral fertilization in the long term Broadbalk wheat experiment in Rothamsted, 1850s-1960s. All quantities are expressed by hectare. Source: adapted from Rothamsted Research (2023)

Jean-Baptiste Boussingault first noticed in crop rotations experiments that nitrogen content in soils seemed to increase following the inclusion of legumes such as pea. This was backed by Lawes' long-term experiments in Rothamsted, England. But controlled experiments in sealed jars, which previously demonstrated the effect of mineral nutrients, failed this time. In these pre-Pasteur times, they could not conceive that this controlled experiment probably killed the symbiotic micro-organisms that could precisely fix the atmospheric dinitrogen.

But the discovery of microbes by Pasteur during the 1860s opened new perspectives. Finally it will be at the end of the XIXth century, that the Germans Hellriegel and Wilfarth will definitively prove that leguminous crops enhance fixation of atmospheric N₂. Micro-organisms role in nitrification and denitrification would also soon be established. The reality of micro-organisms role would also revolutionize the understanding of mineralization, which does not simply occur by decaying physico-chemical reactions, but by active micro-organisms decomposition, paving the way for the emergence of organic agriculture concepts in the early XXth century

2.2.3 Exploitation of N deposits: Peruvian guano, Chilean nitrates, and coal gasification and coking¹⁰

I already described the exploitation of guanos for their phosphorus content in Section 2.1.1.3, and this also holds for their N content (see the legend of Figure 2.17 for some order of magnitude of their composition). While the first imports to Britain began in the 1820s, 50 years later the South American deposits were almost depleted. From 1840 to 1880, 20 Mtons of Peruvian guano were exported to the US and Europe (Gorman 2013c). Colonial nations then turned to another South American deposits: Chilean Sodium Nitrates.



Figure 2.15: With the concurrence of synthetic N fertilizers, Chilean nitrates merchants turned to new marketing strategies, for instance here advertising the "natural" origin of Chilean nitrate, around 1900. Source: Biblioteca Nacional de Chile (2024).

Chilean nitrates were formed when salts in sea sprays were deposited and evaporated in the extremely arid Atacama desert. The economic potential of this resource drove the War of the Pacific (1879–1884), also named the Nitrate War, between, on the one side, Chile, and on the other side, Peru and Bolivia. Backed by the British, the Chilean won, and gained control of the deposits. The help of the British Empire

¹⁰Elements in this chapter come mainly from Gorman (2013b), Gorman (2013c), and Smil (2004c)

was not uninterested: in 1890, 70% of the Chilean nitrates exports were aimed at the United Kingdom (642 out of 927 ktons, Gorman (2013c)).

The nitrate demand grew more and by the early XXth century, it was used for agricultural purpose but also for multiple industrial use: dyeing, metal alloys, photographs... But most importantly, explosives. As in the Renaissance with saltpeter, even the most recent explosives such as nitroglycerin still relied on nitrates derivatives. Explosives were mainly used by mining and railway companies, but were obviously also a concern for nations heading towards industrialized war.

In the early XXth century, estimations of Chilean nitrates deposits were around 240 million tons (Gorman 2013c). This was much more than the 20 Mtons of guano exported during the XIXth century¹¹. But this remained a finite deposit. This concern was retranscribed by Sir William Crookes' 1898 speech to chemists at the British Association for the Advancement of Science, calling to find a way to fix the nitrogen from the atmosphere, out of concern for European wheat production.



Figure 2.16: Coal gasification, in which ammonia sulfate was extracted from coal. This was performed in gasworks (*"usines à gaz"* in French), where town gas (*"gaz de ville"*), also named coal gas, was produced before fossil deposits of methane were exploited. Source: *Drawing the retorts at the Great Gas Works at Brick Lane*, unknown author, 1821, from Wikimedia Commons (2020)

Finally, a last exploitation concerned not directly N deposit but fossil resource containing nitrogen. Nitrogen content in coal is about 1-1.5% (Smil 2004c). Coal also has other impurities such as Sulfur. Nitrogen could be recovered as ammonia sulfate, but mainly when coal was converted to gas through coal gasification. Coal

¹¹These quantities are not directly comparable, as Chilean nitrates have different N concentrations than guanos, which themselves knew decreasing concentrations during the time they were exploited. See Figure 2.17 for direct comparison in terms of N

gasification served to produce methane used as a lighting source. Indeed, in the late XIXth century, extraction of methane in fossil deposit was barely developed, and coal gasification was the major way to meet the demand for methane (hence the denomination of "natural" gas for the methane extracted in fossil deposits). Ammonia sulfate was a gasification by product. It was also extracted from coal turned into coke for metallurgical needs, namely to transform pig iron slag to steel. The disadvantage was that this ammonia sulfate was dependent on the constrained demand for coal-based manufactured gas and coke.

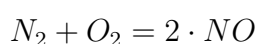
Only Chilean sodium nitrate could be significantly increased, but in the case of a War, its supply could be easily jeopardized with a naval blockade, especially for Germany with less access to the sea than France or England. This drove interest to find a way to directly fix N from the atmosphere and gain Independence.

2.2.4 The first quest for fixation of atmospheric N: cyanamide process and Norwegian electric arc

From the 1900s-1920s, several N fixation processes were being developed, but in the end, the Haber-Bosch process would prevail. These processes include the Norwegian process arc and the Cyanamide process. They are shown in Figure 2.17, alongside with the exploitation of guano, Chilean nitrates deposits, and extraction of nitrogen from coal gasification or coking.

2.2.4.1 Norwegian arc process

Already in the early XVIIIth century, scientists knew how to produce electric arcs and understood that oxidized forms of nitrogen could be produced by this means out of the atmospheric N₂ and O₂. However this was an energy intensive process, as it requires temperatures above 3000°C to produce significant amounts. Only after 100 years of improvement did the first industrial facilities emerge in Norway, in 1903, to produce nitric acid (Leigh 2004a). Even then, it required a temperature of around 2000°C with electric arcs, which could be attained thanks to Norway's large fjords and its abundant hydroelectricity. The first tests were made with an electric power of 3 kW, but soon reached 4 MW in 1905. In 1915, the Norwegian Rjukan–Notodden Industrial had a capacity of 400 MW, an enormous amount for that time (Smil 2004c).



But the Norwegian arc process emerged around the same time as the Haber-Bosch process, which, even then, consumed about 25% less energy (Leigh 2004a). By 1920,

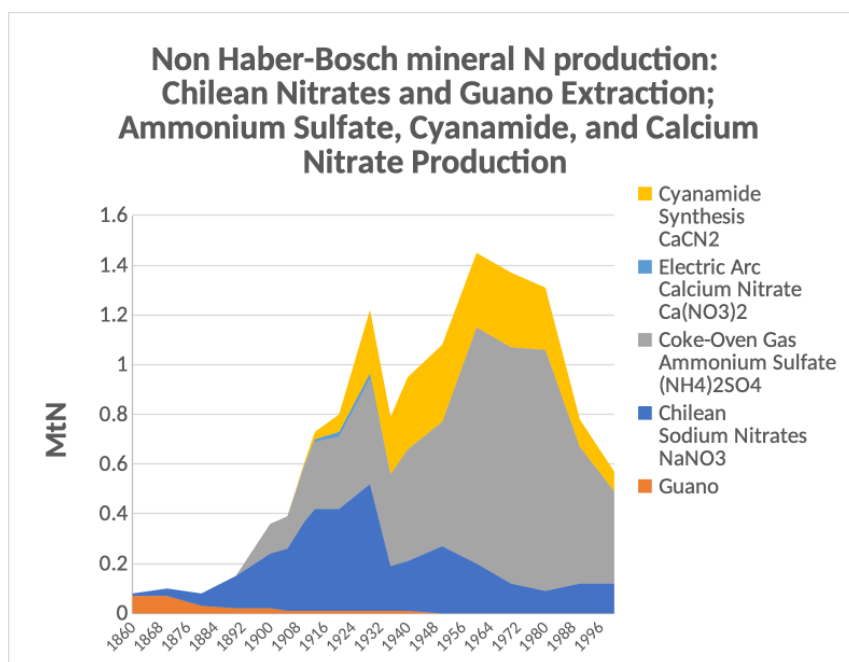


Figure 2.17: Production of the main sources of mineral N, excepted the Haber-Bosch process. Source: Smil (2004a) Appendix F. Smil used the following conversion rates to express all quantities in nitrogen: NaNO_3 , 15% N; guano, 12% N in 1860 and 1870, 7% in 1880, 4% in 1890, 3% in 1900, 2% in 1910–1920, 1% in 1930 and 1940; $(\text{NH}_4^+)_2\text{SO}_4$, 25% N; CaCN_2 , 20% N.

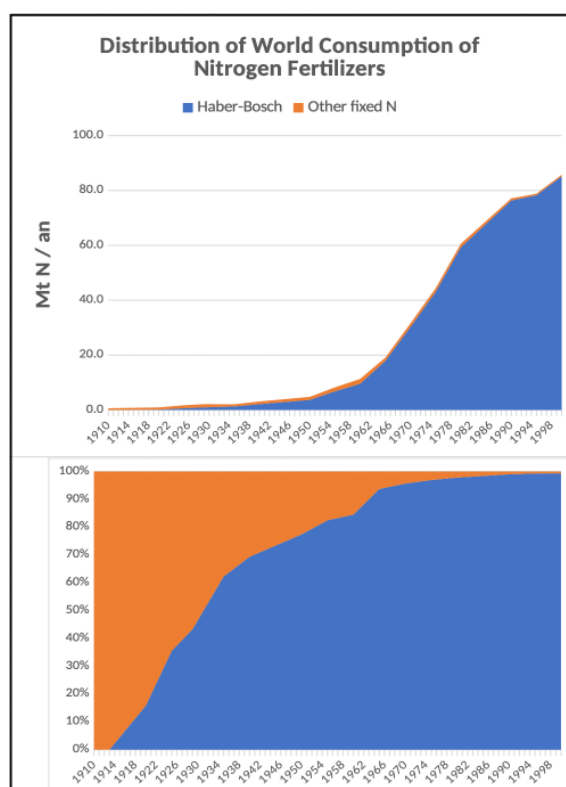
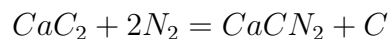
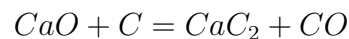


Figure 2.18: Global consumption of mineral N fertilizers, discriminated between Haber-Bosch and non Haber-Bosch. Source: Smil (2004a), Appendix L

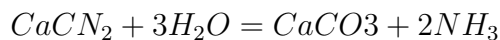
it was clear that the Haber-Bosch process was much more efficient (Table 2.4). By 1930, this process was abandoned, especially after World War I demonstrated the reliability of the Haber-Bosch process to produce nitrates for the German army.

2.2.4.2 Cyanamide process

The reader can find more details about the cyanamide process in Gorman (2013f) and Smil (2004c). The cyanamide process, also named Frank–Caro process after the ones who developed it, uses calcium carbide (CaC_2) reacting with atmospheric N_2 in high temperature ovens at around 1000°C , to produce calcium cyanamide (CaCN_2). But actually the critical and energy-intensive step is the preliminary synthesis of calcium carbide itself, from lime (CaO) and coke (a source of C) in electric arc furnace at more than 2000°C .



Calcium cyanamide (CaCN_2) could be used raw as a fertilizer in the form of powder, but could also be reacted with superheated steam to produce ammonia:



Even though the cyanamide process was more energy-intensive than the Haber Bosh-process (Table 2.4), it was still way more efficient than the Norwegian arc.

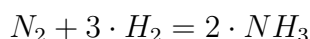
For these reason, this process lasted throughout the XXth century (Figure 2.17), but its production was nonetheless without measure compared to the Haber-Bosch process.

Table 2.4: Energy efficiency of the different N fixation technologies. Source: Smil (2004c) and its appendix K.

Fixation method	Development Stage	GJ/tN
Norwegian arc	early process	700
	later optimized	180-270
Cyanamide	early process	200
	later optimized	130
Haber-Bosch	coal-based	80-100
	methane-based	20-50

2.2.5 The Haber Bosch process and the fixation industry: XX-XXIth century¹²

Numerous attempts at synthesizing NH_3 occurred during the XIXth, century, but it finally succeeded only in 1909 with the Haber-Bosch patent (Smil 2004f). The reaction equation seems pretty straightforward:



But the impediments lied in: the high temperatures ($>500^\circ\text{C}$); high pressure (300-350 atmospheric pressure); a long search for the right catalyst enabling to decrease temperature and increase yield; finding a source for H_2 production; and getting rid of oxygen which could prevent the reducing process.

In 1910 Oppau became the first full scale factory for the Haber-Bosch process. Increasing ammonia plants production capacities became a very concrete industrial challenge during World War I, when the naval blockade prevented Germany from importing the Chilean nitrates essential to produce its gunpowder. Without a solution, Germany would be outpaced. By the end of the war, thanks to its new military industrial complex, most of its nitrates originated from Haber-Bosch factories, prolonging the sufferings of the war. War was a particularly efficient source of selection pressure, and without it, Chilean nitrates may have stayed competitive for some decades more; instead, their production peaked in the 1920s (Gorman 2013c). In 1918, getting the Haber-Bosch process was a key demand of the *Traité de Versailles* by France and England, to ensure their future military power.

During the 1930s, about half of the world capacity of NH_3 production was still in Germany (Gorman 2013f), but facilities began to be emerge in the US or in France, with the infamous AZF plant which blew up in 2001, resulting in 31 deaths. There have been dozens of ammonitrates silos explosions throughout the century, the first one as soon as 1921 in Oppau, killing more than 500 and destroying the homes of more than 7,000 (Smil 2004g); the latest in Beyrouth in 2020, killing more than 200 and injuring more than 6,000. WWII further pushed for increasing NH_3 production capacities, namely in the US, to support the war effort of the Allies (Gorman 2013f). Today there are more than 400 plants in the world.

During much of the first half of the XXth century H_2 feedstock was inefficiently obtained from coal. Switching to natural gas made it way more efficient, and allowed to use methane as both an H_2 and thermal energy provider. Smil (2004d) estimated that in the late 1990s, the best performances reached about 25 GJ/ton of fixed N, about 20% above the theoretical minimal, leaving little room for further

¹²The elements detailed in this subsection are mainly from Leigh (2004e), Smil (2004d), and Smil (2004e)

improvements with the current reactants. But as many facilities around the world continued to use coal (namely in China) and were not state-of-the-art methane based factories, the global mean was 40-45 GJ/ton of N, which is still half the value of the 1950s (Figure 2.19). A equivalent rough approximation is that to fix 1 ton of N, one needs around 1 ton of oil equivalent (= 42 GJ). This is way more than the energy require for other fertilizers. Smil (2004d) reports values of 15 GJ per ton of P (up to 30 for complex compounds) and 19 GJ per ton of K. NH_3 fixation consumes about 1 to 2% of the global energy consumption (Sutton et al. 2013).

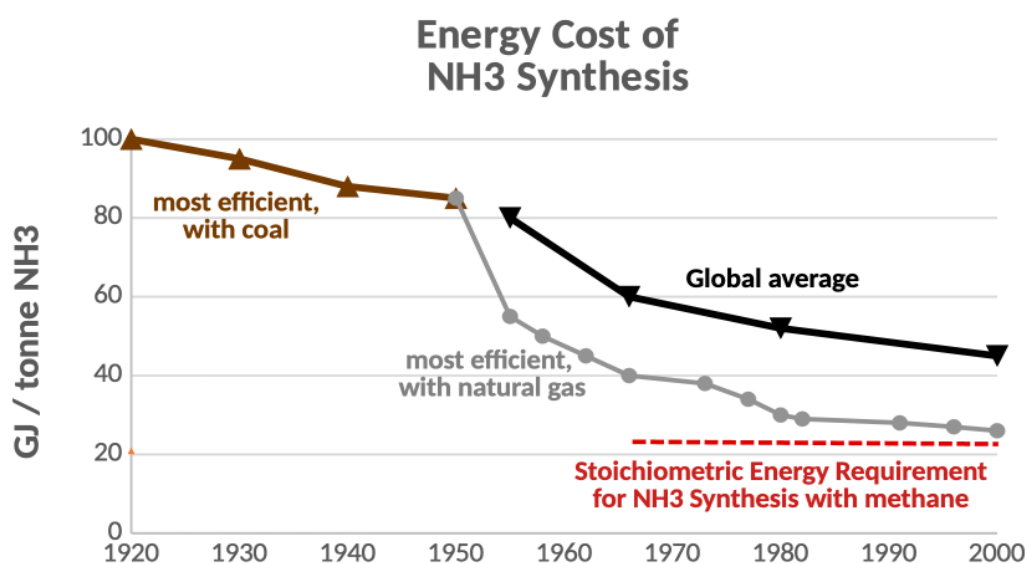
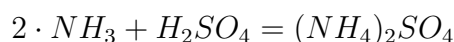
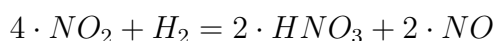
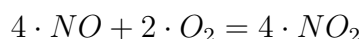


Figure 2.19: Evolution of the energy efficiency of ammonia fixation with the Haber-Bosch process. Source: Smil (2004a) Appendix K, and Smil (2004d)

From the few kgN fixed every day by early plants in the 1910s, multi-100 million \$ modern facilities now fix on the order of 2,000 kg a day. After NH_3 fixation, it still has to be transformed to be adapted to its future uses. A first option is to keep it under its reduced form, as ammonium sulfate, for fertilizer use. The reaction involves sulfuric acid:

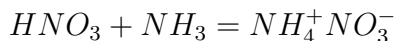


Another option lies in the oxidized form, as nitric acid:



A supplementary step yields ammo-nitrates, which can then be further transformed

to fertilizer, but whose other application can be explosives:



During almost 50 years, from the mid 1920s to the mid 1970s, ammonia fixation has experienced a steady exponential increase (Figure 2.21), stopped only during the late 80s. For more than a decade, it plateaued around 100 MtN, but drastically increased after 2000, mainly due to China, reaching more than 140 Mt in 2022 according to the USGS (2022), while FAOSTAT (2023a) only reports 120 MtN. I could not identify the reason for the discrepancy

The production is much less concentrated in a handful of countries than phosphate production, but still skewed (Figure 2.20). About one third happens in China. Adding Russia, the United States and India represents a bit more than 50% of the global production. NH_3 prices are closely correlated to oil and gas global market prices, with important spikes in the 70s oil crises and in 2022 following the post-COVID inflation and Russian invasion of Ukraine (Figure 2.21).

Nowadays, more than half of the proteins eaten by humans are obtained in the first place from nitrogen generated by the Haber–Bosch process (Erisman et al. 2008; Smil 2004a). But this rate could certainly be lower if the food system was less wasteful, reducing the inputs needs, which could not be met in the current food system without the Haber-Bosch process.

2.2.6 A XXth century unfulfilled promise: the quest for mastered biofixation

Until this subsection, the history of mastering N fixation by humans has been a story of chemistry and physics, while in nature this is mainly a biological process. The contrast is stark: while the (optimized) Haber-Bosch process requires pressures 350 times the atmospheric pressure and several hundred degrees Celsius, in facilities of several 100 millions of dollars each, fixating micro-organisms operate at ambient pressure and temperature (Leigh 2004a). Even if biological fixation is relatively intensive compared to other metabolic processes (need for 8 ATP to fix 1 N, see Section 1.2.2), this is still less than human-made fixation¹³.

This was recognized early by Haber himself in his Nobel Prize speech: *“It may be that this solution is not the final one. Nitrogen bacteria teach us that Nature, with her sophisticated forms of the chemistry of living matter, still understands and*

¹³The hydrolysis of ATP to ADP has a Gibbs free energy of 30.5 kJ·mol⁻¹ of ATP, so the hydrolysis of 8 ATP gives 244 kJ·mol⁻¹ of NH_3 . With a NH_3 molar mass of 17 g·mol⁻¹, this is 14.4 kJ·g⁻¹ of NH_3 , or 14.4 GJ·ton⁻¹ of NH_3 . Compare this with the ~20 GJ·ton⁻¹ of NH_3 for the Haber-Bosch process in Figure 2.19

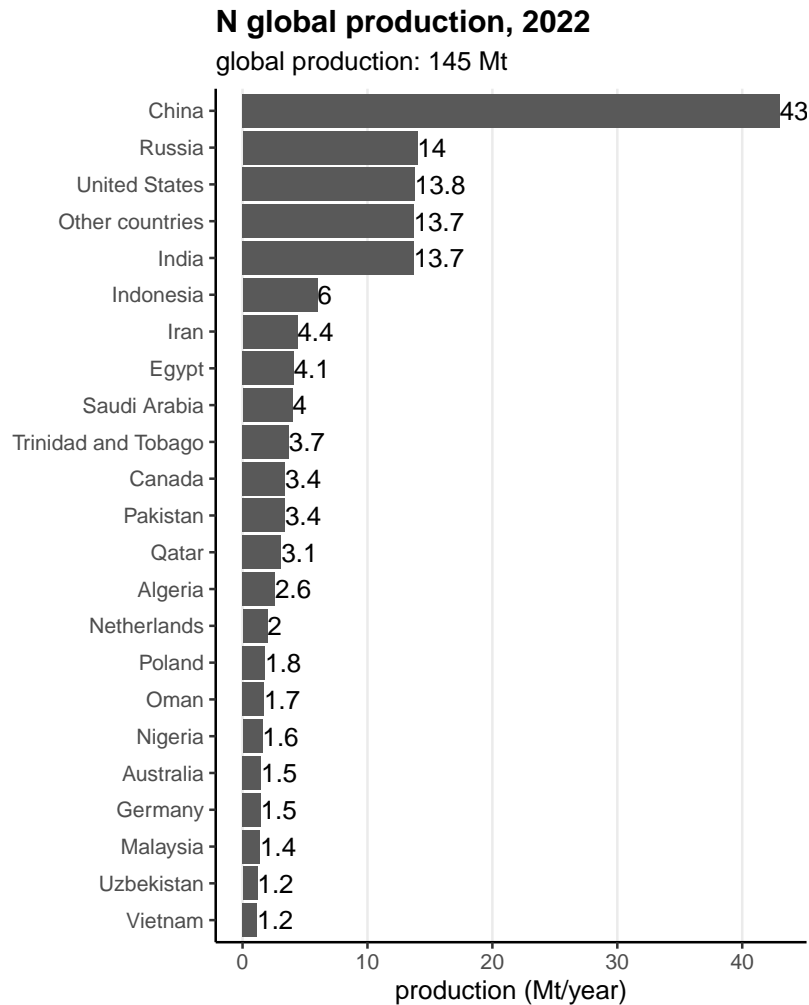


Figure 2.20: Production of N fixed by the Haber-Bosch process in each country for the year 2022. USGS (2022)

utilizes methods which we do not as yet know how to imitate.”

Post-World War II progresses in genetics gave hope to a quick mastering of biofixation. In the 1960s the agri-food company Dupont extracted the enzymes responsible for fixation, the nitrogenases. Early successes were made in transferring the genes coding for nitrogen fixation to *E. coli* bacteria. But then scientists did not manage to ensure that plants transferred with the gene could perform biofixation (Leigh 2004f). This is because it requires specific conditions to operate: some steps in the reaction demand anaerobic condition, while photosynthesis precisely produces oxygen.

Research is still ongoing, but the hype went down. As Leigh (2004a) puts it: *“In 1987, The Times of London was making optimistic reports about the possibility of making nitrogen-fixing plants, but today one can no longer whisper the magic words “nitrogen fixation” to granting bodies and expect to be funded.”*

While breeding N-fixing cereals could diminish farmer’s dependence on mineral

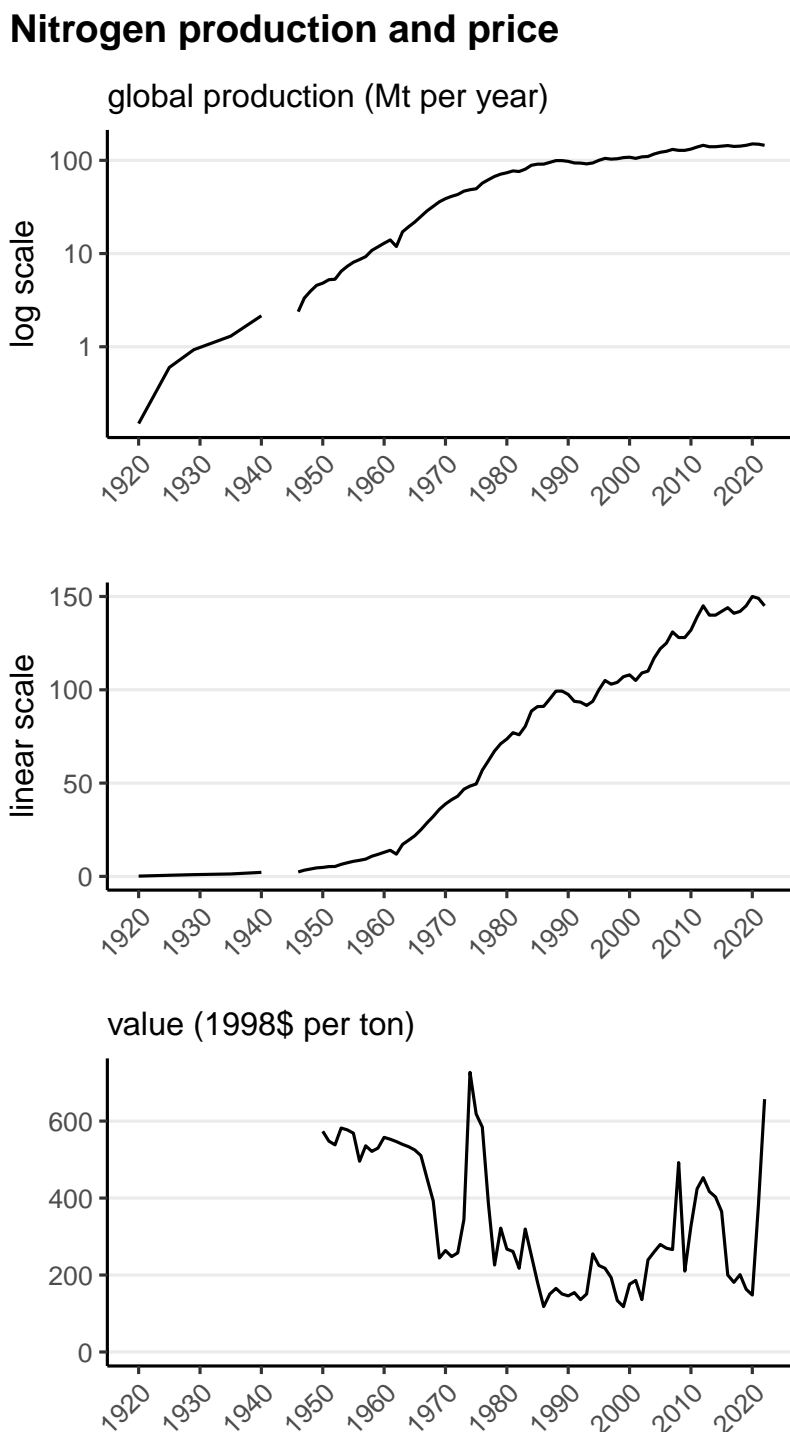


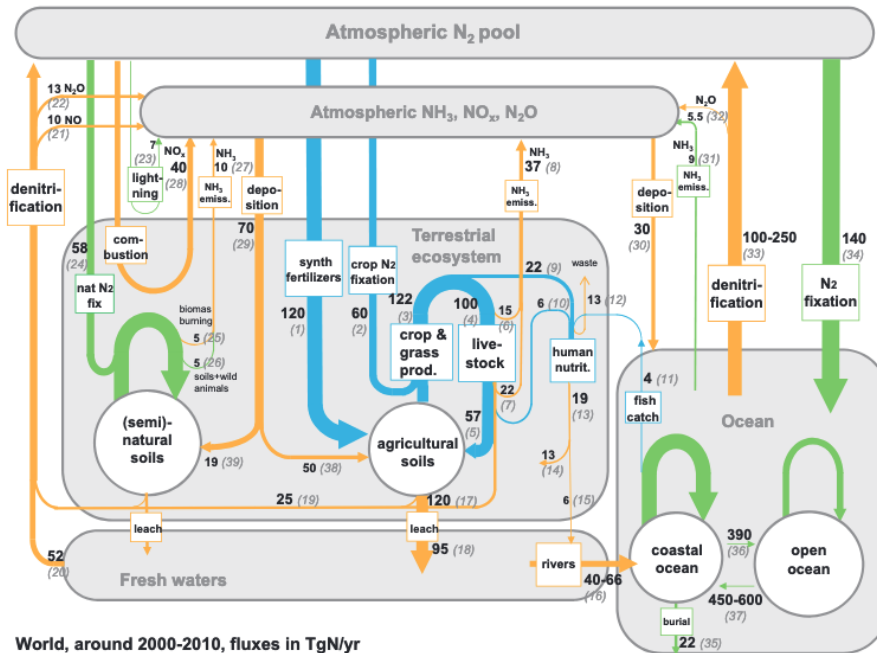
Figure 2.21: Historical N fixation from the Haber-Bosch process, and prices in 1998 constant dollar. Source: USGS (2024b). Before 1945, production values are from Smil (2004a), appendix L.

fertilizers, this would probably simply replace it by a reliance on patented plants, and would not benefit the majority of the world's farmers who may not afford it. Moreover, in many regions of the world, the problem is not so much about too little nitrogen, but too much. Before the industrial revolution, global N fixation flows in

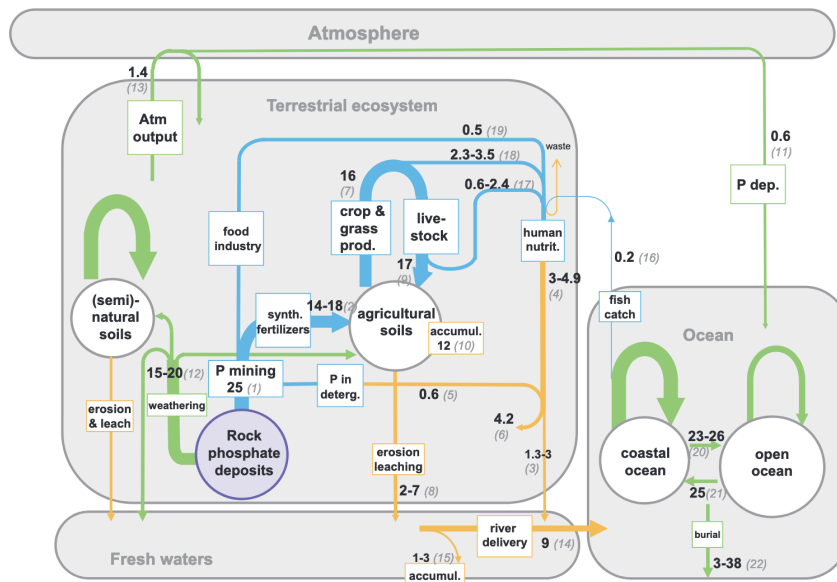
nature were about 100 MtN per year. At the start of the XXth century, human-induced fixation was negligible in front of this quantity, below 1 MtN (Figure 2.17). But now anthropic fixation is roughly of the same order of magnitude (Galloway et al. 2004). Questions and alerts were raised as soon as the 70s, for instance when a popularized article presented a global N budget in *Scientific American* (Delwiche 1970) suggesting that industrial fixation doubled the natural flow. In the past, denitrifying bacteria would compensate new fixation and send back to the atmosphere all this reactive N. But what if they could not keep up with the enhanced flows ? Where would all that reactive nitrogen go ?

2.3 Consequence: N and P disturbed global flows

2.3.1 N and P global flows



(a) Global N flows, around 2000-2010, in MtN per year



(b) Global P flows, around 2000-2010, in MtP per year

Figure 2.22: Global nitrogen and phosphorus flows in the years 2000-2010. Source: Sutton et al. (2013)

The global N and P flows in the years 2000-2010 have been synthesized by Sutton et al. (2013). On Figure 2.22, it is striking that the N cycle is much more complex

than the P cycle, reflecting the many more N forms and transformations, namely gaseous ones, compared to P forms.

As these nutrients are mostly (>80%) used for food production, the concept of Nitrogen Use Efficiency (NUE) and Phosphorus Use Efficiency (PUE) has been developed: out of 100 units of new nutrient inputs to the agricultural system, how many actually ends up ingested by humans ?

A simple synthesis of the nutrient chain for phosphorus, allowing a clear visualization of its full chain P use efficiency, is presented in Figure 2.23 (from Cordell (2010)) for the early 2000s. Depending on whether one only considers P fertilizers applied to croplands (14 MtP) or the P mined before mining and beneficiation losses (17.5 MtP), the 3 MtP ingested by humans translate into a 17-21% full-chain P use efficiency, consistent with the 12-20% reported in Sutton et al. (2013). Most of the losses occurred as soil erosion.

However there are caveats to this interpretation. First, most of the P lost along that chain does not actually ends up in the environment, but is rather stored in croplands soils, and can be remobilized later (a phenomenon labeled "P legacy" (Sharpley et al. 2013)). If we do not consider this stored P as lost, then the full-chain P use efficiency can increase up to 75% (Sutton et al. 2013).

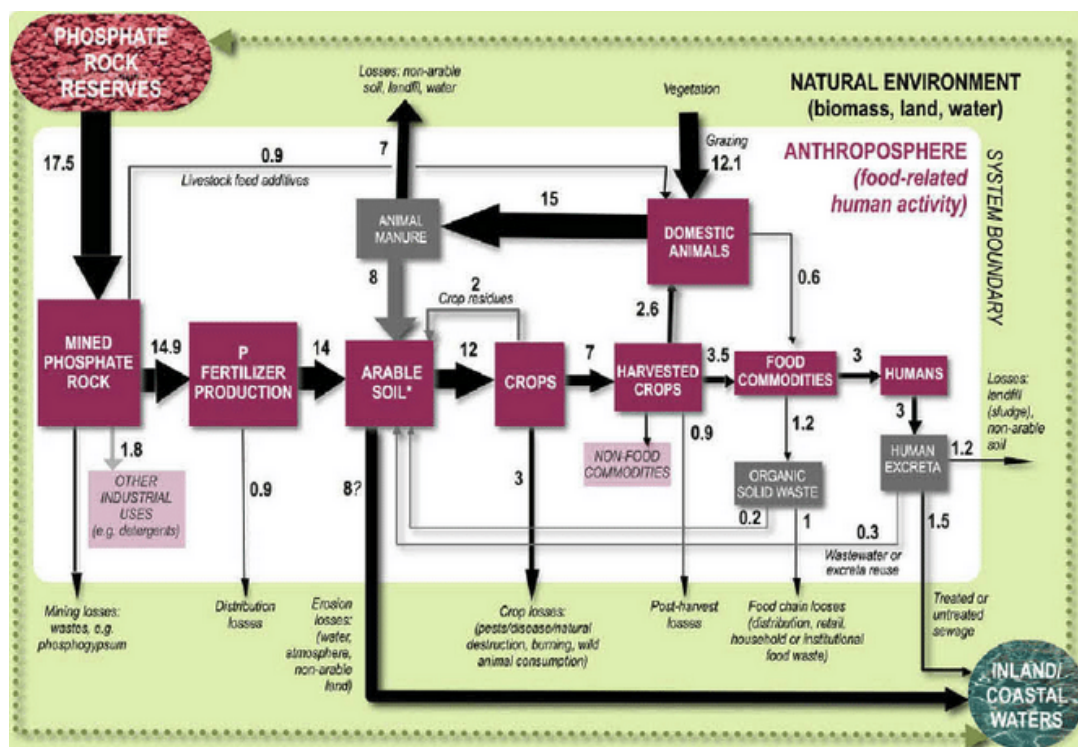


Figure 2.23: Simplified P flows across the food system chain, in the early 2000s. Flows in MtP per year. Source: Cordell (2010). For more detailed and recent assessment, see Scholz et al. (2014).

Second, a global nutrient use efficiency hides major heterogeneity. The most

important source of variation lies in the diet. While the P use efficiency is around 50% for crops, it is only 4-15% for animal products. Sutton et al. (2013) thus reports that globally 80% of the N and P in harvested crops and grass serves to feed livestock, and will finally account only for 15 to 35% of the N and P intakes by humans. Consequently, the doubling in global meat production since the 1960s (Erisman et al. 2008) has deeply impacted the global nutrient flows. Countries with lower animal products intakes will have a better nutrient use efficiency, and thus lower losses. In Europe, about half of the protein intakes come from animal products (excluding fish), compared to 25-30% at the global scale; this adds to the fact that Europeans consumed 70% more proteins than nutritional recommendations.

As for nitrogen, the inputs in 2000-2010 were made of 120 MtN of synthetic fertilizers, and 33 MtN of legumes biological fixation (Sutton et al. 2013). The difference with phosphorus is that about 30-40 MtN of reactive N created by humans comes from fossil fuel combustion, resulting in accidental fixation, and part of it will fertilize croplands following atmospheric deposition. Considering the 19 MtN of human intakes, this translates into a approximate 10% full-chain NUE; but this figure accounts for all the NO_x accidentally fixed by combustion, and incorporates all grasslands, while they are not fully anthropized ecosystems. These 2 factors decrease the full-chain NUE, and I will come back in more details on that matter in Chapter 5. The main N losses estimated by Sutton et al. (2013) were N leaching (95 MtN); NH_3 emissions (37 MtN), mainly from manure; and denitrification (25 MtN), but with large uncertainties.

Building on the European Nitrogen Assessment also allows to compare situations with the global average. Sutton et al. (2013) report crops NUE to be 43% at the global scale vs 58% for Europe; concerning livestock, the respective figures are 6% and 19%. This shows that even though technologies and agronomic practices can improve crops or livestock NUE and thus reduce the respective losses, the major factor affecting the full-chain NUE and losses remains the diet, as even the best meat NUE remains below the worst crops NUE.

With an historical perspective, Sutton et al. (2013) conclude that *"humans have more than doubled global land-based cycling of nitrogen and phosphorus"*. This can be visually represented for nitrogen in Figure 2.24. Consequently, in places where N inputs are excessive, it appears that denitrification of reactive N to inert N_2 cannot keep up, leading to the accumulation of reactive N, referred as the "nitrogen cascade" (Galloway et al. 2004; Galloway et al. 2003).

Based on this simple indicator of global new reactive N inputs, some have tried to define a threshold, named planetary boundary, which would prevent the adverse effects of excess nitrogen.

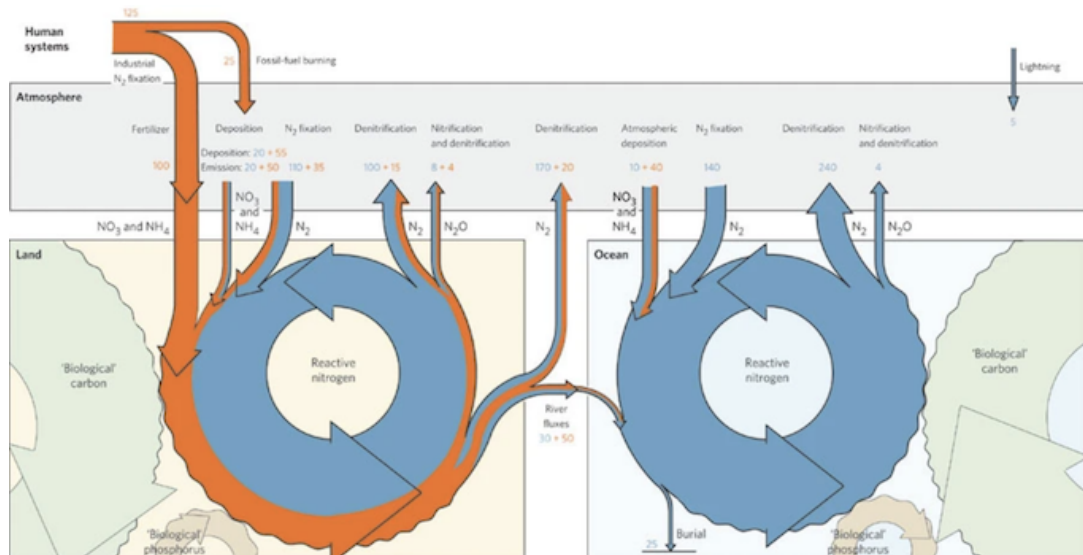


Figure 2.24: Anthropogenic N inputs have more than double the global land-based cycling of nitrogen. Left: terrestrial ecosystems N cycle; right: oceanic N cycle. Orange: human-induced N fixation; blue: natural flows. The diagram also represents the coupling of the N cycle with C and P cycles. Source: Gruber et al. (2008b)

2.3.2 The (moving) nitrogen planetary boundary framework

The concept of planetary boundaries emerged in the late 2000s. Remarking that biophysical variables of the Earth system (CO₂, ozone, nitrogen, phosphorus, share of land dedicated to agriculture...) have been drastically altered by human activities, some authors attempted to determine a safe operating space for the Earth system (Rockström et al. 2009b; Rockström et al. 2009a). Their exact question was *“What are the non-negotiable planetary preconditions that humanity needs to respect in order to avoid the risk of deleterious or even catastrophic environmental change at continental to global scales?”*.

In the following I particularly focus the evolution of the nitrogen boundary. The major disturbance of this element planetary flows was noticed way before the emergence of the concept of the planetary boundary: *“Of all man’s recent interventions in the cycles of nature the industrial fixation of nitrogen far exceeds all the others in magnitude.”* (Delwiche 1970).

2.3.2.1 The seminal 2009 article

While Rockström et al. (2009b) insisted that it is inevitable that the safe thresholds incorporate a normative judgment on how to deal with risk, they also state that *“The thresholds in key Earth System processes exist irrespective of peoples’ preferences, values, or compromises based on political and socioeconomic feasibility”*

Despite this ambitious goal, their actual N boundary was arbitrarily fixed as a

”first guess”. They recommended to limit industrial (Haber-Bosch) and agricultural legumes biofixation of N_2 to $35 \text{ MtN}\cdot\text{year}^{-1}$. The rationale behind this was to limit anthropogenic N_r inputs to one quarter (25%–35%) of what is naturally fixed by terrestrial ecosystems.

The problem with this $35 \text{ MtN}\cdot\text{year}^{-1}$ threshold is that it makes it virtually impossible to feed 10 billion people, a global population plausible within the century. Even if global N intakes are decreased to $3 \text{ kgN}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$, the lower bound of what is recommended by health agencies (see Section 1.1.3), the total food system N use efficiency would have to be somewhere between 50% and 100%, depending on the share of N in human excreta reused in agriculture, if we authorize $30 \text{ MtN}\cdot\text{year}^{-1}$ of anthropogenic fixation, to keep a safe margin from the $35 \text{ MtN}\cdot\text{year}^{-1}$ threshold. (see Table 2.5)¹⁴.

Table 2.5: Required N Use Efficiency (NUE) to feed 10 billion people with $3 \text{ kgN}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$ (i.e. $30 \text{ MtN}\cdot\text{year}^{-1}$) when only $30 \text{ MtN}\cdot\text{year}^{-1}$ of new intentional fixation are authorized in order to comply with the $35 \text{ MtN}\cdot\text{year}^{-1}$ planetary boundary. Results are shown for different recycling rates of N in human excreta.

recycling from the 30 MtN·yr ⁻¹ in human excreta	inputs from excreta	total inputs with 30 MtN·yr ⁻¹ of intentional fixed N	minimal system NUE to produce 30 MtN of food
0%	0 MtN·yr ⁻¹	30 MtN·yr ⁻¹	100%
25%	8 MtN·yr ⁻¹	38 MtN·yr ⁻¹	80%
50%	15 MtN·yr ⁻¹	45 MtN·yr ⁻¹	66%
80%	24 MtN·yr ⁻¹	54 MtN·yr ⁻¹	56%

While the current food system NUE is only about 15-20% because of its wastefulness, the gap is certainly too large to be closed: this requires high crop N use efficiency, an almost universal vegetarian/vegan diet, and an almost complete recycling of the few waste that remains, with almost no losses during storage and transport. This triggered the attention of researchers specialized in food systems, namely De Vries et al. (2013), who proposed a more realistic boundary.

2.3.2.2 2015: a large update.

De Vries et al. (2013) argued that the $35 \text{ MtN}\cdot\text{yr}^{-1}$ boundary was *”too low in view of needed N fixation and also unnecessary low in view of most environmental impacts”*. They first calculated global N boundaries based on critical threshold of the different

¹⁴The actual rate would be partially reduced as some of the proteins would come from fish, but this raises the question of potential over-fishing. Also biofixation from permanent grasslands should not be counted as it is not ”intentionally” fixed; see Section 5.2.3.3 for a discussion on the way to compute the role of permanent grasslands. On the other hand, this does not consider the required fixation for textiles, like cotton or flax

N_r forms and impacts, and translate them into their respective global N fixation threshold:

- for NH_3 : 1 to 3 $\mu\text{g}\cdot\text{m}^{-3}$, that they converted to a new input of 89-115 $\text{MtN}\cdot\text{year}^{-1}$.
- for N_2O : 1 to 2.6 $\text{W}\cdot\text{m}^{-2}$, that they converted to a new input of 20-133 $\text{MtN}\cdot\text{year}^{-1}$.
- for N in surface runoff: 1 to 2.5 $\text{mg}\cdot\text{l}^{-1}$, that they converted to a new input of 62-82 $\text{MtN}\cdot\text{year}^{-1}$.

As N_2O is related to climate change, which is a boundary on its own in the planetary boundary framework, De Vries et al. (2013) chose to focus only on the other 2 forms, and finally chose N in surface runoffs as it as the lowest boundary. The chosen boundary is thus 62 MtN with an uncertainty zone of 62-82 MtN. They find it compatible with the feeding of 9 billion people, which according to them is between 52-80 MtN depending on the food system NUE¹⁵.

The arguments and boundaries value set by De Vries et al. (2013) were then adopted by Steffen et al. (2015), resulting in an almost doubling of the previously set boundary. They nonetheless recognize that a more appropriate indicator should be the actual N leaching to freshwaters, as new N inputs do not necessarily end up as pollution: it could be directly denitrified as inert N_2 (naturally or in wastewater treatment plants) or simply build up the soil organic matter. But they chose the new N inputs as a practical indicator for policy making. They also recognized that the boundary may be larger if N inputs are optimally allocated around the globe.

In 2023, a planetary boundary article (Richardson et al. 2023) continued to adopt the methods and indicators followed in 2015 by Steffen et al. (2015).

2.3.2.3 2023: dropping an approximate indicator.

But a paper published a few months before (Rockström et al. 2023) took a radically different approach and focused not on inputs but rather on N surplus (i.e. cropland inputs minus removals from harvest), which is much more related to pollution, based on the work of Schulte-Uebbing et al. (2022). For comparability and as an indication, they translated this surplus in total inputs, given the current food system efficiency, and found a value of 143 $\text{MtN}\cdot\text{yr}^{-1}$; this means that with efficiency improvements compared to the current wasteful food system, this 143 $\text{MtN}\cdot\text{yr}^{-1}$ threshold could be further increased. It is not easy to compare these "total inputs" to the previous "intentionally fixed (Haber-Bosch + legumes biofixation) indicator, as it depends on the food system nutrient use efficiency.

¹⁵It seems to me that their estimation is still too low, because they included manure fertilization in the computing of the total NUE inputs, whereas it should be counted as an internal flow. See my detailed discussion on that matter in Section 5.2.3.3.

2.3.2.4 Re-localizing nitrogen planetary boundary

I gathered in Table 2.6 the evolution of the nitrogen planetary boundary thresholds. Note that out of consistency, I focused here only on the papers written by the Rockström research team, as he is at the head of the seminal 2009 paper and of the most influential Stockholm Resilience Center which developed the boundary framework. The center webpage (Stockholm Resilience Center 2024) particularly features the Rockström et al. (2009b), Steffen et al. (2015), and Richardson et al. (2023) papers, and I also included the Rockström et al. (2023) paper. It appears that the initial 2009 values as been expanded by a factor ~ 2 in 2015 and even ~ 4 in 2023 if we consider that the total inputs are comparable to the intentionally fixed N.

Table 2.6: Evolution of proposed planetary boundaries for Nitrogen.

source	indicator	boundary value (MtN)
Rockström et al. (2009b)	industrial and legumes fixation	35
Steffen et al. (2015)	industrial and legumes fixation	62 (62-82)
Richardson et al. (2023)	industrial and legumes fixation	62 (62-82)
Rockström et al. (2023)	N surplus	61 (35-84)
Rockström et al. (2023)	total N inputs (indicative, with current NUE)	143 (87-189)

The consecutive expansions of the boundary values shed light on the fact that global and uniform values seem uncertain and non robust. Also, it warrants caution before setting thresholds "irrespective of peoples' preferences, values, or compromises based on political and socioeconomic feasibility", since it appeared that the 2009 threshold was incompatible with the aim of feeding humanity. We shall see in Section 5.3.4.2 that the 62 MtN boundary set in Steffen et al. (2015) and Richardson et al. (2023) also appears to be very restrictive, and published agro-ecological scenarios exceed their thresholds by an important margin.

As stated as soon as 2009, in an accompanying editorial for the release of the first planetary boundary paper (Nature 2009), some of the proposed planetary boundaries (of which nitrogen) have inherently local impacts, and not global ones, contrary to climate change where greenhouse gases are quickly homogenized in the global atmosphere. Maybe a better alternative is to actually monitor and model the local impacts of nitrogen. For instance Gerten et al. (2020) modeled the N concentration (critical threshold of 1-3 mgN/l) on a 0.5° grid cell level, based on current fertilization inputs and practices. This approach particularly highlighted the areas where inputs can be increased (sub-Saharan Africa) and where it could be decreased (East China, India, Europe..). This spatially explicit approach has also been adopted by

Schulte-Uebbing et al. (2022), which served as a basis for Rockström et al. (2023) to determine their new planetary boundary based on N surplus rather than on N inputs.

Chapter 3

Handling Human Excreta

Here I first offer a brief glimpse of the way human excretions have been handled before modern times, and especially how human excretions management in dense cities diverged between Europe (Paris and London) and Asia (Japan's Edo and China) (Section 3.1). Then in Section 3.2 I more specifically focus on the current system, in industrialized countries, based on wastewater treatment plants, and their potential future evolution, namely regarding nutrients recovery. Finally in Section 3.3 I address the topic of source separation of human excretions.

3.1 Historical aspects regarding handling of excretions

Here I present a brief summary of the way human excretions have been handled across the ages, and particularly how they were used (or not) to fertilize crops. The reader looking for more details, in the context of France, can refer to Barles (2005), Esculier (2018), Adler (2020), and Dufour (2024)

One of the oldest form of underground sewers and drainage system can be found during the Neolithic. For instance, Harappa/Indus valley civilization, around 3300-1300 BCE, had a main sewer 1.5 meters deep and 91 cm across (Khan et al. 2014). Sewers could also be found in Minoan Crete, and more generally in the Hellenic world, more than 3,000 years old (Yannopoulos et al. 2017), with some forms of primitive flushing toilets. Besides sewers, there are evidence of domestic wastewater used to fertilize crops in the Neolithic Mesopotamia, the Indus valley, and Minoan (Angelakis et al. 2015). In antic Rome, the city was crossed by the Cloaca Maxima, a major sewer evacuating waste into the Tiber. The Roman emperor Vespasian famously implemented a tax on urine, which was used as a fixing agent for dyes. In Paris, public toilet were named after this emperor ("*les vespasiennes*").

Roman authors like Pliny the Elder Natural History, wrote on the ways to handle

human excreta as manure (Ferguson 2014), though it is not evident from this source how widespread this practice was. It is difficult to know what was the fate of excretions in rural areas during much of history. One can speculate that a non negligible part was excreted in the field, or mixed up with animal manure, where they would fertilize crops. But in French cities, during the Middle Ages, excretions were often simply poured onto the street, even though since 1374 a Parisian law forced inhabitants to have latrines and later a septic tank (Ferguson 2014). Men would collect these during the night, or they would be washed by runoffs.

However, with the industrial revolution and urbanization, this already inadequate urban Western system would reach its limits: during the XIXth century, London population exceeded 2 million inhabitants, and more than 1 million for Paris. Interestingly, in Japan, Edo (now Tokyo) reached the 1 million threshold during the XVIIIth. Yet Japan and China took a complete different path than their French and English contemporaries when it came to handle human excretions in large urban centers

3.1.1 Western capitals in the XIXth and early XXth century: the choice of sewers

3.1.1.1 Before sewers

Underground cesspools before buildings began to replace chamber pots emptied to the street in Paris. These cesspools required regular emptying, usually by night, by *vidangeurs* in Paris or dustmen in London. It was not uncommon for the content to be discharged in the Seine (Ferguson 2014).

During the XIXth century, a "fruitful mutualism" was being developed between some French cities and the surrounding farmland (Esculier et al. 2020). Usually, excretions (feces and urine) were collected in cesspools in front of or behind buildings. Many patents on processes to treat cesspools nightsoil into fertilizers were developed during that period, for instance regarding ammonium sulfate or struvite production (Adler 2020). In Paris by the end of the XIXth century, collected nightsoil had to be treated in a single facility in Montfaucon, where it was dried and transformed to the fertilizing matter "poudrette". Yet, despite good intentions, many (90%) of the N was lost through infiltration or volatilization (Esculier et al. 2020).

Contrary to the volume reductions methods adopted in Paris, Flemish farmers in Northern France directly applied cesspools content and urine onto farmland (Ferguson 2014; Mathey 2019), even though they also practiced volumes reduction through composting. Another notable difference with Paris was that there, farmers were paying landlords to get their excretions. This illustrates the diversity of economic models, depending on the viewpoint adopted: is the person excreting pro-

viding the farmer with fertilizers, or is the farmer taking unhealthy matter away from the city ? In Lille, Mathey (2019) estimates that throughout the XIXth century about 50% of human excretions were returned to agriculture, with substantial uncertainties (25-75%).



Figure 3.1: Flemish spreading. Source: Mathey (2019), from Girardin (1876)

Meanwhile in London, the system struggled to handle the quantities associated with increased urbanization. George (2011a) reports that cesspool emptying reached a shilling, twice a labourer's daily wage. Moreover, watertightness of cesspools was still uncommon during the first half of the XIXth century, leading to leaks to groundwaters and feecal contaminations. People used only a few liters of water per day, usually drawn from neighboring wells. This was the perfect recipe for epidemics. The XIXth century was closely tied to the first cholera epidemics in Europe. Imported from India, they occurred in Paris and London during the 1830s, killing tens of thousands in the following decades. In London, 5 of the 9 water companies were sourcing their drinking water from the Thames, where much of cesspools nightsoils ended.

3.1.1.2 London's linear sewers

Following Edwin Chadwick 1842 report on the Sanitary Condition of the Labouring Population of Great Britain, the Public Health Act was enacted in 1848 (George 2011a). His solution for Public Health was to send all sewage to the Thames, based on the still important miasma theory supporting that disease spread through unhealthy air. He was aware this was polluting the river, but thought this would improve health. Fueled by sewers discharged in the Thames, another cholera epidemics happened in 1854.

Finally, what benefited most to London inhabitants was the summer 1858 Great Stink. An unusually long and dry weather transformed the Thames into a rotting sludge. The unpleasant smell was especially worrying considering the then dominant

miasma theory, supposing that bad odour would carry disease. In just 2 weeks, the parliament extended the powers of the Metropolitan Board of Works. Sewers would still be discharged into the Thames, but way downwards London, in Barking. The problem was simply displaced but Londoners were relieved. The basis for London's sewage network was built from 1858 to 1866 (George 2011a). Similarly, in 1874, Berlin mandated combined sewers (Esculier et al. 2020).

This did not prevented the generalization of sewers to be seen as a huge waste of a precious fertilizing material. A clear illustration of this are these excerpt from Victor Hugo's *Les Misérables* (1862):

"Paris jette par an vingt-cinq millions à l'eau. [...] Le plus fécondant et le plus efficace des engrais, c'est l'engrais humain. [...] Il n'est aucun guano comparable en fertilité au détritius d'une capitale. [...] Employer la ville à fumer la plaine, ce serait une réussite certaine. Si notre or est fumier, en revanche, notre fumier est or. [...] Rendez cela au grand creuset ; votre abondance en sortira. La nutrition des plaines fait la nourriture des hommes."

"La terre appauvrie et l'eau empestée. La faim sortant du sillon et la maladie sortant du fleuve. [...] On expédie à grands frais des convois de navires afin de récolter au pôle austral la fiente des pétrels et des pingouins, et l'incalculable élément d'opulence qu'on a sous la main, on l'envoie à la mer. Tout l'engrais humain et animal que le monde perd, rendu à la terre au lieu d'être jeté à l'eau, suffirait à nourrir le monde."

3.1.1.3 Paris sewage spreading, a relative circularity

Paris also slowly adopted the sewer system, but lagging some decades behind London. The major difference was that the sewers were seen as an opportunity to considerably increase the recycling of human excretions (Esculier et al. 2020), contrary to the English capital.

Before 1850, only 20% of Parisian buildings were equipped with domestic water supply (Musée des Egouts de Paris), and dumping excretions to sewers was forbidden. But gradually equipment and water consumption increased, and simple dumping of domestic water on the streets became impractical. In 1852, disposal of domestic water (not including excretions) to sewers was mandated, which was a first extension from its original role, originally aimed at cleaning the roadway with street fountains and evacuating rainwaters (Esculier et al. 2020). While the sewers network was being extended, namely by Belgrand, some engineers saw an opportunity to use it as a transport highway to agricultural parcels. In 1868, sewage spreading was tested in Paris suburbs.

Finally in 1894 was enacted the *tout-a-l'égout* in Paris, mandating the connection of households and their toilets to sewers. From 1% in the 1880s, the connection rate

increased to 10% in 1895 and 55% in 1905 (Esculier et al. 2020). Meanwhile, in the 1900s, about 75% of sewage water was spread on agricultural land (Figure 3.2). Paris was an outlier, as many large cities considered implementing sewage farming impractical, but some medium-sized cities nonetheless adopted the practice: in the early 1900s, about 30 French and 50 German cities (Gorman 2013c). Some German towns used wastewater for fish farming.

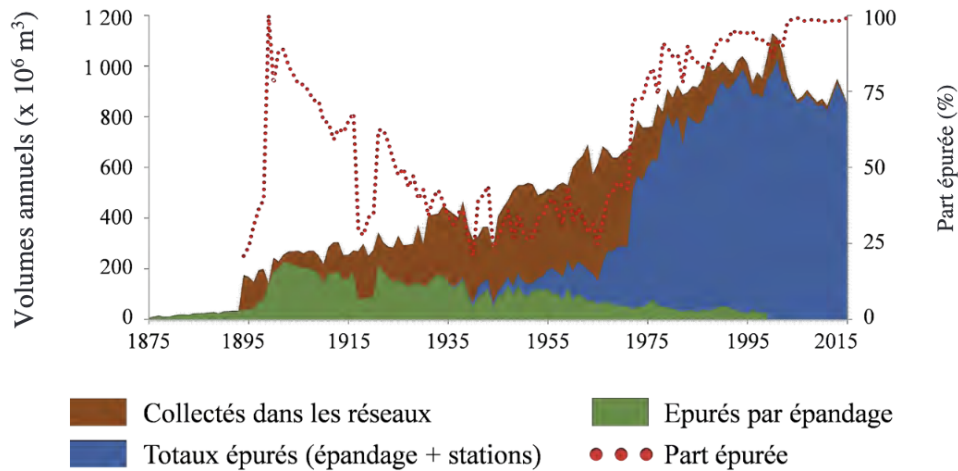


Figure 3.2: Change in volumes of wastewater collected in networks and treated, via agricultural spreading or centralized treatment in wastewater treatment plants, from 1875 to 2015, in the Paris area. Source: Rocher et al. (2017)

Yet in Paris operational costs were way higher than revenues from sewage farms. As commercial fertilizers developed, namely based on the Haber-Bosch process, authorities no longer considered wastewater as resource but rather as a waste, and turned to simple discharge of wastewater to the Seine, which was sacrificed. While some tests were carried out in 1905 of partial treatment with bacterial digestion, real wastewater treatment plants were only generalized decades late.

The total result (Figure 3.3) was an increasing N and P recycling throughout the XIXth century, reaching a peak with the combination of *tout-à-l'égout* generalization and sewage spreading to crops around the 1900s (about 50% circularity for N and 70% for P); followed by a decline during the first half of the XXth, as this practice was abandoned.

3.1.1.4 Did public sewer system decrease water-borne disease ?

With sewers is often associated the idea of hygiene, and namely water-borne diseases. For instance, a poll was conducted among the readers of the British Medical Journal (Ferriman 2007) on the most important medical milestone since 1840. Of

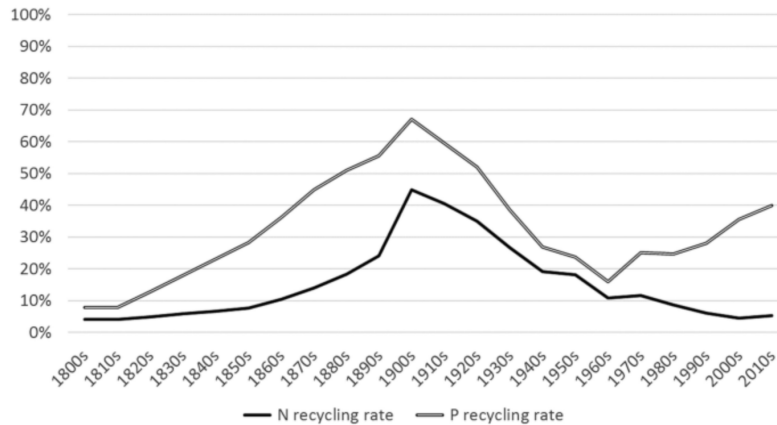


Figure 3.3: Reuse of excreted N and P to agriculture in the Paris conurbation, from the 1800s to the 2010s. Source: Esculier et al. (2020)

the more than 10,000 answers, 16% chose "piped water to people's homes and sewers rinsed by water", very close to antibiotics (15%) and and anesthesia (14%). But can we causally link the decrease of water-borne disease to sanitation facilities and sewers, or was it more due to water supply sanitation? This is one of the question tackled by Minier (2023). Most interestingly, he produced the following Figure 3.4, comparing Cholera mortality (in blue) with the rate of connection to *tout-à-l'égout* (in orange) in Paris. Cholera plummeted way before toilets connected to sewers were generalized, suggesting that other important factors are at play.

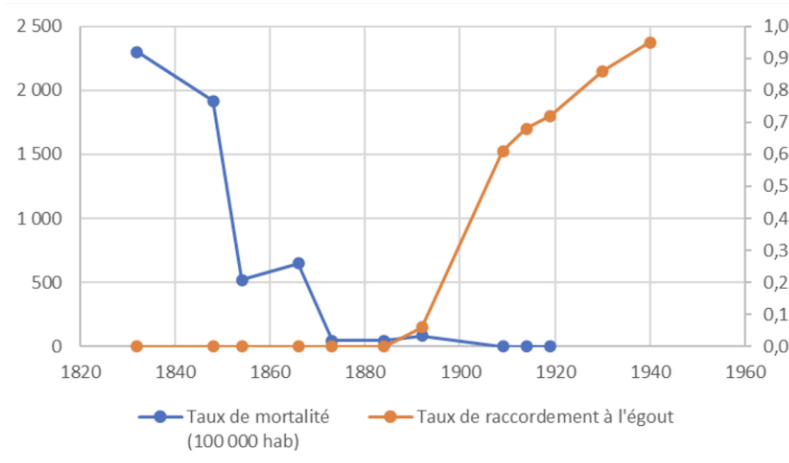


Figure 3.4: Cholera mortality (in blue) and rate of connection to *tout-à-l'égout* (in orange) in Paris. Source: Minier (2023).

I looked for more causal inferences regarding health effect of the sanitation system, with randomized controlled trials in the Cochrane database (specialized in systematic reviews on health issues) for empirical evidence (Cochrane 2024).

As cholera is a much less common disease than it used to be, I could not find a Cochrane review studying the effect of sanitation on cholera (only reviews on

cholera vaccines). I thus focused on another water-borne disease, diarrhea. Results are presented in Table 3.1.

Interventions assessing the effect of access or improvement of sanitation facilities found a reduction of diarrhea prevalence from -0% to -15%, with a low certainty of evidence. Meanwhile, improving the water supply quality reduced diarrhea by -25% to -50%, with generally moderate confidence. Actually, simply promoting hand washing had a higher effect than sanitation facilities, with reductions around -30% (level of confidence from low to high).

Table 3.1: Effect of public health interventions on diarrhea incidence in the Cochrane database. Source: Bauza et al. (2023) for sanitation facilities (50 studies, only the results from the 17 Randomized Controlled Trials (RCTs) shown here); Clasen et al. (2015) for water supply (49 studies, 44 RCTs); Ejemot-Nwadiaro et al. (2021) for hand washing (29 RCTs). A risk ratio of 0.7 means that the intervention reduces diarrhea incidence by 30% compared to no intervention.

intervention		population	risk ratio	level of certainty
Sanitation Facilities	provide access	adults	0.89 (0.73-1.08)	low
	provide access	child <5 y	0.98 (0.83-1.16)	low
	improve	adults	0.89 (0.73-1.08)	low
	improve	child <5 y	0.85 (0.69-1.06)	low
Water Supply	Chlorination	-	0.77 (0.65-0.91)	low
	Flocculation / disinfection	-	0.69 (0.58-0.82)	moderate
	Filtration	-	0.48 (0.38-0.59)	moderate
	Solar disinfection	-	0.62 (0.42-0.94)	moderate
Hand Washing	day-care centres or schools	high income countries	0.70 (0.58-0.85)	high
	day-care centres or schools	low and middle income countries	0.66 (0.43-0.99)	low
	community	low and middle income countries	0.71 (0.62-0.81)	moderate

It is actually not possible to conclude from these randomized controlled trials, because in the studies interventions regarding sanitation facilities range from providing pit latrines to flush toilets with sewers. But it at least reminds that one most important factor is how much fecal contamination is limited just before human in-

gestion, for instance with sanitized water or hand washing. Flush toilets and sewers can limit contact with pathogens at the excretion site and exfiltrate pathogens from the city, but this simply moves the problem downstream, and pathogens are usually not treated in wastewater treatment plants.

3.1.2 Asia and the nightsoil economy

While sewers and flush toilet were generalizing in some urban centers of the Western world, laying their XXth century generalization, some Asian communities were perfecting centuries old recycling of human excretions, even in very dense urban environments. This was not going unnoticed by many European thinkers of the XIXth century. Many of them (for instance Liebig or Victor Hugo) lamented the waste of their cities excretions management system compared to Chinese or Japan ones, as reported in Ferguson (2014). But with the generalization of synthetic fertilizers after WWII, this fascination progressively turned into disdain for these considered backward practices.

3.1.2.1 China

Shirai et al. (2023) argue for evidence of nightsoil use in China as soon as between 1800-500 BCE. While the evidence can be scarce for these ancient times, extensive description of the reuse of nightsoil in China (Figure 3.5), alongside with other organic fertilizers (imported soybean cakes, canal dredgings...), has been documented in the late XIXth and early XXth century, namely by King (1911).



Figure 3.5: Woman carrying buckets of nightsoil in China, 1871. Source Wikimedia Commons (2024a)

The reuse of human excreta in China continued well into the XXth according to

Gu et al. (2015). As late as in 1980, they estimated the reuse rate to be around 90% for both urban and rural settings (Table 3.2). But it quickly declined in urban areas with connection to sewers, reaching a negligible 5% by 2010. Reuse somehow continued in rural areas and began to decline only by the 1990s, reaching 50% in 2010 according to their estimation. Note that these rates relate to the reuse of human excreta in terms of volume, but that important losses of N can still happen during storage through volatilization and leaching.

Table 3.2: Estimates of reuse rates of human excreta in urban and rural China, 1980-2010. Source: Gu et al. (2015), Table S10

	1980	1990	2000	2010
Urban sewage treatment	5%	15%	35%	75%
Rural sewage treatment	0%	1.5%	4%	7%
Urban excretions recycled	90%	70%	40%	5%
Rural excretions recycled	95%	95%	75%	50%

Smil (2004a) produced an estimation of N inputs to China's agroecosystem. Strikingly, it appears that synthetic fertilizers did not "replace" traditional fertilizers, but were rather simply added in absolute terms (Figure 3.6). Human excretions still represented about 15% of the total inputs in the 1950s. But by the end of the XXth century, it was no more than a few percent, due to the enormous synthetic inputs and decreasing reuse rates.

3.1.2.2 Edo/Meiji Japan

The management of human excreta in Japan urban centers, such as Osaka, Okayama, and particularly in Edo (Tokyo) during the Edo/Tokugawa (1603-1868) and Meiji (1868-1912) dynasties is extensively described in Tajima (2005). Notably, Edo already had 1 million inhabitants by the XVIIIth century, but the path chosen to handle such a large amount of excretions was very different to the choices made in Paris or London one century later. In 1649, the shogunate proclaimed the Keian edict, forbidding toilets to be emptied into canals or creeks (Ferguson 2014), and even gave advice as how to not waste the precious night soil "*Farmers should build the toilet with a roof over near the main house, so that even if it rains night soil will not dilute and waste*" (Szczygiel 2016).

Tajima (2005) describes how a symbiosis emerged between the city and surrounding urban-fringe farmers, favored by improved road transports and canals. While using excreta to fertilize crops was not uncommon in ancient societies, few have made an economic good out of it to such an extent. In that system, urban inhabitants sold their shimogoe ("fertilizer from the bottom of a person" in Japan) to neighboring farmers. This was a genuine demand from farmers, who would move to the city

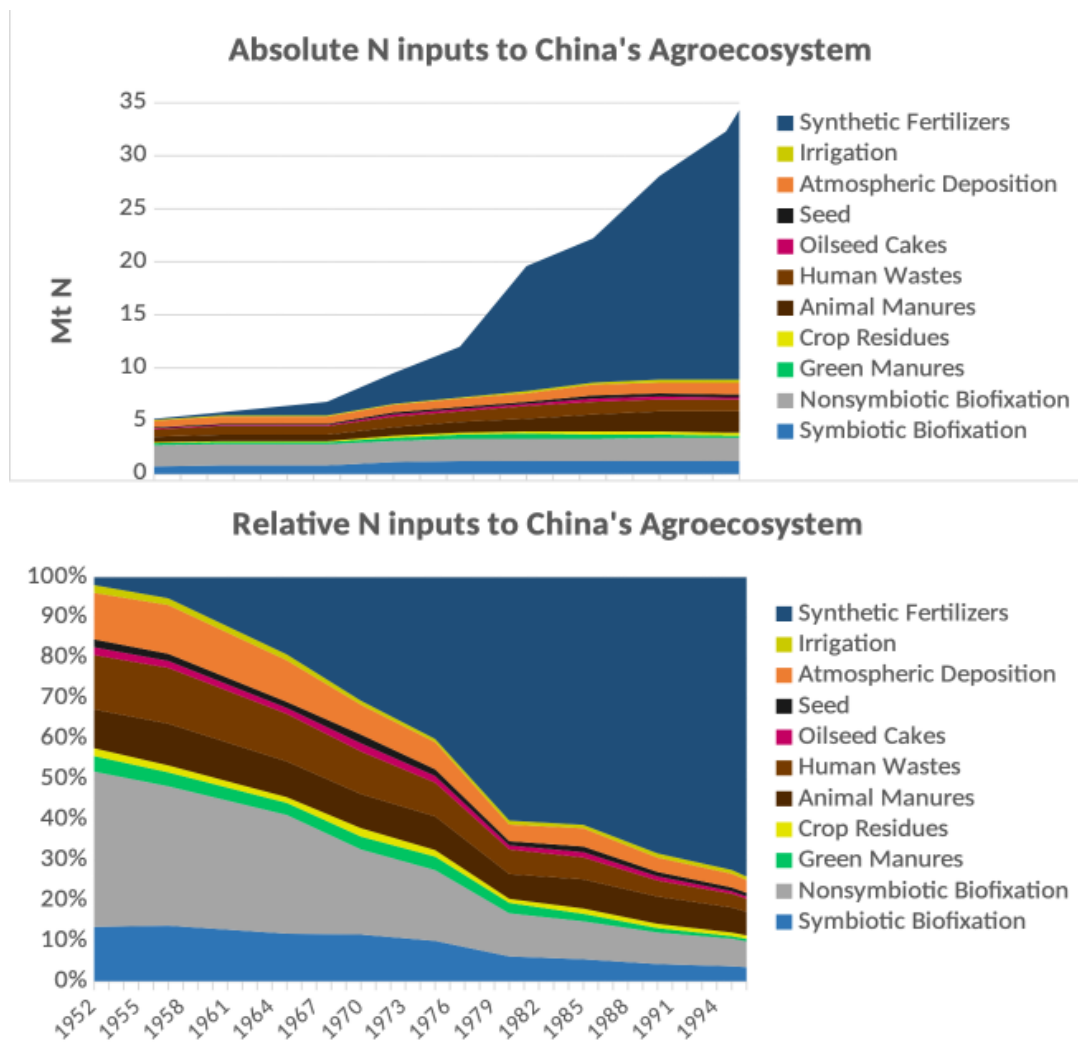


Figure 3.6: N inputs to China's agroecosystem in the second half of the XXth century. Source: Smil (2004a), appendix R.

for collection. As time went on, this collection service was operated by private services. During the XVIIIth-XIXth century, the demand was sometimes so high that it triggered high prices and conflicts between farmers and city landowners. Farmers were generally not more than 10 kms away from the center, as nightsoil contained 95% water, preventing large scale transportation. They specialized in high value perishable products, with several harvests per year, like vegetables, fruits or flowers. The price of this nightsoil commodity would fluctuate during the year, depending on the fertilization demand: from nearly free on winter to high in spring/summer.

As urbanization continued, Tokyo did not escape one of its more dire consequence, as in Europe, and experienced its first major cholera epidemic in 1877. Then, by the early XXth century, several factors hindered this mutualism. First, urban sprawling, which increased the distance to be covered from city to rural areas and replaced some of the closest farms by residential areas. Second the development of commercial concentrated fertilizers: soybean cake imported from China, or chem-

ical like ammonium sulfate or superphosphates, which required less labor to spread on crops.

Human excretions were less and less seen as an economic good for the private actors. In 1900, the Waste Cleaning Act made municipalities responsible for street cleaning. The perceptions had not shifted so much as to consider human excretions as a waste, but that would soon change. Tokyo finally started its own municipal collection service in 1917, and in 1930, night soil effectively became a waste, and not an economic asset anymore. By 1932, the municipality was dumping human excreta to the ocean.

3.1.3 Current global and Western situation regarding sanitation

3.1.3.1 Sanitation access in the world

Many people around the world do not even have access to toilets: The global share of people practicing open defecation went from about 20% in 2000, down to almost 5% in 2022 (See Figure 3.7 from Ritchie et al. (2019)).

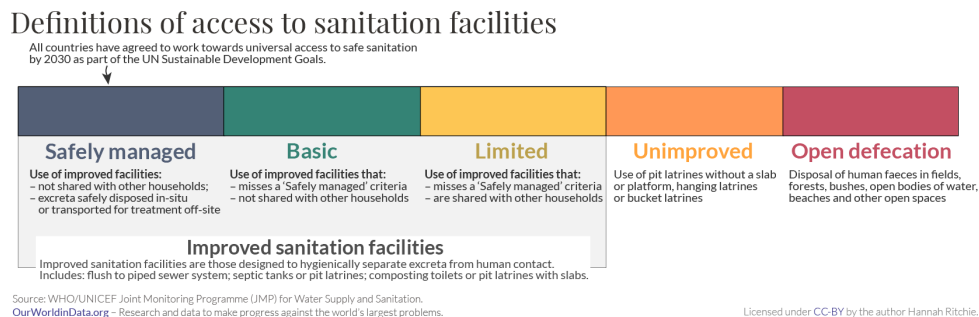
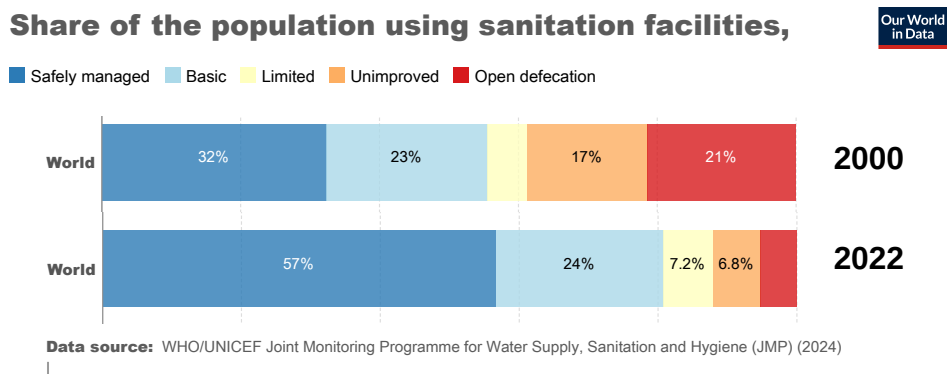


Figure 3.7: Acces to sanitation facilities in the world. Source: adapted from Ritchie et al. (2019)

In absolute population (considering population growth meantime from 6.15 to 8 billion)), this translates into a decrease from 1.3 billion to 0.4 billion. India,

Afghanistan, Cambodia, Laos, Haiti, Papua New Guinea, East Timor have about 10-20% of their population practicing open defecation. This is all the more striking when in 2000 India and Cambodia were around 75-85%. All countries currently above the 20% threshold are sub-Saharan, with rates as high as two-thirds in Niger, Chad, and South Sudan. All the other countries in the world are below 10%.

Open defecation is associated with many health and social adverse consequences. It promotes infectious intestinal diseases as water supplies are more likely to be contaminated, and increased the odds of violence against women and girls (Mara 2017). The Global Burden of Disease estimates that in 2021 unsafe sanitation was responsible for 0.6 million deaths and unsafe water source for 0.8 million deaths (Global Burden of Disease Study 2021)

Besides the sheer lack of sanitation facilities, some people have access to toilets, but still in a very unimproved manner (Figure 3.7), considered by the WHO as not ensuring hygienic separation of human excreta from human contact: pit latrines without a slab or platform, hanging latrines, or bucket latrines. They represented 17% of the world population in 2000, and 7% in 2022. While there has clearly been some important progress in the last 2 decades, hundreds of millions of people around the world still lack improved facilities

Once excretion has happened in the toilets, human excreta can have several fates. Around the mid-2010s, only a bit more than one third (36%) of the global population was connected to sewers, mainly in high-income countries (WHO and UN 2018). About half of the global population used pit latrines or had no facilities. The remaining 15% are connected to septic tanks, collecting blackwater and greywater. Collection is not synonymous of treatment: sewers or septic tanks can simply discharge excretions directly to the environment. WHO and UN (2018) estimates that globally only 60% of domestic waters are safely treated.

Finally, in all the scenarios developed by Van Drecht et al. (2009), more than 50% of the global population would remain not connected to sewers by 2050. So, even if processes to recover nutrient in wastewater treatment plants are developed in the near future, there will still be a need for cleverly handling human excretions for at least half of the global population not connected to sewers. Some of these possibilities will be explored in Section 3.3.3.

3.1.3.2 Sewers, mostly in high-income countries

Sewers make great economies of scale: in large and dense cities, the requirements of meters of sewers per inhabitants are much smaller than in rural areas. Conversely, they are quite expensive in less populated areas. As a result, most countries with sewers still have a non negligible share of their citizens not connected, with their own individual autonomous system. In France, this share is about 15-20% (Ministere de

la Transition Ecologique 2023; Eurostat 2022).

Western sewers rely on large quantity of waters, typically on the order of 100L/cap, to transport waste, but also to prevent malfunctioning: the important flow limits stagnating and malodorous waters. But this water consumption is not suited for many places in the world, with arid climate, and sewers are capital-intensive facilities, not affordable in poor countries.

Sewers can be Separate or Combined Sewer System, if they handle domestic flows and rainwater in the same pipes, thus increasing the volumes treated in WWTP, or separately, as Combined Sewer System, directly discharging the rainwaters to the environment. The advantage of Combined Sewer System is the "flush" effect during rainfalls, which prevents stagnating waters; but a main drawback lies in that during storms, it often has to discharge stormwaters mixed with untreated domestic waters in the environment.

3.2 Waste Water Treatment Plants

In Western countries such as France, most wastewater treatment plants (WWTPs) operate in 3 steps: a primary treatment removes suspended solids and particulate organic matter, a secondary treatment removes dissolved biodegradable matter and part of the nutrients, and a tertiary treatment further removes phosphorus and nitrogen to comply with discharge limit thresholds. General elements regarding WWTP treatments can be found in Nathanson et al. (2018), Tchobanoglous et al. (2003), Gorman (2013c), and Degrémont (2005).

3.2.1 Primary and Secondary Treatments

A proxy of water quality is the oxygen concentration downstream of the WWTP discharge. If a lot of organic matter is discharged by the WWTP, microorganisms will eat it and consume by breathing, which will consume and deplete the oxygen present in the water (Figure 3.8)

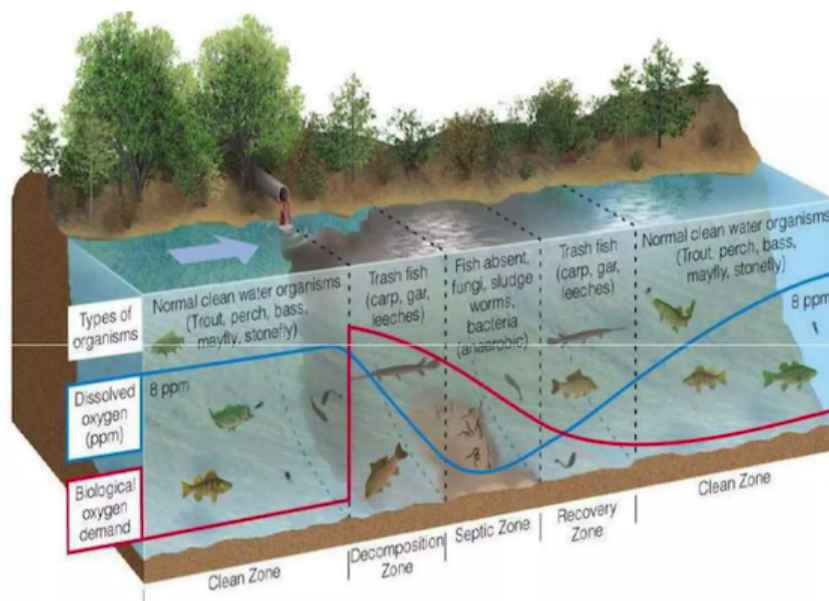


Figure 3.8: Evolution of dissolved oxygen and biological oxygen demand downstream of WWTP discharge, inspired from the Streeter et al. (1925) model.

The aim of the post WWII wastewater treatment plants was thus to remove this organic pollution. The efficiency of this process is assessed by following the Biological Oxygen Demand (BOD), and in particular BOD₅, i.e. the mass of molecular oxygen consumed by micro-organisms in five days in one liter of water at 20°C in the dark. Another indicator is the COD, Chemical Oxygen Demand.

A first primary treatment is used to simply screen for large macro-pollutants, and then let the sludge settle in a primary clarifier, which produces the primary

sludge (Figure 3.9). This can remove 50-70% suspended solid and 25-40% BOD, for a retention time of about 1.5-2.5h. What did not settled is then sent for secondary treatment.

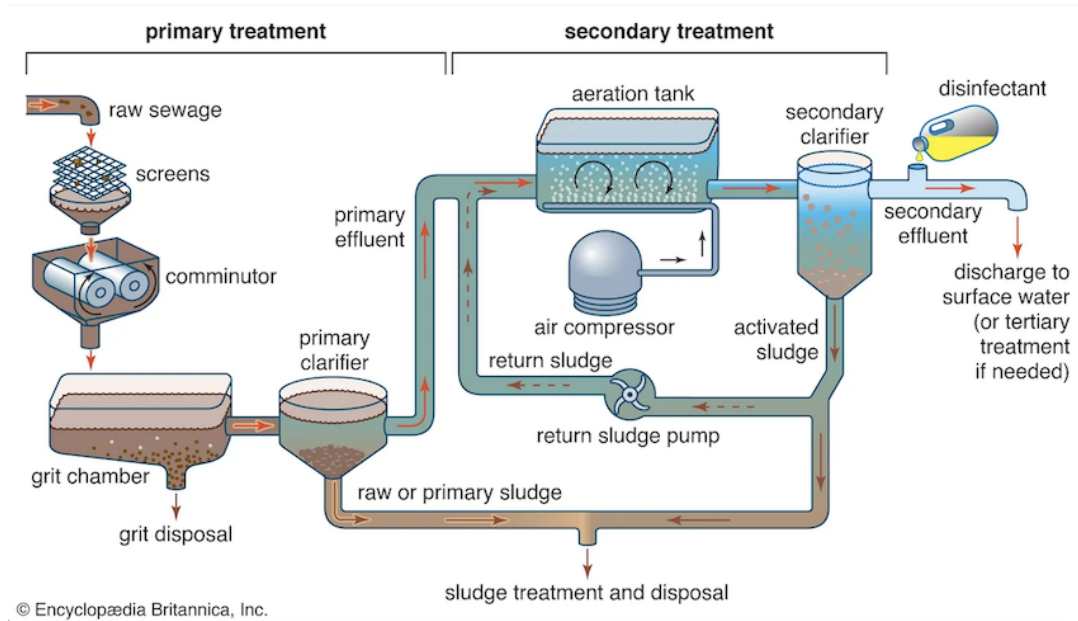


Figure 3.9: Main processes in WWTP for primary and secondary treatments. Note that the final disinfectant treatment is uncommon in most WWTPs. Source: (Nathanson et al. 2018).

For the secondary treatment, the easily degradable organic matter is treated in an aerated basin, through activated sludge. Activated sludge means that the organic pollution passes through bacteria that are "activated" by oxygen aeration blown into the basin. The bacteria eat the organic matter and transform the carbon chains into CO₂ by breathing. Some bacteria can also do it with reduced nitrogen, transforming NH₄⁺ into nitrates through nitrification. This is a simple conversion of nitrogen from reduced to oxidized form: this considerably limits the BOD, but does not remove nitrogen, which can still contribute to eutrophication. In total 70-85% of BOD is removed, with a retention time of about 6-8 hours.

Table 3.3: Historical development of the main WWTP treatment processes. Source: Lesavre (2015)

Date of development	Process	Basin Volume (l/cap)
1950s	Organic Matter Removal (aerated basin)	20-40
1970s	Nitrification, P precipitation (with Fe or Al)	80-140
1980s	Denitrification	100-160
2000s	P biological removal	140-200

A second clarifier lets the sludge settle, which contains many of the activated

bacteria fed in the aeration tank. While most of the carbon has been lost as CO_2 to the air, the bacteria stored some nutrient they used to grow, namely about half of the phosphorus entering the WWTP.

These treatments are sufficient to reach requirement regarding BOD discharge. Even though they remove a good chunk of phosphorus (about 50%), it is not enough to meet P discharge standards, and even less if we consider nitrogen. For these nutrients, a further tertiary treatment is required.

Table 3.4: Order of magnitude of N and P removal for primary, secondary and tertiary treatments. Source: Vigiak et al. (2020) and Van Drecht et al. (2009).

removal efficiency (cumulative)	treatment		
	primary	secondary	tertiary
Nitrogen	10-25%	35-55%	80%
Phosphorus	10-30%	45-60%	90%
BOD	50%	95%	95%

3.2.2 Enhanced N treatment

3.2.2.1 Before N removal: N conversion by nitrifying activated sludge and biofilters

To meet N discharge standards, WWTPs have to switch from N conversion to N removal. This is usually done by denitrifying NO_3^- to N_2 , under anoxic conditions. The process of preliminary N conversion from NH_4^+ to NO_3^- in activated sludge has been described in the previous paragraph. This process, as well as organic matter degradation, require a lot of oxygen, and aeration is actually the main consumer of energy in WWTPs because of the electricity consumption to blow air in the basins.

Another way to nitrify reduced N into NO_3^- is to use nitrifying biofilters. Contrary to aerated basins where the bacteria performing nitrification are suspended in water, here the bacteria are fixed to the biofilter. This makes this technology more intensive and compact than aerated basins. In France, biofilters are almost exclusively used on very large WWTPs, with a capacity superior to 100,000 population equivalent. Activated sludge and biofilters make up 90% of the installed WWTP capacity in France (Figure 3.10).

3.2.2.2 N_2O emissions

Whatever the process, most of the N_2O emissions occurs during nitrification. N_2O emissions are the major post of greenhouse gases emissions in WWTP (Filali et al. 2022). N_2O emissions depend on the quantity of nitrified nitrogen and on the process. There are substantial uncertainties concerning WWTP N_2O emissions, as

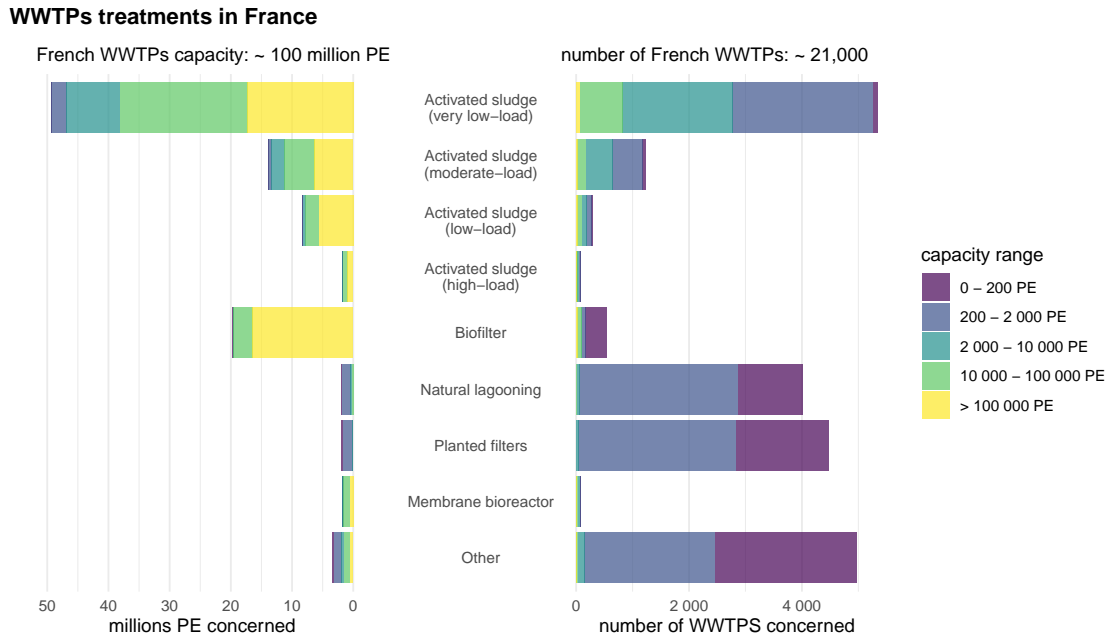


Figure 3.10: Treatment in French WWTPs. Left: in terms of installed capacity, expressed in population equivalent. Right: in terms of number of WWTPs. Colors: capacities of the concerned WWTPs. The database may be less trustworthy for small plants (e.g. concerning the Biofilter category for 0-200 PE). Source: own processing from Portail Assainissement Collectif (2023)

exemplified by IPCC guidelines on that matter, reported by Filali et al. (2022) (Table 3.5). The 2019 IPCC update of their 2006 guidelines multiplied the emission factor by 40.

In the case of France, the ASTEE association (Filali et al. 2022) published its own estimation. Their guidelines are adapted to the French context, where most treatments are activated sludge with low or very low load, and biofilters (these 3 make up 75% of the installed capacity, see Figure 3.10). Contrary to the IPCC, they recommend to estimate N_2O emission based on removed NTK, and not on incoming NTK. They note that the major factor enhancing N_2O emissions is the intensity of the nitrifying process: this gives advantage to low load activated sludge, but increases biofilters emissions.

Note that the ASTEE also discusses indirect N_2O emissions, that happen in the natural aquatic environment where the residual N is discharged, but which I do not detail here.

3.2.2.3 N removal: denitrification

Once nitrogen has been converted to NO_3^- nitrates, it is ready for the final removal step, denitrification, sending nitrogen in the air as inert N_2 . This reaction happens in an additional basin or during non aerated phases, this time under anoxic condition.

Table 3.5: N₂O Emissions Factor (EF) with different WWTP N removal treatments. Source: Filali et al. (2022)

source	treatment	N ₂ O emission factor (EF)	EF in kgN-N ₂ O per kgN-TKN...
IPCC 2006	all	0.037%	incoming
IPCC 2019	all	1.6%	incoming
ASTE 2018	all	0.046%	removed
ASTE 2022	activated sludge	0.06% (0.02-0.11%)	removed
ASTE 2022	membrane bioreactor	0.06% (0.02-0.11%)	removed
ASTE 2022	biofilters	1.65% (1.29-1.96%)	removed

The bacteria performing the reaction need to feed on organic matter. However there is sometimes not enough DBO₅ in the wastewater to fully enhance denitrification and reach legally required performance. In that case, operators add methanol (mostly issued from fossil reserves) to enhance the reaction. In Ile-de-France, the SIAAP consumes 30,000 tons of methanol each year for that purpose (Tabuchi et al. n.d.).

Considering all the steps described before, Maurer et al. (2003) estimate that in WWTPs, the full treatment of nitrogen ending with denitrification consumes 45 MJ of primary energy per kgN, or about 1 kg oil equivalent¹. They report about the same quantity to produce 1 kgN fertilizer with the Haber-Bosch process. For phosphorus, the respective values per kgP are about 50 MJ for precipitation in sludge in WWTP (detailed in Section 3.2.3) and 30 MJ for fertilizer production.

3.2.2.4 The anammox process

During sludge treatment under anaerobic conditions to produce biogas, sludge dewatering produces ammonia rich digester liquid ($>300 \text{ mgN-NH}_4^+ \cdot \text{L}^{-1}$, 15–20% of the N entering the WWTP). These waters can be resent in the main treatment process for further nitrification / denitrification. Yet that requires extending the basins volume to handle this extra load.

Another alternative is to use the anammox process (anaerobic ammonium oxidation). This relies on bacteria that can transform ammonium under anaerobic conditions, discovered during the 90s. Already in 2014, there was more than 100 full-scale anammox installations in the world (Lackner et al. 2014). Compared to the mainstream nitrification/denitrification, the first nitrification process is replaced

¹1 kg oil equivalent = 42 MJ

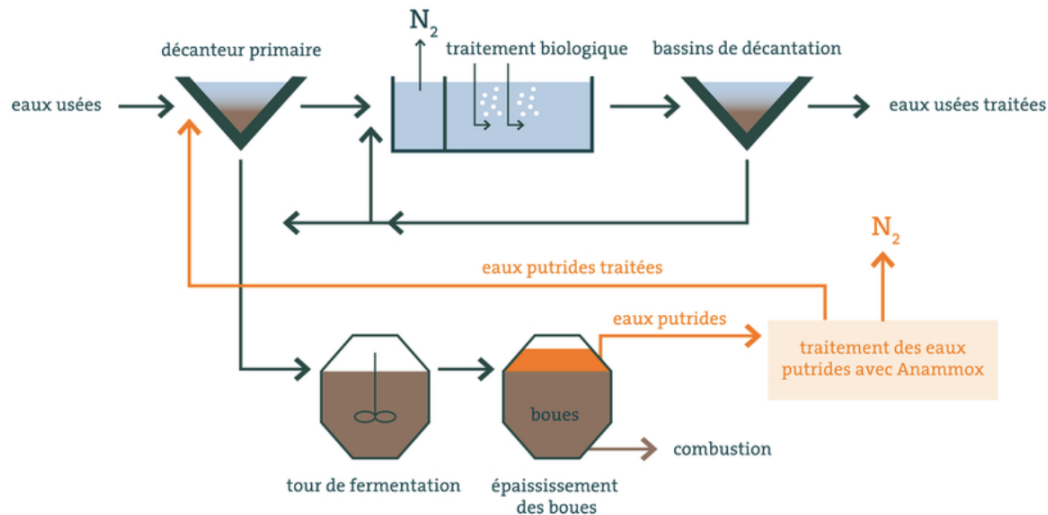


Figure 3.11: N removal in ammonia rich digester liquid with the anammox process. Source: EAWAG (2023)

by a partial nitrification, nitritation (production of NO_2^- instead of NO_3^-) and the final denitrification is replaced by the anammox process. The anammox process produced N_2 with just NO_2^- nitrites and NH_4^+ ammonium. This process requires less oxygen, because the nitrification is replaced by only a partial nitritation (so less energy to ventilate the basins), and less organic carbon, because there is no denitrification process (so no need to add methanol, and more organic carbon left to produce biogas).

While this consumes less energy than aerated sludge, the drawback is that it emits more N_2O during the nitritation step, with an emission factor of 3-7% according to Filali et al. (2022). This can be reduced if nitritation and anammox are combined in the same basin (emission factor <1%) (Siegrist et al. 2008; Carle 2023; Joss et al. 2023). A key challenge in the anammox process is the difficulty of maintaining stable operating conditions.

3.2.3 Enhanced P removal

Removal of phosphorus from municipal waste waters is a common and mastered process. Bacteria used for secondary treatment, which aims at reducing organic loads, typically store around 50% of incoming P into sludge (Table 3.4). To achieve higher removal rates, typical tertiary treatment involve P precipitation with salts (often Fe^{3+} , sometimes Al^{3+}) or enhanced biological phosphate removal (EPBR), using bacteria storing P in polyphosphates. Removal rates of 80-90% is commonly reached for WWTP using these processes. Contrary to nitrogen where most removed N is lost in the air as N_2 , all of the removed P ends up in the sludge and can thus

be recovered.

I did not find up to date figures about the share of chemical or biological treatment for P removal, but it seems that in Europe during the 2010s and most certainly during the 2000s, the majority of WWTP used chemical precipitation (Nättorp et al. 2019; Korving et al. 2019; Paul et al. 2010).

3.2.3.1 Chemical Precipitation of Phosphate Salts

A simple way to remove P is through salts precipitations. This uses cations binding to phosphates. This can be done with Fe, Al, or Ca (Elser et al. 2020e). Chemical Fe^{3+} precipitation is often preferred, due to lower costs (Schipper 2019; Korving et al. 2019), but it strongly binds phosphorus and makes it insoluble, hindering its fertilizing potential once spread on crops. Worst, if there is residual free Fe^{3+} , it can further immobilize the phosphorus already present in the soil. This problem is less present with Al salts.

3.2.3.2 Enhanced Biological Phosphorus Removal

Simple biological removal occurs during primary and secondary treatment, when bacteria deposited in excess sludge store phosphorus in their amino acids or phospholipids. But this can be further enhanced with special bacteria that can accumulate P, sometimes up to 20% of their dry mass, in the form of polyphosphates (Elser et al. 2020e).

The advantage of this method is that P is removed in an organic form, that can be more easily further remobilized on agricultural land, compared to P bound in salts (Schipper 2019). However sometimes high removing rates cannot be reach, especially during winter, which requires a hybrid process mixing EPBR with precipitation (Paul et al. 2010; Varga 2022).

3.2.4 Consequence: Improvement of water status, but not enough to reach a generalized good ecological status

Treatment of municipal wastewater helped to improve in water bodies certain physico-chemical parameters linked to urban pollution discharge. This is exemplified for the case of Paris in Figure 3.13, where improvement for particular nutrients correlates with the timing of the different treatment adoption. For dissolved oxygen and BOD, there is a progressive improvement from the 1970s to the 2010s. However, for NH_4^+ and PO_4^{3-} , the generalization of N and P treatment occurred in the late 1990s and during the 2000s. Thus, their status in water deteriorated until the 1990s, and improved in 2012, reaching a better status than the original values during the 1970s.

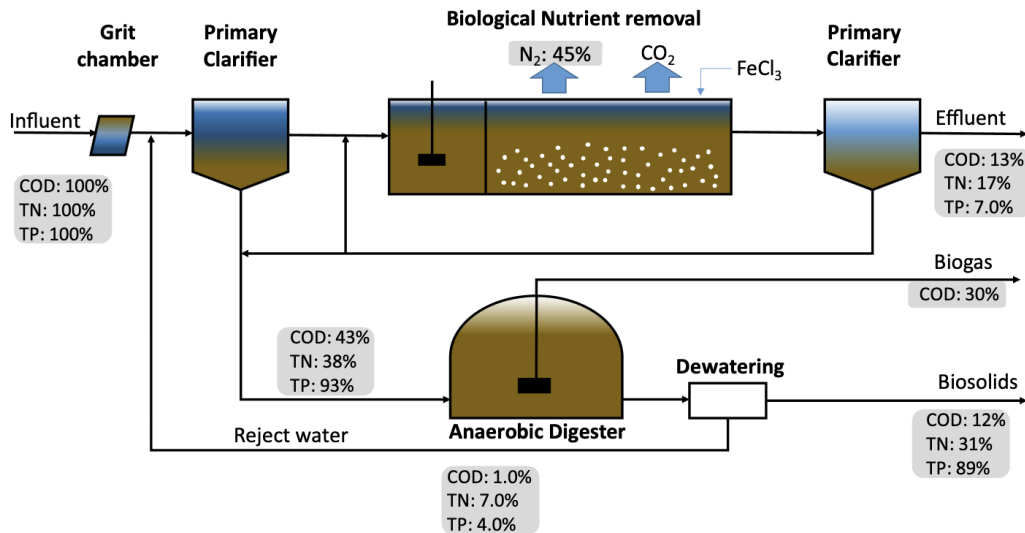


Figure 3.12: Indicative nutrient flows in WWTP with tertiary treatment and high removal efficiencies (80-85% for N and 90-95% for P). Source: Ostermeyer et al. (2022)

For these 4 parameters, the physico-chemical status is good or very good in most areas of the Seine.

This can also be seen in the Rhine river for phosphates PO_4^{3-} , in Figure 3.14. However this Figure also shows that the effect is way less visible for nitrates NO_3^- . This is because most of nitrates originate from diffuse agricultural emissions and not from WWTP point-sources; and contrary to phosphates, nitrates did not benefit from emissions reductions at the source with the phosphate detergent ban.

The improvement of these physico-chemical parameters were not enough to reach what the Water Framework Directives qualifies as "good ecological status" for most water bodies. This is because there are many other parameters involved in the good ecological status, including other physico-chemical parameters (pH, temperature), biological ones (phytoplankton, macrophytes, fished), and pollutants (zinc, arsenic, copper, chrome, pesticides) (MTES 2019).

In France in 2018, among the more than 10,000 watercourses, about 45% had an at least good ecological status; 35% was qualified as medium; the rest was mediocre to bad (Figure 3.15. Results are also presented in Table 3.6 for water bodies and coastal waters.

3.2.5 Quaternary treatment: what is not treated (yet)

The next step in the end-of-pipe treatment framework will probably be to focus on other pollutants. One is the further removal of pathogens, whose contamination can be of particular concern if WWTP effluent is discharged in lakes or for cities down-

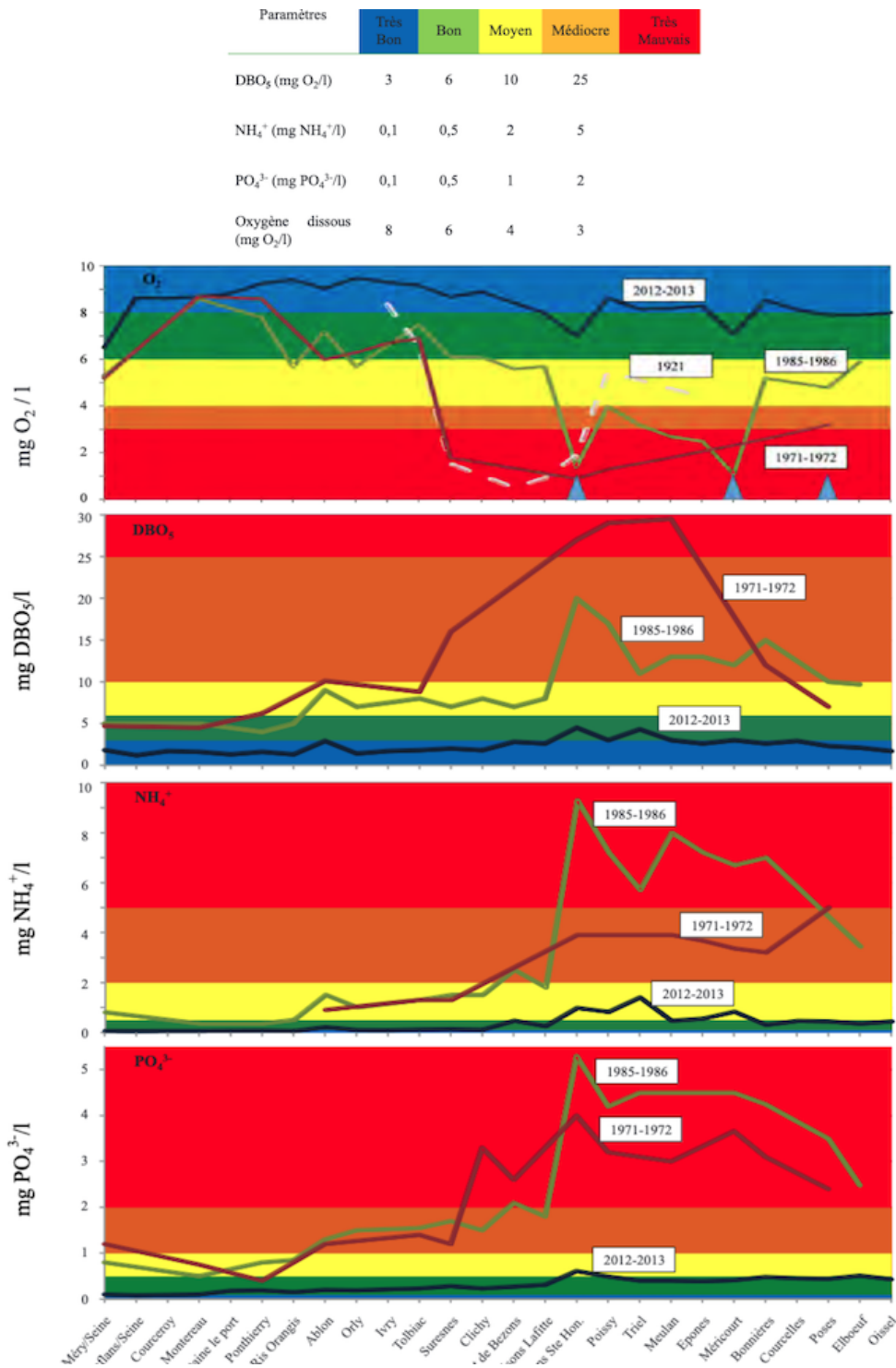


Figure 3.13: Trends in some chemical parameters concentrations in the Seine during the XXth and early XXIth century. Source: Rocher et al. (2017).

stream. Currently less than 2% of French WWTPs treat pathogens, representing about 7% of the total installed capacity Ministère de la Transition Ecologique (2023). There is also the concern of micro-pollutants: pharmaceuticals residues, hormones, micro plastics... Switzerland requires the elimination of these micro-pollutants by 2040.

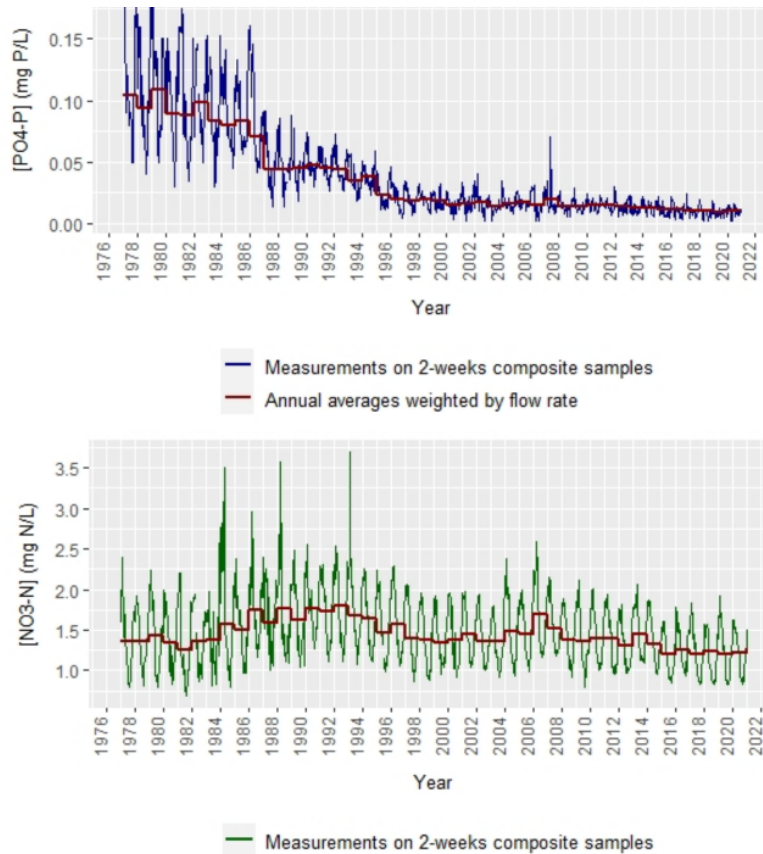


Figure 3.14: PO_4^{3-} and NO_3^- in the Rhine as Basel. Source: FOEN (2022), based on data from the NADUF program (Jakob et al. 2002).

Table 3.6: Ecological status of water bodies and courses in France. Sources: naturefrance (2022a), naturefrance (2022d), naturefrance (2022c), and naturefrance (2022b)

	N	good/very good	medium	mediocre/bad	unknown
coastal water body	179	50%	35%	15%	<1%
estuary, lagoon, deltas	116	40%	25%	30%	5%
water bodies	398	35%	55%	10%	<1%
water courses	10,715	45%	35%	20%	<1%

3.2.6 Sludge: Treatment and Nutrient Recovery

The elements of the following two subsections are mainly from Schipper (2019).

3.2.6.1 Reusing nutrients through simple sludge spreading

The easiest way to recover nutrient and organic matter present in settled sewage sludge is to simply spread it directly on agricultural land, possibly following composting. This is an easy way to reuse nutrients trapped in the sludge, namely phosphorus or potassium, but nitrogen will barely be recycled this way as few of it is retained in sludge.

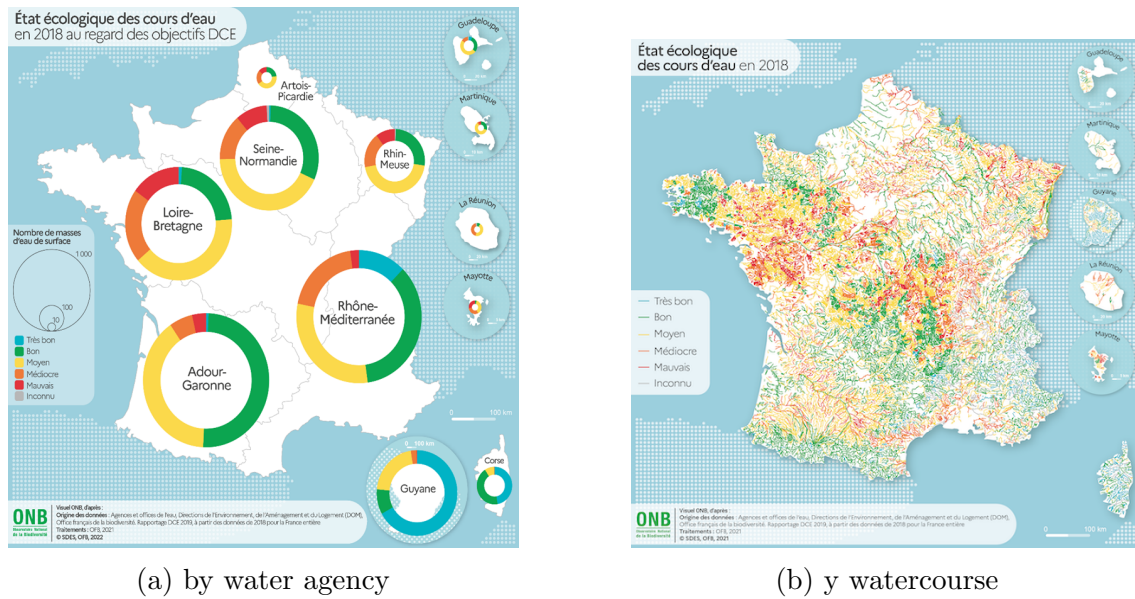


Figure 3.15: Ecological status of French water courses. Source: naturefrance (2022a)

Moreover this method also spreads urban contaminants trapped in the sludge, such as heavy metals, and some countries have banned this practice (Switzerland, Japan...). In countries banning direct spreading, an option is to burn the sludge. But burning sludge volatilizes the organic matter and nitrogen, leaving only phosphorus and some other micronutrients for agricultural recycling. *“Coincineration”* refers to burning sludge with other wastes to recover energy, e.g. in municipal incinerators or cement kilns, but this practice mixes the phosphorus with ashes from other waste, making difficult its recovery. A preferred option for nutrient recovery is *“mono-incineration”*, where the sewage sludge is burnt alone. The phosphorus is then recovered from easily-transportable ashes.

Another option is sludge anaerobic digestion, which produces methane from the sludge organic matter, whose digestate contains the remaining phosphorus and nitrogen. Nutrients are transformed from organic to mineral forms in liquid phase. The drawback of anaerobic digestion and mono-incineration of sludge WWTP is that it does not get rid of heavy metals already present in the sludge.

Usage of sludge varies greatly in Europe. From Eurostat statistics (Figure 3.16, Eurostat (2022)), around 2019, Spain reused about 85% of its sewage sludge to agriculture; Germany incinerated 75% and Netherlands 85% of them, and Switzerland incinerated virtually all of its sewage sludge. In France, about 75% was reused in agriculture, and direct spreading is also relatively high in the United Kingdom. In Japan, as in Switzerland, reuse of sludge to agricultural land is forbidden.

In the Euro zone, there has been a general trend to less and less landfilling. Some countries, like Germany and the Netherlands, moved away from partial direct spreading to more and more incineration, with sometimes recycling as mono-incin-

Disposal of sewage sludge from urban wastewater treatment by method of disposal, 2019

% of total



Note: Denmark, Italy, Portugal, Iceland, United Kingdom: no data or no recent data available

(*): Data for 2018 instead of 2019

(?): Data for 2017 instead of 2019

(*): Data estimated

(*): Data provisional

Source: Eurostat (online data code: env_ww_spd)

eurostat 

Figure 3.16: Fate of wastewater treatment plant sludge in Europe. Source: eurostat (2022).

eration, due to stricter limits.

3.2.6.2 A common P extraction process: struvite precipitation

I will expand in Section 3.2.7.1 on developing P recovery processes, aimed at extracting phosphorus from the WWTP flow and sludge. But I nonetheless mention here a process which is already mature enough to have been implemented on more than 40 full-scale operational WWTP in the late 2010s: struvite precipitation.

The recovery is not necessarily the main goal for implementing struvite precipitations. First, it prevents naturally occurring P precipitations which can clog pipes. Second, when implemented on return waters, it reduces the loads recycled within the WWTP flows, and thus allows smaller scaling, reducing the costs associated with handling sludge (Nättorp et al. 2019).

Struvite formula is NH_4MgPO_4 . It is clear from this formula that it needs 1 mole of NH_4^+ , 1 mole of PO_4^{3-} , and 1 mole of Mg^{2+} . Municipal wastewater stoichiometry requires that Mg, the limiting factor, has to be added to recover P, and that most N will not be recovered that way (since N:P stoichiometry of wastewater is about

10).

This process cannot be implemented on the mainstream line after nitrification to NO_3^- , as N has to be under NH_4^+ form. Before nitrification, NH_4^+ is present, but too diluted. Another impediment is when phosphates are already bound in salts to other cations, such as Fe or Al regularly used in chemical sludge precipitation for P treatments. PO_4^{3-} has stronger affinity to these cations, and make it impossible to recover it with Mg^{2+} .

For these reasons, struvite precipitation has to be implemented in WWTP using enhanced biological phosphorus removal, in the return flow after dewatering of sludge or anaerobic digestion. This is a limitation, as most WWTP use chemical precipitation and not enhanced biological removal.

Also, struvite precipitation barely recovers the P present in organic form. Because of this, while 90% of the inorganic P can be recovered through struvite precipitations, an upper bound of 40% of the total P is actually recovered (Koga 2019); the rest stays in the organic P and insoluble Ca-Al-Fe phosphate minerals. Actual rates of total recovery are actually closer 15 to 20% of the total WWTP P influent (Besson et al. 2021a). Higher recovery rates can be reached by crystallizing struvite on P in sludge ashes after mono-incineration, following leaching with acid to release the phosphate bound to other compounds (Nättorp et al. 2019).

A process very similar to struvite precipitation is the P precipitation with Ca, producing hydroxyapatite $\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$ (for instance described in Okuno et al. (2019)). The main difference is that it does not require NH_4^+ rich waters nor Mg inputs, but rather Ca addition. Like struvite, it recovers around 20% of the total WWTP P influent, and the majority is recovered in sludge.

3.2.7 The future WWTP ?

From the current situation, an industrial vision for the near to mid-term future is the transformation of “wastewater treatment plants” to “resource recovery and water recycling centers” (Soares 2020; Diaz-Elsayed et al. 2019). This vision has already been figured at least since 2006 and the first energy crisis, with expansive energy prices, and most certainly in the early 2020 with the post-COVID and Ukraine invasion triggering energy inflation in Europe. These resource recovery centers should continue to recover P, but in more purified and available form than the current Fe-P salts; transform the carbon in sludge into energy through methane or dihydrogen, to have energy-neutral or positive facilities; and finally recover the currently wasted nitrogen, while reducing the N_2O greenhouse gas emissions. To this is also sometimes added reclaimed water, i.e. producing water sanitized water from WWTP influent. Here I will only focus on nutrients recovery, as nutrients in excretions can

represent a substantive share of nutrients applied to agroecosystems (this will be the object of Chapter 5), while carbon recovery to produce biogas could represent at the very best only around 1% of global anthropic energy consumption (Rittmann 2013). Yet locally, energy represents a major cost for water and wastewater services, and a significant proportion of indirect greenhouse gas emissions. Carbon recovery could offset the WWTP energy consumption.

3.2.7.1 Purified P recovery

A review and typology of P recovery processes in WWTPs can be found in Egle et al. (2016). It can be schematically broken down into 3 categories, depending on where the recovery happens. First, recovery can take place in the aqueous phase, for instance with ion exchange membranes, precipitation or crystallization. Second, some processes aim at extracting and transforming P in the sewage sludge (leaching, oxidation). Third, processes can concern sewage sludge ashes after monoincineration, for instance adsorbed on zeolites.

However recovery rates compared to P from the total WWTP influent substantially differ: for processes in the aqueous phase, they were always below 50% and mostly below 30%; extraction from sludge yielded a 40-70% recovery; and only recovery in ashes reached 65-90% recovery. Consequently, some of these processes cannot be used alone and have to be combined with other techniques to reach high overall recovery.

More specifically, advocates of P recovery with hybrid ion exchange nanotechnology put forward another advantage, in addition to recovering P in the form of soluble phosphate: this process allows to reach very low discharged levels ($<0.1\text{mgP}\cdot\text{L}^{-1}$) (Ownby et al. 2021; Guida et al. 2021a), susceptible to comply with future regulation that current technologies may struggle to reach (currently the threshold required in Europe is only $1\text{-}2\text{ mgP}\cdot\text{L}^{-1}$). We can also note some studies on microbial strains able to biologically produce struvite (Leng et al. 2023), allowing to produce this mineral without chemical precipitation .

If we include struvite recovery, which is already a mature process, there were only 3 facilities recovering purified P in France in 2021 according to Varga (2022), citing inventory data from Kabbe (2023). The most common recovery method is struvite, but is not particularly suited for French WWTP, whose P treatments are mostly based on chemical precipitation (to reach P discharge threshold regulation), while struvite recovery is more suited to purely biological systems (often with digester) Varga (2022).

These developing technologies are in the continuity of already high recovery rate of P with current processes, with incremental improvement: less reactant consumption, recovery of more readily-available P... A real breakthrough would be to recover

N in high amount in WWTPs.

3.2.7.2 N recovery

Many new N recovery processes are being developed, but contrary to phosphorus, they usually recover only a small fraction (<30%) of the total nitrogen entering the WWTP (Esculier 2018; Besson 2020).

A multitude of processes... A review about N recovery processes can be found in Winkler et al. (2019). They note that methods to recover highly concentrated ammonia streams (side-streams effluents or urine) exist and are cost effective: air stripping, steam stripping, or hollow fiber membrane contactors. But in the N diluted mainstream, it becomes uneconomical. N can also be captured in biosolids, for instance with heterotrophic organisms, but they consume COD in higher amounts than the COD/N ratio present in wastewater, and as a result capture only 10-20% of influent N. Phototrophic organism overcome this limit because they can feed on C fixed from CO₂ using photosynthesis. In particular, microalgae can reach high N removal efficiencies (>50%), even with effluents with a low N content typical of municipal wastewater (<100 gN/L) (Cai et al. 2013). But a challenge is the subsequent solid separation to recover the fixed N in microalgae cells. There is a fundamental trade-off: to improve photosynthesis, one prefers having diluted algae, to let light come in water; but this hinders the recovery of these suspended organisms, and consumes space. Techniques are developed to harvest algae (chemical, mechanical, electrical, biological, immobilization), but this consumes energy and/or chemicals.

Another review of N recovery processes from wastewater have been made by Beckinghausen et al. (2020). They particularly note that many of the studied processes do not have energy or costs analysis, and concluding about the feasibility of upscaling is thus difficult. Besson et al. (2021a) estimate that transmembrane chemisorption is one of the most efficient N recovery technique when it comes to energy consumption, with 2.55 kWhelec·kgN⁻¹ recovered, compared to 7.2 kWhelec·kgN⁻¹ for ammonia stripping and 76 kWhelec·kgN⁻¹ for nitrification/distillation.

...which generally necessitate high N concentrations... In the review on ammonia stripping undertaken by Wu et al. (2022), the tested effluent had an ammonia concentration 0.3-5 g·L⁻¹, generally issued from urine or digestates, whereas typical WWTP influent and effluent are only 0.06 and 0.008 gN/L. Perera et al. (2020) underline that for gas-permeable membranes, the cost increase significantly for concentrations below 0.05 g·L⁻¹.

For that reason, N recovery is often implemented on the dewatering liquors from anaerobic digestion, for instance with suspended sludge sequencing batch reactor, moving bed biofilm, or granular sludge sequencing batch reactor (Ochs et al. 2020). Another possibility is to apply the processes directly on urine before flush dilution, but in that case the subject is not WWTP recovery anymore.

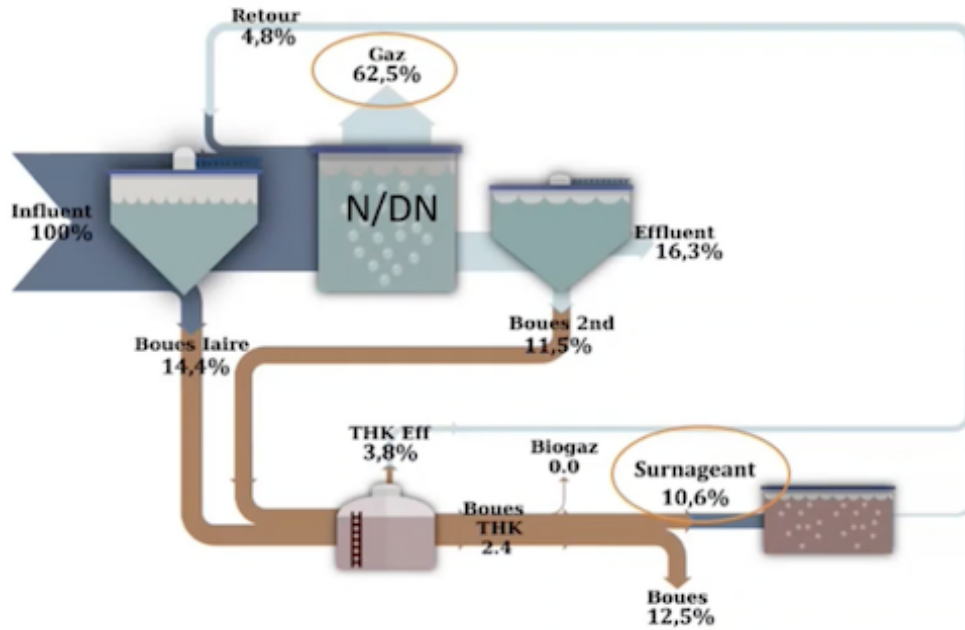


Figure 3.17: Concentrated dewatered stream, where most of N recovery occur, contain only about 10% of the total N entering WWTP. Source: Besson et al. (2019).

...resulting in low total recovery. But the dewatered stream only carries about 10% of the total N entering the WWTP (Figure 3.17); combined with the 10-15% of N recovered in sludge, this explains why total recovery is less than 30%, the rest being mostly lost as denitrified N_2 .

Van der Hoek et al. (2018) reviewed current technologies of N recoveries and applied them to the flows of a typical WWTP. On N-rich effluent, the different processes (air stripping, hydrophobic membranes, vacuum membranes) can have high recovery rate (75-90%); but when considering the totality of the N flows, the recovery compared to the totality of N entering the WWTP is always below 25%. Only urine separation and treatment allowed high total recovery rates, above 60% in their case study.

With some exceptions I nonetheless identified some processes that can operate at low N concentrations; the question is now to what extent they can be economically upscaled in WWTPs in the foreseeable future. Perera et al. (2020) use

a technology precipitating phosphates with Ca, and recovering ammonia through liquid-gas stripping under vacuum, producing ammonium sulfate. This is done with electrochemical pH modulation from 6 to 11. They reach high recovery of N and P (>90%), with concentrations typical of municipal wastewater (<50 mgN/L and <5mgP·L⁻¹). They find it more economical than the current conventional process of fertilizer production + treatment in WWTP, but more energy-intensive (+65%). Another example is a ion exchange process by Guida et al. (2021b), which was able to extract (30-95% efficiency) N from influent typical of what could be found in diluted municipal wastewater (<20 g-NH₄⁺/L).

3.2.7.3 Wastewater reuse

Finally, a way to recover N and P from municipal wastewater could be to simply irrigate crops with them (fertigation), without attempting to remove nutrients in the first place. This is not a new practice, since spreading of WWTP effluent was already practiced around Paris in the early XXth century (see Section 3.1.1.3). However this also spreads pathogens, and all the micropollutants and heavy metals whose presence in wastewater have increased during the XXth, and this practice is now generally prohibited. But it could be revived with the development of wastewater reuse that is currently promoted.

Wastewater reuse consists in sanitizing municipal wastewater to supply agricultural irrigation, industrial uses and even sometimes domestic use requiring drinkable standards. A review of life cycle analyses of water reuse has been undertaken by Maesele et al. (2021). Their conclusion is that wastewater reuse is generally an environmental benefit in settings using non-renewable groundwater, arid regions, littorals where WWTP effluent is discharged into the sea and tertiary treatment not too energy-intensive. But in settings of low water scarcity, inland location and advanced tertiary treatments, wastewater reuse would be less eco-friendly than current water supply mixes.

They also notes that, in the current framework of water reuse, nutrient reuse is generally barely considered, and the projects are simply added downstream of N and P removal in wastewater treatment plants, resulting in nutrient-poor reclaimed waters. Enhancing fertigation would necessitate to stop treating N and P in the considered WWTP.

The main issue with municipal wastewater reuse for fertigation is that crops do not need irrigation nor fertilization all year round, but just for 2-3 months. Fertigation during winter would simply leach nitrates and pollute groundwaters. As municipal wastewater is produced continuously all year long, and the enormous amounts of water produced precludes storing them, this means that less of a quarter of the reclaimed water could be used in agriculture, unless it is used in market

gardening, as in XIXth Paris or Japan' Edo, with continuous cropping all year long. But that would necessitate redesigning agricultural land in order to have vegetables surrounding human settings. So fertigation would be a restrained opportunity of wastewater reuse in certain particular contexts, and necessitate to dynamically adapt WWTPs removal efficiencies depending on the season.

3.3 Current state of source separation

3.3.1 Rationale for source separation

Proposals to divert human excretions from municipal wastewater have been suggested for at least three decades (Jönsson et al. 1997; Larsen et al. 1996; Larsen et al. 1997). The rationale is that urine constitutes less than 1% of the water volume in WWTP, but more than 80% of the N loads (Figure 3.18). For P, this is 50% (Vinnerås et al. 2002), because a bit more phosphorus is excreted in feces, and because phosphates are used in detergent, though their use varies greatly between countries and through time. So, directly recovering excreta yields a solution tens to hundred times more concentrated than wastewater, and less contaminated with urban pollutants found in sewers.

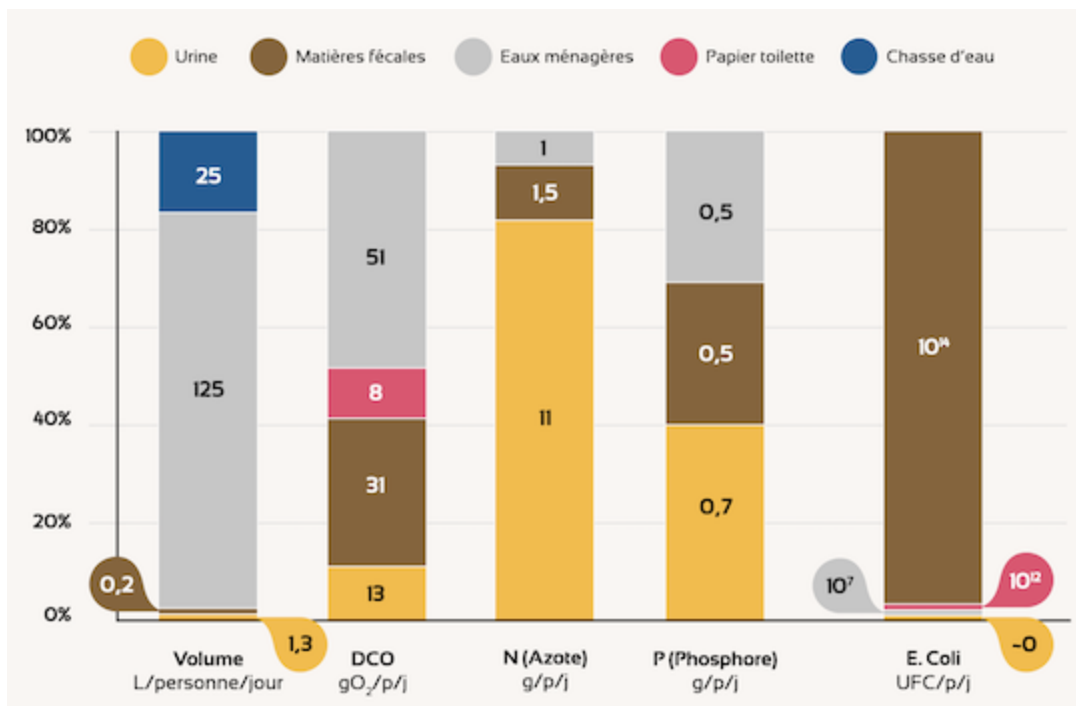


Figure 3.18: Characterization of the various effluents making up domestic wastewater (urine, faeces, toilet paper, household wastewater) in percentages and absolute values. Source: Besson et al. (2021b).

Besides the dilution aspect, some have urged to stop regarding urine as a "waste" and rather as the "liquid gold" of wastewater (Randall et al. 2018). Moreover, many Southern countries do not have a sewer system: worse, in 2022, 5% of the population in low-income countries was still practicing open defecation (down from 20% in 2000), and thus did not even had access to toilets (Ritchie et al. 2019). For those connected to sewers, it may not be suited to water-scarcity situations (Drangert 1998). To enhance the recovery of this liquid gold, Drangert (1998) has

proposed to fight "urine blindness", i.e. the vision of excreta as an unavoidable mix of urine and feces and failure to consider urine on its own. The rationale behind this is that urine carries most of the excreted nutrients, with way less pathogens than feces. Once separated, urine could be used directly to fertilize crops instead of being discharged to rivers, thus suppressing the need for WWTP. Even if one does not want to apply raw urine and rather prefers to extract nutrients, extraction on undiluted urine rather than diluted municipal wastewater will always be more efficient (by 40-70% for nitrogen and 15-35% for phosphorus according to McCartney et al. (2021)), which is also in support to preliminary urine diversion, as long as energy consumed during transport does not cancel the benefit.

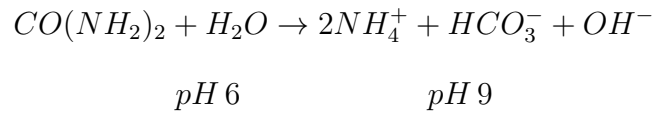
3.3.2 Typology of Processes for Urine-Based Fertilizers

One can identify several goals of urine treatments producing urine-derived fertilizers (Maurer et al. 2006). The reader can particularly consult the review of urine-based fertilizers made by Martin et al. (2022) for a detailed typology, or the didactic sheets presenting different urine-based fertilizers by Agrocapi (2022). Below I propose to study urine treatments with the following typology:

- **stabilization:** to limit ammonia volatilization (acidification, alkalization, nitrification, fermentation).
- **concentration:** to reduce volumes while preferentially keeping all nutrients in the concentrated solution (distillation, evaporation, freeze-thaw, reverse osmosis)
- **extraction:** to target and extract specific nutrient from the liquid (struvite precipitation, ammonia stripping, ion-exchange)
- **hygienisation:** to remove pathogens (storage with pH increase, activated carbon, temperature increase)
- **removal of micropollutants:** specifically targeting pharmaceutical residues and hormones (electrodialysis, nanofiltration, ozonation, activated carbon)

3.3.2.1 Stabilization

In freshly excreted urine, 80% of nitrogen is under the form urea $\text{CO}(\text{NH}_2)_2$ at pH 6, with the rest mostly under organic form and a bit of ammonium NH_4^+ . Under normal conditions, urea is quickly (in a matter of minutes or hours) hydrolyzed by urease enzyme naturally present in urine (Udert et al. 2006), which releases nitrogen in its ammonium NH_4^+ form and increases pH to 9 by releasing OH^- described by the following reaction:



After this hydrolysis, virtually all of urea is converted to ammonia forms. The pH increase during urea hydrolysis triggers salts precipitation, mostly under struvite from $NH_4MgPO_4 \cdot 6H_2O$, but also hydroxyapatite $Ca_{10}(PO_4)_6(OH)_2$ and calcite $CaCO_3$ (Udert et al. 2003). About 1/3 of the phosphorus is precipitated this way in undiluted stored urine (Udert et al. 2006). Potassium and phosphorus are also present in dissolved and bioavailable forms.

This new pH of 9 is very close to the pKa of the ammonium NH_4^+ / ammonia NH_3 equilibrium:



A part of the ammonia gas NH_3 is dissolved in the water, but it also volatilizes in the air especially under windy conditions (usually less than 10% of the N applied as fertilizer on the agricultural parcel, Johansson et al. (2001)), resulting in N losses but also air pollution and odor emissions.

There are several ways to limit this NH_3 volatilization from urine. As we saw the pKa of NH_4^+/NH_3 reaction is 9.2. This means that at pH = 9.2, half of nitrogen is under the dissolved NH_4^+ form, which does not risk volatilization, and the other half is in the NH_3 form. By lowering the pH, the stabilized NH_4^+ becomes the dominant form. Typically, for 3 pH unit below the pKa (so at 6.2), NH_3 represents only 0.1% of the NH_4^+/NH_3 couple. Acidification of the *hydrolyzed* urine is thus one way to reduce NH_3 volatilization. To bring urine to a pH < 6.5, one typically needs to add sulfuric acid in quantities of $30 \text{ gH}_2\text{SO}_4 \cdot \text{L}^{-1}$ (Agrocapi 2022).

Still with pH modification, it is also possible to limit NH_3 volatilization not by modifying the acid/base pair, but by preventing the hydrolysis of urea into NH_4^+/NH_3 from happening in the first place. As we saw, the hydrolysis is due to the urease enzyme and happens in a matter of minutes, so treatment has to take place just after excretion. Urease activity is greatly reduced outside its operating range of pH 4-11 (Krajewska 2018). Acidification below pH 4 of *fresh* urine requires $3 \text{ gH}_2\text{SO}_4 \cdot \text{L}^{-1}$, about 10 times less sulfuric acid than acidification of *hydrolyzed* urine (Hellström et al. 1999). Alkanisation above pH 11 requires lime in amounts of $10 \text{ g} \cdot \text{L}^{-1}$ (Randall et al. 2016). In these 2 cases, nitrogen is kept in the form of urea $CO(NH_2)_2$.

The 2 previous paragraphs used acid-base reactions. Another way to stabilize nitrogen in urine is through nitrification, $NH_4^+ \rightarrow NO_3^-$. This process is similar to

the one operated in wastewater treatment plants, and requires nitrifying bacteria and aeration to provide the oxygen for the reaction (Fumasoli et al. 2016). Nitrification can also be performed passively on activated carbon ("pitribon" process).

One can also use urine lactic acid fermentation to decrease pH (around 4 to 5) and inhibit the action of urease (Andreev et al. 2017). Finally, addition of urine to organic residues (compost, wood, biochar, compost...) can incorporate mineral nitrogen to the organic matter and thus reduce losses, but the process has to be finely controlled in order to avoid potentially important volatilization (Martin et al. 2023b).

Table 3.7: N composition of urine-based fertilizers and synthetic fertilizers. Source: Martin et al. (2020)

fertilizer type		organic	nitric	ammonia	urea
		N_{org}	NO_3^-	NH_4^+/NH_3	$CO(NH_2)_2$
urine-based	fresh urine	15%	-	5%	80%
	stored urine	20%	-	70%	10%
	acidified stored	10%	-	90%	-
	acidified fresh	15%	-	5%	85%
	alkynized fresh urine	10%	-	-	90%
	(partially) nitrified	-	50%	50%	-
	+ wood chips	75%	-	15%	5%
synthetic	ammonium nitrate	-	50%	50%	-
	nitrogen solution	-	25%	25%	50%
	urea	-	-	-	100%

3.3.2.2 Volume Reductions: concentration, drying, extraction

The previous processes were aimed at preventing N losses, primarily ammonia emissions that can occur in the field. Here I describe processes aimed at reducing the volumes that have to be handled during transportation or by farmer. Some of these processes benefit from preliminary N stabilization. For instance, nitrification of ammonium to nitrates reduces N volatilization during concentration obtained by distillation.

Concentration: Numerous concentration processes exist, for instance described by Ek et al. (2006). Evaporation (Zanchetta et al. 2015; Antonini et al. 2012; Simha et al. 2020) and distillation (Udert et al. 2012) of urine can be used to concentrate nutrients, usually after first stabilizing nitrogen under the nitrate form. This consumes energy but solar evaporation can be a way to consume less. An alternative is freeze-thaw: when water freezes, chemicals are not included in ice, thus concentrating the elements in the remaining liquid water when subsequently thawed (Noe-Hays et al. 2021). This requires less energy than evaporation. Other

processes include osmosis (Zhang et al. 2014) and membrane distillation (Volpin et al. 2020). Typically these concentration processes concentrate the nutrients in a volume about ten times smaller than the original urine.

Drying to produce solid product: Pushing evaporation even further can result in complete drying of the urine-based fertilizer. Such a process is described in Simha et al. (2018) and Simha et al. (2020). After stabilization of urine under alkaline conditions (obtained with wood ash or alkalized biochar), they evaporated urine water until obtaining a solid product, recovering all of P and K and >70% of N. A drawback of this process is the relatively high energy consumption to evaporate water.

Extraction: These are essentially the same processes that were already described for WWTP waters rich in nutrients (see Section 3.2.7): stripping of ammonium to produce sulfate ammonium, struvite MgNH_4PO_4 precipitation after Mg addition (otherwise only 1/3 of P precipitates), or precipitation of P under apatite after adding calcium hydroxide. Facilities currently developing these processes in WWTPs could be used in the future as a centralized treatment of collected urine. A drawback of extraction is that it only recovers the targeted nutrients. Extractions of sulfate ammonium only recovers nitrogen and not phosphorus nor potassium. Struvite precipitation recovers P but a small part of nitrogen. And besides the major nitrogen or phosphorus compounds, extraction does not recover micronutrient such as S, Zn, Cl, Cu...

3.3.2.3 Hygienization and micropollutants removal

In simply stored urine, spontaneous hydrolysis raises the pH, which kills most bacteria Richert et al. (2010). Ammonia (issued from hydrolyzed urea) is also a sanitizing factor killing bacteria (Etter et al. 2015). Pasteurization can be a quicker and more energy-intensive sanitization technique, for instance 70°C for 30 min or 80°C for 1.5 min as required in Vermont permitting requirements (Love 2020). These pasteurizing temperatures are typically reached during distillation for instance. Treatments that use dehydration (e.g. extraction of crystallized struvite) also strongly reduce bacteria prevalence (Senecal et al. 2018), as low moisture content is a major factor inactivating bacteria (Etter et al. 2015).

Besides pathogens, another concern is micropollutants such as pharmaceuticals residues found in urine. Simple storage and dehydration results in a small reduction of pharmaceutical residues quantity (Jaatinen et al. 2016). A way to reduce further their prevalence is to use activated carbon, where the bacteria fixed on the carbon matrix will metabolize these organic (Duygan et al. 2021).

While these treatments are interesting, one should note that micropollutants are not systematically treated in the current system with WWTP, and so WWTP

sludge spread on croplands also contains pharmaceutical residues.

Table 3.8: Recommended duration of raw urine storage for safe use as fertilizer. Source: Richert et al. (2010) based on WHO (2006)

storage T	storage time	recommended crops for application
4°C	>1 month	food and fodder crops that are to be processed
4°C	>6 months	food and that are to be processed
10°C	>1 month	food that are to be processed, fodder crops
10°C	>6 months	all crops

3.3.3 Examples of source separation projects

Projects of urine source separation gained momentum in Sweden during the 90s, and then spread to Scandinavian and German Europe during the 2000s (Martin 2020; Esculier 2018). In France it particularly took off during the mid-2010s.

A story map of current source separation projects in France has been developed (Toilettes fertiles 2024). It features the projects categories based on the type of separated flow (urine, feces, urine + feces, urine + feces + domestic waters) and the type of facility (individual household, collective households, office, public facility...). The interested reader will find in this source feedback sheets for some projects. Below I give some selected examples of how source separation and nutrient recovery can concretely be implemented.

3.3.3.1 Stabilization with nitrification followed by concentration

Some actors of source-separation can seek to produce commercial fertilizers out of treated excretions. In Switzerland, near Zurich, researchers have developed the VUNA process (Fumasoli et al. 2016) and implemented it in their own building, in EAWAG university (Etter et al. 2015). The final product is a concentrated liquid fertilizer approved for commercial use in Switzerland.

Urine is collected using urine-diverting toilets and dry urinals in the office (about 100 liters per working day). The first step is partial nitrification, yielding a solution of half NO_3^- and half NH_4^+ . This partial nitrification drops pH around 6², well below the $\text{NH}_4^+/\text{NH}_3$ couple pKa, and stabilizes the reduced N mainly as NH_4^+ , preventing ammonia losses. Starting the process takes 45-60 days, but then operates in steady flow. The preliminary nitrification prevents volatilization during the subsequent distillation, and so only 1% of N is lost during that step.

²This done by two bacterial strains, one oxidizing ammonia to nitrites, the other pursuing by transforming nitrites to nitrate: pH control is important to balance the 2 reactions and not overproduce toxic nitrites. Nitrifying more than 50% of the original reduced N may drop pH below 5.5, which would limit nitrifying bacteria development.

Distillation can be energy-intensive, but distillers with vapor compression can recover 90% of energy. After distillation, the nutrients are concentrated more than 10 times. In EAWAG, treated urine has an approximate N concentration of $2.2 \text{ gN}\cdot\text{L}^{-1}$; after treatment the concentration is $50 \text{ gN}\cdot\text{L}^{-1}$ (and 2.1 and 15 for phosphorus and potassium), or a concentration by 20-25. Distilled water is a by-product.

Table 3.9: Features of the nitrification/distillation process. Source: Etter et al. (2015)

Nutrients recovery	>99%
Water removed	97%
Nitrification rate (NH_4^+ to NO_3^-)	$0.4\text{-}0.8 \text{ gN}\cdot\text{NH}_4^+\cdot\text{L}^{-1}\cdot\text{d}^{-1}$
Electricity consumption for distillation	$80 \text{ Wh}\cdot\text{L}^{-1}$ urine
Electricity consumption for nitrification	$50 \text{ Wh}\cdot\text{L}^{-1}$ urine
Temperature in distiller (0.5 bar)	$80\text{-}85^\circ\text{C}$

The process also treats pathogens, which are mostly killed during the 80°C distillation lasting at least 30 minutes. During the aerobic nitrification, the heterotrophic bacteria only partially break down pharmaceuticals and hormones. A subsequent activated carbon treatment is added for that reason, which removes 90-99% of the monitored compounds (Duygan et al. 2021).

The final concentrated product recovers almost all of the micronutrients and the nitrified nitrogen limits volatilization and odors during spreading on crops. For now, it is more energy-intensive than the current optimized WWTP followed Haber-Bosch process, but yields better results on other Life Cycle Analysis indicators (eutrophication, acidification, greenhouse gas emissions...) (Martin et al. 2023a).

A similar process will probably be adopted for a large-scale source separation project in intramural Paris, the Saint-Vincent de Paul neighborhood. It aims at recovering urine of about 600 households on 3.4 ha, with about 1,000 urine-diverting

Table 3.10: Elements concentrations in the final distilled product. Source: Etter et al. (2015)

Element	Concentration ($\text{g}\cdot\text{L}^{-1}$)
N	50
P	2.1
K	15
S	1.5
Ca	0.4
Mg	0.04
Fe	0.5
Cu	0.3
Zn	15
B	16

toilets, corresponding to 2,000 liters of urine a day (Legrand et al. 2021). Feces will go to sewers and wastewater treatment plants via the conventional system, while urine will be transported via independent gravity and pressured networks, to be treated in-situ. The project is backed by the Paris municipality, and the concentrated fertilizer will be used for the city gardens for proof of concept.

3.3.3.2 Low-tech recovery of urine

While concentration can be interesting in densely populated areas (Paris center can reach 40,000 hab/km² on some places), this is not necessarily always relevant. This is exemplified in another project in the Paris urban area, but this time in a lower density environment, in the EPA Paris-Saclay. In this context, volumes reduction is less of an issue, especially given that it is surrounded by agricultural fields. Office will be equipped with men dry urinals to collect urine, which will then be stored in a 20 m³ tank with activated carbon to treat micropollutants (Figure 3.19). A local farmer will empty the tank once to twice a year to fertilize its field.

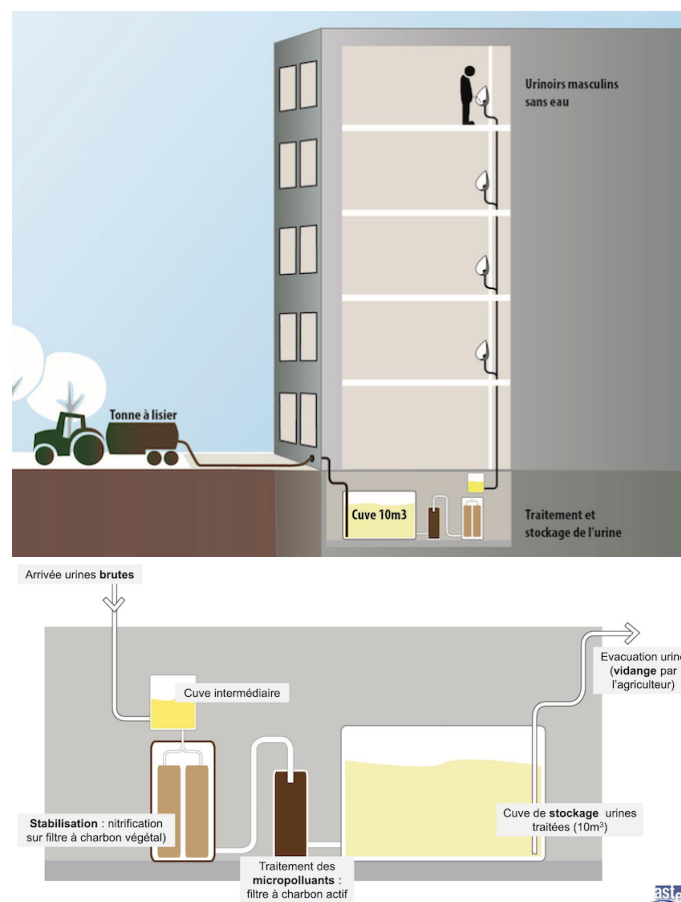


Figure 3.19: Urine separation by men dry urinal in offices, in EPA Paris-Saclay project. With activated carbon, urine is nitrified and its micropollutants are treated, and the simply stored in a tank, before emptying by a farmer. Source: ARCEAU (2023a)

Low tech handling of excretions obviously include dry toilets, for which there exists a great variety of typologies: separating urine from feces or not, with or without addition of carbon, in situ treatment or off site storage... Dry toilets are described extensively in the chapter 4 of Minier (2023)).

Low tech nutrient extraction projects also exist, and have been tested in low to middle income countries, for instance in Nepal (Etter et al. 2011). The aim is to produce a low-cost struvite fertilizer. The project took advantage of the fact that in the municipality the acceptance of dry urine diverting toilets is already quite high. Moreover the mountainous landlocked situation makes fertilizers imports quite complicated. The drawback is that struvite precipitation requires an external magnesium addition, in the form of Mg salts or waste like bittern. In order not to waste nitrogen, the remaining urine after struvite precipitation still has to be spread, since more than 90% of nitrogen is not precipitated.

3.3.3.3 Vacuum toilets for blackwaters recovery

Finally, a source separation option, similar to sewers but with much less water consumption, is the blackwater (urine + feces) diversion by vacuum, treated in centralized or semi-centralized facilities (Meinzinger 2010). Vacuum aspiration avoids the important dilution experienced with flush toilets. Excretions can then be treated aerobically (storage, liquid composting) or anaerobically digested. In that latter case, nutrients are mineralized (and possibly extracted from the liquid phase) and energy production through biogas is a by-product of the process (Wendland et al. 2007). This also decreases pathogens levels, and the liquid phase can still be treated or volume reduction, as described in Section 3.3.2

3.3.4 Challenges of source separation

3.3.4.1 Technical Maintenance of Toilets

One major issue in urine networks is the clogging of pipes, due to precipitation. The main precipitating salt is struvite $\text{NH}_4\text{MgPO}_4 \cdot 6\text{H}_2\text{O}$, impeding the recovery of some of the P and the N, enhanced by the increase in pH following urea hydrolysis. Hydroxyapatite $\text{Ca}_5(\text{PO}_4)_3(\text{OH})$ or calcite CaCO_3 precipitation can also happen. In conventional networks, this precipitation is prevented by the flush dilution and the bigger diameter of the pipes.

One cannot only rely on the infrastructure conception to prevent this clogging, and adequate maintenance are required. Brun (2019) considers that the current reference on how to handle a urine network is the guide "Urine Diversion: One step towards sustainable sanitation" (Kvarnström et al. 2006). His literature review and

experience feedbacks identified several recurrent issues and recommendations: use inclined pipes to limit urine stagnation; limit residence time by using the shortest network possible and limit bending in the network; do not use pipes with too small a diameter (at least 25-50 mm); never ventilate, to prevent high volatilization of NH_3 ; do not use metal pipes or tanks to prevent metals ions precipitations; and favor plastic materials.

For maintenance, he recommends regular cleaning by flush with hot demineralised water and acid, and occasionally cleansing with basic products for the organic matter accumulation. The maintenance is higher than with flush toilets, and consequently training of maintenance staff is an important stake.



Figure 3.20: Mineral deposit inside a PVC pipe for urine diverting toilets. Source: Brun (2019)

3.3.4.2 Appropriation by users

Etter et al. (2015) describe the case of the South African eThekweni municipality which in 2002 installed dried sanitation in the form of more than 80,000 urine diverting toilets. Ten years later, the municipality commissioned a study for feedback.

Before delving into details, the readers should be aware that the results are to be interpreted with cautious, because there is no control group to compare the urine diverting toilets to. For instance, in a randomized control trial studying the effect of subsidies given to people to maintain pit latrines in Northern Ghana, people practicing open defecation increased from 25% to around 50% after almost 2 years (Trimmer et al. 2022). This can be seen as a failure, but in the control group (not receiving any subsidies) the rate increased even higher, from 25% to almost 70%. This illustrates how difficult it can be in poor areas to maintain a functioning toilet infrastructure.

Back to the eThekweni municipality, 10 years after the installation of the dry urine diverting toilets, a major concern was odors, with a quarter of users complain-

ing about toilets smell. A quarter to a third of people were satisfied with their dry urine-diverting toilets, but more than half were not satisfied. 80% of toilets were not maintained properly, with broken items not repaired or improperly repaired, and most of them declared not having received education on these toilets usage.

The aspirations for flush toilets remained very high, with revealing statements (reported in Etter et al. (2015)): *"I am waiting for the Municipality to come and change those urine-diverting to flush toilets"*. The influence of role models is also perceptible: *"If it was such a good toilet then why is our councilor not using it or even our president"*. This is exacerbated by the historical apartheid background in South Africa (George 2011b). Flush toilets were associated to white settlements, while blacks had bucket latrines. A waterborne sewerage was thus associated to high status, even though it might not be the more adapted system in a water-stressed country.

More than 90% of people saw the urine-diverting toilets as temporary toilets. A key finding was that urine-diverting toilets were more accepted by younger people, who also were more committed in toilet maintenance.

At the other end of the chain of the source-separation chain are the farmers themselves, studied for instance in the case of Ile-de-France, characterized by large cereals crops (Brun et al. 2020; Brun 2018). They usually preferred solid fertilizers, requiring less volumes, closer to their current practices, which are expected contain less micropollutants, and with more standardized contents. But they could be eager to switch to liquid fertilization. A key expressed concern was the volumes to be transported and the storage capacities needed. Farmers were eager to get returns from real-world full supply chain, a proof through example, and to gain more hindsight. They report having numerous criteria when they evaluate urine-based fertilizers: cost, odors, perception by consumers and their neighbors, pathogens and micropollutants, norms and regulations. Finally, they would like institutions to support the practice, be it through discourse but also with clear regulations frameworks.

3.3.4.3 Pathogens and Micropollutants

Pathogens: Urine has a low load in pathogens, but it can be cross contaminated by contact with feces (Höglund et al. 1998). Regarding these pathogens concerns, the World Health Organization recommends dry excreta and fecal sludge to be stored 1-2 years, depending on the temperature, before safe use in the field (WHO 2006, executive summary, table 2). For urine alone, the recommended storage is 6 months (WHO 2006, executive summary, table 3). This 6-months threshold is seen as a limit for a safe use on all kind of crops; between 1 and 6 months, the WHO considers urine utilization on crops supposed to be processed or used as animal feed. The shorter storage duration for urine than for feces reflects the lower contamination of

urine with pathogens.

More practical guidelines for the safe use of Urine and Faeces in Ecological Sanitation Systems are described in Schönning et al. (2004). Besides treatments to reduce pathogens, a useful and more holistic framework is the WHO multi-barrier framework (Figure 3.21). Reduced risk can be reached by limiting contact at all stages: with adequate source separation systems, by applying excreta preferentially on non-food crops or on crops that are processes and cooked before ingestion, by respecting a withholding period...

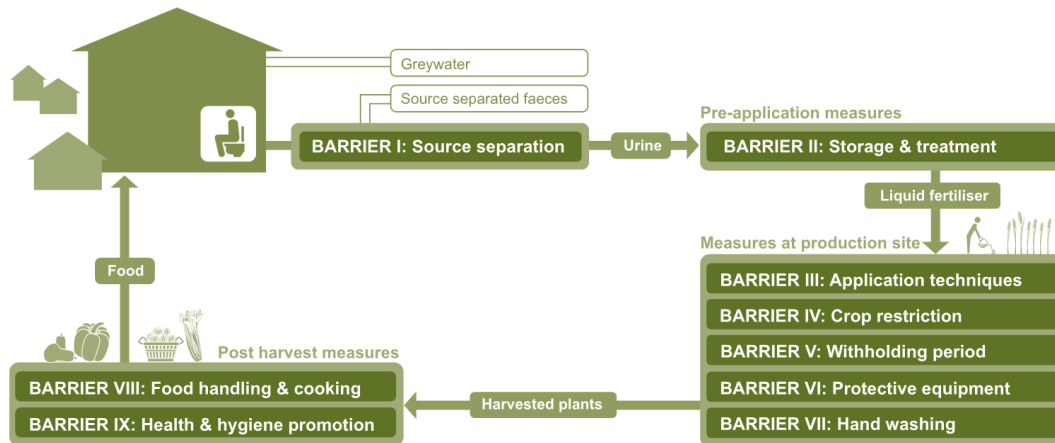


Figure 3.21: WHO multi-barrier framework. Source: Richert et al. (2010)

Drug residues and hormones: Drugs can be directly excreted but are often transformed in the body and are released in another form, named metabolite. Lienert et al. (2007a) and Lienert et al. (2007b) report that about two-thirds of pharmaceutical micropollutants are excreted via urine, after treatment by kidneys, but with high variability between people. A simple storage can reduce the amount of some micropollutants, but with a high variability, and some drug residues are barely impacted. To reach more systematic concentration decrease, further treatments are required, like activated carbon, which reduced all the tested drug residues by 90 to 99% in Etter et al. (2015).

The WHO considers that these micropollutants "*negative effect on the quantity or quality of crops are assumed to be negligible*" (WHO 2006, p120). Micropollutants in urine spread on crops could be transferred to surface and underground waters, to plants (and ultimately ingested by humans) or stay in the soil. Winker (2009), who produced a PhD on the matter of pharmaceutical residues in urine and their potential risks related to usage as fertilizer in agriculture, recommends "*that urine from people under medication should not be used for fertilization of food crops*".

But one needs to compare the relative risk of micropollutants in human excreta to other existing pollutants, already largely present in field crops, for instance pesticides. Moreover, drug residues and hormones are already present in livestock

excretions used to fertilize crops, sometimes in higher amount than in human excreta (Hammer et al. 2007), even though the drug used can be very different than human treatments (we can think of cancer treatment). Currently, there are no regulatory thresholds on drugs residues in livestock manure or wastewater treatment plant sludge.

Another reflection is to consider the current fate of micropollutants in the current system, where they are not treated in wastewater treatment plants. They are partially recovered in wastewater sludge, and can already reach agricultural fields by this mean, but most of them are simply discharged to surface waters. Yet these micropollutants tend to be way more metabolized and abated in the soil than in waters. This is because soils are better aerated than waters, promoting microbial activity and degradation (the same principle as activated carbon). UV rays from the sun also contribute to abating pharmaceuticals residues, as well as the high specific surface of soil particles (Richert et al. 2010).

Finally, if micropollutants are to be treated to prevent releasing them in the environment, it would probably be easier to treat them in concentrated excretions than after their dilution by a factor 100 in municipal wastewater. Finally, urine has very low heavy metals concentrations compared to amounts found in wastewater treatment plants.

3.3.4.4 Farming technical operation

Air pollution and greenhouse gases: When the ammonium/ ammonia equilibrium occurs, for instance in non-stabilized urine-based fertilizer, there are important risks of ammonia volatilization if the product is simply spread on the field. Existing methods to reduce volatilization with manure include using trailing shoe and open-slot injection machines (mean volatilization reduction of 65% to 80%), or trailing hose (35% mean reduction), but in each case with many variability (Webb et al. 2010).

Besides air pollution through ammonia, N-based fertilizers can also produce greenhouse gases. This is not specific to urine-based fertilizers and also concerns conventional synthetic fertilizers or livestock effluents. N_2O emissions happen during nitrification and denitrification. When the soil is well aerated, under aerobic conditions, NH_4^+ is first transformed into nitrates by nitrifying bacteria $NH_4^+ \rightarrow NO_3^-$. This process emits small amounts of N_2O . The second case when N_2O emissions happen when oxygen is scarce, under anoxic conditions, typically when soils are saturated in with water. In this situation, denitrifying bacteria can transform the nitrate into dinitrogen $NO_3^- \rightarrow N_2$, which also emits small amounts of N_2O . This process is exemplified in the experience of Martin et al. (2023b) on urine fertilizers, when N_2O emissions peaks occurred just after urine spreading (when NH_4^+ is

nitrified) and after rain events (NO_3^- denitrification under anoxic conditions).

Logistic issues: Then main challenge for farmers using non-concentrated urine based fertilizers is the mass concentration of nitrogen in urine, about 0.6% (6 gN/L), or even less if it has been diluted by some flush in toilets. But if the urine-based fertilizer is dehydrated, this will require minimal to no change in current farmers' practices. Synthetic mineral fertilizers like ammonium nitrate have a nitrogen mass concentration of 20-45%, meaning a farmer only needs to spread 0.35-0.9 tons per ha. By comparison, for a typical conventional fertilization in a cereal field of 180 kgN/ha, the farmer would need to spread about 30 tons of raw urine, or one to two orders of magnitude higher than with synthetic fertilizers. More realistically, farmers using urine would either: be integrated in lower inputs systems (like organic farming), and thus fertilize with less than 180 kgN/ha; or still bring a total of 180 kgN/ha, but urine would contribute only partly.



Figure 3.22: Top: slurry spreading with a slurry tanker, whose heavy weight can compact soil and prevent the tractor from entering the field if the soil is too wet. Bottom: slurry spreading from fixed slurry tanker, using a dragline, limiting soil compaction. Sources: Wikimedia Commons (2023c) and youtube (2023)

For conventional winter wheat, a typical cash crop in France, a starter fertilization generally occurs in late winter / early spring during march or April. The main fertilization occurs one month later during the growing stage, and a final one is used

to reach high protein content. Fertilizing using non-concentrated urine, with a slurry tanker on the field border connected to the tractor (Figure 3.22, bottom), can be difficult during the third application, when wheat has already significantly grown. Spreading urine during the first application is possible, but can be challenging depending on the weather conditions: if soils are wet, they will difficultly support a machinery with a 30 tons tanker (Figure 3.22, top), resulting in soil compaction. The second application may be more favorable, as it occurs during warmer times. Other crops fertilized during spring or summer include maize or rapeseed. Typical application rates for 1 spreading are around 50-100 kgN/ha.

Liquid urine-based fertilizers are similar to biomethane digestates or slurry: effluents rich in mineral nitrogen but typically less concentrated than commercial mineral fertilizers. One can imagine that urine can benefit from the structuring of the sector to handle digestates issued for methanizers, or the current practices to spread slurry, namely on the ground of farming equipment and practices.

Table 3.11: N concentration of urine-based fertilizers and quantity needed to bring 100 kgN/ha.

	N concentration		mass to bring 100 kgN/ha
	% of mass	kgN/t	tons of product/ha
non concentrated urine	0.1-1%	1-10	10-100
concentrated urine	1-10%	10-100	1-10
dehydrated urine	5-25%	50-250	0.4-2
synthetic fertilizers	20-45%	200-450	0.2-0.5

Reception in organic farming: In organic agriculture, where synthetic mineral fertilizers are forbidden, looping the nutrient cycle through excreta recycling seems virtuous on the biogeochemical stance. Yet the views on that matter from organic farming stakeholders are contrasted. While some agree with the nutrient looping and the absence of exogenous inputs philosophy, others think that organic farming should minimally rely on mineral fertilization (Marandet 2022). Most of the fertilization should be under organic form, and a living soil would release nutrients through mineralization.

If this reluctance does not impede urine usage in organic agriculture, it could complete the current organic farming practices. Organic agriculture mainly relies on organic fertilizers, in which nitrogen has first to be released through mineralization before plant uptake. However, in late winter/early spring, this mineralization process is limited by low temperature, and so crop growth is nutrient limited (crop growth is also limited by the low temperature itself). A mineral fertilizer such as urine could boost biomass production at this early stage and increase the final yield. Another possibility is that fertilizers based on human excreta could be an additional fertilization source in organic rotations, allowing an additional cash crop successions,

before the soil fertility is replenished with legumes fostering nitrogen biofixation in agro-ecological rotations.

3.3.5 Benefits of source separation

3.3.5.1 Agronomic

Unsurprisingly, as urine-based fertilizers contain nitrogen mainly in mineral form (urea, NO_3^- or NH_4^+ depending on the processes, see Table 3.7), its properties in the field are very similar to commercial mineral fertilizers (Richert et al. 2010). Martin et al. (2023b) found nitrogen fertilizer replacement value on the order of 70-120% compared to ammonium nitrate. The lower values were for simply stored urine, and the gap was mainly explained by NH_3 volatilization, which can reach up to one third of the spread nitrogen in the worst case, but typical values are more on the order of 10%.

3.3.5.2 Water savings

Depending on the type of toilets used for urine source separation, avoided impact also include different amounts of water that are not used for the flush. Current toilets flush are about 3 liters for urination and 6 liters for defecation, even though older systems in the 80s consumed about 10 liters, and sometimes more than 18 liters for even older toilets (Arocha et al. 2013).

There can be variability, but one can consider that with the SAVE urine-diverting toilets, out of the 3-6 liters flush, 0.2L goes into the urine compartment, which recovers 80% of urine, and the remaining 20% goes with feces into the sewers (Besson et al. 2021a). With 5 urinations and 1-1.5 liters of urine a day, this results in a factor 2 dilution of urine. With urine-diverting toilets, maybe people will mostly not flush for simple urination, resulting in less water consumption, but it is difficult to anticipate.

Concerning dry urinals and dry toilets, they will consume negligible amounts of water compared to current 3-6 liters per flush.

3.3.5.3 Impact on WWTP operations in hybrid systems

Urine represents 80% of the nitrogen inflow to wastewater treatment plants (Larsen et al. 1996), and nitrogen treatment has the strongest environmental impact in the facility operations. So wastewater treatment plants operations will strongly be influenced by even partial urine diversion. Several studies have looked at the consequence of urine diversion deployment on WWTP (De Faria et al. 2015).

Matar et al. (2022) compared a scenario where 40% of urine was source-diverted to the current system of WWTP in a large urban watershed. They hypothesized weekly collection of urine by trucks on a mean 10 km distance and a WWTP with nitrifying biofilter, operating with the French electricity mix. After urine recovery, they studied 3 treatments: partial nitrification + distillation; struvite precipitation and ammonia recovery by microbial electrolysis cell ; struvite precipitation followed by the nitrogen recovery by transmembrane chemisorption. Note that these analyses all include a urine treatment following collection. Simple spreading of raw urine necessitates handling of larger volumes, but has less environmental impact according to a life cycle analysis undertaken by Martin et al. (2023a).

Matar et al. (2022) found that a 40% urine diversion would result in a 30% ammonium load reduction in the WWTP. Consequently, 30% less aeration is needed for nitrification, also driven by a more favorable shift in COD:N ratio for denitrification (from 8.1 to 10.3). For denitrification, 35% less methanol is required (the studied WWTP normally consumes 3.4 kg of methanol per kg of nitrate denitrified). Additional benefits are less chemical reactant consumption, namely FeCl_3 for phosphorus treatment. The main gain was global warming potential due to avoided emissions of N_2O in WWTP and N fertilizer synthesis. But this is in the particular case of a nitrifying biofilter, with relatively high emissions of N_2O compared to activated sludge. In the case of a European electricity mix and a comparison to activated sludge, the benefit is dwarfed and even non-existent with the nitrification/distillation process. But one must remember that the current WWTP system + Haber-Bosch synthesis is already deeply optimized, while urine treatment processes could benefit from further optimization.

The originality of Matar et al. (2022) work was that they dynamically modeled the WWTP inflows on an hour to hour basis. This allows to see that urine diversion results in less absolute variability, allowing more stable processes in the WWTP. Finally, they argue that urine source separation would possibly allow "to avoid a potential expansion of the wastewater treatment plant, thus retrofitting it into a water resource recovery facility".

Besson et al. (2021a) went further and compared urine source separation treated with transmembrane chemisorption and struvite precipitation to a water resources recovery facility, a future more efficient WWTP producing energy, within the current European electricity mix. The water resources recovery facility has a better environmental impact than current WWTP, namely a 14% decrease in greenhouse gases emissions. Urine source separation has similar environmental performance as the water resources recovery facility, except on 2 main points: nitrogen recovery and climate change. With known technologies, the water resources recovery facility still fails to recover most of the nitrogen in municipal wastewater, because it is too

diluted. It also continues to emit N_2O during its N treatment processes. In the end, urine source separation with recovery emits about half as much greenhouse gases compared to the water resources recovery facility, also benefiting from avoided impacts from N fertilizers production.

3.3.6 Regulatory issues

3.3.6.1 In conventional agriculture

The legal status regarding excreta used as fertilizer in agriculture is unclear in France (ARCEAU 2023b). Urine and feces are not explicitly mentioned, and several legal frameworks can be considered if they are considered as a waste.

- First, the water law, dealing with sludge from sanitation systems, in particular sewage sludge from individual autonomous systems. This considers excreta as a waste, and it would be regulated as such. Namely, above a certain threshold, it would need to be integrated to a spreading plan.
- Second, the legal frame regarding fertilizing materials, which includes sewage sludge from wastewater treatment plants, with requirements regarding trace metal elements. This frame would also consider excreta as a waste.
- Another option is to use a treatment removing excretions from the waste status, for instance by urine storage which removes most of the pathogens (WHO 2006), or feces composting to produce an organic amendment.
- The final option is to transform them into a commercial product with marketing authorization, following a normalization process (for instance the VUNA fertilizer in Switzerland).

3.3.6.2 In organic agriculture

The case of human excreta in organic agriculture has been studied in depth by Marandet (2022). Her interpretation is that the European legislation prevents from using human excreta in organic agriculture: if a substance is not explicitly listed as authorized for organic agriculture, it is forbidden by default. To be included in the list, 2 conditions must be fulfilled: get a marketing authorization (or be exempted based on article L255-5 of the Code Rural et de la Pêche Maritime) and be listed in Annex II of Regulation 2018/848. The procedure to get the full authorization is summarized in her figure 3. But this interpretation has been challenged by other legal experts (Fabien Esculier based on Benoît Schmaltz, personal communication). Like for conventional agriculture, the legal framework for human excreta use in organic farming thus seems to be unclear.

While reusing and cycling nutrients is aligned with organic agriculture objectives, Marandet (2022) identifies some challenges, owing to the "spirit" of organic farming. The first one lies in concerns regarding micropollutants (drug residues, hormones...) in human excretions. Some treatment such as distillation or activated carbon could overcome this issues.

The second is the strongly mineral character of urine, when organic agriculture principles might prefer organic fertilizer. According to the logic "nourish the soil to nourish the crops", fertilizer in organic farming first have to be mineralized by microorganisms before plants can uptake nutrients, a stage skipped in the case of mineral fertilizers which directly nourish the plant. Marandet (2022) cites the article 5 of Regulation 2018/848 to support this view point, stating that external inputs in organic farming should be limited to "poorly soluble mineral fertilizers". Such a logic is valuable, but she identifies some contradictions with already enacted practices. Slurry or methanization digestates, both strongly mineral, are currently authorized in organic agriculture, as well as manure, if issued from non industrial livestock, even though they can receive and excrete drugs. If because of its mineral form, urine is not accepted as such in organic agriculture, one solution might be to authorize it after transformation, for instance incorporation in compost, in order to raise the organic matter content and respect the organic principle of "poorly soluble mineral fertilizers".

On the other hand, in favor of human excreta use, the article 5 of Regulation 2018/848 states that a general principle in organic farming is to use natural resources internal to the management system. The perimeter of the "management system" is not defined, but it can be argued that recycling human excreta is coherent with this logic.

Conclusion of Part I

Part I of this thesis explored the critical roles, global disruption, and current excreta management practices concerning nitrogen (N) and phosphorus (P), two essential nutrients for life and agriculture. These three chapters aimed to provide a comprehensive scientific context for the subsequent analyses of nutrient flows and their potential recovery in human excretions for agricultural use, which will be examined in Part II.

Chapter 1 opened with an exploration of the origins and biological significance of nitrogen and phosphorus. Both elements play indispensable roles in living organisms: nitrogen is a key component of amino acids and proteins, while phosphorus is critical for DNA, RNA, and energy transfer molecules such as ATP. The chapter examined their abundance in the universe, emphasizing phosphorus's relative scarcity compared to nitrogen. The focus then shifted to their ecological roles, particularly in nutrient cycling within ecosystems. Phosphorus, often bound in mineral forms, tends to be less mobile in the soil than nitrogen, which can readily leach into water systems due to its soluble nitrate form. Both nutrients, when present in excess, contribute to severe environmental issues. This includes eutrophication for N and P, and soil acidification, air pollution, water pollution, and greenhouse gas emissions for N. This chapter concluded by stressing the delicate balance between their essential biological roles and their potential as pollutants when mismanaged.

Chapter 2 explored how human activities have profoundly disrupted the natural biogeochemical flows of nitrogen and phosphorus, with a focus on historical and technological developments. The chapter detailed the XIXth-century emergence of industrial phosphorus fertilizers, derived from phosphate rocks, and traced the growth of the modern phosphorus industry. It emphasized the limited availability of high-grade phosphate reserves, the relevancy of the concept of *peak phosphorus*, and the geopolitical tensions surrounding this fossil resource. On the nitrogen side, the discovery and commercialization of synthetic nitrogen fertilizers were examined, with a particular focus on the Haber-Bosch process, which enabled mass production of ammonia from atmospheric nitrogen. While these industries drastically increased agricultural yields, they have also led to significant nutrient imbalances, resulting in losses and pollution. The chapter concluded by highlighting the resulting

planetary-scale disturbances in nitrogen and phosphorus cycles, and the concept of the *N planetary boundary*.

Chapter 3 provided a historical and modern overview of human excreta management, tracing practices from ancient civilizations to contemporary wastewater treatment systems. Historically, there is evidence of excreta recycling for agricultural use, as seen in the nightsoil practices of East Asia and parts of Europe. However, with the advent of sewer systems and urbanization in the XIXth and XXth century, human excreta began to be viewed as a waste rather than a resource. The chapter examined modern wastewater treatment plants (WWTPs) and their nutrient removal efficiencies, emphasizing the limitations of conventional treatment systems in nutrient recovery. The concept of source separation was also introduced, where urine and feces are separately collected and processed for nutrient recovery. The chapter ends with the benefits and challenges of source separation implementation.

This Part I has provided a comprehensive foundation for understanding the biological importance, ecological roles, and human impacts on nitrogen and phosphorus cycles. It highlighted how current agricultural and waste management practices have contributed to nutrient imbalances, often resulting in pollution. Importantly, the chapters have underscored the untapped potential of human excreta as a nutrient resource. This context sets the stage for Part II, where the thesis will present original research results on nutrient flows in France (Chapter 4), assess the agronomic potential of nutrient recovery from human excreta (Chapter 5), and explore the logistical feasibility of implementing circular nutrient management strategies (Chapter 6).

Part II

Personal Results: Towards a circular management of N and P in human excreta

Chapter 4

French Sanitation system: current fate of excreted nutrients

Within the 3 chapters comprising this part II, I will focus the following questions: How much of the nutrients in human excretions is currently reused (Chapter 4) ? What would be their maximum agronomic potential (Chapter 5) ? And what are the (logistical) constraints to their use (Chapter 6) ?

Rough assessments of the fate of human excreta have already been made as part of national N and P mass balance flows (Häußermann et al. 2021; Hayashi et al. 2021; Hutchings et al. 2014; Papangelou et al. 2021), which do not focus only on the sanitation system but on all of the flows involved in a country: farming system and food supply, imports, exports...

Here I present what is, to my knowledge, the first N and P budget of a national sanitation system based on a complete dataset of the 20,000 wastewater treatment plants in France. In addition to the global budget, my detailed dataset allows an in-depth analysis of the temporal evolution and factors influencing wastewater treatment plants removal efficiencies.

Due to data availability and WWTP monitoring, I only focus on N and P; but ideally, one should expand the analysis to other nutrients (K) and micronutrients (S, Zn, Cl, Cu) (see Esculier (2018) Section 2.1.5 for a discussion on that matter).

The results presented here have been published in 2 papers: one for nitrogen (Starck et al. 2024a) and one for phosphorus (Starck et al. 2024b). The dataset underlying the results and its cleaning is stored on a Zenodo repository (Starck 2023).

abbreviations WWTP: WasteWater Treatment Plants; IAS: Individual Autonomous System; N: Nitrogen; P: Phosphorus.

4.1 Methods and sources

In this section, I describe the large datasets used for my N and P budgets (Table 4.1). I also present the individual parameters used as inputs for my nutrient budgets (Table 4.2). I discuss the uncertainties associated to each nutrient flow and these uncertainties are then combined using Monte-Carlo procedure; the obtained uncertainty values are summarized in Table 4.3, and are also presented on the main results Figure 4.11.

The core of my work consisted in analyzing the data of the 20,000 French WWTPs, issued from the 6 French Water Agencies and their respective watersheds: Artois-Picardie, Rhin-Meuse, Seine-Normandie, Loire-Bretagne, Rhône-Méditerranée, Adour-Garonne (see them in Figure 4.13a). From these datasets, a N and P mass balance is performed for each of the 6 French water agencies basins, then gathered to obtain the national balance. The budget focuses on metropolitan France (including Corsica but excluding overseas territories).

The procedure to generate, cleanup, and analyze the data is available as an interactive website¹ whose code is stored on a git repository². It is also stored on a permanent repository on Zenodo³.

All the nutrient flows, their sources, and the way to compute them are summarized in Figure 4.1 for greater clarity.

4.1.1 Datasets sources

There are over 20,000 WWTPs in France, and roughly 1,500 industrial facilities disclose discharging N and P in sewers. My estimates of nutrient flows discharges in sewers by industries, WWTPs' inflows and outflows, as well as the various destinations of WWTP sludge, rely on comprehensive datasets detailed hereafter (Table 4.1).

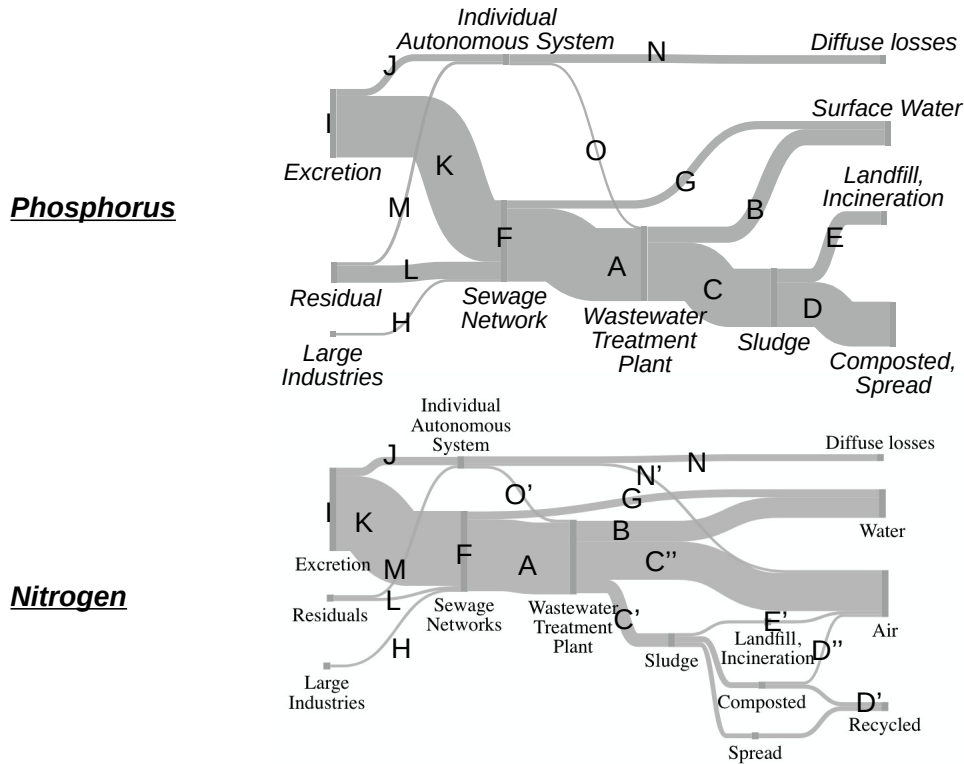
4.1.1.1 Data corrections

I scrutinized the datasets regarding sludge production, industry discharges and WWTPs inflows and outflows for obvious outliers. For each of the 6 water agency basins, I aggregate the nutrient flows at the basin scale for each year and look for unexpected large peaks on specific years. This allowed to detect obvious outliers. When it was possible to identify that the peak was simply due to a misplaced comma for that particular year (factor 10, 100, or 1,000), I corrected the entry; otherwise,

¹<https://thomas-starck.github.io/n-p-sanitation-flows/>

²<https://github.com/Thomas-Starck/n-p-sanitation-flows>

³<https://zenodo.org/doi/10.5281/zenodo.7990171>



Phosphorus

Nitrogen

Equation

- A: $\sum_{WWTP} Incoming\ Nutr_{WWTP}$
- B: $\sum_{WWTP} Outgoing\ Nutr_{WWTP}$
- C: $A - B + O$ C': $C * ratio_{N:P} + O$
- C'': $A - B - C'$
- D: $C * \%_{composted + spread} = C * \frac{\sum_{WWTP} sludge_{composted + spread}}{\sum_{WWTP} sludge_{total\ produced}}$
- D': $C * (\%_{compost} * (1 - \%_{compost\ volatil}) + \%_{spread})$
- D'': $C * \%_{compost} * \%_{compost\ volatil}$
- E: $C - D$ E': $C - D' - D''$
- F: $\frac{A}{1 - \%_{sewer\ discharge}}$
- G: $F * \%_{sewer\ discharge}$
- H: $\sum_{industries} Discharged\ Nutr_{industries}$
- I: $\sum_{sex, age} Population_{sex, age} * Ingested\ P_{sex, age}$
- J: $I * \%_{pop\ indiv\ autonomous\ system} * (1 - \%_{excretion\ not\ at\ house})$
- K: $I - J$
- L: $F - K - H$
- M: $L * \frac{\%_{pop\ indiv\ autonomous\ system}}{1 - \%_{pop\ indiv\ autonomous\ system}}$
- N: $(J + M) * \%_{underground\ diffuse\ losses}$ N': $(J + M) * \%_{volatilization}$
- O: $J + M - N$ O': $J + M - N - N'$

Dataset or Parameter source

- Outgoing $Nutr_{WWTP}$ Different water agencies databases
- Incoming $Nutr_{WWTP}$ Different water agencies databases
- O added to C flow to ensure mass conservation at all steps. Since O is very small, does not influence result.
- $ratio_{N:P} = 2$ French literature review on sludge content (Fuchs 2014)
- $\sum_{WWTP} sludge$ Portail Assainissement Collectif (2023) database
- $\%_{compost\ volatil\ for\ N} = 30\%$
- $\%_{sewer\ discharge} = 10\%$ Estimation from different water agencies
- Discharged $P_{industries}$ GERE database
- Ingested $P_{sex, age}$ INCA3 study (Dubuisson et al. 2019, data.gouv 2021)
- Population $_{sex, age}$ INSEE (2022)
- $\%_{pop\ indiv\ autonomous\ system} = 15\%$ weighted average of the 6 water agencies figures reported in their *Etat des Lieux* assessments
- $\%_{excretion\ not\ at\ house} = 20\%$ Estimation
- $\%_{underground\ diffuse\ losses\ N} = 55\%$ $\%_{volatilization\ N} = 20\%$
- $\%_{underground\ diffuse\ losses\ P} = 85\%$ Catel et al. (2017); Risch et al. (2021)

Figure 4.1: Equations used to establish the N and P mass balance, and the datasets and parameters associated.

Table 4.1: Data sources used for the N and P budgets.

parameter	source	
Nutrients discharge to sewers from each industry facility	GEREP database, nonpublic extension of the open access georisque French database, part of the European Pollutant Release and Transfer Register (E-PRTR).	
Population by age and sex for each French city	INSEE (2022)	
Sludge production and destination for each WWTP.	Portail Assainissement Collectif (2023)	
Nutrients flows in and out of each individual wastewater treatment plants, for each water agency basin)	Artois–Picardy basin years 1992–2021	Artois–Picardy agency website
	Rhine–Meuse basin, years 1996–2021	Rhine–Meuse agency website
	Seine–Normandy basin, years 2015, 2016, 2018, 2020	Etat des lieux (status reports) data, only for 2015, 2016, 2018, 2020 communicated through mail. We also have the SIAAP data over 2007–2021, consisting of the 6 largest WWTP representing 50% of the basin nutrient flows.
	Loire–Britanny basin years 2005–2021	Communicated by email, publicly shareable
	Adour–Garonne basin years 2000–2020	Adour–Garonne agency website, link for nutrient flows discharge and link for WWTP description
	Rhone–Mediterranean basin years 2009–2020	Rhone–Mediterranean agency website

the outlier was not considered in the analysis and the entry was discarded from the final dataset. The outliers identification and correction process is detailed on my website repository. See for instance the process for the Adour-Garonne basin on *WWTP flows preparation – > 05 Adour-Garonne – > Data Cleaning – > outliers correction*.

For P flows in and out of WWTPs, I identified a dozen obvious outliers out of the 20,000 French WWTPs, over a decade or more (depending on the basins). I also identified a dozen outliers for N. For sludge quantities production, I identified 25 obvious outliers for the 20,000 WWTPs over the 7 years of reported data. For industry discharge, I found 3 obvious outliers for the 1,000 facilities over 1 decade of data.

4.1.1.2 Measurement uncertainty

A French report on extended measurement uncertainties on automatic sampling operations in wastewater found that uncertainties for phosphorus and nitrogen were $< 10\%$ (Lepot et al. 2022). That is my base rate uncertainties for flows whose estimation relies on direct measures of N and P. This concerns nutrients in and out of WWTPs (flows A and B on Figure 4.1) and industries discharge to sewers (flow H). In addition, there is a broader uncertainty for each flows, due to reporting errors and perhaps incomplete reporting. I discuss the uncertainty specific to each flow in the following section, and how it modifies my $< 10\%$ measurement uncertainty base rate.

4.1.1.3 Nutrients inflows and outflows of wastewater treatment plants (flows A and B)

Flow value. France counts over 20,000 WWTPs, with data on the annual average nutrients inflows and outflows for each station. The temporal availability of the data varies for each basin, but spans at least one decade (maximum span 1992–2020 for Artois–Picardy, minimum span 2009–2020 for Rhone–Mediterranean) except for the Seine–Normandy basin, where data was only available for the years 2015, 2016, 2018 and 2020. For this basin, I retrieved additional data over 2007–2020 from the SIAAP (Syndicat Interdépartemental pour l’Assainissement de l’Agglomération Parisienne), the syndicate in charge of 6 of the largest WWTPs in the Paris region, which handle half of the Seine-Normandy’s sewage nutrients.

In the following, nutrient inflow refers to nutrient entering the WWTP, in ton per year (flow A on Figure 4.1). Nutrient outflow refers to nutrient discharged by the WWTP in water (flow B on Figure 4.1). Phosphorus stored in sludge (flow C) is assumed to be the difference between phosphorus inflow and outflow: $P_{\text{sludge}} =$

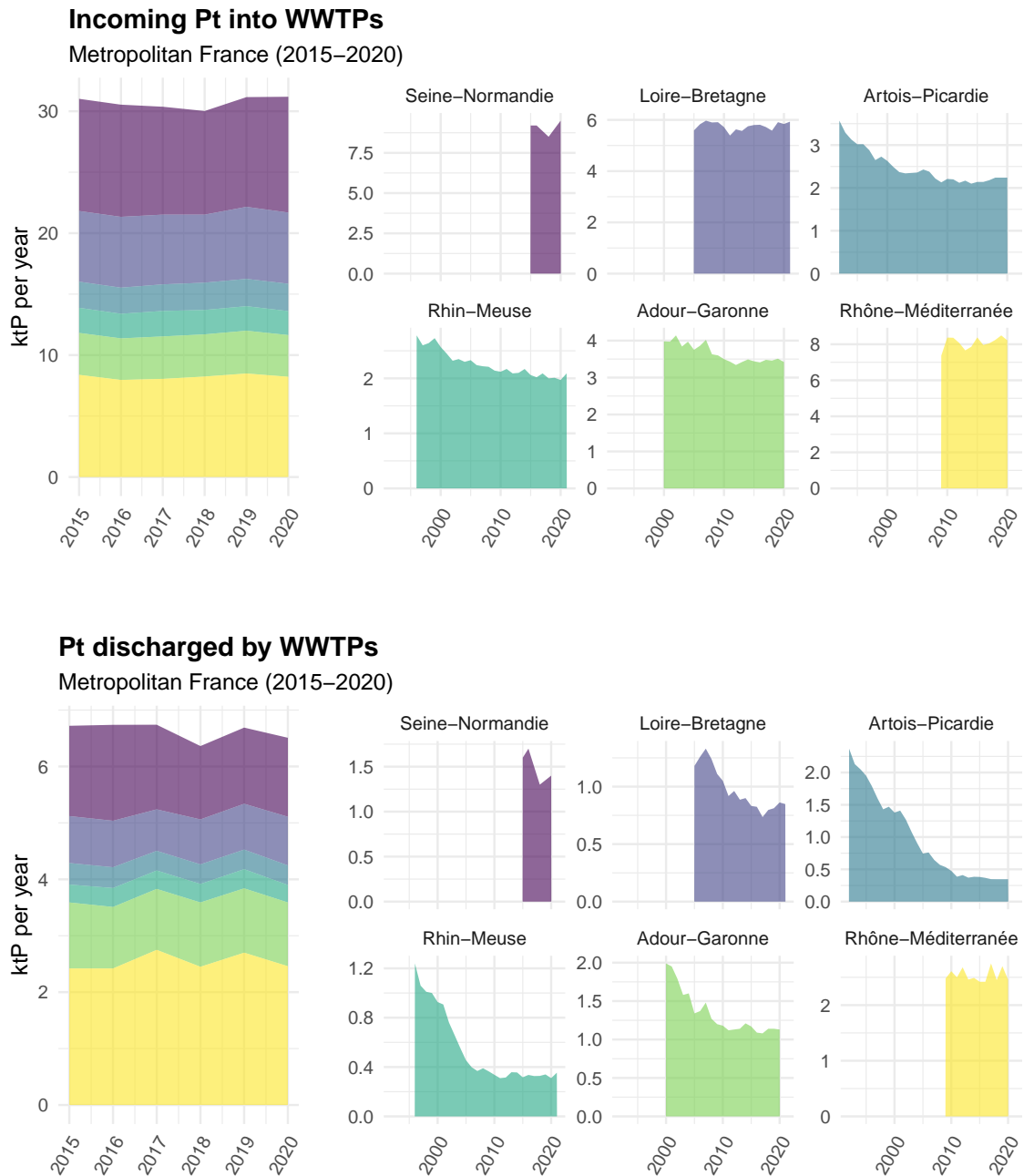


Figure 4.2: Top: Reported incoming P flows into Waste Water Treatment Plants (WWTPs) at the national scale (left) and in the 6 water agencies (right), after data cleaning. For the national scale, only the reference period (2015-2020) is shown; for the 6 basins, all available data is shown. Actual flows may differ as reporting rates increase over time. Bottom: same but for discharged P flows. Note the different vertical scale.

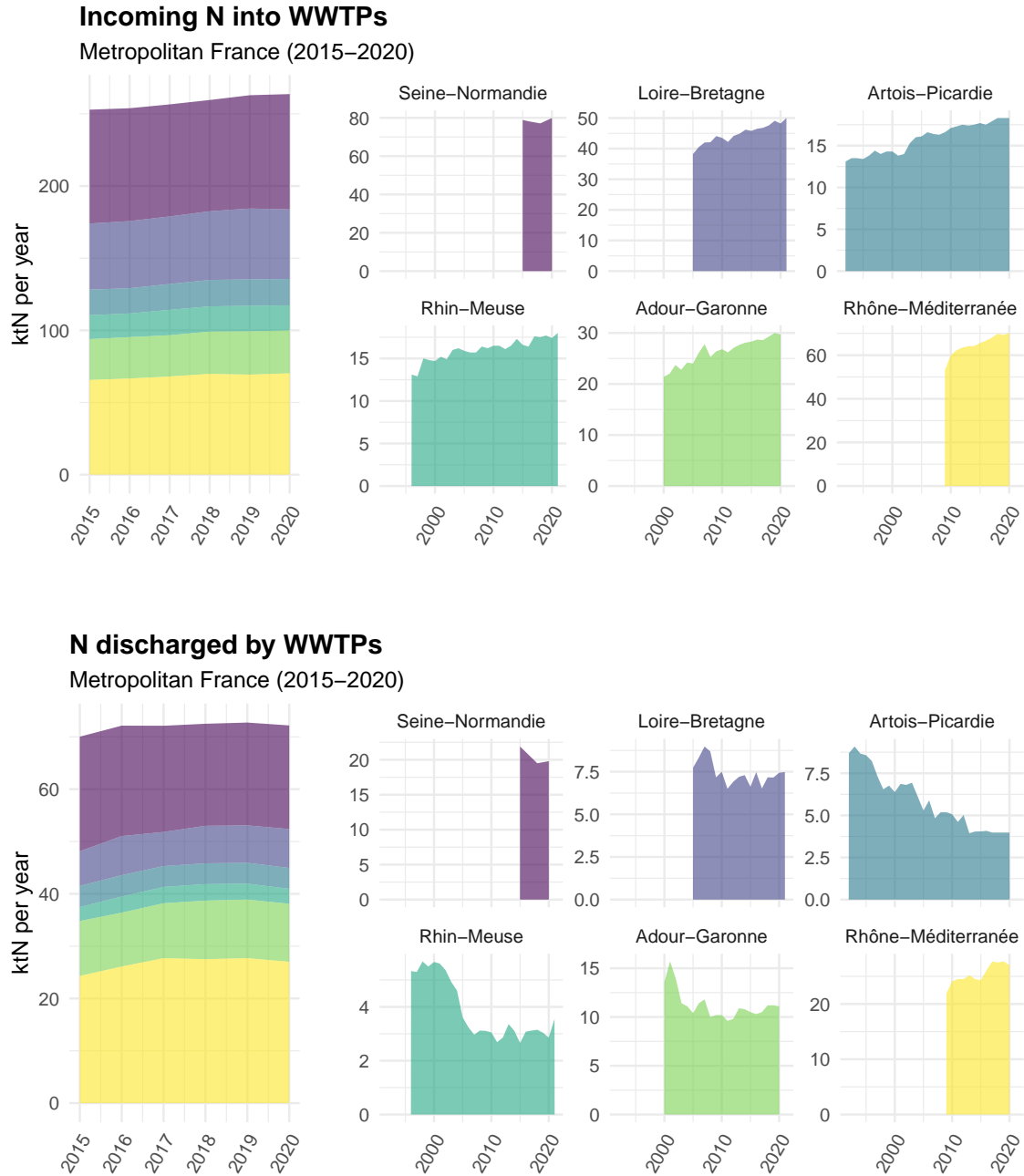


Figure 4.3: Top: Reported incoming N flows into Waste Water Treatment Plants (WWTPs) at the national scale (left) and in the 6 water agencies (right), after data cleaning. For the national scale, only the reference period (2015-2020) is shown; for the 6 basins, all available data is shown. Actual flows may differ as reporting rates increase over time. Bottom: same but for discharged N flows. Note the different vertical scale.

$P_{\text{in}} - P_{\text{out}}$. Deducting nitrogen amount in sludge (flow C') is not as straightforward, as most of the treated N following denitrification is sent back in the air as dinitrogen (flow C''). This is detailed in Section 4.1.2.3.

The nutrient removal efficiency of a WWTP is defined as

$$\text{Efficiency}_{\text{removal}} = 1 - \text{nutrient}_{\text{out}} / \text{nutrient}_{\text{in}}$$

Flow uncertainty. In the dataset used to quantify the WWTP nutrient flows, the largest stations, with a capacity >100,000 population equivalent, are monitored at least every week, and often every day (Légifrance 2015). They represent ~50% of the total French WWTP capacity. Facilities from 10,000 to 100,000 population equivalent, representing 30% of the total capacity, are monitored at least every month. So, for 80% of the national flow, there is a high certainty about the yearly-averaged reported value.

After data cleaning, the year-to-year flows variability at the basin scale is small and is on the order of 10% maximum (Figure 4.2 and 4.3). Considering this and the < 10% measure uncertainty on nutrient flows in wastewater (Lepot et al. 2022), I use 10% as the uncertainty for incoming and outgoing flows.

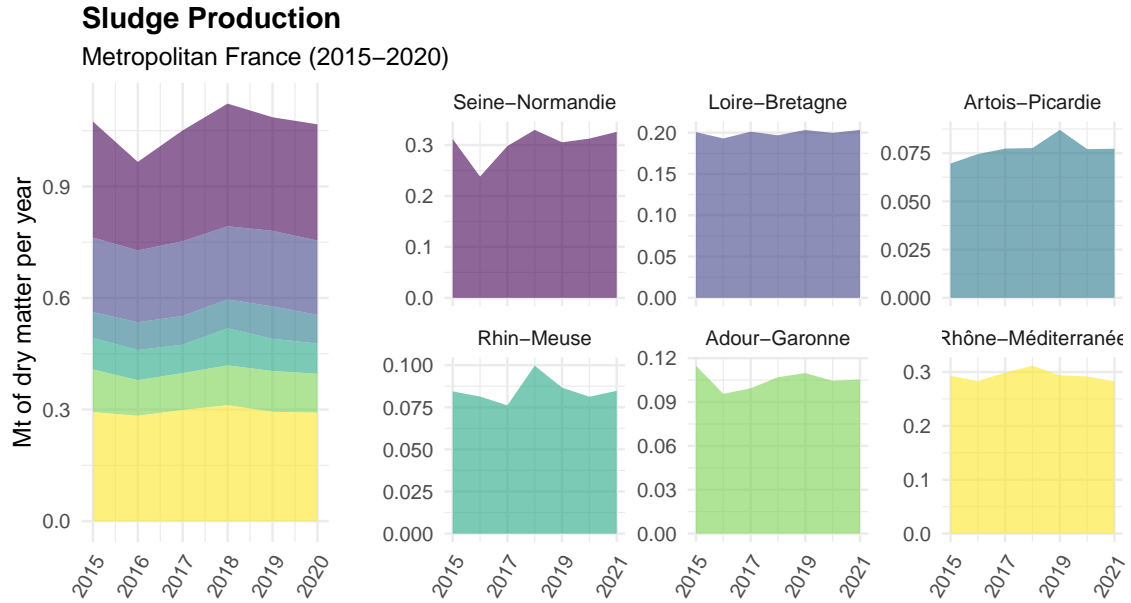
4.1.1.4 Sludge production and destination (flows C, D, E)

Flow value. Annual sludge production (without chemical reagents) for each WWTP and their destination (direct spreading, compost, incineration or landfill) is reported for each facility in the French sanitation portal database (Portail Assainissement Collectif 2023), and at the national scale in Eurostat (Eurostat 2022). National sludge production is consistent between these 2 sources over the whole period, around 1-1.1 Mtons of dry matter per year.

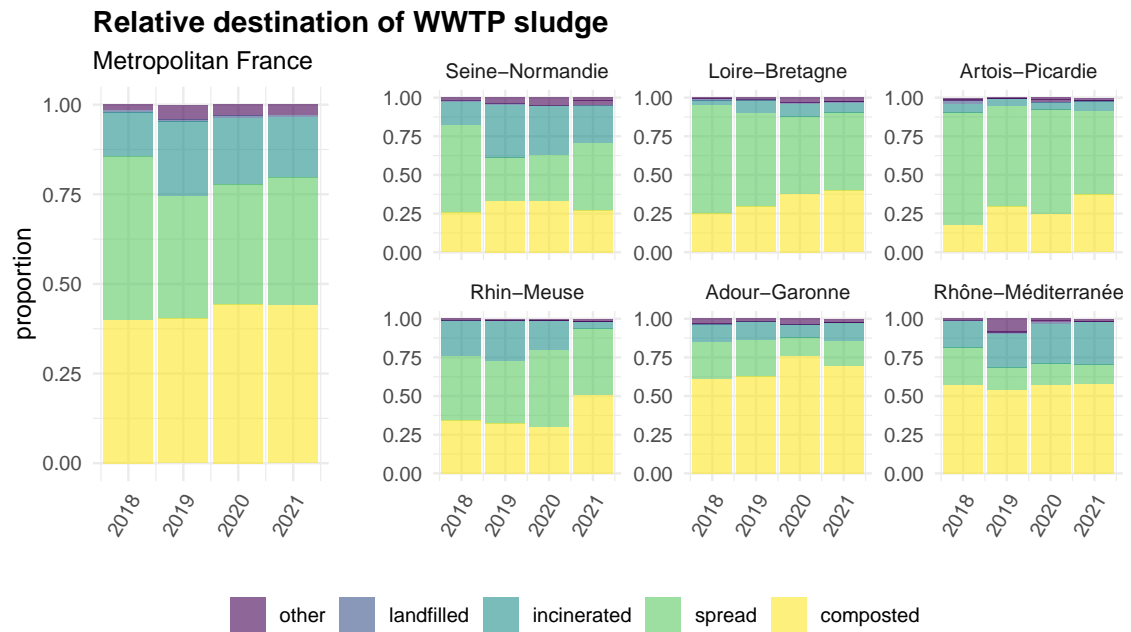
From 2019 to 2021, where sludge destinations reporting is stabilized and coherent (Figure 4.4b), approximately 45% of sludge is composted and 30% directly spread on crops at the national level (flows following flow C and C' in Figure 4.1).

I averaged destination amounts over the 2018-2021 period because data before these dates is barely reported and inconsistent.

Flow uncertainty. After correction, the year-to-year sludge production variability at the basin scale is small, on the order of 10% maximum, except in 2016 for Seine-Normandie (Figure 4.4a). The relative destination of sludge is also rather constant starting 2018, with about 45% of sludge composted and 30% directly spread on crops at the national scale (Figure 4.4b). I thus use a 10% relative uncertainty for sludge production and destinations.

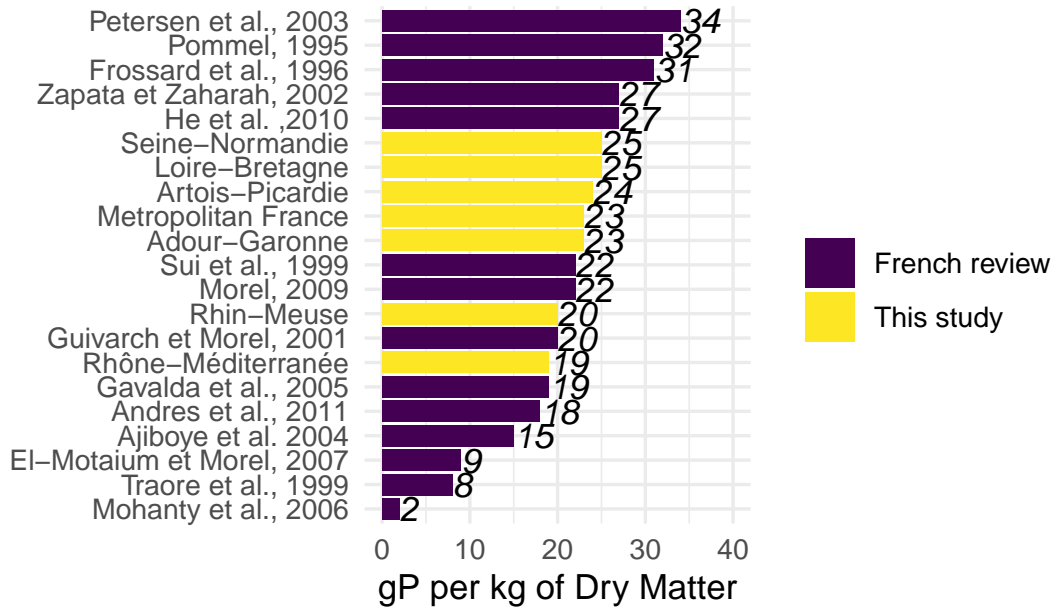


(a)

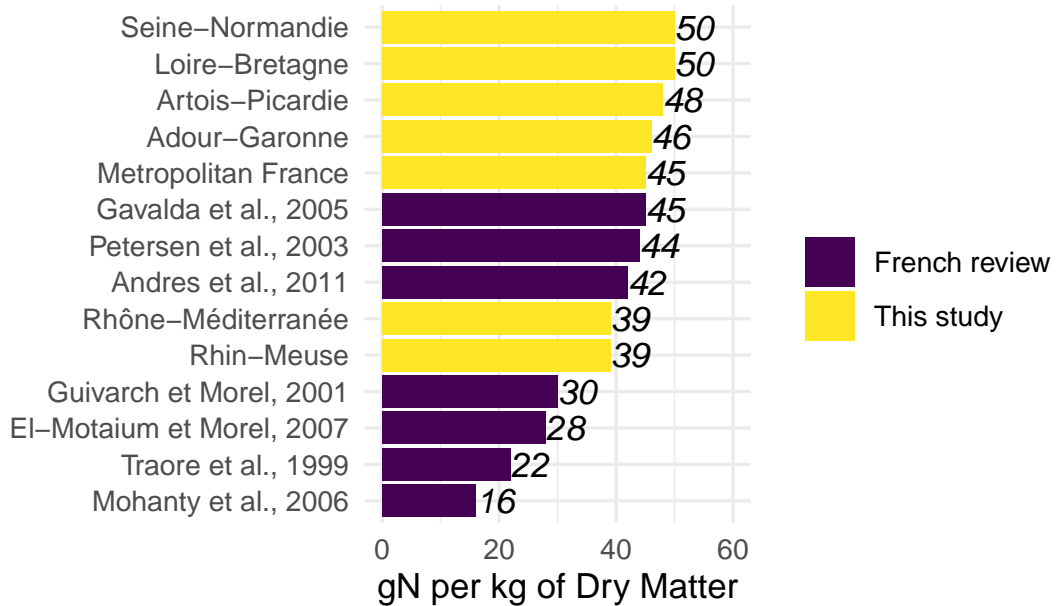


(b)

Figure 4.4: Top: Reported Waste Water Treatment Plants (WWTPs) sludge production at the national scale (left) and in the 6 water agencies (right) over the 2015–2020 study period, after data cleaning. Bottom: Reported Waste Water Treatment Plants (WWTPs) sludge relative destination at the national scale (left) and in the 6 water agencies, after data cleaning. Only the 2018–2021 period is shown, as previous years have very scarce and incoherent data.



(a)



(b)

Figure 4.5: Top: Comparison of sludge P content in Houot et al. (2014) (purple bars) and the value obtained from our P budget (yellow bars), at the national scale and in the 6 water agencies basins. Bottom: same for N.

The sum of the reported sludge destination quantities $r = \sum_i d_i$ does not cover the whole production: only 0.9–0.95 Mt were reported in 2018–2021 across all destinations, compared to 1–1.1 Mt for sludge production p . Therefore, I extrapolate the total amount of sludge sent to each destination i from the total production quantities, d_i^p , considering that the fate of missing sludge follows the same distribution as the reported quantities.

My results are comforted by the comparison to a French collective scientific expertise review on sludge composition (Houot et al. 2014). Combining my results on sludge production quantities and P in sludge ($P_{\text{sludge}} = P_{\text{in}} - P_{\text{water}}$) allows to estimate the mean P sludge concentration, for each basin and at the national scale. The values are within the median range of the ones reported by Houot et al. (2014) (Figure 4.5a).

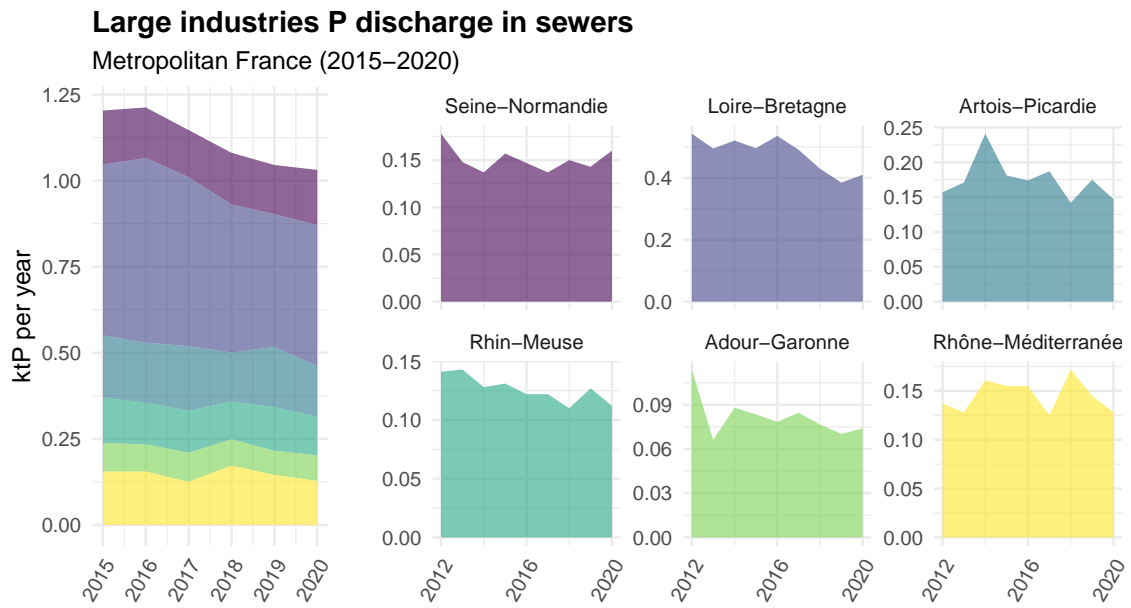
As the N treated in WWTP can end up in sludge but also in the air, N_{sludge} cannot simply be deducted from N_{in} and N_{out} . I estimate it from a sludge N:P molar ratio = 2 based on Houot et al. (2014) review, detailed in Section 4.1.2.3. The obtained values for N are consistent with the upper range reported by Houot et al. (2014) (Figure 4.5b).

4.1.1.5 Large industries discharge in sewers (flow H)

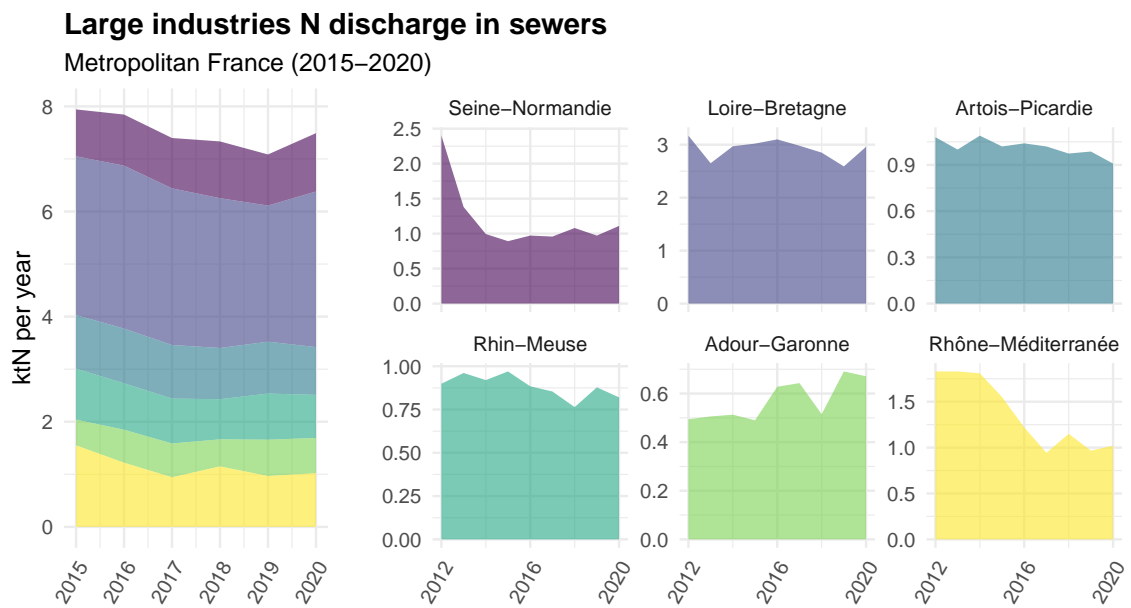
Flow value. The industries nutrient annual discharge to sewers (flow H in Figure 4.1) was estimated using the GERE database, provided by the Direction Générale de la Prévention des Risques from the Ministry of Ecological Transition. The database reports around 1,500 industrial facilities discharging nitrogen and phosphorus to sewers.

I chose to use the non-open GERE database, provided by the French *Direction Générale de la Prévention des Risques*, rather than the open-access “Géorisques” database (Géorisques 2023) that also reports industrial facilities discharging pollutants as part of the European Pollutant Release and Transfer Register (E-PRTR). “Géorisques” data only reports facilities discharging more than 50 tons of N or 5 tons of P per year (Légifrance 2008), but the non-public GERE data also includes facilities below these threshold. While “Géorisques” only reports a few dozen facilities discharging N and P in sewers, the GERE database reports about 1,500 facilities, which increases the N and P flow roughly by a factor 2.

Flow uncertainty. There is little year-to-year variability over the 2015–2020 period after data correction (Figure 4.6). Based on this little variability and the nutrient flows measurement uncertainties of < 10% (Lepot et al. 2022), I chose a 10% uncertainty for this flow.



(a)



(b)

Figure 4.6: Reported P (top) and N (bottom) flows discharged by large industries at the national scale (left) and in the 6 water agencies (right), after data cleaning. Note the different vertical scales.

4.1.1.6 French population (impacts flow I)

Flow value. I used data from the French National Institute of Statistics and Economic Studies (INSEE 2022), which provides information on the population in 2018 in each city together with the age distribution. Combined with data on excretion of N and P capita (Section 4.1.2.1), this allows to obtain national N and P excretions (flow I).

Flow uncertainty. The following uncertainty analysis for population was performed by Tanguy Fardet. Between 2015 and 2020, the French population grew from 64.3 to 65.3 million residents. In the analysis, we used the population census from 2018, with 64.8 million residents, hence a maximum error smaller than 1%.

Every year, France receives around 70 million tourists who come to visit the country, staying on average a week⁴. To this, one can add around 10 million tourists who transit through France from non-neighboring countries and around 100 million more who cross the border from a neighboring country, in both cases for a single day. Taken together, this represents an increase of around 9 days worth of excretion per capita over a year. This is partly compensated by the number of French residents going abroad for their holidays, which would represent around 250 millions days before COVID-19 so roughly 4 days per capita.

Overall it means that we may underestimate the national input to the sanitation systems from excreta by 2%. I will neglect this uncertainty, as it is negligible compared to the other flows uncertainties, and especially to the uncertainty of N and P excretions per capita.

4.1.1.7 Reference years for the budget

I calculated the nutrient balance flows for each water agency, using averaged quantities of industries discharge to sewers and WWTP nutrient inflow and outflow over the 2015–2020 period. For sludge destination, I average over the years 2018–2021, as data before this period is inconsistent and barely reported.

4.1.2 N and P budget components and parameters

In this section I describe the different punctual parameters of the N and P balance (Table 4.2), the hypotheses made for estimating some of them and the associated uncertainties, estimated by qualitative comparison to literature values.

⁴<https://www.insee.fr/fr/statistiques/1374552>

Table 4.2: Parameters used for the nutrient mass balance of the sanitation system.

Parameter	Value	Source
Direct discharge and losses from the sewers at the basin scale (% of N or P flow entering the WWTP)		
Artois-Picardie	20%	Water Agencies
Rhin-Meuse	20%	
Seine-Normandie	10%	
Loire-Bretagne	15%	
Adour-Garonne	7%	
Rhône-Méditerranée	7%	
France	10%	Combined basins
Total population (million) and % not connected to sewers		
Artois-Picardie	4.8M, 15%	Water Agencies
Rhin-Meuse	4.3M, 6%	
Seine-Normandie	19M, 7%	
Loire-Bretagne	13M, 24%	
Adour-Garonne	7.8M, 30%	
Rhône-Méditerranée	16M, 30%	
France	65M, 18%	Combined basins
Individual Autonomous Sytem (IAS) mass balance for <i>Phosphorus / Nitrogen</i>		
air	0% / 22%	based on
sludge	15% / 5%	Catel et al. 2017 and
underground diffuse losses	85% / 73%	Risch et al. 2021
Ingestion by French people <i>gP/day ; g protein/day</i>		
female, ≤10 years old	0.96 / 56	INCA3 study Dubuisson et al. 2019 data.gouv 2021
female, 11–18 years old	1.09 / 68	
female, 18–44 years old	1.11 / 71	
female, 45–64 years old	1.12 / 74	
female, ≥65 years old	1.03 / 69	
male, ≤10 years old	1.01 / 58	
male, 11–18 years old	1.32 / 84	
male, 18–44 years old	1.41 / 94	
male, 45–64 years old	1.42 / 96	
male, ≥65 years old	1.32 / 96	
Other parameters		
Conversion factor gN per g protein	1/6.25	Macleon et al. 2003
N:P molar ratio in sludge (basin and national scale)	2	Houot et al. 2014
For people not connected to sewers, share of excretions in sewers, (i.e. excretion in public spaces connected to sewers)	20%	estimation

4.1.2.1 Nutrient excretion (urine and feces) from French population (flow I)

Flow value. Data related to protein and phosphorus daily ingestion by French citizens is reported in the INCA3 study (Dubuisson et al. 2019; data.gouv 2021), disaggregated by age and sex categories. I convert the protein contents to nitrogen using a 1/6.25 factor for the mass N/protein ratio (Maclean et al. 2003). I considered no N nor P accumulation in adult bodies and, that all ingested N and P is excreted (Leach et al. 2012). Excretion out of the sanitation system is neglected (diapers, skin and breath volatilization, open urination, etc). For context, an assessment in the Ile-de-France region estimated that diapers for babies and old people may represent 3% of the total excretions (Villaseñor Yapur et al. 2022); this flow is generally not used in agriculture and directly incinerated.

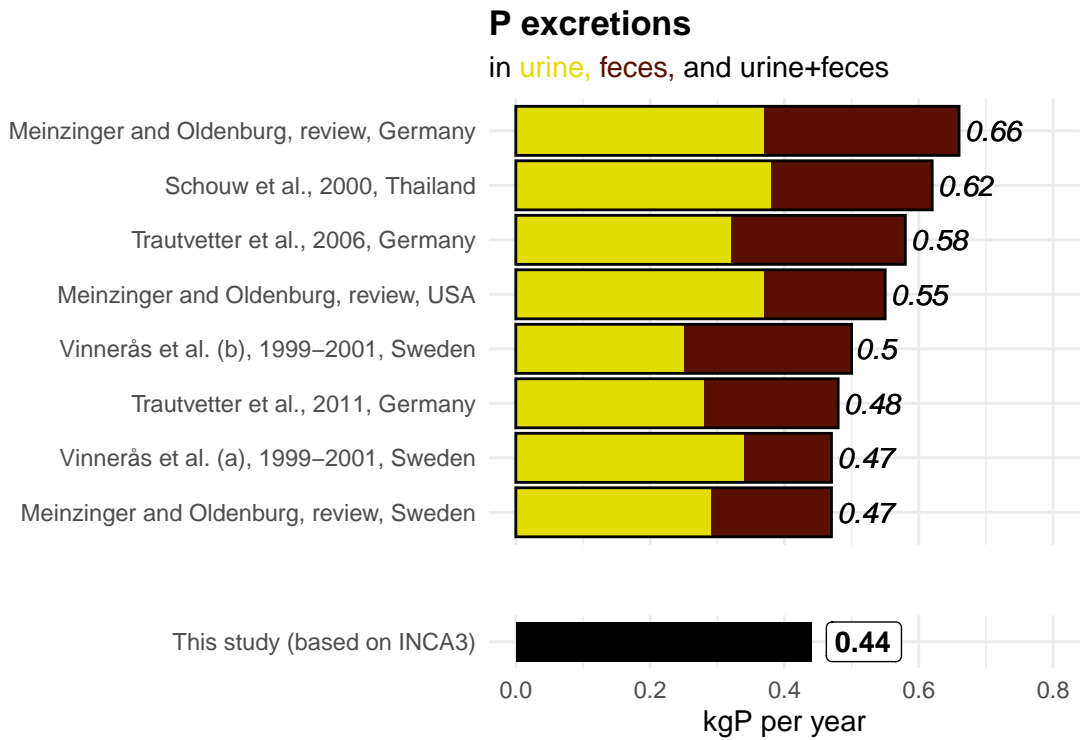
This ingestion data is coupled with INSEE data describing the French population by city and by age (INSEE 2022), to determine N and P excretions for each water agency basin and for metropolitan France (flow I in Figure 4.1). This results in an averaged national excretion of $0.44 \text{ kgP}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$ and $4.6 \text{ kgN}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$.

Flow uncertainty. The following uncertainty discussion is based on the work of Tanguy Fardet. The calculation of N excretion by the French population is subjected to two main sources of uncertainty: conversion from protein to nitrogen content and the approximation that all nitrogen intake is excreted. For P only that latter approximation comes into play.

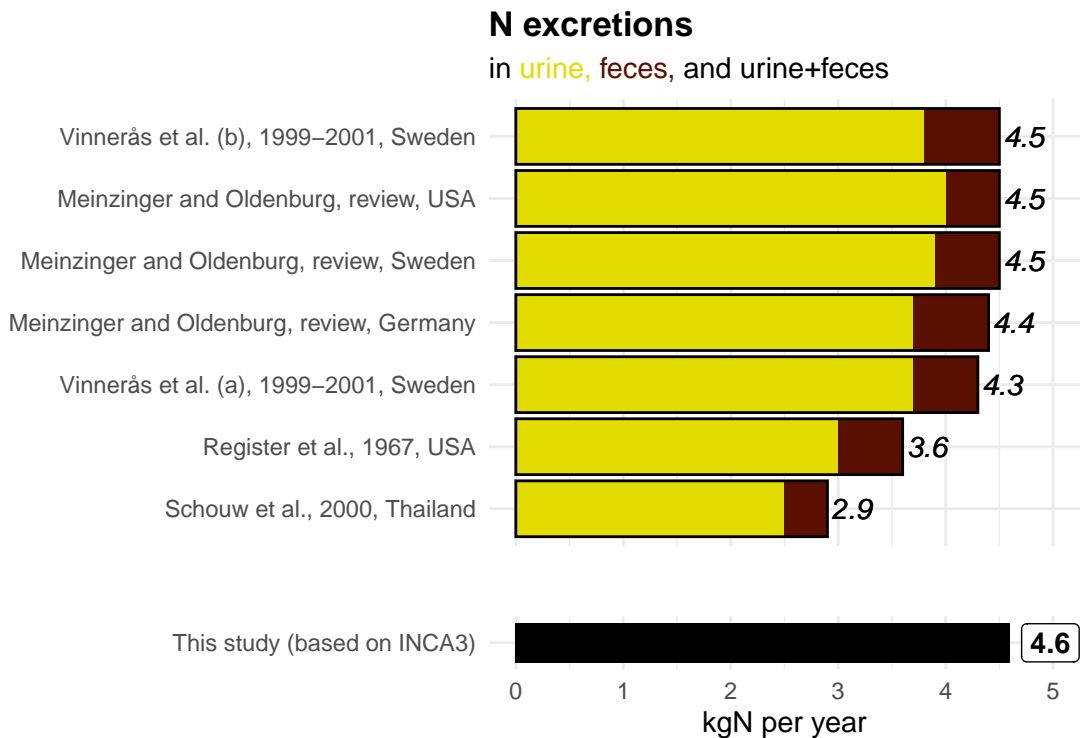
For protein to nitrogen conversion, we chose to keep the “standard” 6.25 ratio as more recent publications criticizing this factor focused on the reverse operation (computing protein from nitrogen content), whereas the 6.25 ratio seems adequate to quantify nitrogen from animal protein (Mariotti et al. 2008), which is the dominant protein source in France. Searching for nitrogen to protein ratios in the literature we found only one reference quantifying nitrogen and amino acids in cereals (Fujihara et al. 2008), which gave ratios between 6 and 6.45 for the most common varieties eaten in France (rice, wheat, flour, oatmeal, quinoa), with the exception of corn flour at 7.

Regarding nitrogen retention in the body, we neglect it for adults, which should be a reasonable approximation on average. We make the same approximation for children and adolescents, for which (Jönsson et al. 2004) found a mean storage rate of 2% between 2 and 17 years. This retention during growth translates into a mere 0.4% total retention over a 80 year life.

Part of the excreted N is also lost from other miscellaneous pathways (sweat, nails, hair, sloughed skin, body secretion), estimated at 8 mg/kg/day (Pellet 1990). For a mean national body weight of 60–70 kg, this would give $\sim 0.5 \text{ gN}\cdot\text{day}^{-1}\cdot\text{cap}^{-1}$ or



(a)



(b)

Figure 4.7: Top: Comparison of our study value for P in excretions (black bar) to actual measurements in the literature, discriminated between urine (yellow) and feces (brown). Bottom: same for N.

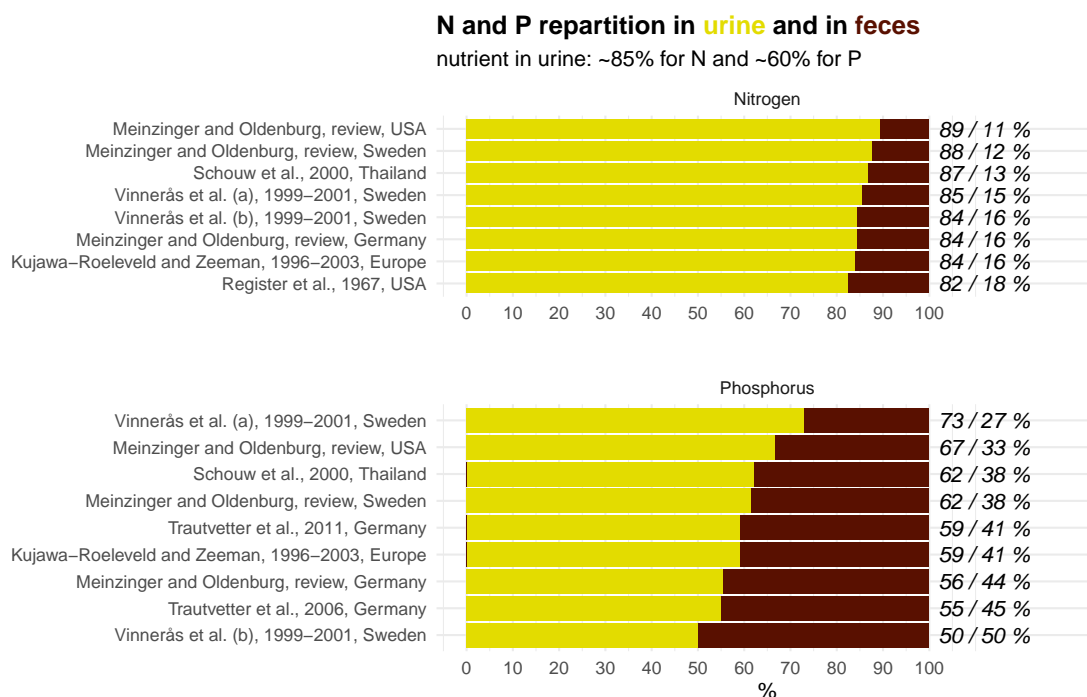


Figure 4.8: Distribution of excretions between urine and feces in our literature review for Nitrogen (top) and Phosphorus (bottom).

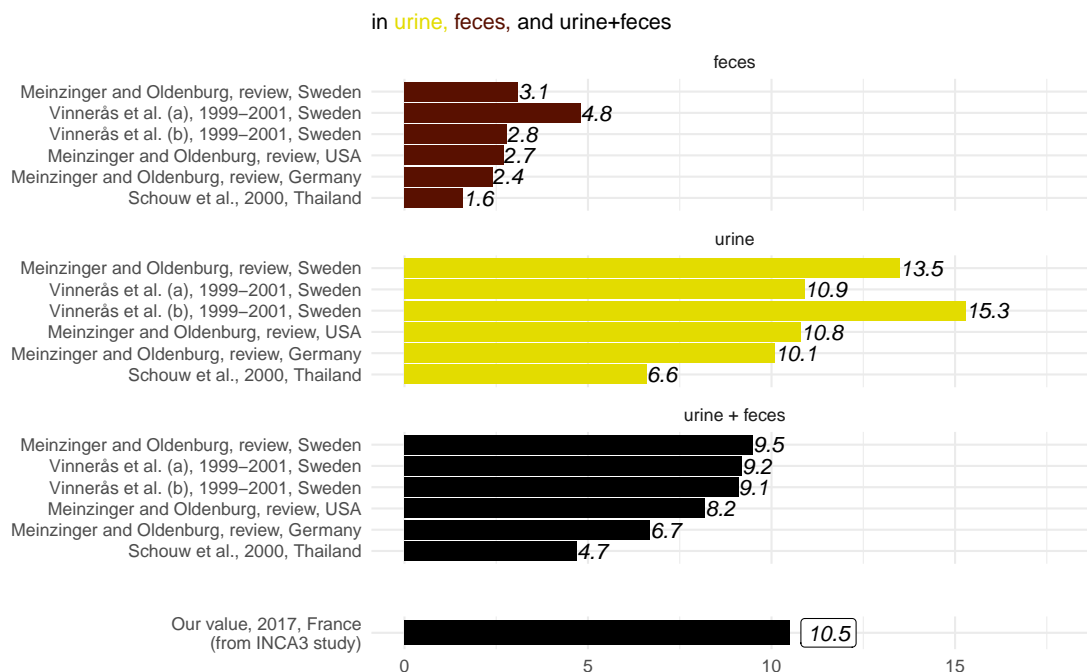


Figure 4.9: Molar N:P ratio in our literature review in feces (brown), urine (yellow), and total excretions (black). The bottom value is the one of our study, based on the INCA3 survey.

4% of daily intakes, part of which would still go in sewers when people wash themselves. Overlooking miscellaneous N losses and N body storage tend to overestimate N excretions by a few percent.

In the end, our value for averaged N excretion at the national scale ($4.6 \text{ kgN}\cdot\text{year}^{-1}\cdot\text{cap}^{-1}$, corresponding to $12.6 \text{ gN}\cdot\text{day}^{-1}\cdot\text{cap}^{-1}$ or in terms of protein $78.5 \text{ g}\cdot\text{day}^{-1}\cdot\text{cap}^{-1}$) is in line with our literature review on the matter (Figure 4.7b). The most distant values are from an old USA estimate in the 60s, and survey from Thailand not necessary relevant of France in the 2010s. For context, an N budget of Belgium in 2014, (Papangelou et al. 2021) used an N consumption (and excretion) of $4.32 \text{ kgN}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$, close to our value of $4.6 \text{ kgN}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$

Considering the alignment of our values with the real excretion measures reported in (Figure 4.7b), we estimate the uncertainty on nitrogen excretion to be $\sim 10\%$.

For phosphorus, we assumed no P accumulation in adults' bodies and that all ingested P is excreted. The same approximation was made for children and adolescents, for whom Jönsson et al. 2004 report a mean storage rate of 6% between 2 and 17 years, which is consistent with medical data on bone growth (Bass et al. 1999) and P intake from food. This is equivalent to 1.3% over an 80-year lifespan, consistent with estimates from Scholz et al. 2014 and higher than the 0.5% from Liu et al. 2008. Based on a mean body P content of 500–650 gP and daily intakes of $1\text{--}2 \text{ gP}\cdot\text{cap}^{-1}\cdot\text{day}^{-1}$ (Baccini et al. 2012; Elser et al. 2020f) over 80 years, the resulting annual population-level storage rate is also 1–2%. Our estimate for the average P excretion at the national scale ($0.44 \text{ kgP}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$, or $1.2 \text{ gP}\cdot\text{cap}^{-1}\cdot\text{day}^{-1}$) is consistent with the lower range of our literature review on P excretions (Figure 4.7a). This is exactly the same value as for Belgium in Papangelou et al. 2021, also based on food intakes survey.

Considering our review on Figure 4.7a, we estimate that the estimation from Thailand is not representative of France, an industrialized country. Based the range of values, we consider that the highest value for Germany ($0.66 \text{ kgP}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$) is an outlier for our situation. We thus consider a plausible range of $0.44\text{--}0.68 \text{ kgP}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$. This constitutes our uncertainty range.

Based on this P excretions literature review, we estimate the uncertainty range of P excretions to be $0.44\text{--}0.58 \text{ kgP}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$.

4.1.2.2 Individual Autonomous Systems (flows O and N)

Flow value. Part of excretions reach individual autonomous systems (flow J in Figure 4.1). The share of people not connected to sewers and with IAS are retrieved for each of the 6 French basins, based on their different experts' assessments *Etat des lieux*. The weighted average for Metropolitan France is 15%, close to the 18% reported by Eurostat (2023). For these people not connected to sewers, I considered

that 20% of their excretion happens in public spaces connected to sewers (workplaces, schools, and other public spaces), as the midpoint of the uncertainly range detailed hereafter.

The mass balances of individual autonomous system (flows N, N', O and O' in Figure 4.1) was estimated from Catel et al. (2017) and Risch et al. (2021) with the following fate for N: 22% in air, 73% in underground diffuse losses and 5% in septic tank sludge. For P, the values 85% as underground diffuse losses, and 15% in septic tank sludge. All of the septic tank sludge is assumed to be subsequently directed to WWTPs.

Flow uncertainty. According to the Ministry of Ecological Transition, 15 to 20% of the population is not connected to sewers and relies on Independent Wastewater Treatment (Ministere de la Transition Ecologique 2023). Compared to the Eurostat figure of 18%, which I use for metropolitan France (Eurostat 2023), I estimate the uncertainty range of this parameter to be 15–20% .

We considered that for people in individual autonomous systems, 5-35% of their excretions happen outside their home, in public spaces connected to sewers: 5% would correspond to elder people who rarely leave their house while the 35% corresponds to people who work (210 days on average, in France) in a workplace connected to sewers and excrete on average 60% of their nutrient flow during their time at work.

The uncertainty related to N and P mass balance of individual autonomous system was estimated using the different values of LCA analyses of a septic tank with sand filter from Risch et al. (2021) and a French report (Catel et al. 2017).

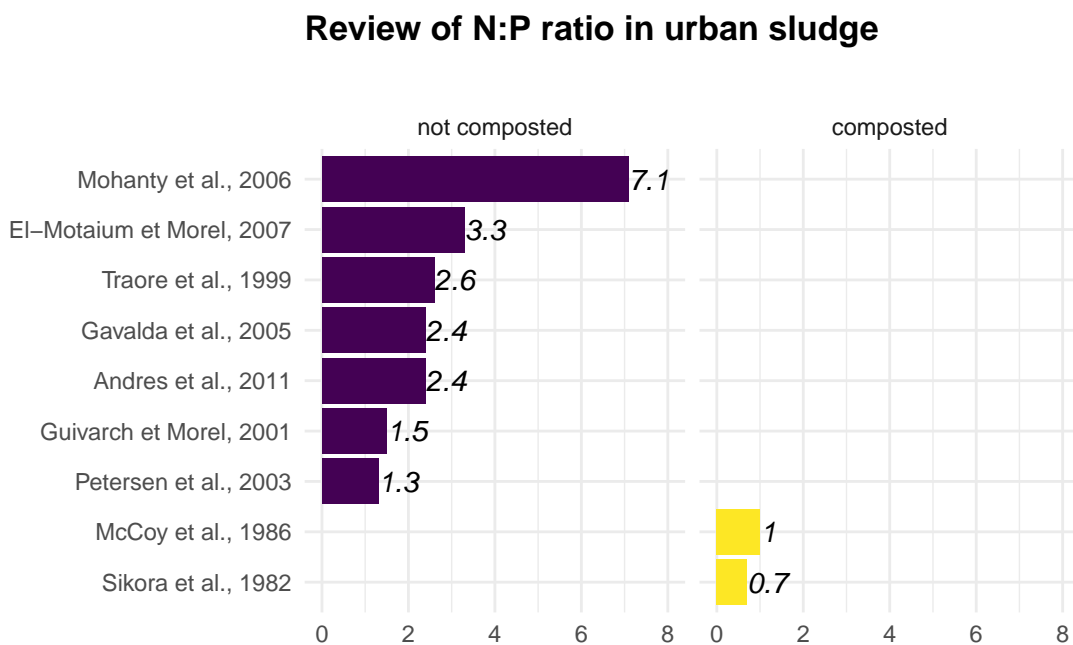
The N destination for Risch et al. (2021)) is 3% in air, 92% in groundwater and 5% in sludge but 22%, 73%, and 5% respectively in Catel et al. (2017). Since in both cases 5% of the N ends up in sludge, the final global circularity figure is unaffected. Only the distribution between groundwater and air emissions may change. I estimate the uncertainty range for the different N destinations to be 0-5% for sludge, 70-90% for groundwater and 5-25% for air.

For the P, destinations in Risch et al. (2021) is 83% in underground and 17% in sludge, while Catel et al. (2017) give slightly different figures of 90% and 10% respectively. I assume an intermediate distribution of 85% (80-90%) and 15% (10-20%) and that all sludge is directed to wastewater treatment plants. This is also the distribution used in (Linderholm et al. 2012), but lower than the 25%/75% used by Senthilkumar et al. 2014.

4.1.2.3 Sludge N:P molar ratio for sludge N content and denitrified N (flows C' and C'')

WWTP data only provides the nitrogen input (N_{in} , flow A in Figure 4.1) and the amount (N_{water} , flow B) of nitrogen rejected in the effluent. The flows of N to the atmosphere (N_{atm} , flow C'') and to sludge (N_{sludge} , flow C) are computed based on the phosphorus data: since there is no gaseous form for P, we know that $P_{sludge} = P_{in} - P_{water}$. I then use an average N:P molar ratio of 2 in the sludge to determine $N_{sludge} = 2 * P_{sludge}$, and then compute $N_{atm} = N_{in} - (N_{water} + N_{sludge})$. I considered a 30% N volatilization during the composting process (Zhong et al. (2013), flow D''). This is consistent with trial results reported by Denes et al. (2015), where N losses during composting amounted to 20-45% depending on the conditions.

The N:P molar ratio of 2 was estimated using a French collective scientific expertise review (Houot et al. 2014). When focusing only on studies reporting both N and P sludge content, the mean N:P molar ratio is 2.2 (Figure 4.10). For comparison, Papangelou et al. (2021) used a similar method but with an N:P ratio of 1.25–1.4 for their N budget in Belgium. Based on this elements, and considering the Mohanty et al. value on Figure 4.10 as an outlier, I choose to use a N:P molar ratio of 2, but consider a large uncertainty range of 1-3.



Source: French Collective Scientific Expertise on Organic Fertilizers, 2014

Figure 4.10: Sludge molar N:P ratio in sludge, in Houot et al. (2014) review.

4.1.2.4 Direct discharge losses before WWTPs (flows G and F)

Flow value. Part of the excreted nitrogen and phosphorus never reaches the WWTPs due to incorrectly connected pipes, sewers overflows, and sewer leaks (flow G in Figure 4.1). Based on individual WWTP data and the different water agencies' *Etat des lieux* assessments, I obtained a national mean of 10% for pre-WWTP loss (see Table 4.2 for each basin rate).

Flow uncertainty. Data are non-existent concerning sewers leaks, and very scarce for sewers overflows. Adour–Garonne and Loire–Britanny basins report data from a sample of the largest WWTPs, where direct discharges represent respectively 7% and 15% of the flows finally entering the WWTPs. For these 2 basins, I extrapolate the loss rate of this WWTPs sample to the whole basin. No data was available for the Rhone–Mediterranean basin, so I used the loss rate of the other French Southern basin, Adour–Garonne. For the three other basins (Rhine–Meuse, Seine–Normandy and Artois–Picardy), I use estimates from their respective experts' assessment *Etat des lieux* (respectively 20%, 10% and 20%).

The resulting national mean (weighted by incoming nutrient flow quantity) is 10%, and, based on the diverging values among the different basins, I estimate the actual value to be in the uncertainty range of 5–20%. For comparison, Scholz et al. 2014 estimate wastewater losses due to leakage before reaching WWTPs in the EU15 at 5–10%.

4.1.2.5 Residual inflow

From the reported nutrient flows entering the WWTPs and my estimation of sewer losses I estimated the theoretical nutrient flows entering sewers. This quantity is higher than the sum of human excretions and large industries discharges in sewers. I call this difference “residuals” (flow L in Figure 4.1), which includes several elements: household detergents, household kitchen leftovers, small industries not required to declare discharges into sewers, surface runoff entering the combined sewer networks...

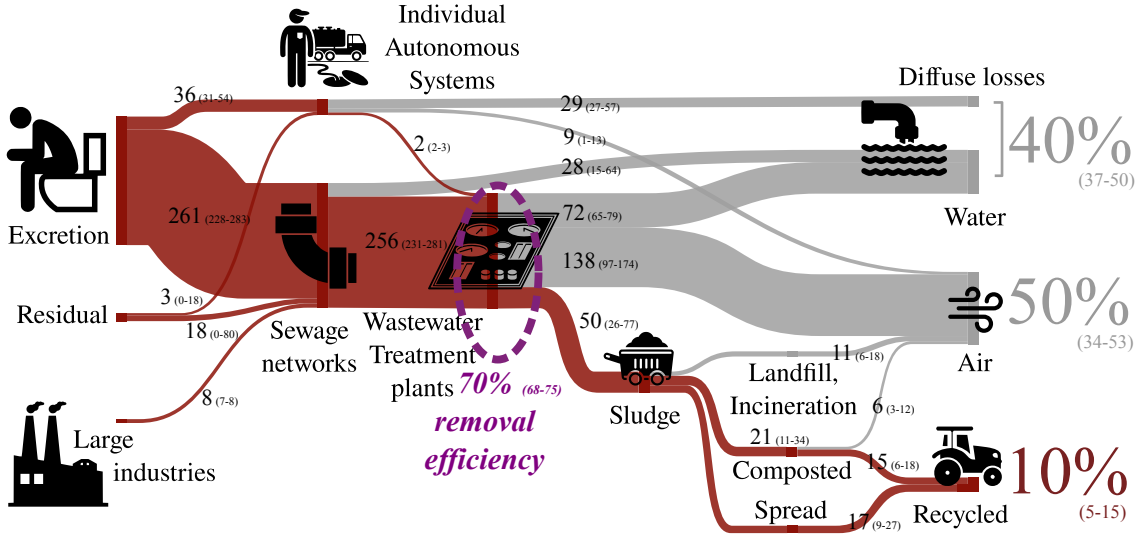
To estimate the residual flow entering individual autonomous systems (flow M in Figure 4.1), I made the assumption that the residual flow of the networks is essentially due to domestic discharges, and applied the same residues/excretions ratio to individual autonomous systems (still taking into account that people with individual autonomous system excrete 20% of their P and N in public spaces connected to sewers, see Section 4.1.2.2).

Table 4.3: Parameters values and their associated uncertainties to compute nutrient flows detailed on Figure 4.1. The "value" column reports our parameter best estimate. For point estimates of single parameters, we consider a uniform distribution with a range of values, based on our discussion in the previous sections. For measured flows with multi-year values, we consider a 10% uncertainty, based on reported measurement uncertainties (Lepot et al. 2022) and year-to-year variability. We consider that the $\pm 10\%$ range contains 95% of the values, i.e. $2 * \sigma / \mu = 10\%$

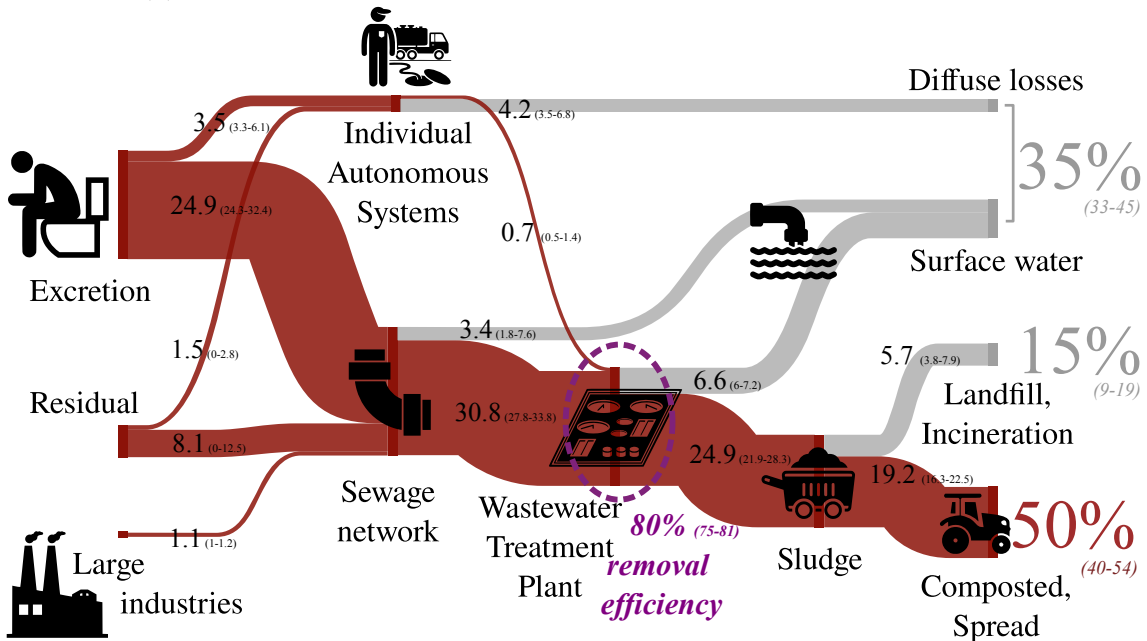
parameter	used in flow on Fig-ure 4.1	value	distribution	range for uniform or $2 * \sigma / \mu$ for normal
ktP coming to WWTPs	A	30.8	normal	10%
ktP discharged from WWTPs	B	6.6	normal	10%
ktN coming to WWTPs	A	256	normal	10%
ktN discharged from WWTPs	B	72	normal	10%
% sludge composted or spread	D	77%	normal	10%
% sewers discharge	F, G	10%	uniform	5-20%
ktP industries discharge to sewers	H	1.1	normal	10%
ktN industries discharge to sewers	H	8	normal	10%
kgP·cap ⁻¹ ·year ⁻¹ in human excretions	I	0.44	uniform	0.44-0.58
kgP·cap ⁻¹ ·year ⁻¹ in human excretions	I	4.6	normal	10%
% people in IAS	J, M	15%	uniform	15-20%
% excretions outside home	J	20%	uniform	5-35%
% P IAS to diffuse loss	N	85%	uniform	80-90%
% P IAS to WWTP	O	15%	uniform	10-20%
% N IAS to diffuse loss	N	73%	uniform	70-90%
% N IAS to air	N'	23%	uniform	5-25%
% N IAS to WWTP	O	5%	uniform	0-5%
N:P molar ratio in sludge	C'	2	uniform	1-3

4.2 Results

4.2.1 French sanitation system N and P budget



(a) Yearly N flows in the French sanitation system, in ktN per year.



(b) Yearly P flows in the French sanitation system, in ktP per year.

Figure 4.11: Nutrient mass balance in the French sanitation system, averaged for 2015-2020. Discrepancies between the sum of the incoming and outgoing flows are due to rounding. Ranges indicate the 95% percentiles obtained with a Monte-Carlo procedure, using the uncertainties detailed in Table 4.3. Percentages on the right refer to the share of total inflows. The removal efficiency percentage refers to the share of WWTP inflow that is not discharged into waters.

For each water agency basin, I produced an N and P balance flow, using the computed quantities averaged over 2015-2020. These are summed to produce the

national N (Figure 4.11a, Table 4.4) and P (Figure 4.11b and Table 4.5) budgets.

4.2.1.1 Nitrogen

Over the course of one year during 2015-2020, French N excretions are estimated to be about 300 ktN, large industries discharges in sewers represent ~ 8 ktN, and the residual N account for ~ 21 ktN (from kitchen waste, runoff entering combined sewers, small industries. . .) (Figure 4.11a). The ~ 330 ktN from these 3 sources enter the sanitation system through sewers or individual autonomous systems. In the end, 50% (165 ktN) ends up in the atmosphere, mostly as N_2 following denitrification, 40% reaches surface water or is diffusely lost in the ground (130 ktN). Less than 10% of the initial nitrogen (32 ktN) is recovered and used as fertilizer on crops.

Table 4.4: N Flows best estimate values and uncertainties. Best estimate is simply obtained by applying equations reported in Figure 4.1 to the values reported in Table 4.3. Uncertainties are obtained by applying a Monte-Carlo Procedure to the distributions reported in Table 4.3.

Flow description	Letter in Fig 4.1	Best esti- mate (ktN \cdot year $^{-1}$)	95% percentiles
N entering WWTPs from sewers	A	256	231-281
N discharged to waters by WWTPs	B	72	65-79
N trapped in sludge	C'	50	26-77
Sludge recycled to agriculture	D'	32	15-45
Sludge landfilled or incinerated	E'	11	6-18
N entering sewers	F	284	246-354
N lost in sewers or stormwaters	G	28	15-64
N discharged by large industries to sewers	H	8	7-8
N excreted by French population	I	297	228-283
Excretions going to IAS	J	36	31-54
Excretions going to sewers	K	261	259-337
Residual N in sewers	L	18	0-80
Residual N to IAS	M	3	0-18
N in IAS to underground water	N	29	27-57
N in IAS to air	N'	9	1-13
N trapped in IAS sludge sent to WWTP	O	2	2-3

85% of the atmospheric emissions comes from nitrification/denitrification in WWTPs, with the remaining 15% coming from individual autonomous systems, sludge composting process, and sludge incineration. Denitrification in WWTPs and losses outside WWTPs explain that, even though 3/4 of French sludge are used to fertilize crops, the whole sanitation system recycling rate is only around 10%.

Note that here I did not discriminate between losses as inert N (N_2) and reactive

N (like NH_3 or N_2O), because I focus on the mass balance of the nitrogen resource. Even though most of the air emissions occur as inert N_2 , the small air emissions (on the order of 0.1-1%, see Section 3.2.2.3) of N_2O has an important greenhouse effect, accounting for the majority of the warming greenhouse gases emissions in an operating WWTP. Higher emissions of NH_3 , affecting air pollution occur during composting and in individual autonomous systems.

Finally, despite the large relative uncertainty ($\sim 50\%$) for the N contained in sludge used to fertilize crops, since the absolute recycling rate is low ($<10\%$), this translates in an absolute uncertainty of only a few percentage points, and the final recycling value is between 5% and 15%.

4.2.1.2 Phosphorus

Over 2015–2020, French P excretions totaled 28.4 ktP per year, with large industries discharging 1.1 ktP into sewers and residual P (e.g. detergents, kitchen waste, runoff, small industries) amounting to 9.6 ktP (Figure 4.11b). The 39.1 ktP resulting from these three flows enter the sanitation system through sewers and individual autonomous systems. Of this, 19.2 ktP (50%) is used as fertilizer on crops (30% as direct application of WWTP sludge and 20% after composting), 14.2 ktP (35%) reaches surface water or is diffusely lost in the ground, and 4.2 ktP (15%) is incinerated or lost in landfill.

P discharged by WWTP to surface waters is 6.6 ktP (6–7.2 ktP). The combined losses from individual autonomous systems – 4.2 ktP (3.5–6.8 ktP) – and direct discharge from sewers – 3.4 ktP (1.8–7.6 ktP) – add up to 7.6 ktP (2.5–8.7), thus exceeding WWTP discharge, but with much higher uncertainty. Because of these losses, despite a national WWTP removal efficiency of 80%, and 75% of French sludge being used as crop fertilizer (combined recovery of $80\% \cdot 75\% = 60\%$), the whole sanitation system recycling rate for P is only 50%.

4.2.2 WWTP removal efficiency through space and time, influenced by regulation on sensitive areas

N and P removal efficiencies have been calculated for each individual WWTP, and aggregated over each water agencies (Figure 4.12 and 4.13). Removal efficiencies increased in all 6 French water agencies during the 2000s, and then leveled off in the 2010s, reaching a national mean of 70% for N and 80% for P. Note that the timing of the removal efficiency increase and stagnation correlates the "Borloo plan" during 2007-2013, which aimed at ensuring compliance of the largest WWTPs performance.

The final stagnation of performance is closely correlated to the classification into N sensitive areas. Adour-Garonne and Rhône-Méditerranée basins, largely consid-

Table 4.5: P Flows best estimate values and uncertainties. Best estimate is simply obtained by applying equations reported in Figure 4.1 to the values reported in Table 4.3. Uncertainties are obtained by applying a Monte-Carlo Procedure to the distributions reported in Table 4.3.

Flow description	Letter in Fig 4.1	Best esti- mate (ktP·year⁻¹)	95% percentiles
P entering WWTPs from sewers	A	30.8	27.8-33.8
P discharged to waters by WWTPs	B	6.6	6-7.2
P trapped in sludge	C	24.9	21.9-28.3
Sludge recycled to agriculture	D	19.2	16.3-22.5
Sludge landfilled or incinerated	E	5.7	3.8-7.9
P entering sewers	F	34.2	31-40
P lost in sewers or stormwaters	G	3.4	1.8-7.6
P discharged by large industries to sewers	H	1.1	1-1.2
P excreted by French population	I	28.4	28.4-37.2
Excretions going to IAS	J	3.5	3.3-6.1
Excretions going to sewers	K	24.9	24.3-32.4
Residual P in sewers	L	8.1	0-12.5
Residual P to IAS	M	1.5	0-2.8
P not trapped in IAS	N	4.2	3.5-6.8
P trapped in IAS sludge sent to WWTP	O	0.7	0.5-1.4

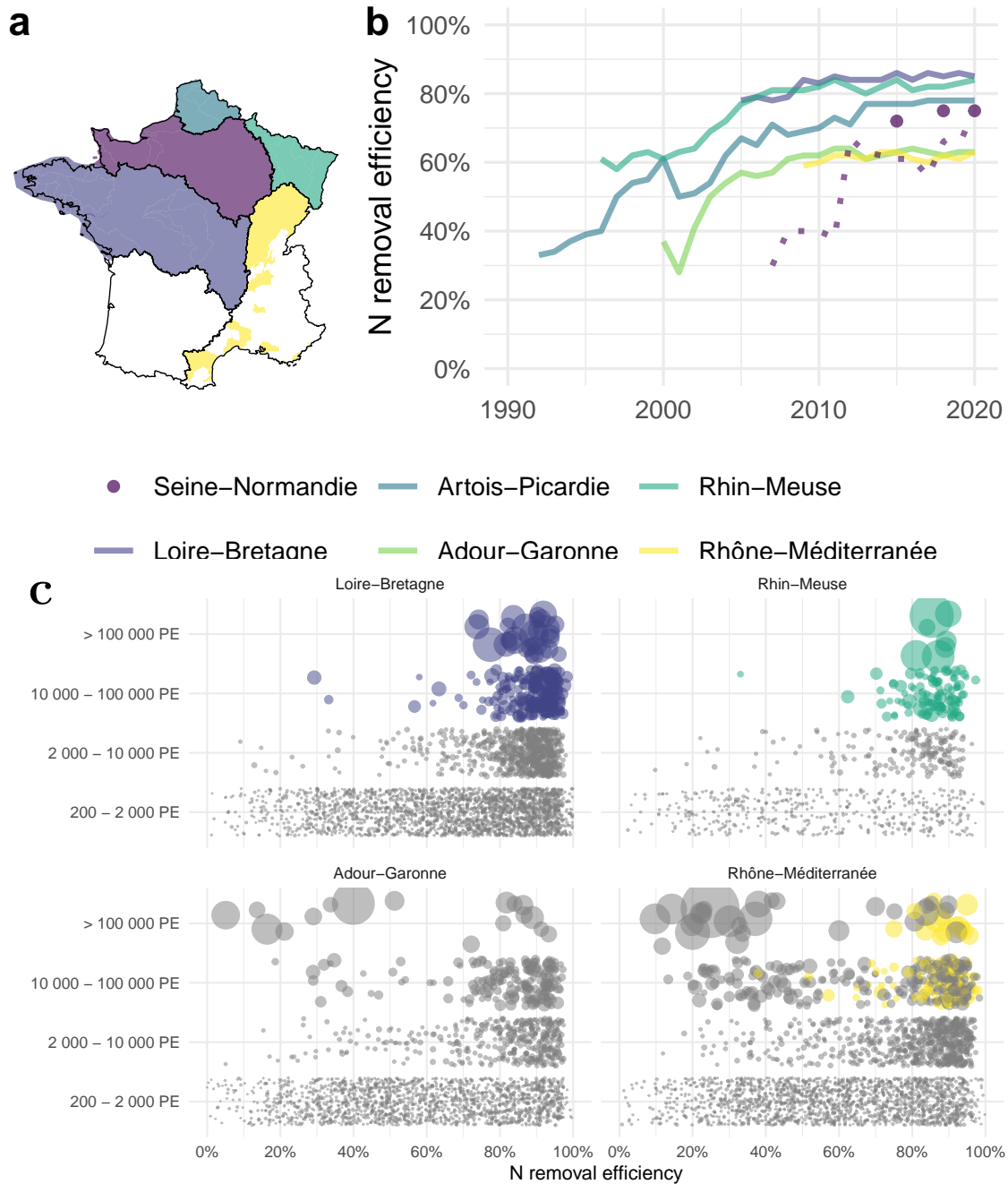


Figure 4.12: Influence of areas classification as N sensitive on N removal efficiency at the basins scale (top) and on individual wastewater treatment plans (bottom). a) Current Nitrogen sensitive areas of the 6 French water agencies basins, as of 2024. Colored areas are considered N-sensitive, white areas are considered non sensitive. See Figure 4.14 for the temporal evolution. b) Temporal evolution of N removal efficiency for the 6 basins. For Seine-Normandie, complete data was available only in 2015, 2018 and 2020 (points). The dotted line represents the partial data from the 6 largest Seine-Normandie facilities, handling about half of the total sewage. c) Individual WWTP N removal efficiencies, averaged over 2018-2020, in 4 of the French water agencies basins. Each dot is a WWTP, size is proportional to nominal capacity in population equivalent (PE). Dots are colored if the WWTP must reach minimal performances due to N sensitive area classification (see a) and text) or gray otherwise.

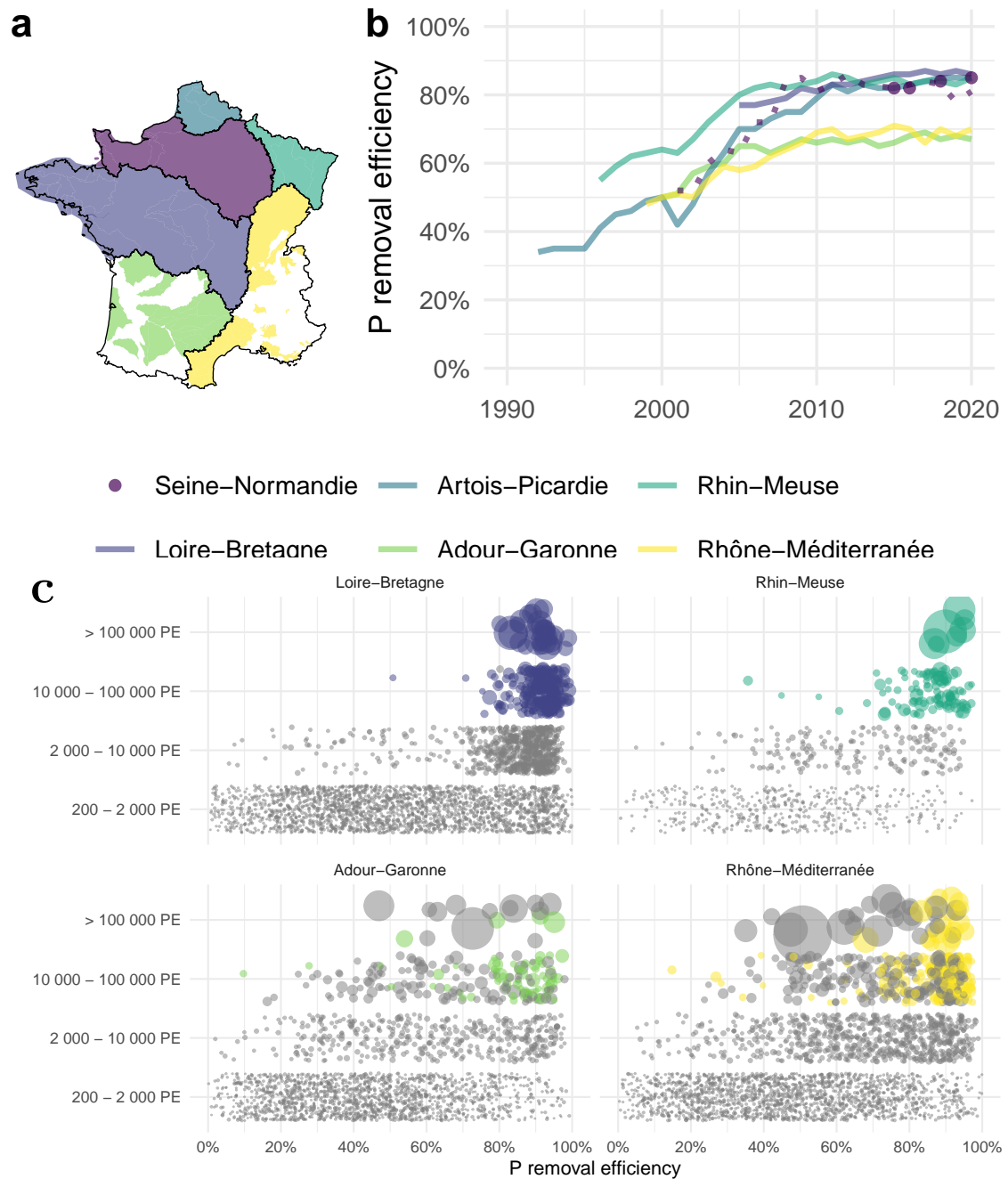


Figure 4.13: Influence of areas classification as P sensitive on P removal efficiency at the basins scale (top) and on individual wastewater treatment plans (bottom). a) Current Phosphorus sensitive areas of the 6 French water agencies basins, as of 2024. Colored areas are considered P-sensitive, white areas are considered non sensitive. See Figure 4.14 for the temporal evolution. b) Temporal evolution of P removal efficiency for the 6 basins. For Seine-Normandie, complete data was available only in 2015, 2016, 2018 and 2020 (points). The dotted line represents the partial data from the 6 largest Seine-Normandie facilities, handling about half of the total sewage. c) Individual WWTP P removal efficiencies, averaged over 2018-2020, in 4 of the French water agencies basins. Each dot is a WWTP, size is proportional to nominal capacity in population equivalent (PE). Dots are colored if the WWTP must reach minimal performances due to P sensitive area classification (see a) and text) or gray otherwise.

ered “non-sensitive to N” (Figure 4.12a), have a global N removal efficiency around 60–65%, while the other basins reach 75–85% (Figure 4.12b). The same applies to P, with slightly different values: 65–70% for Adour–Garonne and Rhône–Méditerranée basins (Figure 4.13a), while the other basins achieve ~80–85% efficiency.

This difference is mostly due to removal efficiencies of large WWTPs. Indeed, the EU regulation concerning WWTP in N sensitive zones requires a maximum outflow concentration of 10–15 mgN/l or an annual N removal efficiency of 70–80% for facilities larger than 10,000 population equivalents (the French decree has translated the 70–80% of the European directive (EU 2012) into a less restrictive 70% (Légifrance 2015)). For P, the required removal performance for these facilities larger than 10,000 population equivalent is 80% annual P removal or an outflow concentration below 1–2 mgP/l. These large facilities represent only 5% of French WWTP but handle 80% of the flows. There is no specific requirement regarding N for smaller WWTPs (<10,000 population equivalent), or the ones that are not in N sensitive zones.

Figure 4.12 (bottom) illustrates this effect of the European directive. Facilities larger than 10,000 population equivalent and in N sensitive areas (i.e., colored dots) generally have removal efficiencies above 70%. On the contrary, large stations above 100,000 population equivalent but not classified as N-sensitive (in grey), e.g., in Adour–Garonne and Rhône–Méditerranée, can have removal efficiencies well below 50%. Figure 4.13 (bottom) shows the same for P. Large WWTP in P-sensitive areas tend to have removal efficiencies >80%. However, in Adour–Garonne and Rhône–Méditerranée basins, large facilities not classified as P sensitive frequently have lower removal efficiencies.

From official data (data.europa (2021), based on Sandre (2020)) I produced the N and P sensitive zones temporal evolution (Figure 4.14), from 2006 to 2024. I could also adapt an official map of the 1994 decree (WikiODR 2017), but could not produce the map of the 1999 decrees. For these 2 latter dates, there was not yet a distinction between N or P sensitiveness, only a global “sensitive area” classification, and the local prefect would decide the required minimal performance. Then, in the following years, these areas became classified sensitive to N, P or both.

The partial glimpse in Figure 4.14 shows the regulatory battle that occurred during the last 2 decades. The first areas labeled sensitive were the regions bordering Germany, North-East, followed by a progressive extension to North-West, finally incorporating all the North basins. From then, patches of the 2 Southern basins are gradually included in the sensitive zone classification. Only the South-Eastern Adour–Garonne basin is still considered completely non sensitive to N-induced eutrophication, even though it is partially labeled P-sensitive.

Finally, WWTPs smaller than 2,000 population-equivalent take a wide range of

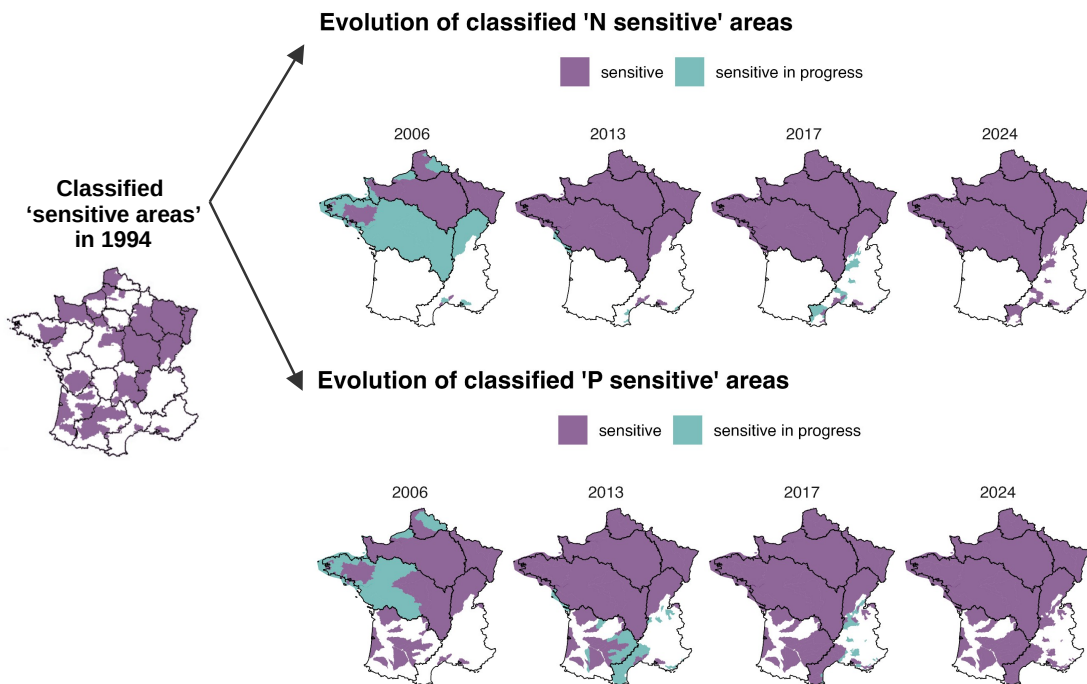


Figure 4.14: Temporal evolution of the classification as sensitive area for N (top) and P (bottom), based on the different decrees years. Minimal performance thresholds for large wastewater treatment plants are required in such areas (see Section 4.2.2 for more details). Purple: zones where the regulation is effectively in place in the considered year. Blue: zones where the regulation will be in place in the subsequent year (e.g., in 2013 for 2006 yellow zones). I could not produce the exact map for the 1999 decrees, but was able to adapt an existing official map for the 1994 decree (WikiODR 2017). In 1994, there was no distinction between N and P sensitive zones, and the required minimal WWTP performance was defined locally by the prefect.

values, but intermediate WWTPs (2,000 to 10,000) tend to have relatively high removal efficiencies even though they are not subjected to national legal requirements. This is because local prefectural orders require individual WWTP to reach a certain performance that can be more stringent than what the national legislation asks for.

4.3 Discussion

4.3.1 Consistency of nutrient flows and removal efficiencies

Removal Efficiencies and Recycling: Eurostat (2023) and national public data report the share of primary, secondary and tertiary WWTP treatment in France. Vigiak et al. (2020) and Van Drecht et al. (2009) each propose typical values of N and P removal efficiencies for these 3 treatments. Combining these sources allows to estimate the expected order of magnitude of national WWTP removal efficiency. The result of the calculation is presented in Table 4.6. The obtained national WWTP removal efficiencies are 70-75% for N and 80-85% for P, very close to our values of 70% and 80%. This is also coherent with water treatment textbook (Degrémont 2005), which reports removal efficiencies for tertiary treatment of 70–80% for N and 80–90% for P. This shows that simple assessments based on typical values can get results similar to my in-depth analysis.

Table 4.6: Estimate of expected national nitrogen and phosphorus WWTP removal efficiency, based on the distribution of primary, secondary and tertiary treatment in France, coupled with typical removal efficiencies for these 3 treatments.

	prim. treat- ment	second. treat- ment	tert. treat- ment	weighted	source
distribution of French WWTPs	1-2%	15-20%	80-85%	-	Eurostat (2023)
Nitrogen					
proposed efficiencies	25%	55%	80%	75%	Vigiak et al. (2020)
proposed efficiencies	10%	35%	80%	72%	Van Drecht et al. (2009)
				70%	this study
Phosphorus					
proposed efficiencies	30%	60%	90%	85%	Vigiak et al. (2020)
proposed efficiencies	10%	45%	90%	82%	Van Drecht et al. (2009)
				80%	this study

Regarding the temporal evolution of removal efficiency, Van Drecht et al. (2009) suggested a P removal efficiency of 44% in 1990 and 59% in 2000 for Europe. For basins with data at those times, my values are coherent, with $\sim 40\%$ in 1990 and 50–63% in 2000.

At a more regional scale, Esculier et al. (2019) focused on Paris urban area (10 million inhabitants) from a historical perspective. They found only marginal improvement on excreted N recycling, which has stayed around 5–10% for the last decades, after a peak over 40% around 1900. Figures for France in 2006 reported in

Le Noë (2018) confirm the low recycling rate I found. It proposed an N budget for French agri-food system in 2006. The rough estimation (based on N sludge content) was that 7% (26 ktN out of 388 ktN) of excreted N was spread on crops, a figure similar to my 10% for the situation more than a decade later.

N sludge/atmosphere distribution: An important uncertainty of my national budget is the N air/sludge distribution in WWTPs output, which does not rely on empirical data, but only on my estimated N:P molar ratio of 2. To assess my results, I compared my output to other studies.

For large European cities in the 2000s, assuming an 80% removal efficiency through denitrification, Svirejeva-Hopkins et al. (2011) estimated an output distribution from the plants of 58% to air, 21% to water and 21% to sludge, close to my figures of 53%-28%-19%⁵. I also reviewed national sanitation budgets in Table 4.8. I delve into more details in Section 4.3.2, but for now we can just consider the distribution at the outlet of WWTPs. If we exclude the particular case of Denmark in 1990, when almost no wastewater was treated for N, all countries recover 10-20% of the incoming N into sludge, similar to my 20% rate. The rest is simply distributed between air denitrification and water discharge, depending on the treatment removal efficiency, whose rate ranges from 50% to 85%. For instance, if 20% of N ends up in sludge and the removal efficiency is 85%, this means that $100\% - 85\% = 15\%$ ends up in discharged water, and the remaining $100\% - 15\% - 20\% = 65\%$ ends up in the air following denitrification.

Domestic Emissions: For the 2000s in Europe, Van Drecht et al. (2009) proposed a human N emissions in wastewater (domestic + industries) of $5.7 \text{ kgN}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$, higher than my value of $5.1 \text{ kgN}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$, but Hutchings et al. (2014) for Denmark in 2010 report a closer value of $4.9 \text{ kgN}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$. Most of this N load (> 90%) are due to excretions, highlighting the influence of dietary nitrogen consumption on WWTP activity (Rautiainen et al. 2023). In my budget, the residual quantity is the N flow entering sewers that could not be explained by excretions nor by large industries discharges to sewers and is estimated to be 18 ktN. This could be due to kitchen waste entering the sink, other gray waters, rainwater runoffs entering combined sewers, or small industries not declaring discharge to sewers. If we consider that the residual quantity is mostly due to households, this would mean the N emissions not related to excretions would represent about 5% of domestic emissions, in line with literature values consistently reporting rates below 10% (Friedler et al. 2013; Vinnerås et al. 2002). However, given the uncertainty on the excretion input,

⁵Established from Figure4.11a: out of the incoming 256 ktN, 138 ktN (53%) goes to the air, 72 ktN (28%) goes to the water, and 50 ktN goes to sludge (19%)

most of these residual inputs may also simply come from an underestimation of the N contained in human excreta.

For P, the discussion about domestic emissions is more complicated, since it should include detergents containing phosphates, whose quantities have drastically diminished during the last decades. I tackle this in Section 4.3.5, more focused on P in detergents.

4.3.2 High removal efficiency does not ensure high recycling rates.

Other studies performed nutrient mass balances of the sanitation system, usually as part of the larger frame of national N or P budget. For the sanitation system perimeter, these studies often rely on rough assessments for their sanitation sub-system, as in Section 4.3.1 and Table 4.6. Nutrient loads coming to WWTP or removal efficiencies are often part of the models assumptions, and not based on comprehensive operational data. Losses from sewers and combined sewers overflows are almost never assessed.

I am not aware of another study assessing a sanitation system at the national scale based on such comprehensive data as the ones presented here. I performed a quick review of national N and P budgets studies, and focused on their sanitation sub-systems, to get sense of the removal efficiencies and nutrient recycling in other countries. While their assessments are not precise, they should give the right order of magnitude. The results are synthesized in Table 4.7 for Phosphorus and Table 4.8 for Nitrogen.

For phosphorus, almost all the reviewed countries for the period 2000-2020 have high WWTP P removal efficiencies, often at least 80%. However this is not sufficient to ensure a high reuse of P in agriculture, because WWTP sludge are not consistently recycled on agricultural soils. Consequently, it appears that the higher recycling rate is for France during the 2015-2020 period. All the other countries have recycling rates consistently below 50%, often around 20-40%, or even below 20% when sludge utilization in agriculture is forbidden. Note that all these countries are rich industrialized countries, with a widespread use of sewers.

Even high recycling rates of sludge would be a partial solution. My detailed analysis for France shows that as many losses happen outside WWTPs as inside, because of sewer losses and individual autonomous systems. Thus, future measures regarding P pollution may need to look beyond WWTPs and address the whole sanitation system.

The situation is even worse with Nitrogen. Despite often high removal efficiencies, above 70%, the recycling rate is consistently below 10%. In this case, even

Table 4.7: Comparison of our detailed result for phosphorus in France with coarser assessments found in the literature for other countries. When a study focused on a period over several years, I took the median year. Sources: Papangelou et al. (2021) for Belgium; Egle et al. (2014) for Austria; Cooper et al. (2013) for the UK; Linderholm et al. (2012) for Sweden; Jedelhauser et al. (2015) for Germany; Smit et al. (2015) for Netherlands; Matsubae et al. (2011) for Japan; Roca et al. (2017) for Spain; Jedelhauser et al. (2015) for Switzerland; Senthilkumar et al. (2012) for France; Jeong et al. (2009) for South Korea.

Country	Year	P Removal Efficiency	Sludge Recycling	P Recycling
Belgium	2014	80-90%	15-20%	15%
Austria	2006	80-90%	25-30%	20-25%
UK	2009	50-60%	80-85%	45-50%
Sweden	2010	90-100%	20-25%	20-25%
Germany	2010	80-90%	25%	20-25%
Netherlands	2011	80-90%	0%	0%
Japan	2002	70-80%	60-65%	35%
Spain	2012	60%	70%	40-45%
Switzerland	2006	80-90%	35-40%	30-35%
France	2006	50%	60%	30%
South Korea	2009	60-70%	65%	40-45%
This study	2018	80%	75%	50%

Table 4.8: Comparison of our detailed result for nitrogen in France with coarser assessments found in the literature for other countries. When a study focused on a period over several years, I took the median year. Sources: Papangelou et al. (2021) for Belgium; Häußermann et al. (2021) for Germany; Olsthoorn et al. (1998) for Netherlands; Hayashi et al. (2021) for Japan; Hutchings et al. (2014) for Denmark.

Country	Year	N Removal Efficiency	WWTP % Air/ Water/ Sludge	Sludge Recycling	N Recycling
Belgium	2014	80-85%	60/20/20	15-20%	0-5%
Germany	2012	70%	55/30/15	25%	5-10%
Japan	2015	60%	40/40/20	-	-
Netherlands	1995	50%	30/50/20	-	-
Denmark	1990	5-10%	0/95/5	0-5%	0-5%
Denmark	2010	80%	70/20/10	70%	5-10%
This study	2018	70%	50/30/20	75%	10%

improving sludge recycling will not change the situation, as only 10-20% of incoming N is recovered in sludge. Denmark is the perfect example of this: between 1990 and 2010, it increased its N removal efficiency from less than 10% to 80%, and its sludge recycling from almost nothing to 70%. Yet the total N recycling barely increased and remained below 10%. The current sanitation system based on nitrification / denitrification for N treatment actively destroys this resource.

The current main indicator used to monitor the performance of sanitation systems, namely WWTP removal efficiencies, appears to be incomplete. First, it is blind to losses happening outside WWTPs. Second, while it indeed monitors the removal of part of the water pollution, it does not inform on nutrient recycling rates.

4.3.3 Perspectives for the evolution of WWTPs and recycling

4.3.3.1 Nitrogen

In France, N removal efficiencies closely match classification into N sensitive areas and European directives requirements. First, with the temporal increase in the 2000s for each basin and the stagnation levels in the 2010s (Figure 4.12a); second with the effect on large WWTP when they are in sensitive areas. This illustrates the point made by Gorman (2013g):

“The ambient concentrations of some naturally occurring compounds (including nitrogen compounds), long regulated by natural processes alone, had now come to be regulated by human institutions.”

In our N budget, N discharged from WWTPs to waters is 72 ktN (65-79 ktN uncertainty interval based on the 95% percentiles of the Monte-Carlo procedure), while combined losses from individual autonomous systems (29 ktN) and sewers (28 ktN) represent 57 ktN, with higher uncertainty (respective 95% intervals of 27-57 ktN and 15-64 ktN).

Another option would be to shift the main goal of the sanitation from minimizing N discharge to maximizing N recycling to agroecosystems. This would still lead to minimize N losses but would also enhance food security.

4.3.3.2 Phosphorus

In the case of France, currently 75% of WWTP sludge are spread on agricultural land. Recycling all P in produced sludge could increase the sanitation recycling rate of P by 15 percentage points, or 30% in relative terms (Figure 4.11b). Increasing WWTP removal efficiencies is another lever, especially in the Southern basins. At best, hypothesizing 100% removal efficiency, it would increase recycling by 6.6

ktP (6–7.2 ktP), or about 15-20 percentage points. The potential improvements are thus similar as the P lost from sewers (leaks and storm waters overflows) and Individual Autonomous System together, but these flows are overlooked in current policies. Finding ways to recover P from individual autonomous systems and reducing sewer losses should be considered by policy makers if the aim is to further improve circularity.

4.3.3.3 Limitations to recycling within the current framework

However activating all these levers would still leave some issues. First, this will not enhance nitrogen cycling in the sanitation system, which, contrary to phosphorus, is dissipated in the air as N_2 and N_2O when treated in WWTPs. Second, future regulation and lower thresholds for contaminant (especially heavy metals, pharmaceutical residues) in WWTP sludge may prevent high recycling rates. Source separation of excreta, before they reach sewers, could be one way to address these issues (Larsen et al. 2013a).

4.3.3.4 Future regulation

Future EU regulation will change the requirements for WWTPs (Halleux 2024). By 2035, all stations larger than 100,000 PE (whether or not in sensitive zone) will have to reach a minimum N removal efficiency of 85%, or a 6 mgN/L discharge. For stations between 10,000 and 100,000 PE and in sensitive zone, the deadline will be by 2040. The same principle applies for phosphorus, with removal efficiency of 90% or a 5 mgP/L discharge limit. The impact will be most felt for large stations in areas classified as "not sensitive" and which can currently have relatively low removal efficiencies, as in the Southern basins.

Moreover, the future EU directive considers implementing stricter monitoring and treatment of micropollutants in WWTP sludge, which represent a risk for health (Lamastra et al. 2018). This might impact local regulations concerning sludge recycling to agriculture (Pivato et al. 2022; Hudcová et al. 2019), following the path of countries like Switzerland which already implemented a ban on raw sewage sludge spreading to agricultural land, decreasing nutrient recovery for fertilization.

4.3.4 Consequences of current WWTP treatments on fertilization, compared to urine-based fertilizers

In Western societies, low recycling rates of N excretions have not always been the norm. In Paris, this rate exceeded 40% in the beginning of the XXth century. This rate was achieved by a combination of high recycling of human excreta collected

from cesspools and sewage farms, enabling to return to agricultural land the excreta of people connected to sewers (Esculier et al. 2020). More recently, it is estimated that urban excretions recycling rate in China were as high as 90% in 1980, 70% in 1990, 40% in 2000, and below 5% after 2010. For rural settings, the estimation was 95% in 1980, 77% in 2000 and just 50% in 2010 (Gu et al. 2015).

Only focusing on end-of-pipe recycling would have some drawback. First, domestic consumption of water is around 100–150L/cap/day, while urine and feces production is around 1–1.5L/cap/day, resulting in a dilution by a factor 100. Recovering highly diluted nitrogen faces strong technological locks that hinder the sewer rationale for a circular economy of human excreta nitrogen, because of the dilution by flush toilet (Besson 2020). While sewers with high water consumption can be adapted to humid climate, a large part of the population lives in regions where high water consumption is not sustainable. In all the scenarios developed by Van Drecht et al. (2009), less than 50% of the global population would be connected to sewers in 2050. And finally, only focusing on end-of-pipe misses the losses that happen before the WWTP, namely in the form of sewers leaks or storm waters overflows.

The rationale for urine-based fertilizers have been described in seminal works as soon as the 1990s (Jönsson et al. 1997; Larsen et al. 1996; Larsen et al. 1997). Currently, WWTP nutrient removal produce sludge bringing human excreta N:P molar ratio back to the value of feces: 1.5–3 in WWTP sludge and feces (Figure 4.9), vs 8–9 in wastewater entering WWTP (Figure 4.15). This is done by removing - and losing - N through denitrification.

If urine is diverted but feces still go to sewers, this would nonetheless greatly reduce the need for denitrification in WWTPs, because incoming wastewater composition would be closer to feces N:P ratio, which is already similar to the produced WWTP sludge after treatment. This would decrease WWTP energy consumption, mainly due to nitrification needed before denitrification.

Finally, human urine has a low heavy metal contamination compared to sludge, but also to animal manure or biowaste (Winker et al. 2009). One important challenge would be the storage, transport and application implementation of urine-based fertilizers, 2 orders of magnitude less concentrated than industrial fertilizers (Larsen et al. 2013b). An option to tackle this issue is to use concentrated urine-based fertilizers but this has to be balanced with the resource consumption of such processes to find the best solution depending on the local context. Concerns regarding micropollutants in urine will also have to be better assessed (Martin et al. 2023a; Winker 2009) when using unprocessed urine. In the meantime, many pharmaceuticals present in urine can be removed by carbon-based filtration to alleviate this issue (Martin et al. 2022).

4.3.5 Regulations on P in detergents contributed to the decrease of P discharge by reducing P inputs to WWTPs

Figure 4.13b shows the improvements in WWTPs P removal efficiencies, which was an important factor in reducing P discharge to water streams in the last 2 decades. Another factor of similar importance was the progressive ban on phosphate in detergents (laundry and dishwasher) which, in the 90s, was a flow of similar magnitude as P in excretions.

At least three successive limitations on P in detergents were taken in France and in Europe. In France, phosphorus in laundry detergents were banned in 2007 (Légifrance 2007). In 2013 the European Union banned consumer laundry detergents with standard doses >0.5 gP, and in 2017 automatic dishwasher detergents with standard doses >0.3 gP (EU 2012). As roughly 75% of the P in detergents came from laundry (Van Drecht et al. 2009), most of the effect of the EU ban is thus expected to be seen before 2013. Supposing a consumption of 1 or 2 “standard doses” per capita and per week for both dishwasher and laundry would give an upper limit P detergent emission of 0.04 – 0.08 $\text{kgP}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$ after 2017, or 7–13% of our computed national emissions (0.6 $\text{kgP}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$).

For the 2000s in EU25, Van Drecht et al. (2009) proposed a human P emission in wastewater (domestic + industries) of 0.9 – 1 $\text{kgP}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$, 40% of which were due to detergents from laundry and dishwashers (0.4 $\text{kgP}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$). Despite large variations between European countries, this is in line with Vigiak et al. (2020), based on Bouraoui et al. (2017), reporting a detergent consumption of 0.37 $\text{kgP}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$ in 2005 for France. These values imply a non-detergent human P emission (domestic + industries) of ~ 0.6 $\text{kgP}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$. This corresponds to our figure of 0.6 $\text{kgP}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$ for 2015–2020 in France, after the ban on P detergents.

Our estimation of residual P discharge is 9.6 ktP, or 0.15 $\text{kgP}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$ (Figure 4.11b), but this comes with high uncertainties, combined from those on sewer losses and human excretions. Following our Monte-Carlo procedure, the 95% uncertainty range associated with residual emission goes from 0 to 15.2 ktP (0 – 0.23 $\text{kgP}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$).

From our data, it is not possible to directly observe the impact of P detergent ban through the temporal evolution of P emissions per capita. Reporting rates of WWTP data increase through time, reaching full levels in the 2010s, but are still incomplete before. This complicates the estimation of the temporal evolution of the number of people connected to sewers associated with the reported incoming sewage. Data about sewers overflows and leakage are also non-existent before 2015–2020.

However, we can normalize incoming P flows by the nominal capacity of the

reported WWTP data (in 2020 France total nominal capacity was 100 million population equivalents, for 54 million people effectively connected to sewers). We also normalize by incoming N flows (Figure 4.15). In both cases, most of the decrease in incoming P loads happens before 2010, and by 2015 the stabilization is over, consistent with the EU directive. These 2 ratios could be used in further work as inputs to model emissions of P detergents for 1990–2020 in France. Note that the trends are consistent between the independent datasets of different basins, which suggests that the results are reliable.

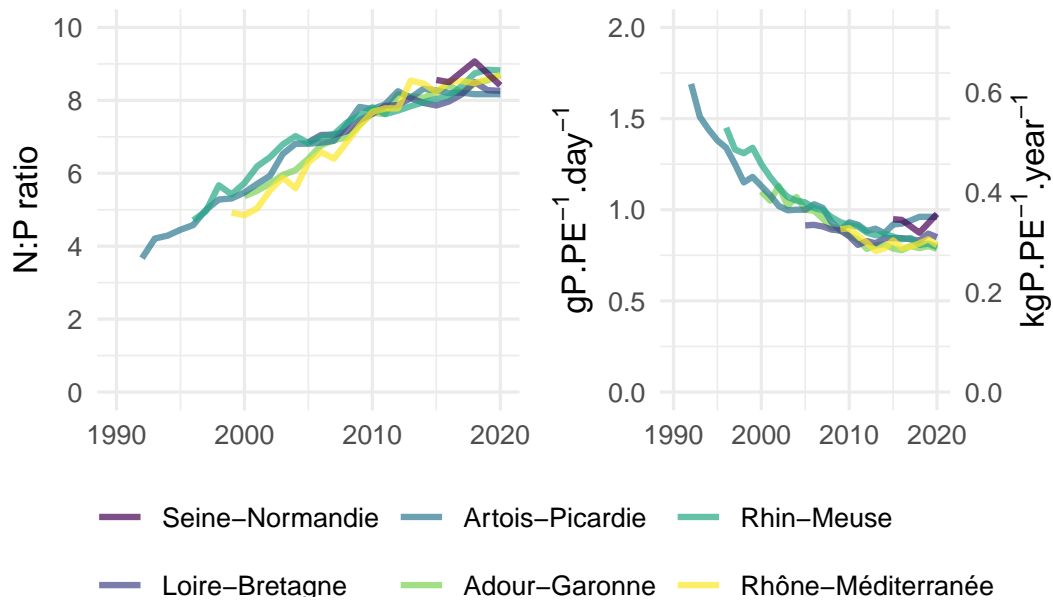


Figure 4.15: For each of the French 6 water agencies basins: a) N:P molar ratio of incoming flows in WWTPs and b) incoming P in WWTPs, normalized by WWTPs capacity, in population equivalent (PE). In 2020, the total French capacity was 100 million PE, for 52.5 million inhabitants connected to sewers. The dynamics highlight that less and less phosphorus are present in incoming wastewater flows, reflecting the European progressive ban on P in detergents during the 2000s.

4.3.6 Indication of NH_3 volatilization in sewers ?

In the previous section, I estimated that currently P in detergents could represent $0.04\text{--}0.08 \text{ kgP}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$. Considering that out of the 65 million inhabitants in Metropolitan France, 85% are connected to sewers, this corresponds to 2.2–4.4 ktP entering the sewers because of detergents, out of the total of 8.1 ktP in Figure 4.11b. This implies a P entering sewers and not due to detergents of about 3.7–5.9 ktP.

Compared to the residual 18 ktN entering sewers, this implies a N:P molar ratio of residuals of 3.1–4.9, substantially lower than the N:P ratio of 8–10 in excretions. If we take the (disputable) hypothesis that the non detergent residual N and P entering sewers has a ratio similar to excretions, this would mean that around 20–40

ktN is missing. But this is largely within the uncertainty bounds of the estimated 18 ktN (0-80), so this should be interpreted only as a hint of a missing quantity.

I insist that all this is highly speculative, since it relies on very rough estimation of detergent P emission, a disputable hypothesis about N:P molar ratio, and very high uncertainties regarding N and P residuals entering sewers. But the hints pointing at missing N in the residual flow could imply some NH_3 volatilization in sewers, on the order of 20-40 ktN, or 7-14% of the N entering sewers⁶.

For context, official estimates of NH_3 emissions in France are about 500-550 ktN (Citepa 2022a), so our estimate would represent 4-8% of national emissions. But as 90-95% of national emissions are due to agriculture, sewers emissions would represent a large part of urban emissions. Some actual measurements above sewers suggest high loads of NH_3 in the particular context of Barcelona (Reche et al. 2012), but more empirical studies should be carried out in sewage systems to confirm this.

4.3.7 Another nutrient: the case of potassium

Potassium (K) is another nutrient present in human excreta and useful for agricultural fertilization, but not analyzed in this study. While the available data did not allow to perform an analysis on potassium as detailed as the one carried out for phosphorus and nitrogen, it is still possible to get an order of magnitude of its fate in the sanitation system.

An initial rapid analysis of the Sillage database⁷ (data not shown here) listing the composition of French WWTP sludge indicates a K content of the order of 3-5 gK per kg of dry matter. On the basis of 1 Mt of sludge dry matter produced in France, three-quarters of which is reused in agriculture, this represents 2 to 4% of the 70 ktK excreted by the French every year (combination of INCA3 data for ingestion and INSEE data for population). Esculier (2018) had estimated the potassium recycling rate at 2% in the Paris metropolitan area, taking into account the non-negligible K present in drinking water.

4.3.8 Limitations and perspectives

The main limitations of this study are due to the uncertainty in the individual autonomous system nutrient mass balance, the rough estimation of losses in sewers, and the N content of sludge. I also could not appropriately estimate the N or P flow due to runoffs entering the combined sewers networks. This issue is also present in other N and P budgets and further investigation on this would be required to

⁶Note that on Figure 4.11a

⁷SILLAGE is a tool from the French Environment Ministry to monitor the spreading of organic matter, especially WWTP sludge.

reduce the uncertainties around direct losses upstream of WWTPs (flows G, N and O), especially since they may represent one of the main source of N pollution to water bodies from the sanitation system.

One of the most critical parameters for the estimation of French N recycling rate is the N quantity in sludge as opposed to the one denitrified. My comparison to literature review supports the general order of magnitude of our sludge / air distribution, but a large relative uncertainty remains (factor 2). However, this translates in a small absolute uncertainty for the final sanitation system recycling rate, which is probably between 5 and 15%. A way to improve this would be to focus on systematic empirical data of French sludge composition. I tried to use some data transmitted by the French ministry, but could not get a coherent result out of it.

Uncertainty in individual autonomous systems will be difficult to reduce, unless a large part of these facilities (about 5 million in France) is regularly monitored, which is implausible. Even the number of people with IAS is not very reliable, the ministry simply specifying that it concerns "15 to 20%" of the population.

The issue is different for sewers direct discharge, some of which are monitored. Increasing reporting rates may help reduce my current factor uncertainty on this matter. Finally, there seemed to be higher uncertainties concerning P ingestion and excretion as compared to N, which could be further assessed (see discussion in Section 4.1.2.1).

4.4 Conclusion

In France, about 50% of the P in human excretions is used to fertilize agricultural land, but that rate is only 10% for N. At the national scale, the WWTP removal efficiency is about 80% for P and 70% for N. But while the removed P ends up in sludge, that is generally spread on agricultural land (three quarters of the 1 Mt of dry matter of sludge), the removed N is denitrified and lost to the air. The main sources of N and P pollutions from the French sanitation system are: what is not treated in WWTPs; what is lost in sewers due to leaks and overflows, upstream of WWTPs; and individual autonomous systems.

The complete dataset of WWTP allowed to have a closer look at the factors affecting their performance. Removal efficiency increased during the 1990s and 2000s, but stagnated in the 2010s. This concerns mostly the largest WWTPs (> 10,000 PEs), handling the majority of N and P flows, located in areas considered sensitive to N or P eutrophication. WWTPs which are smaller or not located in sensitive areas usually do not reach similar performances.

Though France has a higher P reuse in agriculture than most industrialized countries, this is not primarily due to a will to recycle but to a will to limit P discharge in surface water, combined with a permissive legislation on sewage sludge. Enhancing WWTP P-removal in basins where surface water is currently considered “non sensitive” to eutrophication could both reduce water pollution and increase P recycling rate.

However, this strategy will not enhance N recycling with current technologies. In addition, sewage sludge use in agriculture also presents drawbacks due to health and environmental impacts, notably from various pollutants and heavy metals. Because of this, future legislation may restrict its use, thus reducing nutrient cycling. Source separation of human excreta is therefore a method to investigate, as a way to further increase nutrient recycling while preventing it from mixing with contaminants in the sewers.

More precise assessments of the losses occurring in sewage networks – notably through more systematic direct monitoring of sewers – would be necessary to reduce the uncertainties on the nitrogen and phosphorus mass balance.

Chapter 5

Agronomic potential of N in excretions

5.1 Research question

When presenting the results of Chapter 4 during my PhD in congresses and meetings, a question I was regularly asked was "You just showed that N recycling of human excreta is low in France, but were we to totally recycle it, what would be the total *potential* for fertilization?". Or alternatively "How much of the Haber-Bosch fertilizers could be substituted by urine-based fertilizers?".

There is a lot to unpack here. It makes sense to chemically compare urine-based fertilizers to synthetic Haber-Bosch fertilizers, since many urine-based fertilizers are highly mineral and have very similar chemical and agronomic properties (Martin et al. 2021)¹. The questions are also relevant because they do not stop to the observation that urine-based and synthetic fertilizers have chemical similarities concerning their N fraction, but want to dig further and compare absolute quantities: if the amount of N in human urine represented a negligibly small fraction of the synthetic fertilizers consumed in the country, this would be of little agronomic interest.

However there are several limitations with framing such questions. First, I noticed that total N fertilization is often confounded with synthetic fertilization. Yet livestock manure also represent a significant share of N fertilization in agroecosystems, as well as biological fixation and atmospheric deposition. Moreover, only focusing on synthetic fertilizers is not relevant for countries which barely use them because they rely on slash-and-burn (for instance some parts of sub-Saharan Africa and Southeast Asia) or for agricultural systems based on legumes rotation as their main source of N input (e.g. organic agriculture).

Second, the *potential* of human excreta depends on 2 terms: on the numerator,

¹But note that contrary to synthetic Haber-Bosch fertilizers, urine is a multi-element fertilizer, containing P and K but also Ca, S, Mg and micronutrients such as Fe, B, Mn...

i.e. the quantity of excreted nutrients; and on the denominator, i.e. the nutrient inputs in the agricultural system. The potential will thus strongly depend on each country's particular context: does it rely on intensive agriculture or low inputs agriculture? Are there important N losses? Is it an importer or an exporter of agricultural commodities? Simply focusing on the potential substitution of current fertilizers with N in excretions does not question the wastefulness of agricultural system itself, and the room there is for decreasing the system N inputs.

The aim of this chapter is to assess the order of magnitude of the maximum potential contribution of human excreta to agricultural production. I first study this maximum contribution in the current system, in numerous countries: this allows to explore a variety of situations and context, and characterize the factors affecting the potential. I then go further this limited frame and assess the potential when the food system itself is changed to make it more frugal in terms of N inputs.

In the following, the recovery rate for human excreta is assumed to be 100%. For an operational translation in real conditions, an additional effective rate of recovery of human excreta <100% should also be applied; as determining credible rates of human excreta recovery is not the subject of this chapter, I let the reader apply whatever factor they find credible.

The data to produce this chapter results was prepared in a GitHub repository² associated with a permanent DOI on Zenodo³, and is featured on a user-friendly interactive website⁴.

²<https://github.com/Thomas-Starck/potential-human-excretions-fertilization>

³<https://doi.org/10.5281/zenodo.14591081>

⁴<https://thomas-starck.github.io/potential-human-excretions-fertilization>

5.2 Methods and Sources

5.2.1 Hypothesis of current low reuse of N in excretions

My interest in the potential of nutrients in human excreta only makes sense if the current recycling rate is low: were it to be 100%, this chapter would simply be a description of the current flows, and not an assessment of the potential. For that reason, I will not focus on phosphorus, as Table 4.7 in Chapter 4 highlighted that it is usually not negligible, mostly between 20 and 40% in industrialized countries, while estimates vary between 10% and 60% at the global scale (Cordell et al. 2009; Liu et al. 2008; Scholz et al. 2014; Smil 2000).

On the contrary, I found in Chapter 4 that in France the recycling of nitrogen in human excreta is currently only around 10%. Table 4.8 gave a tentative review of recycling rates in rich industrialized countries characterized by a sanitation system centered on sewers and an end-of-pipe treatment in wastewater treatment plants. Even when these countries reach high recycling rate of their sewage sludge, like the 75% rate in France, very few of the incoming N ends up in sludge with the current technologies, as it is mostly sent to the atmosphere following denitrification. This impedes recycling through sludge spread on croplands. Untreated N ending up in wastewater treatment plants effluents could be directly spread on crops, but fertigation with wastewater is generally not allowed, and has other technical limits (see my discussion on that matter in Chapter 3 Section 3.2.7.3). This ensures that all these industrialized countries currently have low N recycling rates, on the order of a few percents.

The assessment is more complex for countries in which most of human excreta is not handled by a wastewater collecting system (see Figure 5.1). More than half of the global population is not connected to sewers (Van Drecht et al. 2009). For those countries, the fate of human excreta is less clear, and depends on local practices. This uncertainty is reflected in the wide uncertainty of the attempts to estimate the global recycling rate of phosphorus in human excreta (10-60%), which reflects uncertainty about how human excreta are handled. In countries without sewers, if local practices generally recycle human excreta, a priori mainly in rural areas, it is reasonable to expect higher recycling rates than in industrialized countries. However, nitrogen recycling would be lower than for phosphorus, because of the N volatilization and infiltration that can occur during storage. One country with rough estimates is China, which experienced a steady decline in its recycling rate of human excreta during the last decades: from possibly >90% reuse of nutrients in human excreta in the 1980s, down to 5% and 50% in respectively urban and rural areas in the 2010s (according to Table S10 in Gu et al. (2015)). The estimate made

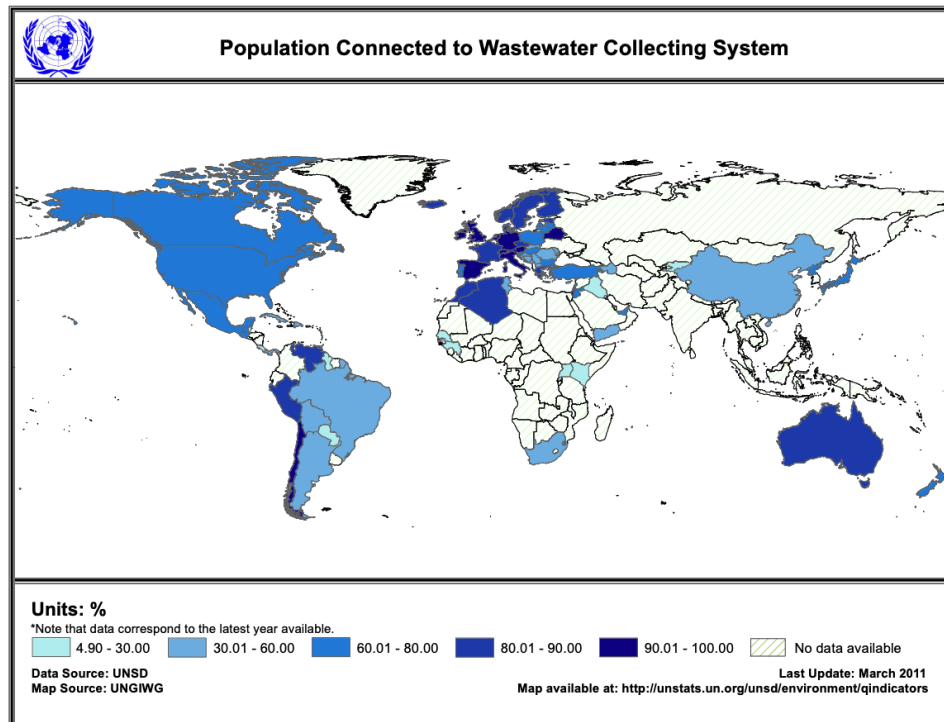


Figure 5.1: Share of population connected to Wastewater Collecting System, circa 2010. Wastewater collecting system (sewerage) is defined as a system "that may deliver wastewater to treatment plants or may discharge it without treatment to the environment". Source: UNECE (2011). More recent data from UNSD is available in a spreadsheet form.

by Chatzimpiros et al. (2023) is that N recycling of human excreta at the global scale decreased from 30% in the 60s to a mere 5% currently.

In Table 5.1, I give tentative estimates of current reuse rate of N in excretions at the global scale. To do so, I focus on the different world regions and their population, use their share of urban and rural population, and estimate a range of plausible local reuse rate for urban and rural areas.

Based on my review in the previous Chapter 4 Table 4.8, I estimate that recycling in urban and rural areas for rich industrialized regions (Europe, Oceania, and North America) to be around 0-10%. I also apply this rate for urban areas in South America, which is another region quite connected to sewers in Figure 5.1. I estimate the rate for rural South America to be also low (0-10%): its very low rural population (15%) is the sign of a generally highly mechanized agriculture with large *latifundia* of often hundreds or thousands of hectares; I hypothesize that this deep industrialization of the agricultural sector is correlated with low reuse rates.

In the remaining regions, for urban areas, I also apply a 0-10% recycling rate. Indeed, it seems implausible that a current urban system handling human excretions would recycle a significant share (say >20%) at a continental scale without being properly documented, given that past systems have been extensively described in

France, Japan and China (see Section 3.1). In the case of urban China, this is supported by the 5% recycling rate estimate in the 2010s by Gu et al. (2015).

For rural regions, the same authors estimate Chinese rural rate to be 95% in 1980, 77% in 2000 and 50% in 2010; continuing the trend would give 25% in 2020, while a stabilization would stay at 50%, and I chose to use these 2 rates as a range of plausible values for rural areas in this region. The most uncertain rates are for India, the rest of Asia and Africa, in rural areas. Due to the lack of data (at least in my knowledge), I purposely chose a wide range for these rural regions, from a low 10% to a high rate of 50%. Note that this 50% rate is not simply the recovering rate of human excretions, but also includes the N losses that can occur during storage through volatilization or leaching, and is thus quite high.

Combining all of these estimates yields a global rate of 5-25% (Table 5.1), with the high end requiring 50% N recovery in about three quarters of the global rural population (Asia and Africa). A more probable global recovery rate might be around 10-15%.

Table 5.1: Estimation of the recycling of N in human excretions at the global scale, circa 2020. Population data from Our World in Data (2024). See text for reasoning.

region	population		N in excretions reuse rate		
	billion	% urban	urban	rural	total (computed)
China	1.4	55%	0-10%	25-50%	11-28%
India	1.4	35%	0-10%	10-50%	7-36%
rest of Asia	2	57%	0-10%	10-50%	4-27%
Africa	1.5	45%	0-10%	10-50%	6-32%
Europe	0.75	80%	0-10%	0-10%	0-10%
North America	0.6	80%	0-10%	0-10%	0-10%
South America	0.4	85%	0-10%	0-10%	0-10%
Oceania	0.05	70%	0-10%	0-10%	0-10%
World (computed)	8.1	55%	0-10%	10-45%	5-25%

This Table suggests that the hypothesis of current low recovery rate is valid for much of Europe, Americas and Oceania. This is more uncertain for Asia and Africa, particularly locally.

5.2.2 Countries of interest

My following estimates of the potential of nitrogen in human excreta used as fertilizers assume a current negligible recycling rate. For this reason, I will preferentially highlight rich industrialized countries, where we know this hypothesis to be true. I thus chose to focus on Western Europe (UK, Ireland, Portugal, Spain, France, Italy,

Germany, Switzerland, Belgium, Poland, Austria, Netherlands, Denmark, Norway, Finland, Sweden, Greece), North America (USA, Canada), rich industrialized Asia (Taiwan, Japan, Korea) and Australia and New Zealand.

However my aim is to feature results in a variety of agricultural context, so I chose to also focus on other countries where it is not sure this hypothesis is true, and the reader should keep this limitation in their mind when reading the results. As the graphs featuring my results could not clearly display the nearly 200 existing countries, I needed a criteria to sample countries outside the rich industrialized group. I fixed an arbitrary threshold of 50 million inhabitants, excluding less populated countries. This yielded 20 countries. Once added to the rich industrialized countries, the total counts 6 billion inhabitants, or 80% of the 7.6 billion global population in 2015-2020. This ensures representativeness of the global situations. I will also present the results for the entire World, based on global population and agricultural data.

5.2.3 Agricultural system limits and N input flows

5.2.3.1 Description of Agricultural land

Comparing the potential of human excreta to current N inputs in agriculture requires to carefully define what constitutes these N inputs. This depends of the definition of the "*agricultural system*". Agricultural land is generally divided between croplands (which include arable land and permanent crops such as vineyards and orchards, see for instance definition in FAOSTAT (2022b)) and permanent grasslands. In the world, the 1.6 Gha of croplands and 3.2 Gha of permanent grasslands represent respectively 1/3 and 2/3 of the total agricultural land (FAOSTAT 2022e). I will follow the FAOSTAT (2022b) methodology and consider temporary grasslands (<5 years) part of croplands, by opposition to permanent grasslands (≥ 5 years)

The underlying reason of this division between croplands and permanent grasslands is that permanent grasslands are semi-natural ecosystems, in which humans appropriate only a small portion (5-30%) of the ecosystem biomass. By contrast, croplands are fully anthropized ecosystems, in which the aim is to maximize human appropriation of biomass (often >80%); for more details on the concept of human appropriation of ecosystems net primary production, see Haberl et al. (2007) or Fetzel et al. (2017).

Figure 5.2 synthesizes the input flows in the whole food / excreta system. Out of simplification for our subject of interest, it does not represent the N losses (volatilization, leaching...); see Billen et al. (2014) for a more complete representation of N flows in the food system. At the cornerstone of croplands and permanent grasslands lies the livestock system, whose inputs are the feed outputs of croplands and per-

manent grasslands. The main output of the livestock system is manure, which is an important input of both grasslands and croplands. The smallest but most important output of livestock is the animal products (meat, milk, eggs...).

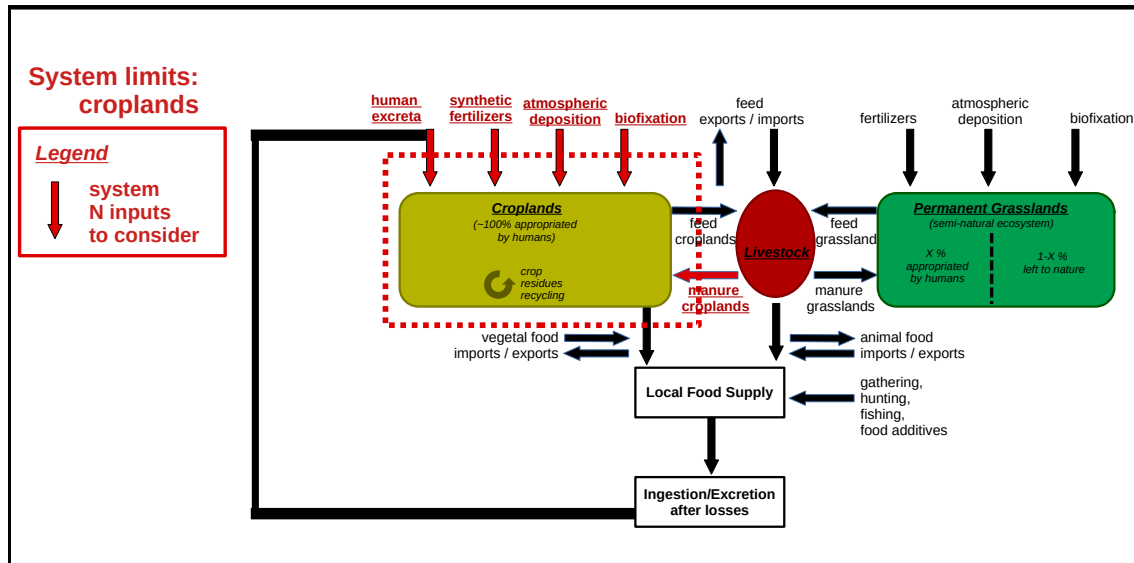


Figure 5.2: N flows in the entire food / excreta system, limits of the "croplands" system, and associated input flows, in red, considered by FAOSTAT (2022b) (FAO does not consider human excreta). As it is not my main interest here, N losses (volatilization, leaching) are not represented, out of simplification. For a more complete representation of N flows, see the GRAFS (Generalized Representation of Agro-Food Systems) approach (Billen et al. 2014).

In traditional farming systems, livestock, and especially large ruminants have several goals: prime movers for exhausting field work; convert non-edible feed like cellulose to human edible animal products; and above all transfer fertility from grazed grasslands to croplands, through manure (Smil 2018; Mazoyer et al. 2017). In modern intensive systems, they increasingly convert vegetal human-edible food to more palatable animal food: according to Chatzimpiros et al. (2023) (in their extended Table 2), the share of crops used for feed at the global scale increased from 48 to 57% from 1961 to 2013.

5.2.3.2 System: Croplands, for all countries

If the considered system is only croplands⁵, this makes the analysis straightforward (Figure 5.2). The croplands inputs to compare human excreta to are: synthetic fertilizers; atmospheric deposition; legumes biofixation; and livestock manure applied to croplands. Note that manure is not a "real" new inputs, as it is issued in the first

⁵In FAO's definition (FAOSTAT 2022b), croplands include permanent crops (coffee, cocoa, vineyards...) and arable land, and excludes permanent grasslands. In more details, arable land is made of temporary (<1 year) crops and temporary (<5 year) meadows, pasture, fallow. Consequently, grasslands are considered permanent if they have not been sown for 5 years or more.

place from synthetic fertilizers, biological fixation or atmospheric deposition ending up in feed eaten by livestock. Considering manure as an input is a consequence of limiting the system boundary to croplands. This is addressed in the following subsection focusing on all agricultural land, including croplands and grasslands.

The cropland inputs methodology is standardly used by the FAO in its cropland nutrient balances methodological note (FAOSTAT 2022b). The FAO reports, for each country, the following N inputs: synthetic fertilizers, manure, legumes biofixation, and atmospheric deposition (see Section 5.2.4).

Note that mineralization / immobilization from the soil organic matter can also contribute to crops growth. It is usually not considered in N balances, and approximated to be about 0, but is not always negligible. If the harvested flow is close or superior to the inputs flows (N Use Efficiency (NUE) $> 100\%$ ⁶), this suggest non-negligible net mineralization, and that N mining from soils occurs. This is the case for instance for Argentina over 1960-2010 in Lassaletta et al. (2014) or in the US Great Plains in the late XIXth / early XXth century (Cunfer 2021). On the contrary, low NUEs can sometimes hide enrichment of N in the soil organic matter, instead of losses through leaching or volatilization.

5.2.3.3 System: All Agricultural land (Croplands + Grasslands), only at global scale

Another option is to focus on all agricultural land (Figure 5.3), made of both croplands and grasslands. This analysis is less straightforward than for croplands, and data less accessible, so I will undertake the analysis only on a worldwide scale.

In that system boundary, livestock manure on croplands is no longer an input and becomes an internal flow of agricultural land, similarly to crop residues recycling in croplands. However one has to consider the imports for livestock feeding. As I will only consider the whole world for this system, feed imports/exports will be null. Cropland inputs other than manure are unchanged, but the subtlety lies in the flows from permanent grassland. I will use 2 different methods to account for these flows.

Method 1: grassland inputs corrected for human appropriation The first method focuses on grasslands inputs, and is detailed on Figure 5.3, top. As in croplands, inputs to grasslands are synthetic fertilizers, atmospheric deposition, and biofixation. Grasslands synthetic fertilization (10 MtN) is deducted by removing cropland synthetic fertilization (100 MtN, FAOSTAT (2022a)) to the total synthetic fertilizer consumption for agricultural use (110 MtN, FAOSTAT (2022c)). For atmospheric deposition, I use the global $8 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$ reported in Chatzimpiros et al. (2023).

⁶the N Use Efficiency NUE is computed as $NUE = N_{in}/N_{out}$

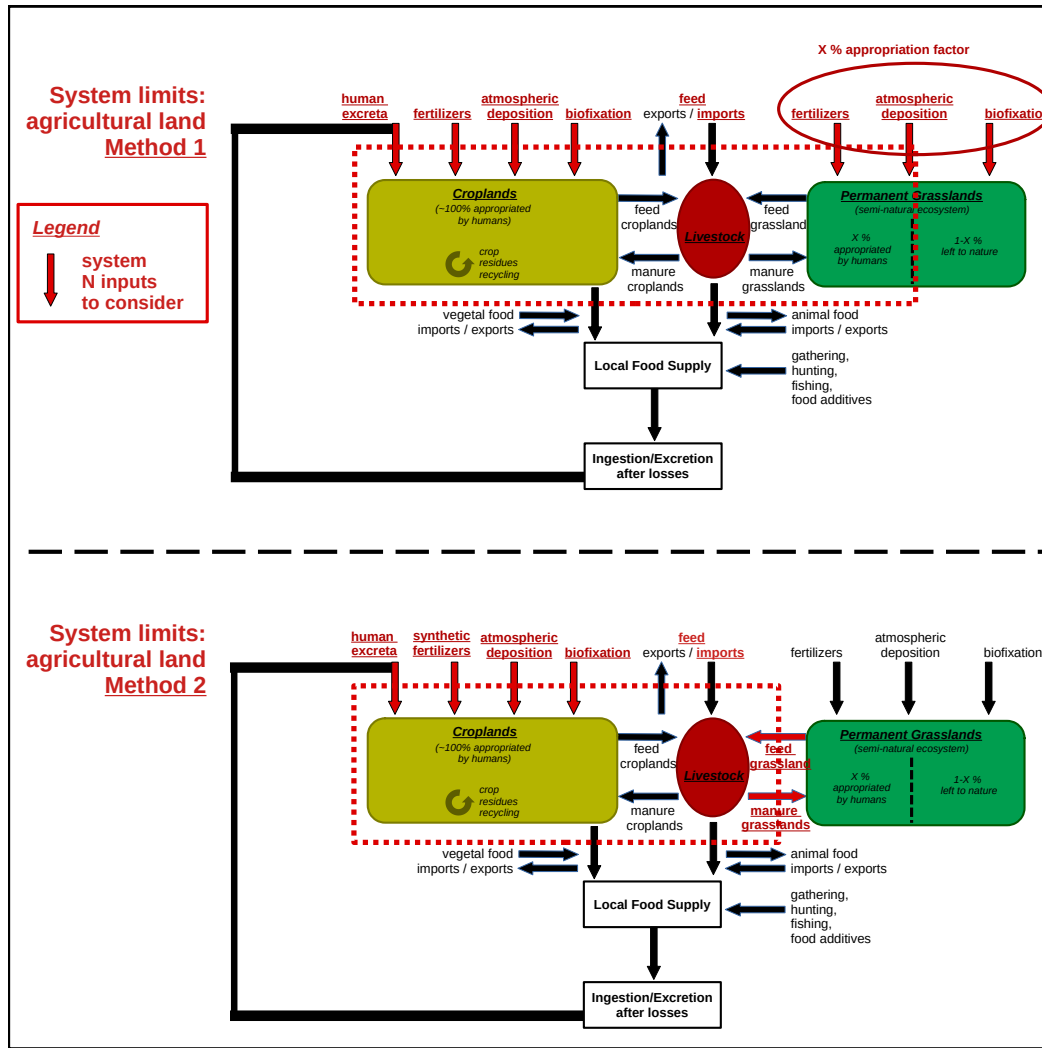


Figure 5.3: Limits and N inputs of the whole agricultural land system, for different methods. Top: counting all N inputs to grasslands, corrected by the estimated share appropriated by humans through livestock grazing. Bottom: counting only the net appropriation of grasslands through grazing (grazing minus manure fertilization, see text for details).

There are much more uncertainties for grassland biofixation. Cleveland et al. (1999) assessed biofixation in ecosystems, and found a central estimate of 80 MtN for grasslands but with large uncertainties: from 40 to 120 MtN (see their table 13, for categories: tall/medium grassland; short grassland; tropical savanna; arid shrub land; wet savanna; and Mediterranean shrub land). But contrary to croplands, only a portion of permanent grasslands is appropriated by humans, and a corrective factor should in all rigor be applied to reflect this reality.

Tentative assessments have estimated the grazing intensity of permanent grasslands, defined as the appropriation of net primary production (NPP) by grazing livestock, and found a rate of about 10% at the global scale (see table 2 in Haberl et al. (2007): human appropriation of 41 out of 433 $\text{gC}\cdot\text{m}^{-2}\cdot\text{year}^{-1}$ for grazing land). A

study specially focusing on grasslands developed a grazing index, with a similar rate of about 15% (Fetzel et al. 2017) but once again with strong uncertainties (ranges from 6% to 30%). In addition, this figure only concerns carbon NPP, yet livestock may find N-rich grass more or less palatable, changing the N-appropriation rate.

Based on these elements, I will use a 15% appropriation rate as an order of magnitude to correct the total inputs. Coupled with a total N inputs of 75-155 MtN in grasslands, this results in a N appropriation of grassland inputs from humans of 10-25 MtN (Table 5.2).

Table 5.2: Parameters and results of global grasslands inputs, for method 1 (see Figure 5.3, top).

Variable	Value	Source
grassland appropriation by humans	15%	Fetzel et al. (2017)
grasslands area	3.2 Gha	FAOSTAT (2022e)
atmospheric deposition rate	8 kgN/ha	Chatzimpiros et al. (2023)
synthetic fertilization on grasslands	10 MtN	FAOSTAT (see text)
atmospheric deposition on grasslands	25 MtN	computed
biofixation on grasslands	40-120 MtN	Cleveland et al. (1999)
total inputs on grasslands	75-155	computed
<i>method 1</i> : appropriated grassland inputs	10-25 MtN	computed

Method 2: net extraction from grasslands My *method 2* tries to reduce the previous uncertainties regarding grassland fixation and human appropriation, by only focusing on the net N extraction from grasslands, easier to monitor through livestock grazing and excretions. For that I will consider a slightly different system, in which I only consider the net appropriation from grasslands by livestock (Figure 5.3, bottom). I define the net appropriation from grasslands as the following:

$$N_{net\ appropriation\ from\ grassland} = N_{grazed} - N_{net\ return\ to\ grassland}$$

with

$$N_{net\ return\ to\ grassland} = N_{manure\ left\ on\ grassland} - N_{manure\ leached\ / \ volatilized\ on\ grasslands}$$

With that definition, the N in manure excreted on grasslands that is eventually leached or volatilized is considered "appropriated" by humans. This is consistent

with the way inputs are considered in croplands: out of the 100 MtN of synthetic fertilizers spread on croplands, a part will be lost through leaching and volatilization, but is still computed as inputs.

N in manure left on grasslands and the associated leached and volatilized quantities are reported in FAOSTAT (2022f), but N grazed is not reported. To estimate it, I use a N mass balance of livestock:

$$N_{grazed} + N_{feed\ from\ croplands} = N_{excreted} + N_{livestock\ production}$$

i.e.,

$$N_{grazed} = N_{excreted} + N_{livestock\ production} - N_{feed\ from\ croplands}$$

The total N excretions are reported by in FAOSTAT (2022f). Regarding feed from croplands and livestock production, I use data from extended figure 3 in Chatzimpiros et al. (2023). All the computation is summarized in Table 5.3.

Table 5.3: Parameters and results of net extraction from grasslands, for method 2 (see Figure 5.3, bottom)

Nitrogen Flow	MtN	Source
excreted manure	120	FAOSTAT (2022f)
animal production	10	Chatzimpiros et al. (2023)
animal feed from croplands	40	Chatzimpiros et al. (2023)
grazing	90	computed
manure left on grasslands	90	FAOSTAT (2022f)
manure volatilized and leached	45	FAOSTAT (2022f)
<i>method 2</i> : net extraction from grasslands	45	computed

5.2.4 N inputs to agriculture

Now that I defined the system limits and input flows to consider in Section 5.2.3, I detail these flows and their data sources. Even though this section only describes N flows and the method to get them, I present numerous graphs, which are not results of my work *per se*, but only ways to visually get a sense of FAOSTAT data.

5.2.4.1 N synthetic fertilizers consumption

For synthetic N fertilizers consumption, I use the data reported on FAO's page *Fertilizers by nutrient* (FAOSTAT 2022c). It reports agricultural use of N, P and K mineral fertilizers for each country. This agricultural use corresponds to the country's production adjusted for imports/exports, but does not consider stocks variations, assumed negligible. The relation $agricultural\ use = production + imports - exports$ has small discrepancies (<10% for France or at the global scale): this may be due to the fact that FAO's agricultural use does not consider synthetic N used for animal feed additives or industrial use (FAOSTAT 2023b) ⁷.

Agricultural use of fertilizers accounts for fertilizers spread on all agricultural land, be it croplands or permanent grasslands. For most countries, fertilization of croplands accounts for the vast majority of total fertilization, and permanent grasslands are barely fertilized. For these countries, total fertilization can be approximated by croplands fertilization. But there are some rare countries which strongly rely on grasslands fertilization and where the approximation does not hold. This is discussed in more details in the following sections and Figure 5.8.

5.2.4.2 N in manure

Table 5.4: Simplification of FAO's livestock categories

FAO's simple category	FAO's detailed category	My ruminants category
Cattle	Cattle, dairy	Ruminants
Cattle	Cattle, non-dairy	Ruminants
Poultry Birds	Chickens	Non ruminants
Poultry Birds	Ducks	Non ruminants
Poultry Birds	Turkeys	Non ruminants
Mules and Asses	Mules	Non ruminants
Mules and Asses	Asses and Hinnies	Non ruminants
Camels and Llamas	Camels	Non ruminants
Camels and Llamas	Llamas	Non ruminants
Sheep and Goat	Goats	Ruminants
Sheep and Goat	Sheeps	Ruminants
Swine	Swine, breeding	Non ruminants
Swine	Swine, market	Non ruminants
Buffalo	Buffalo	Ruminants
Horses	Horses	Non ruminants

Manure excreted FAOSTAT reports N flows concerning livestock manure for each country on its page *Livestock Manure* (FAOSTAT 2022f). Their methodology

⁷the exact relation is $production + imports - exports = agricultural\ use + other\ uses$. If no data is available on other uses, FAO considers it equal to zero, and $agricultural\ use = production + imports - exports$ is exact. Otherwise, there are small discrepancies. At the global scale (so with no imports/exports, FAO reports for 2020 a production of 121.5 MtN and an agricultural use of 114.5 MtN, implying 7 MtN (or about 5%) of "other uses".

for emission factors (FAOSTAT 2023c) is based on IPCC guidelines. FAO's estimates of excreted N is obtained by multiplying the number of livestock heads by coefficients depending on animals typical mass and N excretions, specific to each country. I aggregate FAO's livestock categories into ruminants and non ruminants following the simplification detailed in Table 5.4.

For most countries, ruminants excretions represent the majority of livestock manure (Figure 5.4). The only exceptions are Taiwan and Denmark. Japan, Indonesia, Korea and Vietnam are around 50/50%. However one cannot simply consider only excretions, as the amount collected and finally spread on agricultural land differs between ruminants and non ruminants.

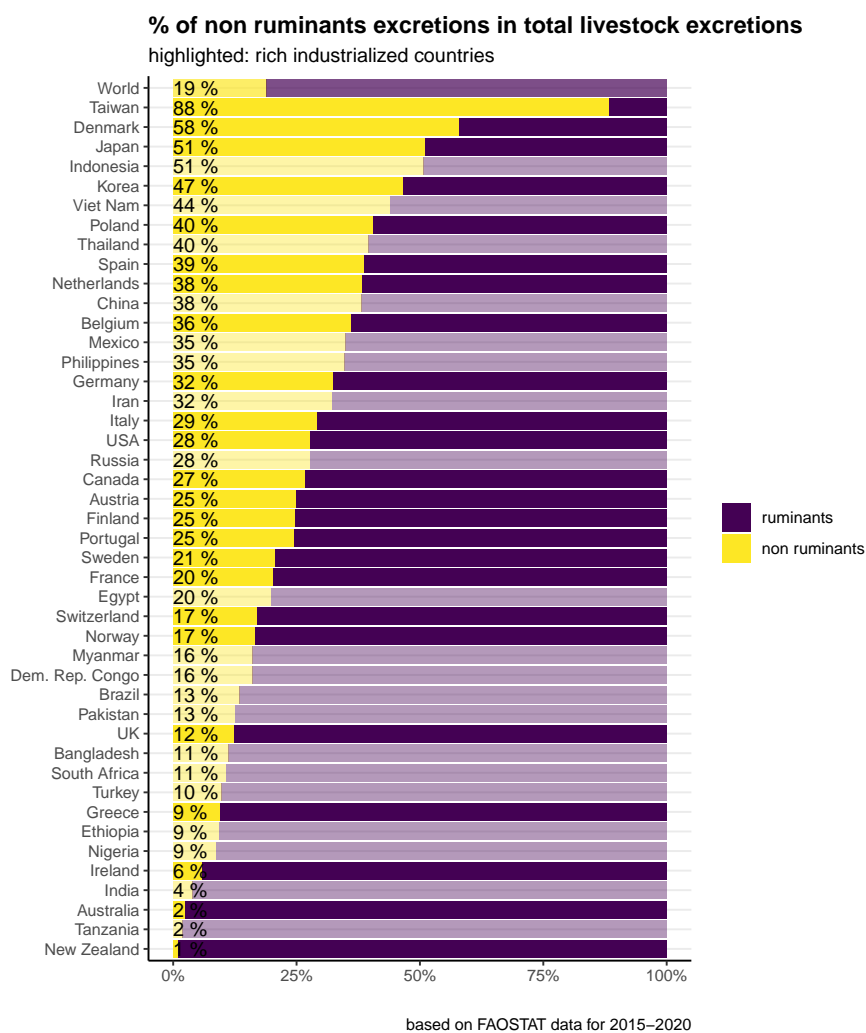


Figure 5.4: Non ruminants excretions as a share of total livestock excretions, based on FAOSTAT (2022f) data.

Manure applied to agricultural land The FAO methodology then discriminates between manure collected by farmers (called *manure management or manure treated*) and manure left on pasture, range and paddock. Only the collected manure

can eventually be spread on agricultural land. The share of collected manure depends on the country practices but also on livestock types: pigs in intensive productions are virtually always confined, ensuring high collection rates whereas ruminants graze on pastures and have lower collection rates. Volatilization losses then occur in collected manure, for instance during storage, diminishing the quantity finally spread. If manure is poorly stored, leaching can add to volatilization during storage.

The absolute distribution of these 3 categories (left on pasture, collected but volatilized, collected and finally spread) is exemplified in Figure 5.5 for some selected countries, for ruminants (right) and non ruminants (left). Representing absolute quantities highlights that even though ruminants generally excrete higher amounts of N than non ruminants, the quantity finally spread can be roughly similar for both categories. This is because of lower collection rates in ruminants which spend more time on grasslands, in opposition to non ruminants which are more often confined in the barn.

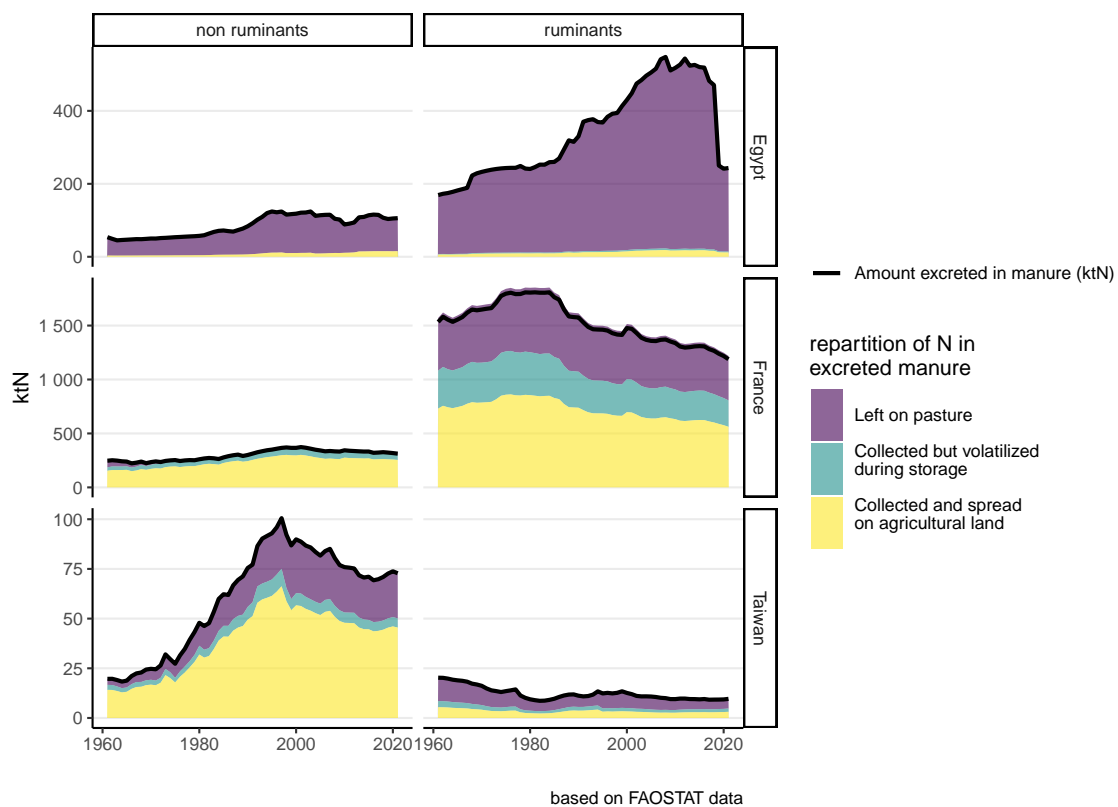


Figure 5.5: Fate of excreted manure in non ruminants (left) and ruminants (right) over 1960-2020, illustrated in contrasted countries. Ruminants generally excrete more than non ruminants (France and Egypt), but the amount finally collected and spread are usually comparable due to lower recovery rates for ruminants which spend more time in pastures, ranges and paddocks (see Figure 5.6). It also illustrates that collection rates can vary widely between countries, here for France and Egypt. In some rare countries like Taiwan, non ruminants excrete more than ruminants.

Figure 5.6 presents the relative destination of excreted manure for all of our countries of interest. In rich industrialized countries (highlighted), livestock production is more intensive, with animals more confined than in low and middle income countries. Consequently, in rich industrialized countries, a higher share of animals excretions is collected and finally spread. The collection rate across all livestock is usually close to the rate of ruminants, because these latter represent the majority of excretions.

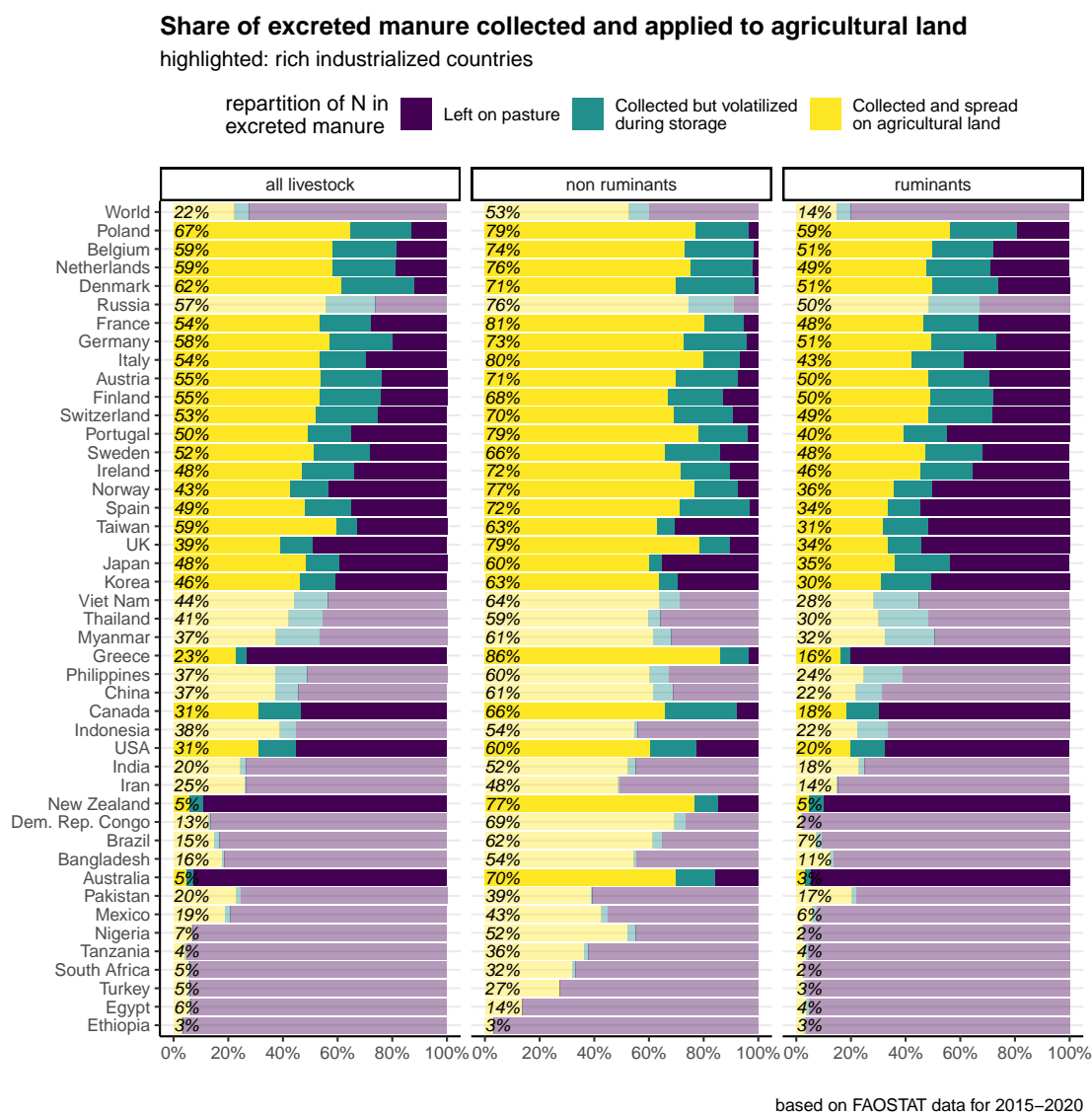


Figure 5.6: Share of excreted manure that is either left on pasture, range and paddock (purple); collected but volatilized indoor (blue); or collected and finally spread (yellow, mainly on croplands but sometimes grasslands). Numbers inform on the share finally spread. For all livestock (left), non ruminants (middle) and ruminants (right). Figures for all livestock are usually close to figures for ruminants, because these latter represent the majority of livestock excretions, except in some countries (see Figure 5.4)

Note that FAO also estimates losses once manure is spread, through volatilization and leaching. I do not consider these losses here, because there can be substantial

variations depending on more or less virtuous spreading techniques in each country. Moreover, there is also a variety of fertilizers based on human excreta, with different chemical properties resulting in different volatilization rates once applied to soils (Martin et al. 2020). Comparing human excreta to animal manure applied to soil would necessitate to consider the numerous possible combinations of practices and products, resulting in numerous analyses, hence my choice not to consider losses after spreading. Similarly, that would require estimating N losses after synthetic N applications, which is not assessed by FAO. References on losses in the field can be found in EMEP/EAA (2023) and Martin et al. (2023a).

5.2.4.3 Total N inputs to croplands

A limitation of only focusing on synthetic fertilizers or manure is that agricultural systems also rely on other sources of N inputs, namely biological fixation through legumes and atmospheric deposition. FAO provides an estimate of these 2 latter inputs, but only for croplands (FAOSTAT 2022a). It also reports seed inputs, which are comparatively small, and that I will neglect in my analysis. Figure 5.7 highlights the historical N inputs flows to croplands for some illustrative countries.

Logically, cropland fertilization should be a fraction of fertilization on total agricultural land, which includes croplands and agricultural land. This is indeed the case for manure, but not for synthetic fertilizers, where FAO's reported cropland fertilization is sometimes higher than total agricultural land fertilization (Figure 5.8). This is because for croplands fertilization, FAO combine its own data with International Fertilizer Association (IFA) figure, which can have slightly different methodologies (Section 5.2.4.1). This highlights the uncertainties in reported data.

Keeping these uncertainties in mind, rates of cropland to total agricultural land fertilization lower than 100% on Figure 5.8 indicate that some fertilization occurs on grasslands. At the whole world scale, 90-95% of the synthetic fertilizers and collected manure are applied to croplands. For most countries, FAO estimates a rate of 80 to 100%. The main exceptions are Ireland and New Zealand, where 80-90% of the fertilization occurs on permanent grasslands, because these countries are specialized on extensive ruminant production on grasslands. Intermediate countries are Finland, Switzerland and Netherlands, where croplands represent only 50-70% of total agricultural land fertilization. So for most countries, focusing on croplands allows to grasp the large majority (80 to 100%) of the country total fertilization by farmers.

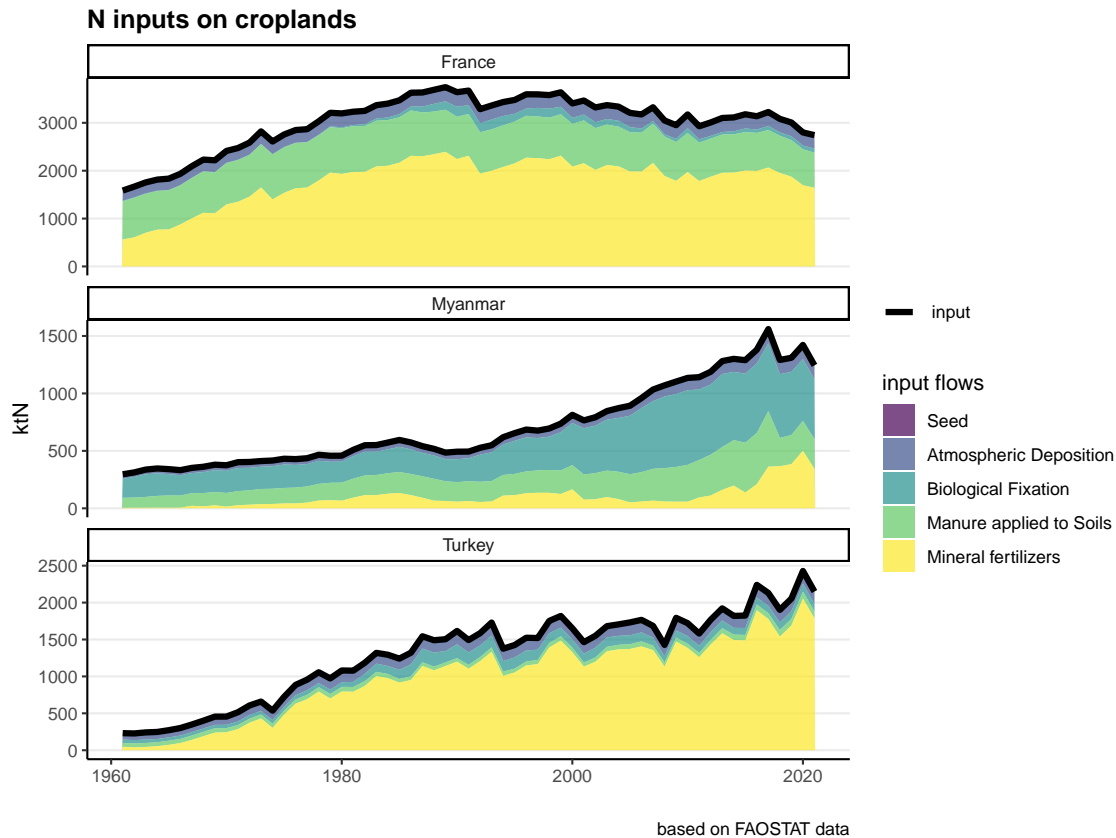


Figure 5.7: N inputs on croplands, 1960-2020 for illustrative contrasted countries. Myanmar (discussed in the following) relies extensively on biofixation, while Turkey’s inputs are almost only composed of mineral fertilizers. Seeds are not visible on the graphs because they are very small compared to the other inputs. In my following analysis I will neglect the seeds inputs.

5.2.5 N in total excretions and urine

N in food supply I use the FAO food balances (FAOSTAT 2022d) for each country in the world, which report protein available for human consumption (food supply)⁸. I convert proteins to nitrogen using Jone’s factor of 6.25 (100g proteins = 6.25 gN, Maclean et al. (2003)). The resulting N food supply, in kgN per capita per year, is presented on Figure 5.9 (right), which also discriminates between animal and vegetal products (left).

Contrary to Chapter 4 with France and INCA3 data on ingestion (Dubuisson et al. 2019; data.gouv 2021), there is no systematic international compilation of actual ingestion, but FAO reports the quantity available for human consumption for every country. This quantity does not account for the subsequent food losses and waste not ingested by humans. As reported by FAO’s definitions and methodological notes on food balance sheet (FAOSTAT 2022g), the food availability ”*refers to the quantities*

⁸This does not report protein in wild game, so the reported quantities may underestimate the supply in countries where hunting is a significant protein provider.

Cropland N fertilization as % of Agricultural Land N fertilization, for...

highlighted: rich industrialized countries

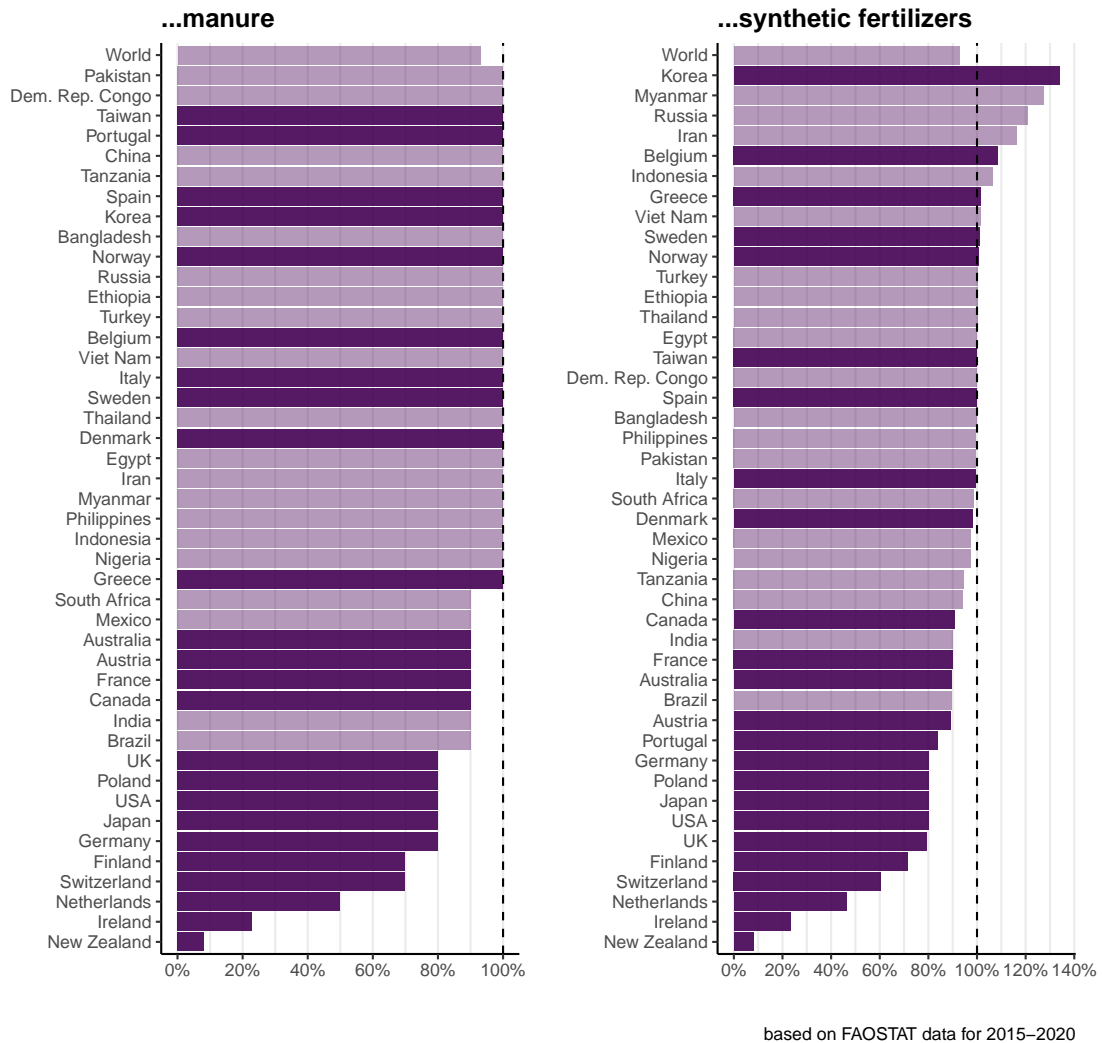


Figure 5.8: Cropland fertilization as a share of Agricultural Land (i.e., croplands + grasslands) fertilization, for manure (left) and synthetic fertilizers (right). The percentages are calculated by N_i on croplands/ N_i on agricultural land, with i standing for manure or for synthetic fertilizers. The rates higher than 100% for synthetic fertilizers are impossible, and reflect uncertainties owing to FAO's sources and methodology. Keeping these uncertainties in mind, rates lower than 100% indicate that 1-100% of the collected manure or synthetic fertilizers are applied to grasslands.

of food available for human consumption at the retail level". Losses pre-retail level are thus already subtracted from FAO's reported food supply, as highlighted by FAO's definition of losses for its food balance sheets: "loss most closely aligns with post-harvest/post-slaughter loss, representing those quantities of food that leave the production/supply chain at any stage from post-harvest up to the retail level (the level of the supply chain at which food availability is defined)".

So to estimate what is actually ingested and excreted, I need to subtract food waste and losses at the retail and consumer level from FAO's reported food supply.

Repartition of vegetal and animal products in N food supply

highlighted: rich industrialized countries

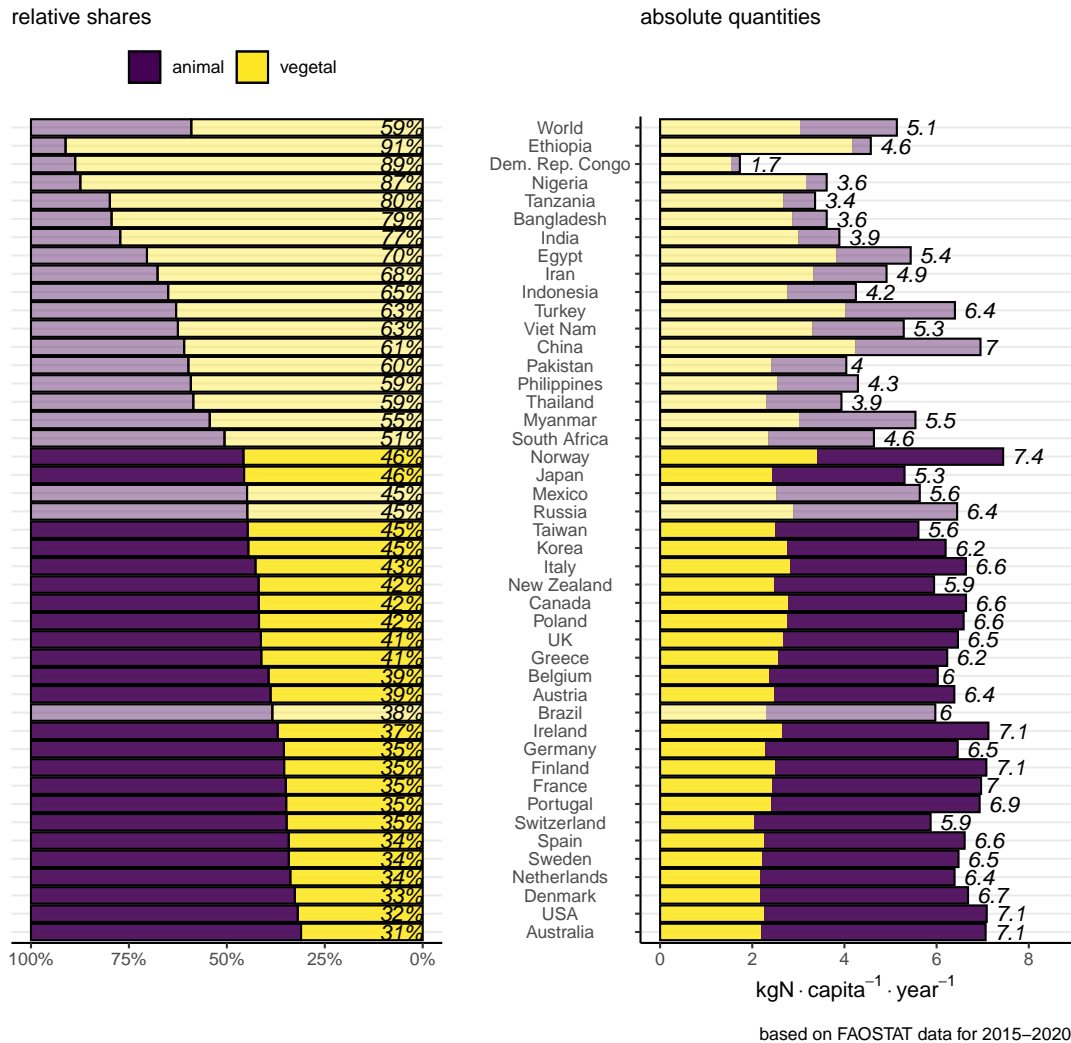
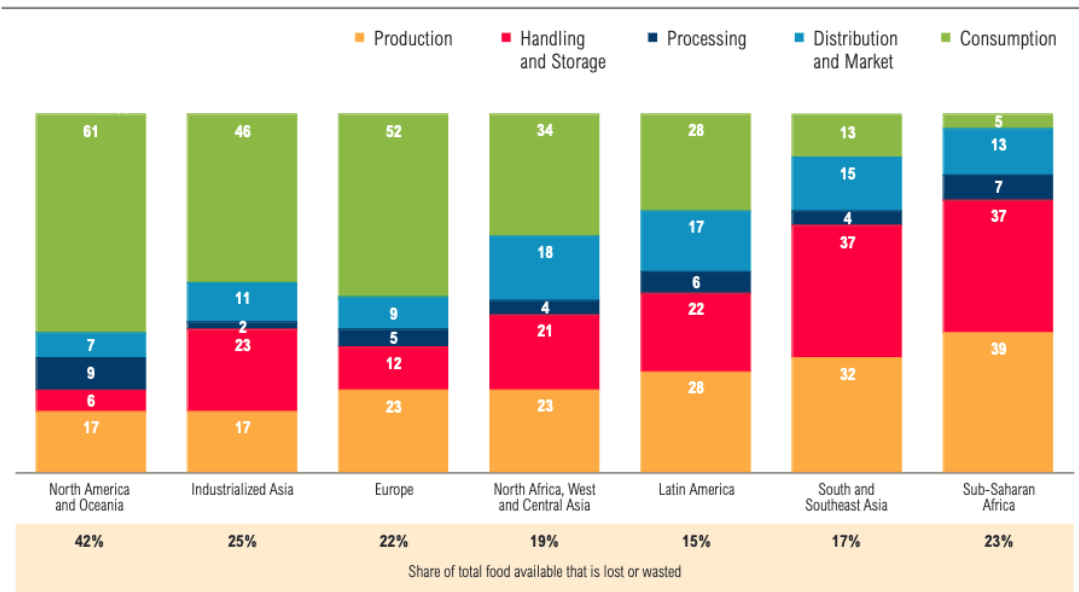


Figure 5.9: N food supply for human consumption per capita and per year, discriminated between animal and vegetal products. Numbers informs on the total N food supply. Food supply accounts for food losses during storage and transportation, but not for food losses and waste at the retail and consumer stages.

Food waste and losses A quite old but widely cited figure on food waste is that about "30% of the food produced for human consumption globally is lost or wasted somewhere along the food supply chain" (Rezaei et al. (2017), based on FAO (2011b)). I will use an analysis of the World Resource Institute (Lipinski et al. 2013) based on these same data from FAO (2011b), which details wasted and lost calories across the supply chain by world region (Figure 5.10).

To conform with FAO's definition of food supply detailed above, I only focus on the last 2 steps of the supply chain: "distribution and market" and "consumption". This yields a waste and losses rate after food supply ranging from 29% in North America to 4% in Sub-Saharan Africa. For my analysis, I will thus use a range of

Figure 6 | **Food Lost or Wasted By Region and Stage in Value Chain, 2009**
(Percent of kcal lost and wasted)



Note: Number may not sum to 100 due to rounding.

Source: WRI analysis based on FAO, 2011. *Global food losses and food waste—extent, causes and prevention*. Rome: UN FAO.

Figure 5.10: Food losses across the food supply chain, in kcal, from the World Resource Institute (Lipinski et al. 2013) based on FAO (2011b). Only focusing on waste and losses at the "distribution and market" and "consumption" stages yields the following percentages (from left to right): 29%, 14%, 13%, 10%, 7%, 5%, 4%.

5-30% as an uncertainty interval, and a central estimate value of 25%. This will make my estimate of excretions more conservative, and also more consistent with the few Western countries in which I have actual measures of N in excretions (see next section and Figure 5.11)

My central estimate of 25% food waste at the supply stage will tend to slightly overestimate N ingested and excreted in North America where the actual rate is more around 30%. On the contrary, this will lead to an underestimation of excreted N in low-income countries where food losses after supply are closer to 5-10%. A more precise estimate would of course need to discriminate the food loss and waste by world regions, but also between food commodities, since they do not all have the same protein content. The aim of this chapter is not to get the precise potential of human excreta in each country, but rather some relevant order of magnitude, and I did not have the time to pursue a more precise assessment.

Total excretions and Urine, after ingestion I hypothesize that all ingested N is finally excreted, assuming no storing in the body (see discussion in Section 4.1.2.1). This is a relevant assumption for adults who stopped growing up, but is less accurate for children and adolescents. Consequently, this hypothesis will slightly overestimate



Figure 5.11: Comparison of actual data on N excretions (points, see numbers in Figure 5.11 for more details) and my estimate after subtracting 25% food waste and losses (white) to FAO’s N food supply (total column). My proxy for excretions is the purple column. Uncertainty bars show the result when applying 5-30% food waste and losses. Rich industrialized countries are expected to be on lower end of uncertainty, low and middle income countries are expected to be on the middle or upper end of the error bar.

excretions in non industrialized countries which tend to have younger populations.

I compare my estimates using the above approximations to the few countries of our literature review where N in excretions were measured (Figure 4.7 to see original literature review). On Figure 5.11, the columns correspond to FAO’s N supply, discriminated between ingestion (purple) and the 25% food losses (white). The error bars correspond to the plausible range of ingestion/excretions considering the 5-30% food waste. This is compared to the actual measures of excretions, represented by the points (for France, I used the INCA3 survey on ingestion (Dubuisson et al. 2019; data.gouv 2021)).

When comparing excretions to N synthetic fertilizers, I only focus on urine and exclude feces. This is because only the urine part is highly mineral (Martin et al. 2021), with chemical properties very close to synthetic fertilizers, contrary to feces which is more organic. I consider that 85% of ingested N is excreted in urine (and thus 15% in feces), based on our literature review on Figure 4.8.

Potential recoverable share of excretions Once all the previous N flows are computed, I assess the potential of human excretions by comparing N in urine to synthetic fertilizers and total human excretions to all N inputs to croplands. The underlying assumption is that 100% of excreted N by humans is recovered and applied, which seems implausible. Yet I insist that the aim of this chapter is not to determine the plausible fertilization potential of human excreta, but rather its maximum theoretical potential. The reader can apply whatever recovery rate they find plausible.

I also compare human excretions to excreted manure, before recovery. In that case the 100% recovery rate assumption is not an issue. The only instance where the assumption is not adequate is when I compare human excretions to manure applied to agricultural land, which lets aside manure left on grasslands and lost during storage. In that case a 100% recovery rate would give an unfair advantage to human excretions. Thus, only in that particular instance, I have used livestock's best recovery rate of 80% for non ruminants (see Figure 5.6), applied to human excretions.

5.2.6 Uncertainties and approximations

This chapter aims at providing some rough estimates of the potential of human excreta, and I will round all the results to the nearest 5%, to focus on the orders of magnitude.

My main factor of uncertainty is the share of food losses after food supply. My 25% midpoint with a 5-30% range translates in a relative uncertainty in ingestion / excretions of -7% to +27%. All the other figures come from FAO's official data, which can have own inherent uncertainties. I mentioned it in the context of Figure 5.8 (right), where rates higher than 100% reflect inconsistencies in the estimation of total synthetic fertilizers consumption. However I did not find a systematic uncertainty estimate for each flow reported by the FAO. Consequently, I will only focus on the uncertainty on food waste and losses.

5.3 Results and Discussion

In the following graphs, the range of percentages between brackets (right part of the graph) represent the uncertainty relative to ingestion/excretion, due to the 5-30% possible values for food waste. The main percentage value is obtained with a 25% food waste. On the left part of the graphs, red dots are the values for a 25% food waste, and red bars (not always visible due to scaling) represent the uncertainty due to the 5-30% range of possible food waste.

Note that all the percentages presented in the following refer to human excreta as a percentage of current N inputs. Let's call it X . This is different from the potential contribution of human excreta to current N inputs. Let's call it Y . There can be 2 approaches for this potential: either human excreta *replace* some of the current inputs (Y_{replace}) or they are *added to* the current inputs (Y_{added}). If one envisions an agricultural system where human excreta *replace* some of the current inputs, then the potential is $Y_{\text{replace}} = X$. However, if one envisions a farming system where human excreta are *added to* current inputs, the potential considering this new input is $Y_{\text{added}} = X/(1 + X)$. If X is already low, then $X/(1 + X) \sim X$ and thus $Y_{\text{added}} \sim Y_{\text{replace}}$. For cropland inputs (Figure 5.13), this is the case for instance in France where $Y_{\text{replace}} = 10\% \sim 9\% = Y_{\text{added}}$; but the approximation does not hold for Japan where $Y_{\text{replace}} = 100\% \neq 50\% = Y_{\text{added}}$.

It is reasonable to consider that in countries with low N inputs, the aim should be to add human excretions to current inputs. On the contrary, in countries with already high N inputs, the aim should be to simply replace part of the current inputs, or even replace an diminish current inputs by modifying the food system, which even further increases the potential of human excreta. It is difficult to model such a food system change for all the countries I study in the following, so I chose to focus on the potential of human excreta in the *current* food system. I nonetheless explore such a change at the global scale in Section 5.3.4, and also for France in next Chapter 6.

5.3.1 Comparison of N in urine to synthetic fertilizers

Figure 5.12 compares N in urine to synthetic fertilizers consumption, in the current farming systems. Many countries rely on imports of synthetic N fertilizers, as Haber-Bosh units are concentrated in a handful of producers. Even when some Haber-Bosch facilities are located on its territory, a country might still rely on imports of natural gas, which is widely used to convert N_2 to NH_3 (but some country like China use coal). Comparing N in urine to synthetic fertilizers informs on the gains in terms

Comparison of N in urine to synthetic fertilizers consumption

highlighted: rich industrialized countries

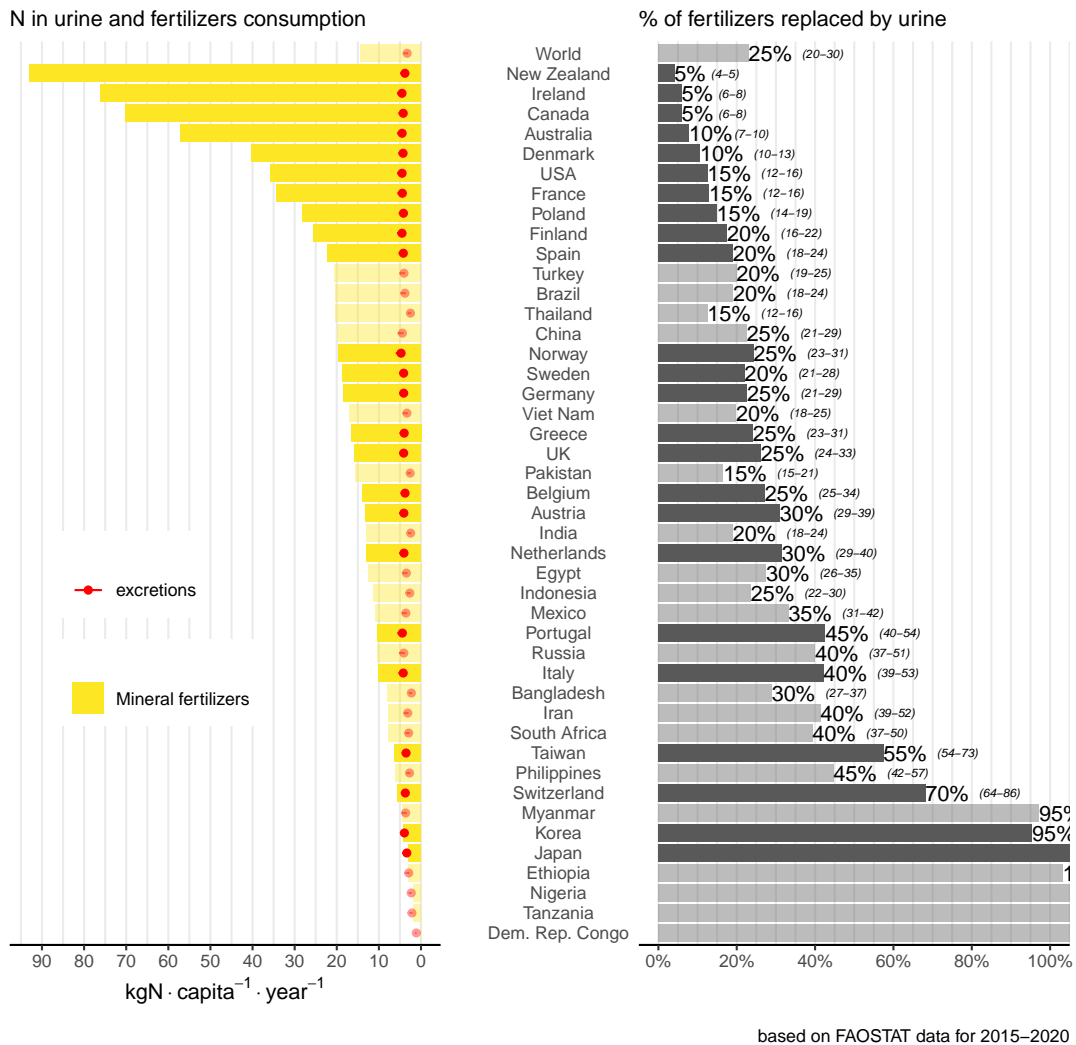


Figure 5.12: Left: N in human urine (dots) compared to synthetic fertilizers consumption (bars), in kgN per capita per year. Right: N in human urine as a share of synthetic fertilizers consumption. Values cut for for greater scaling clarity are: 105% for Ethiopia, 135% for Nigeria, 125% for Tanzania, and 540% for Dem. Rep. Congo.

of energy and fertilizers self-sufficiency⁹. This adds to the environmental benefit, positive under the majority of scenarios comparing urine-based fertilizers to the current Haber-Bosch + wastewater treatment plant (Martin et al. 2023a).

Figure 5.12 shows that for most countries, synthetic fertilizers cannot be fully replaced by urine-based fertilizers: the gains in terms of independence to imports are thus relative, and a full nutrient self-sufficiency would require more structural changes in the food system.

⁹For instance, from the 1970s to the late 2010s, France generally produced 1.3 to 1.7 MtN on its territory, for a agricultural use of 2-2.5 MtN. But self-production fell below 0.5 MtN in the 2010s and almost 0 MtN starting 2020, while consumption is still around 2 MtN (FAOSTAT 2023a).

For most rich Western industrialized countries, N in urine represents 15-30% of their synthetic N fertilizers consumption, with Italy and Portugal a bit higher at almost half (40 and 45%). Way higher, at 50 to 100%, are Asian rich industrialized countries which have largely outsourced their agricultural production (Taiwan, Korea and Japan), and Switzerland. The low end of the spectrum (5-15%) is occupied by countries which export a large share of their agricultural production (Canada, USA, France, Australia, Denmark). Even below, around 5-10%, are countries specialized in ruminant production on fertilized grasslands (New Zealand, Ireland).

When focusing on large countries not belonging to the rich industrialized group, the typical range is shifted towards slightly higher values, around 15-40%. Way higher are the sub-Saharan countries (Ethiopia, Nigeria, Tanzania and Democratic Republic of Congo) where N in urine could cover more than their current consumption of synthetic fertilizers, reflecting their low N inputs to agricultural systems (see Figure 5.13 in next section on that matter). One outlier is Myanmar, which has high N inputs but where N urine is almost equal to synthetic fertilizers, and whose case will be detailed in the following section.

Considering the whole world, N in urine represents around 25% of the synthetic fertilizers consumption in the current farming system. Table 5.5 summarizes the typical orders of magnitude according to countries' typology.

Table 5.5: Comparison of N in urine to synthetic N fertilizers in the current farming systems, by country typology. See text for countries examples.

country typology	N in urine as % of synthetic N fertilizers
rich Western industrialized	15-30%
rich Asian industrialized	50-100%
agricultural exporter	5-15%
sub-Saharan	> 100%
low and middle income	15-40%
World	25%

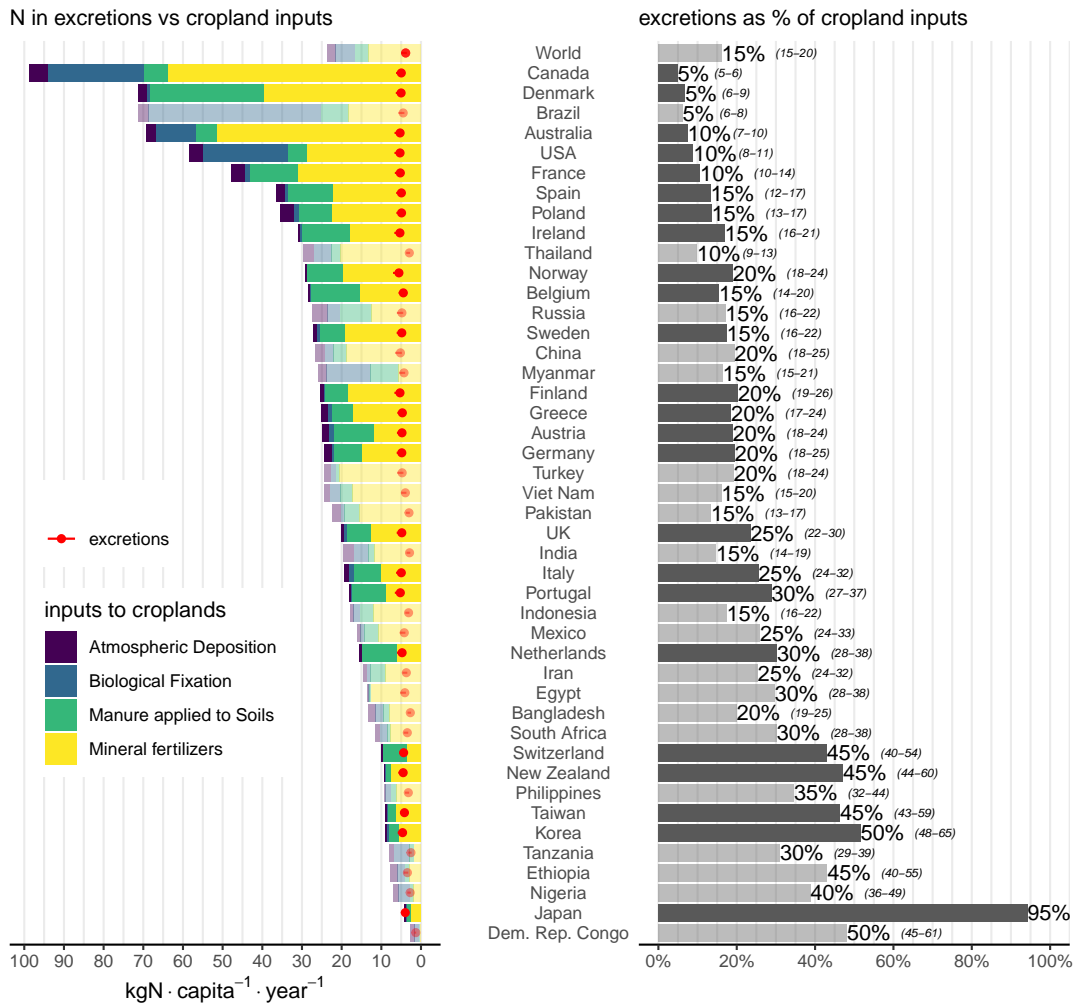
5.3.2 Excretions compared to N inputs in croplands

Only focusing on synthetic fertilizers gives a sense of their possible substitution, but provides a partial view of the potential contribution of human excreta to the country's total fertilization, as it ignores biofixation, atmospheric deposition and manure fertilization. For that reason I also compare total human excretions to all N inputs in croplands in Figure 5.13.

The results are in the same order of magnitude as the previous paragraph for urine vs synthetic fertilizers, but slightly lower, generally by 5 to 10 percentage

Human excretions compared to croplands N inputs

highlighted: rich industrialized countries



based on FAOSTAT data for 2015–2020

Figure 5.13: Left: comparison of N in human excretions (points) to N inputs to croplands by category (bars), in kgN per capita per year. Right: N in human excretions as a share of total N inputs to croplands.

points. Several factors are at stake in this slight change. First, it concerns total human excreta and not only urine which is only 85% of N in human excreta (see Section 5.2.5), thus increasing the potential. Second, in this section synthetic fertilization only concerns croplands, in opposition to the previous section where it concerned all agricultural land. Cropland synthetic fertilization is usually 80-100% of total synthetic fertilization (Figure 5.8), except for some large outliers (10-20% for Ireland and New Zealand) and smaller outliers (50-60% for Netherlands and Switzerland). This also tends to increase the potential. Finally, synthetic fertilization usually represents about 50-80% of total cropland inputs (Figure 5.13, left), this time decreasing human excreta potential. There are a few exceptions below this 50-80% rate: sub-Saharan African countries, because of their little synthetic inputs; and Myanmar and Brazil, due to their relatively important legume biofixation.

Consequently, human excreta tend to represent 15-30% of cropland inputs in many rich Western industrialized countries. Rich Asian industrialized countries are around 50%, as Switzerland and New Zealand, and Japan almost 100%, once again reflecting their outsourcing of cropland production. In the low end, around 5-10%, are exporters and countries with intensive livestock: Canada, Denmark, Australia, USA and France.

In sub-Saharan Africa, human excreta represent around 30-50% of the current cropland inputs, reflecting the low external N inputs to agroecosystems. Most of low and middle income countries are around 15-30%, as for rich industrialized countries, with the exception of Brazil (about 5%) because of its large soybean production for exports. Myanmar is a particular case. It has low synthetic fertilizers inputs, around $5 \text{ kgN}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$, but contrary to sub-Saharan or rich industrialized Asian countries, Myanmar is not characterized by low total N inputs to cropland, experiencing levels similar to Germany or China. Its low synthetic fertilizer consumption genuinely reflects a farming system based on legumes biofixation and manure recycling. Contrary to Brazil, this legume production is not centered on soybean exported for animal feed, but rather on beans, of which only one third is exported (FAOSTAT 2022d).

Finally, when considering the world as a whole, human excretions represent an amount of N equal to 15% of current total N input to cropland. The orders of magnitude by country typology are summarized in Table 5.6.

5.3.3 Excretions compared to livestock manure

I first started my comparisons with synthetic fertilizers, which represent most of cropland inputs, and then with total cropland inputs. Comparing human excretions to these 2 flows give a good sense of the potential contribution of human excretions

Table 5.6: Total human excreta compared to cropland N inputs. See text for countries examples.

country typology	human excretions as % of current cropland N inputs
rich Western industrialized	15-30%
rich Asian industrialized	50%
agricultural exporter	5-10%
sub-Saharan	30-50%
low and middle income	15-30%
World	15%

to agricultural production in the current agricultural systems.

I now choose to compare human excretions to livestock manure. Livestock manure generally represents a lower share on cropland inputs compared to synthetic fertilizers (Figure 5.13, left). However I find it is useful to compare humans to livestock because, even though their excretions are similar from the viewpoint of an agroecosystem, human excretions are treated as a separate category, and often not even considered. While livestock excretions are not systematically collected (Figure 5.6), it would seem absurd to seek to actively *destroy* this collected N; yet, for human excretions, this is considered a standard treatment in wastewater treatment plants¹⁰.

I operate 2 distinct comparisons. First, total human excretions compared to total livestock excretions. This is useful to compare the metabolisms of humans and livestock in the different countries' agroecosystems. However for most countries, the majority of ruminants excretions are not collected and are simply excreted on permanent grasslands (Figure 5.6, right), constituting an internal flow into the grasslands system not contributing to net nitrogen export from agroecosystems to food products (see reasoning in Section 5.2.3.3). For that reason I will also compare human excretions to the share of livestock manure that is actually collected and finally spread on croplands. It gives a better sense of the amount of manure actually used to fertilize croplands. To ensure a fairer comparison, and only in that case, I will not consider a 100% recovery rate of human excretions, but only 80%, corresponding to the best recovery rates for non ruminants (Figure 5.6, middle, and Section 5.2.5).

¹⁰But in regions with a high density of livestock, for instance Brittany specialized in swine production, denitrification of slurry is sometimes proposed as a response to the overload of pigs slurry inducing local pollution.

5.3.3.1 Total excretions

A first interesting observation in Figure 5.14 (left) is that human excretions (red dots) are often similar or higher than non ruminant excretions (yellow bars). The main large deviations from this general rule are countries specialized in intensive non ruminant production: Denmark, Netherlands, and, to a lesser extent, Spain.

For most rich Western industrialized countries, human excretions represent about 20-40% of total livestock excretions (Figure 5.14, right), except for two outliers groups beyond and below that range. Above are rich industrialized Asian countries (Japan, Korea, Taiwan), which have outsourced their animal production, and where human excretions is close or above livestock excretions. On the other extreme lie some countries specialized in animal production, with high amounts of livestock excretions and in which human excretions is consequently small compared to manure, around 5-10%: New Zealand, Australia, Ireland and Denmark.

There is no clear typology for low and middle income countries, and the range of values is quite large, with human excretions typically representing 20-80% of livestock excretions. The main exceptions, around 5-10%, are countries with a higher number of ruminants: Brazil, Ethiopia, and Tanzania.

At the global scale, human excretions represent around 25% of all livestock manure, but 125% of non ruminants excretions.

5.3.3.2 Manure spread on croplands

Now I only consider manure collected and finally spread on croplands (Figure 5.15). This corresponds to my previous analysis on cropland inputs (Figure 5.13), but this time only focusing on manure.

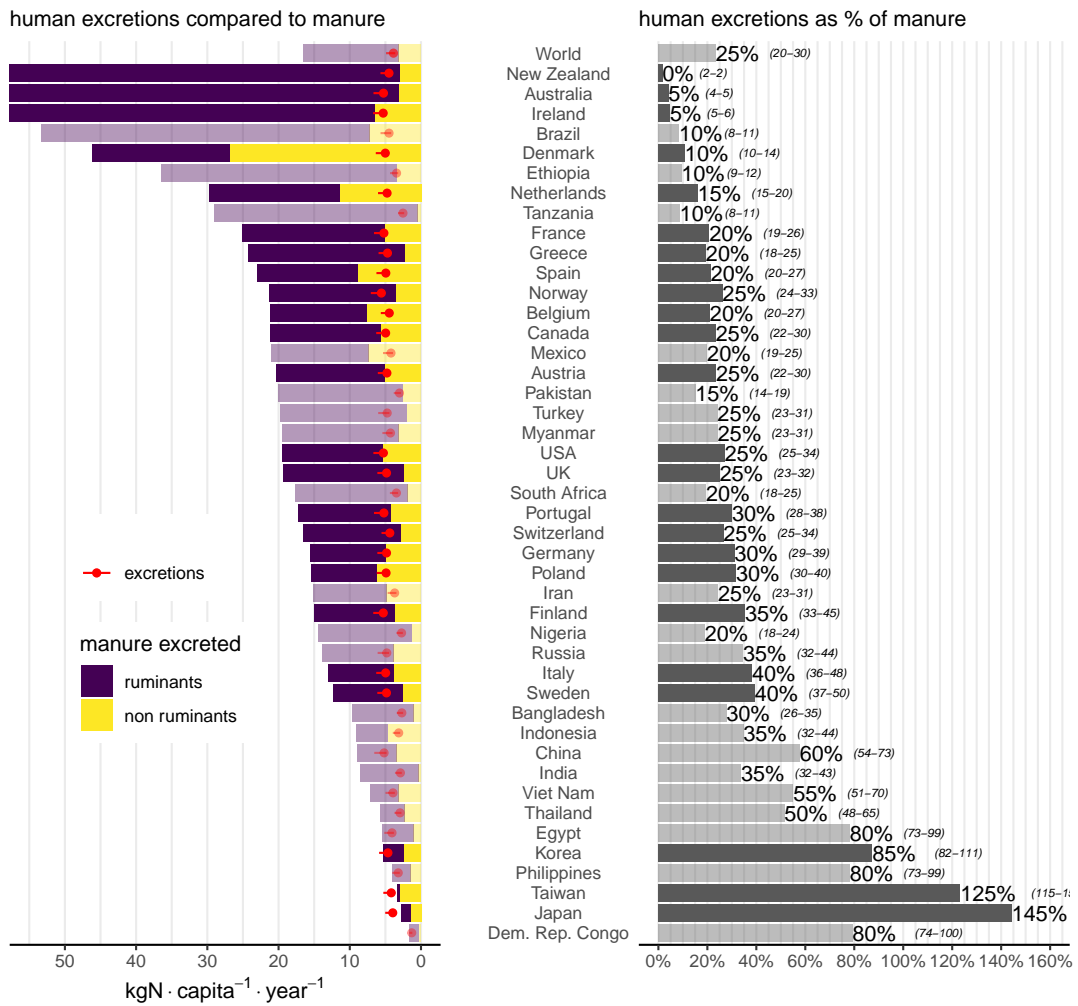
This yields a different result than simply comparing human excretions to total livestock manure. The main trend is that the rate in rich industrialized countries is lower than for low to middle income countries. This is mainly due to higher recovery rates of manure in rich industrialized countries (Figure 5.6), and to a lesser extent also more livestock excretions in these countries (Figure 5.14, left). Higher collection rates reflect the more intensive breeding in rich countries, where livestock, and especially non ruminants, are often confined.

In most rich industrialized countries, humans excretions represent 30-90% of manure applied to croplands. Above 100% are rich Asian countries, which have little livestock. New Zealand is also above 100% because, despite its numerous ruminants, they typically are bred on grassland and their manure is not recycled to croplands. Below, at 15%, is Denmark because of its high number of confined monogastrics, namely swines.

For most low to middle income countries the rate is close to 100%, or above, but

Human excretions compared to manure excretions

highlighted: rich industrialized countries

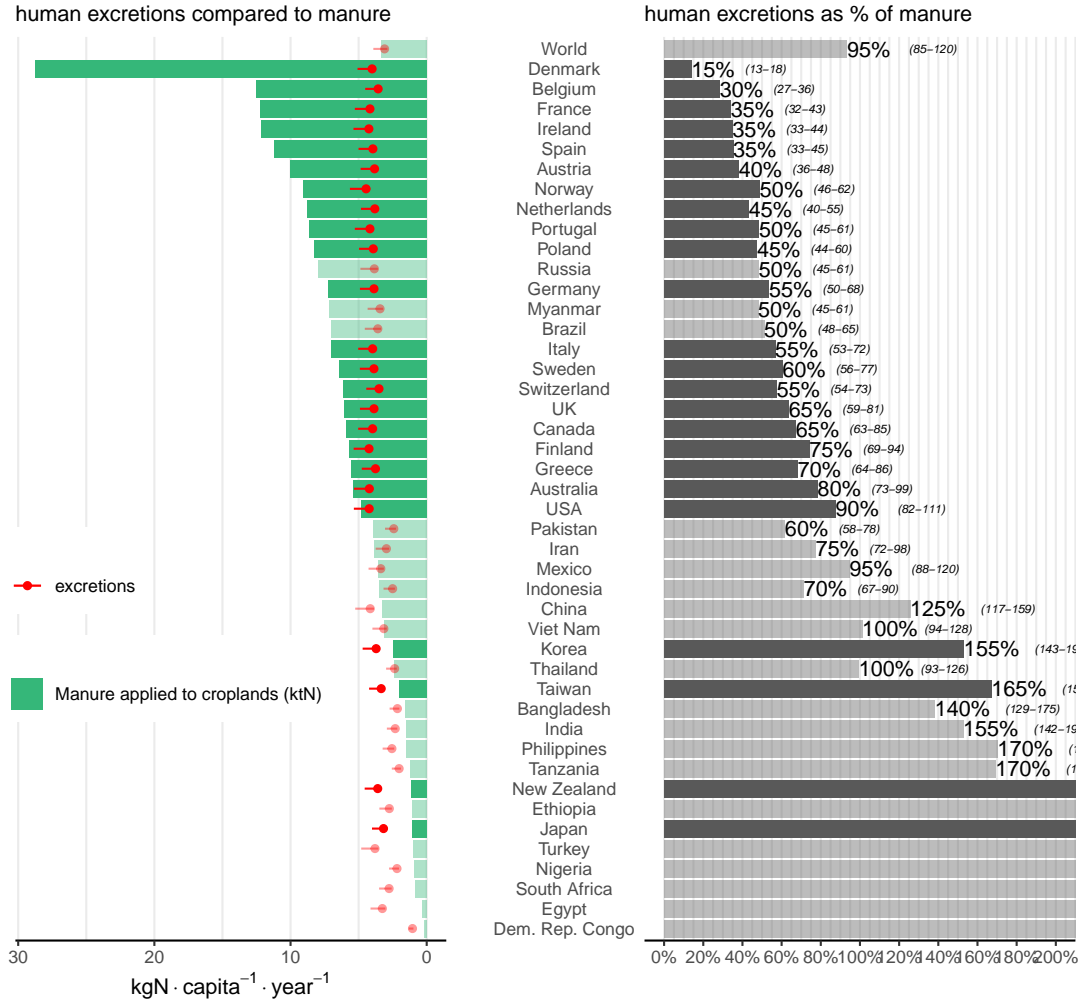


based on FAOSTAT data for 2015–2020

Figure 5.14: Left: comparison of human excretions (red dots) to animal excretions (bars) for ruminants (purple) and non-ruminants (yellow), expressed in kg of N per capita per year. Right: human excretions expressed as a share of total livestock excretions. Highlighted countries are rich industrialized countries. Bars for New Zealand, Australia and Ireland are cut for greater clarity (respective values 270, 125 and 110 kgN/cap/year).

Human excretions (80% recovery) compared to manure applied to croplands

highlighted: rich industrialized countries



based on FAOSTAT data for 2015–2020

Figure 5.15: Left: human excretions with 80% recovery rate (red dots) compared to livestock manure collected and applied to croplands, expressed in kgN per capita per year. See Figure 5.6 for more details on manure recovery rate. Right: recovered human excretions as a share of manure applied to croplands. Values cut for greater clarity are: 305% for New Zealand, 255% for Ethiopia, 300% for Japan, 375% for Turkey, 420% for Nigeria, 325% for South Africa, 1,000% for Egypt, 500% for Dem. Rep. Congo.

it has less meaning than for rich industrialized countries. Were the low to middle income countries recovering their animal manure as consistently as rich countries, the rates would be more similar.

At the global scale, hypothesizing a 80% recovery rate of N present in human excretions would yield a similar quantity to the manure currently spread on croplands.

5.3.4 Considering global Agricultural Land: current situation and agro-ecological scenario

5.3.4.1 Contribution of human excretions

Combining FAOSTAT data on N inputs in croplands (FAOSTAT (2022a) featured in Figure 5.13), and my estimates from grassland contributions with different methods (Tables 5.2 and 5.3), I get several estimations of total N inputs in the whole agricultural land, at the global scale (Table 5.7). This gives a range of estimations for human excretions as a proportion of the current system inputs.

The order of magnitude is 15-20%. This is broadly similar to when I compared human excretions to global cropland inputs (15-20%, Figure 5.13), which pleads for the consistency of the results. Indeed, at the global scale, when there is no feed imports / exports, the analyses for croplands and the agricultural land is approximately similar. With agricultural land boundaries, the manure spread on croplands becomes an internal flow and disappears from the inputs, but is replaced by the flows from the grassland compartments. In a first order analysis, manure spread on croplands is simply a transfer of fertility from grasslands to croplands (see Figure 5.3 and Figure 5.2).

Note that this 15-20% rate simply corresponds to the full-chain N use efficiency (NUE) of the food system: out of an input of 100 kgN in the agricultural system, how much is finally ingested (and excreted) by humans. Chatzimpiros et al. (2023) find a global NUE of 20% (in their figure 3a), but this rate stops at the production rate, before waste and losses. Applying a subsequent global 30% loss rate (see discussion in Section 5.2.5) gives an actual full-chain NUE of about 15%, similar to my result.

Chatzimpiros et al. (2023) also study the full-chain NUE under range of scenarios (their figure 3b), depending on 3 main parameters: the share of cropland production dedicated to feed animals, the recycling rate of manure to croplands, and croplands N use efficiency. Their share of cropland production dedicated to feed animals indirectly influences the share of animal proteins in diets, which varies between 20% to 60%, compared to currently 40% at the global scale. So, contrary to many scenarios related to food systems, the share of animal products in the diet is not a model input, but only a consequence of another input parameter. Their 20% to 60%

Table 5.7: Computation of human excretions as % of N inputs for the current world agricultural system

N inputs	value (MtN)
cropland inputs (FAOSTAT 2022a)	
synthetic fertilizers	100
biofixation	35-40
atmospheric deposition	15
total	150
grassland contribution (see Tables 5.2 and 5.3)	
method 1: appropriated grasslands inputs	10-25
method 2: net inputs from grasslands	45
total system inputs (computed)	
method 1: appropriated grassland inputs	160-175
method 2: net extraction from grasslands	195
total human excretions	
	29 (27-35)
human excretions as % of system inputs	
method 1: all grasslands inputs	18% (15-22)
method 2: 15% of grasslands inputs	15% (14-18)

range for animal proteins in global diet spans a generalization of mostly vegetarian diets (20% is close to current India) to a generalization of Western diets (60% is the UK value).

I adapt their results in Table 5.8, considering a current 30% food losses and waste or an improved 15%, coherent with the Sustainable Development Goal to halve it (Arora et al. 2022), and also the rate for the currently best performing sub-continent, Latin America (Rezaei et al. (2017) and see Figure 5.10).

Note that only focusing on NUE allows to ignore other parameters considered by Chatzimpiros et al. (2023): rate of biofixation by legumes and absolute amount of protein consumption. These parameters will influence the amount of N inputs, and thus the amount of population sustained and the N pollution due to losses, but not the full-chain NUE.

From these results, it appears that both drastically improving the food system efficiency or shifting to more vegetarian diets improves the full-chain NUE by 10-15

Table 5.8: Left column: global food system N Use Efficiencies (NUE) in different scenarios, before food losses and waste, based on results in figure 3b in Chatzimpiros et al. (2023). Rates after food losses and waste (2 right columns) inform on the upper bound of human excretions as a share of N inputs in agriculture. In one cell, low and high ends of the percentage ranges correspond to different diets regarding animal proteins consumption. High end of NUEs ranges typically corresponds to 20-30% animal proteins in diet, compared to 40% currently at the global scale. Much improved food system efficiency: global recycling of manure to cropland increases from 35% to 80% and global croplands NUE increases from 50% to 70% .

	Food system NUE across diets		
	before food loss and waste	after current 30% waste	after improved 15% waste
Current food system efficiency	20-35%	15-25%	17-30%
Much improved food system efficiency	35-55%	25-40%	30-45%

percentage points (Table 5.8)¹¹. From the current 15-20% global rate, it would be thus challenging to reach a 50% value. More probable values for frugal and efficient systems would probably be around 30-40%, effectively doubling the current rate. This gives a sense of the potential contribution of human excreta in a closed system, without imports nor exports.

With another approach, Billen et al. (2024) propose an agro-ecological scenario for Europe, and find a full-chain N use efficiency around 40%¹², but once again this stops before food waste and losses, which they set equal to 30%. Applying this subsequent loss results in a full-chain NUE of about 28% for their agroecological Europe. This is in contrast with their estimate for current Europe, which is 31% * (1 - 30%) = 22%. This further suggests that current wasteful food systems are around 15-20%, and more frugal systems around 30-45%.

For comparison, in the agro-ecological scenario for the Paris area developed by Esculier (2018) (cf his Figure page 431 of his thesis), the 3.9 kgN·cap⁻¹·year⁻¹ of intakes are issued from a total of 14.3 kgN·cap⁻¹·year⁻¹ of agricultural inputs in the food system, resulting in a total NUE of 27%¹³, close to the 30% by Billen et al.

¹¹The figures detailed in Table 5.8 do not consider the recycling of food wastes. Even 100% recycling of food waste leaves the order of magnitudes I present unchanged. For instance, in the last column of Table 5.8, recycling all of the 15% waste in the case of a 30% NUE gives a new NUE of $30/(100-30*0.15) = 31.4\%$. But it would be interesting to also focus on the "inevitable" losses occurring before consumption, for instance during processing.

¹²Their total NUE is 41%, but their scenario includes 25% recycling of human excretions (or 0.35 MtN), applied to market gardening. If we hypothesize 0% recycling and add the 0.35 MtN to the inputs, the new NUE is 39% (see their supplementary excel).

¹³I do not count his biowaste flow issued from food waste and losses of 0.5 kgN·cap⁻¹·year⁻¹, as

(2024).

Another notable relevant work has been carried out by Barbieri et al. (2021), but I did not have time to adequately analyze their results and properly translate them into a global NUE. They present the global N flows for the year 2000, as well as global organic scenarios. A key difference with the previous works is that while they find that providing adequate N intakes to the global population in a 100% organic scenario is feasible, the caloric supply would not be sufficient.

5.3.4.2 N losses and pollution

Reasoning in relative terms with NUE can hide the absolute losses, which are actually the primary interest since they often result in reactive N pollution. In the following, I will use NUE_{system} , whose scope goes down to the final ingestion and thus includes food waste and losses, contrary to the scopes presented in Figures 5.3 or the NUE reported in Billen et al. (2024) and Chatzimpiros et al. (2023). From this NUE_{system} , N inputs requirements and N losses can easily be deduced with only 2 parameters, the N ingested and excreted ($N_{\text{ingestion}}$) and the rate of recovery of human excretions to fertilize crops ($\%_{\text{N excretions recovery}}$):

1. $N_{\text{inputs requirements}} = N_{\text{ingestion}} / NUE_{\text{system}}$
2. $N_{\text{losses}} = N_{\text{new inputs}} = N_{\text{inputs requirements}} - N_{\text{ingestion}} \cdot \%_{\text{N excretions recovery}}$

Safe levels of global pollution have been proposed as thresholds, known as N planetary boundaries: 35 MtN in Rockström et al. (2009b) and then 62 MtN in Steffen et al. (2015) and Richardson et al. (2023), with a zone of uncertainty and increasing risk between 62-82 MtN (see discussion in Section 2.3.2). These pollution levels are approximated by anthropogenic new inputs of reactive N, made of (Steffen et al. 2015):

- anthropogenic industrial fixation of N via the Haber-Bosch process
- intended biological N fixation
- unintended N fixation of NO_x from transport and industry

Note that human excretions spread on crops are rightly not considered, since it is not a new reactive N flow. Unintended N fixation of NO_x currently contributes to fertilization through N deposition, but is not considered in the methodology of the N planetary framework, which only focuses on biological N fixation and industrial

I consider it as an internal cycling flow like animal manure; in any case, this would only marginally impact the NUE, down to 26%.

fixation (see discussion in appendix of Steffen et al. (2015))¹⁴. For my scenarios, I will consider that these 2 flows constitute all of the new reactive N inputs, since NO_x mostly comes from fossil fuel combustion, which should almost disappear in a post fossil-fuel society.

With these elements, I apply equations 1 and 2 to compute the new N inputs in a much improved and frugal food system (30-40% $\text{NUE}_{\text{system}}$, see previous section) and levels of N excretions recovery of 0 or 80%, and compare it to the N planetary boundary (Table 5.9). I hypothesize a global population of 10 billion, which is plausible by the end of the century. I also compare it to published agroecological scenario (Billen et al. 2024; Esculier 2018) using another methodology to check if my results are obviously inconsistent.

For Billen et al. (2024), I adapt their results, applied to Europe and its 538 million inhabitants, to the global situation. In their scenario, human ingestions and excretions are $3.5 \text{ kgN}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$ after food waste (or 1.9 MtN), of which 25% is recovered for fertilization. Total new inputs, excluding fertilization with human excretions, and thus corresponding to N losses, are 9.2 MtN, or $17.1 \text{ kgN}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$; but an adjustment is needed, since only 72% of the agricultural production is consumed, the rest being exported. Applying this ratio, the locally attributable new inputs are $12.3 \text{ kgN}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$. I then convert these per capita values to global values, using the 10 billion population. For context and comparison, Billen et al. (2024) scenario NUE before food waste is around 40% and their $\text{NUE}_{\text{system}}$ after food waste is around 30%, so in the low end of my 30-40% range.

For Esculier (2018), this is more straightforward as the scenario is represented in terms of N imprint per capita (Figure p 431 of his thesis), and is directly adaptable. The intakes/excretions are $3.9 \text{ kgN}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$, of which about 60% are recovered and spread on agricultural land (0.6 via sewage sludge and 1.8 via collected urine). His new reactive anthropogenic inputs are $11.9 \text{ kgN}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$, all coming from biofixation. His NUE is 27%, also close to the low end of my 30-40% range

The results in Table 5.9 show that only the highest $\text{NUE}_{\text{system}}$ (40%) and lowest N ingestions ($3 \text{ kgN}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$) allow to stay below the 62 MtN planetary boundary, at the condition of at least 50% N excretions recovery. For context, within the framework developed by Chatzimpiros et al. (2023), a $\text{NUE}_{\text{system}}$ around 40% requires:

- mostly vegetarian diets (20-30% animal products, typical of India cf Figure 5.9)
- high recovery rate of animal manure (80%) not necessarily compatible with free-ranging animal important for their welfare

¹⁴For context, in 2000-2010, at the global scale, Haber-Bosch fixation was 120 MtN, leguminous crops biofixation was 60 MtN, and NO_x from combustion was 40 MtN.(Sutton et al. 2013)

Table 5.9: Global N new inputs (Haber-Bosch + biofixation) compared to planetary boundary, in the current situation and agro-ecological scenarios. See text for details.

	N planetary boundary threshold	
	kgN·cap ⁻¹ ·year ⁻¹	global
Rockström et al. (2009b)	-	35 MtN
Steffen et al. (2015) and Richardson et al. (2023)	-	62 MtN
Current situation circa 2020		
with ~10-15% excretions recovery		
	kgN·cap ⁻¹ ·year ⁻¹	global
Global Population	-	8.1 billion
Ingestion/Excretion	4	33 MtN
N new inputs Richardson et al. (2023)	23.5	190 MtN
Scenario with 30-40% NUE_{system} and 0, 50 or 80% excretions recovery		
	kgN·cap ⁻¹ ·year ⁻¹	global
Global Population	-	10 billion
Ingestion/Excretion	3-3.5	30-35 MtN
N new inputs (0% recovery)	7.5-11.6	75-116 MtN
N new inputs (50% recovery)	6-10	60-100 MtN
N new inputs (80% recovery)	5.1-8.8	51-88 MtN
Adapted agroecological scenario (Billen et al. 2024)		
with 25% excretions recovery		
	kgN·cap ⁻¹ ·year ⁻¹	global
Global Population	-	10 billion
Ingestion/Excretion	3.5	35 MtN
N new inputs	12.3	123 MtN
Adapted agroecological scenario (Esculier 2018)		
with 60% excretions recovery		
	kgN·cap ⁻¹ ·year ⁻¹	global
Global Population	-	10 billion
Ingestion/Excretion	3.9	39 MtN
N new inputs	11.9	119 MtN

- decreased food losses and waste
- a global cropland NUE of 70% (similar to current France) instead of 50%
- finally, a 3 kgN·cap⁻¹·year⁻¹ intake, which is below most health agencies recommended intakes of 3.1-3.4 kgN·cap⁻¹·year⁻¹, even though there is a security margin in the way these thresholds are determined (Section 1.1.3.3).

A limitation of my estimates is that it does not consider fish products in N

ingestion, which would decrease the need for agricultural production; on the other hand, it only considers agricultural production for food, while a post fossil world would probably rely on fiber crops for textile, which would increase the fertilization needs. Note that the Billen et al. (2024) and Esculier (2018) scenarios do consider fish intakes.

5.3.4.3 Questioning the N planetary boundary threshold

I already discussed in Section 2.3.2 the revised and moving boundary threshold in 2009 and 2015. The results above highlight that published agro-ecological scenarios, whose low N leaching respect drinking standard, are largely above the N planetary threshold, as it is currently defined. This pleads for more local and context dependent verification of the impact of N, rather than a global figure. This also supports my remark made in Section 2.3.2 about the statement by De Vries et al. (2013), who said that feeding 9 billion people with just 52-80 MtN·year⁻¹ new reactive inputs might rely on improper accounting of internal flows like manure.

Within the planetary boundary framework, N pollution is approximated by new anthropogenic inputs. But these inputs can be lost as inert N₂, or build up the soil organic matter. Namely, in Esculier (2018), out of the 11.9 kgN·cap⁻¹·year⁻¹ new inputs from biofixation, 3.5 kgN·cap⁻¹·year⁻¹ actually build up the soil organic matter, and 1 kgN·cap⁻¹·year⁻¹ is denitrified as N₂ in wastewater treatment plants, so his actual pollution with reactive nitrogen is only 7.4 kgN·cap⁻¹·year⁻¹.

The approximation of the N surplus by Rockström et al. (2023) is probably a better proxy to account for N pollution. Their boundary of 61 MtN·year⁻¹ of global surplus translates for 10 billion inhabitants to 6.1 kgN·cap⁻¹·year⁻¹, much closer to Esculier (2018) agro-ecological surplus of 7.2 kgN·cap⁻¹·year⁻¹.

And, in my opinion, the most relevant approach is probably to locally compute nitrates leaching and ammonia volatilization, in the spirit of the work undertaken by Gerten et al. (2020). But with this approach, the notion of a unique and global "planetary" figure is lost.

5.4 Summary of the results and main findings

Here I propose to synthesize the main findings related to the potential of urine-based fertilizers and human excretions. The reader should keep in mind that for the following order of magnitudes, the relevancy and certainty is higher for rich industrialized countries where there is currently a low recycling rate of excreta.

For most countries, N in human urine represents 15-40% of their synthetic fertilizers consumption. Above are rich industrialized Asian countries and sub-Saharan Africa (100% or more). Below are agricultural exporters (5-15%, France around 15%). The figure for the whole world is around 25%.

Comparing total human N excretions (urine + feces) to total cropland N inputs yields similar but slightly lower results for most countries, with a rate of 15-30%. Asian rich industrialized countries and sub-Saharan Africa are around 50%, reflecting low inputs and agricultural imports, and agricultural exporters are around 5-10% (around 10% for France). The figure for the whole world is around 15%.

For most countries, the quantity of N in human excreta quantities is very often similar or higher than domestic non ruminant excretions. At the global scale, human excreta represent 25% of all livestock excretions (20% for France), and 125% non ruminants excretions (in France they are roughly equal). But a large part of livestock excretions is simply excreted by ruminants on grasslands. If we only consider the manure collected and applied to croplands, recoverable human excretions are roughly equivalent to that quantity at the global scale; but this differs widely between countries, because of very different collection rate of manure.

Finally, beyond croplands, I also considered the global agricultural land (croplands + permanent grasslands), where flows are more uncertain. My best estimate is that currently human N excretions represent 15-20% of the world N inputs to the farming system, a rate similar to the one for cropland inputs. Based on theoretical modeling, and with much less certainty, I find reasonable to consider a 50% rate as an absolute upper bound threshold for the potential contribution of human excretions to N fertilization, with more probable values for an autonomous, optimized and frugal food system scenario around 30%, maybe 40%.

All these percentages consider an implausible 100% recovery rate of N in human excretions. The rate should be adapted with recovery factors; but estimating a plausible rate of recovery was not the aim of this chapter, and would depend on many socio-technical factors.

To conclude, we saw in previous Chapter 4 that currently nitrogen in human excretions is negligibly used in agriculture, at least in urban industrialized countries. In more rural countries, especially low and middle income ones, the recycling is much more uncertain, but probably low. In this Chapter, we saw that the potential is non

negligible, especially in more frugal food systems with less losses and pollutions. The following Chapter will inform on how to unlock this potential, focusing on the logistical aspect of spatial matching between excretions and crops fertilization.

Chapter 6

Geographical matching between human excretions and agricultural fertilization

The previous chapter attempted to estimate the maximum potential contribution of human N excretions to agricultural fertilization, without considering any constraint. Of course, in reality, many obstacles can hinder a 100% recovery rate of human excreta. Farmers might not want to use excreta, and citizens might be reluctant to consume products fertilized with excretions; farming machinery, adapted to mineral fertilizers in intensive agriculture, might not be suitable to spread liquid fertilizers; farmers might want urine to fertilize their crops, but the supply chain from excretions to the farmer might not exist; alternatives to flush toilets might not be in place, etc (see discussion in Section 3.3.4, or in Joveniaux et al. (2022)).

In this chapter, I propose to focus on one particular constraint, in the case of France: the geographical matching of the supply side, human excretions, to the demand side, the fertilized fields. Indeed, previous Life Cycle Analyses suggest that the environmental benefit of urine-based fertilizers depends on the distance to be covered during their transport to the agricultural field (Martin et al. 2023a). The fundamental trade-off is due to the relative low concentration of nutrients in urine. Because of that, large volumes of water have to be transported to deliver a given amount of nitrogen. One solution to this is to increase urine N concentration, but this process also comes with its own energy consumption, which can exceed transport energy consumption on short distances (Figure 6.1).

To inform on this trade-off, I will assess the distances to be covered in the current French farming system if all human N excretions were to be recovered. Indeed, one can easily imagine a mismatch between excretions in large metropolitan areas and surrounding agricultural fields. Moreover, it is not necessarily desirable to spread all

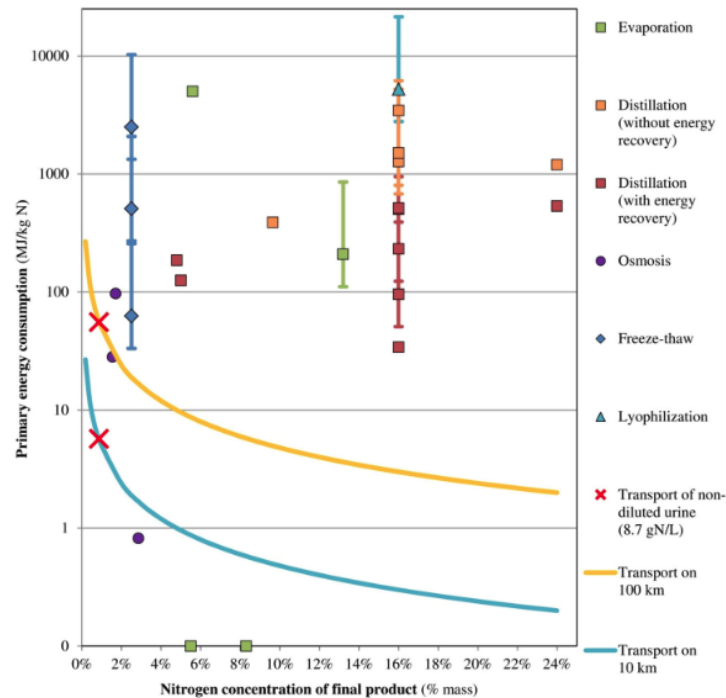


Figure 6.1: Energy consumption and nitrogen concentration of final product for the different N concentration treatments of urine (points and vertical bars). Yellow and blue line present the energy required to transport N at different concentrations for 100 km (yellow) or 10 km (blue). Red crosses are transport values for raw urine. Figure from Martin et al. (2020).

excretions on the nearest agricultural fields, since it would induce local N surplus and pollution, mirroring the current surpluses in regions with important livestock load, like Brittany. It is essential to limit total fertilization to acceptable levels preventing pollution, considering human excretions, livestock manure, and synthetic fertilizers all together. Taking into account these fertilization limits further increases the transport distances. I will identify the regions where mismatches might occur, were the use of human excretions to be generalized.

Finally, as we saw in the previous chapter, the demand side, i.e. how much fertilization there is on a given territory, is not a fixed constant, and can vary a lot depending on the contexts. France is particular because it has high N inputs on its croplands. I explore another situation, which is an hypothetical agro-ecological France with no synthetic fertilizers and where human excretions can be spread at a maximum rate of $50 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$ on non legumes crops (this value is established with a reasoning detailed in the following). In this scenario, I also consider that permanent grasslands are not fertilized with human excretions. This is because my aim is to enhance short-distance synergies between human excretions and crops eaten by humans, and permanent grasslands are only grazed by ruminants

I will focus on nitrogen, because its current reuse rate is low and $<10\%$, contrary

to phosphorus whose recovery rate is around 50% (cf Chapter 4). Given that phosphorus in excretions is already relatively reused as fertilizer through sewage sludge spreading, it would be interesting to study the actual distance covered by trucks carrying sewage sludge. I was not able to perform such a study, but this could be useful to compare my results to real world data.

The data to produce this chapter results was prepared in a GitHub repository and is featured on an interactive website.

An important intermediary result of this Chapter is the production of a map of N, P and K fertilization (and excretion) in France at fine resolution scale. The code to produce this is hosted on a Github repository¹ and associated with a DOI on a Zenodo repository²; this is rendered as user-friendly website³, from which are issued the maps featured here. The complete results, which could not be uploaded to Github because of their large size, are hosted on a second Zenodo repository⁴; for more information, see the README file on the Github page.

¹<https://github.com/Thomas-Starck/spatial-fertilization-excretions>

²<https://doi.org/10.5281/zenodo.14605711>

³<https://thomas-starck.github.io/spatial-fertilization-excretions/>

⁴<https://doi.org/10.5281/zenodo.14605243>

6.1 Methods and sources

This section describes how I obtained, on the one hand, a detailed dataset of N fertilization at the agricultural parcel scale, for the current situation and for an agro-ecological scenario (Section 6.1.1); and on the other hand, a dataset of human N excretions, at a 1 km² resolution for metropolitan France (Section 6.1.2). Then I describe the procedure to distribute human excretions to the nearest agricultural plots, considering the fertilization rates of each crop (Section 6.1.3).

6.1.1 Spatially explicit N fertilization in France

Several French research teams already produced spatial datasets of agricultural parcels at the field level (Crouy-Chanel et al. 2023; Lardot et al. 2022). They combine different complementary French databases to achieve this goal, as no single database reports all of the agricultural parcels. Générmont et al. (2022) go beyond and enrich this spatial data with much detailed fertilization practices. However, for their modeling needs, they do not keep the individual parcels and combine them at the ~ 700 French small agricultural regions level.

In the following I propose a methodology inspired by these previous studies to get a spatial dataset of fertilization in France at the agricultural parcels level. Compared to Générmont et al. (2022), the spatial resolution will be at the parcel level and not averaged over French small agricultural regions; however my description of fertilization will be coarser, only informing about nutrient quantities and not about spreading practices, timing, or number of spreading.

I describe first how I get a spatial dataset of all agricultural parcels in France (Section 6.1.1.1), and then how I attribute a fertilization rate to each of these parcels (Section 6.1.1.2), depending on the crop grown on it.

6.1.1.1 Spatial distribution of agricultural parcels. Resolution: agricultural parcel

My main source to localize each agricultural field in France and what is grown on it is the *Registre Parcellaire Graphique (RPG)*, or Graphical Parcel Register in English (géoservices.fr 2023b; IGN 2023b). It describes the location and areas of agricultural parcels from farmers who have submitted a declaration for the Common Agricultural Policy aid. For most crops, this database reports virtually all of the associated agricultural fields, including permanent grasslands. However it is incomplete for some crops, namely vineyards and orchards. Indeed, Crouy-Chanel et al. (2023) compared the difference in areas reported in the RPG and the official *Recensement Agricole* (Agricultural Census), and found at the Metropolitan scale a difference of

only -1% for corn, 1% for straw cereals and 3% for oilseeds; but the difference was -58% for orchards and -35% for vineyards.

Consequently, for vineyards and orchards, I use a second geographical database, which is the TOPO database (géoservices.fr 2023a; IGN 2023a). Among many other items such as buildings, vegetation or rivers, the TOPO database reports vineyards and orchards, more systematically than the RPG database. The difference in area reported in the TOPO database and the official *Recensement Agricole* is only -1% for vineyards and 3% for orchards (Crouy-Chanel et al. 2023).

The detailed description of agricultural parcels is constituted of more than 300 items and 24 simplified groups in the RPG dataset. For TOPO the only 2 items are vineyards and orchards. I gather these 2 datasets and use my own simplified crop labels. These simplified crops labels (30 items) and their attribution to the more than 300 RPG and TOPO original items are detailed in Appendix A. I use 4 main categories: major field crops (which also includes permanent grasslands), vineyards, orchards, and vegetables and floral crops. The process specific to each one of these 4 categories is detailed below.

Major field crops: RPG database I gather the RPG data for the years 2015-2020. Before 2015, the RPG reporting categories were different and less precise: data were not reported at the agricultural field level, but rather with grouped agricultural fields (called "îlots anonymes")⁵.

The data are reported as a shapefile for each of the 13 French metropolitan regions. I merge these 13 files together to create a national file. With a spatial join, I also assign each agricultural plot to one of the 22 old French region, before the 2015 reform, because I will use this classification to attribute fertilization values (see Section 6.1.1.2). The attribution of the detailed RPG crop codes to my simplified categories is described in Appendix A.1, Table A.1.

The final modified RPG dataset describes each agricultural plot with the following attributes: plot ID, French region (22 old regions and 13 new regions), year (from 2015 to 2020), detailed RPG crop (> 300 items), my simplified label (30 items), and a broad crop category. This broad crop category is made of the 4 main items: major field crops; vineyards; orchards; vegetable and floral crops.

The RPG dataset faithfully reports field crops and vegetable and floral crops. However they only partially report vineyards and orchards. For these, I remove the vineyards and floral agricultural plots partially reported in the RPG database and use the TOPO database instead.

⁵These "îlots anonymes" gathered several contiguous agricultural parcels from 1 same farmer, and consequently had a lower spatial resolution.

Vineyards and Orchards: TOPO database The TOPO database systematically reports vineyards and orchards when their area is greater than respectively 2000m² and 5000m². I attribute each orchard and vineyard plot to a French region, with a spatial join. The map on Figure 6.2 displays the spatial distribution of orchards and vineyards in the TOPO database.

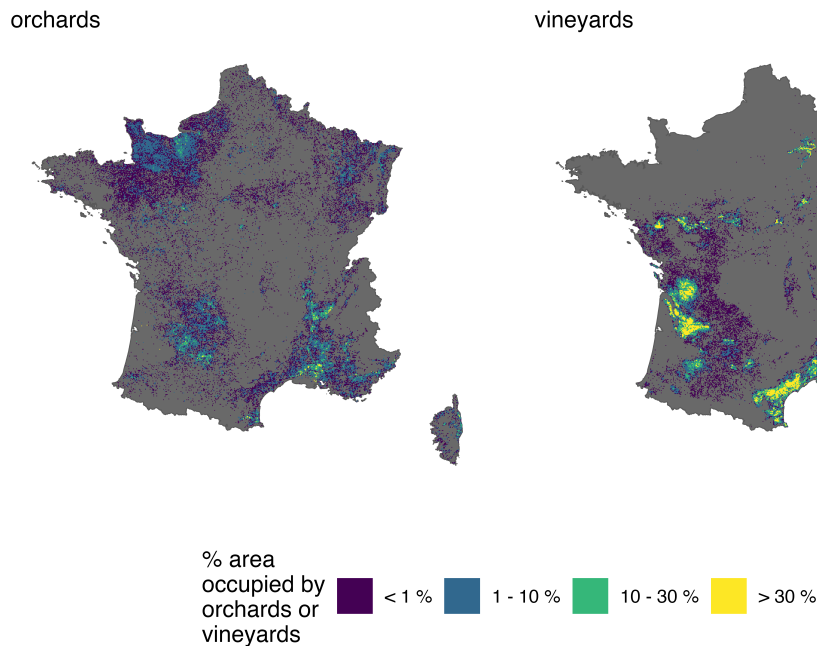


Figure 6.2: Spatial distribution of orchards (left) and vineyards (right) parcels in France. Gray: no reported orchards or vineyards on that area. Data source from TOPO database.

Combined datasets and map resolution I then combine the RPG (minus vineyards and orchards) and TOPO datasets, obtaining about 2.2 million agricultural parcels. To plot the maps, I aggregate the agricultural parcels area at a 4 km² resolution, with the following procedure.

Both RPG and TOPO datasets report agricultural parcels detailed polygons. I simplify them by using only their centroids, which yields more memory-efficient files and is more than enough for my analyses. My results, in terms of area and fertilization, are displayed in 2 manners: as points, with their geographical coordinates and associated area and fertilization; and in pixels, which aggregate the values of all the points inside them. Figure 6.3 details the procedure to transform polygons into points and rasterized pixels.

One should not choose too small a resolution for these pixels, otherwise the aggregated parcels (represented by their points) can have a higher area than the

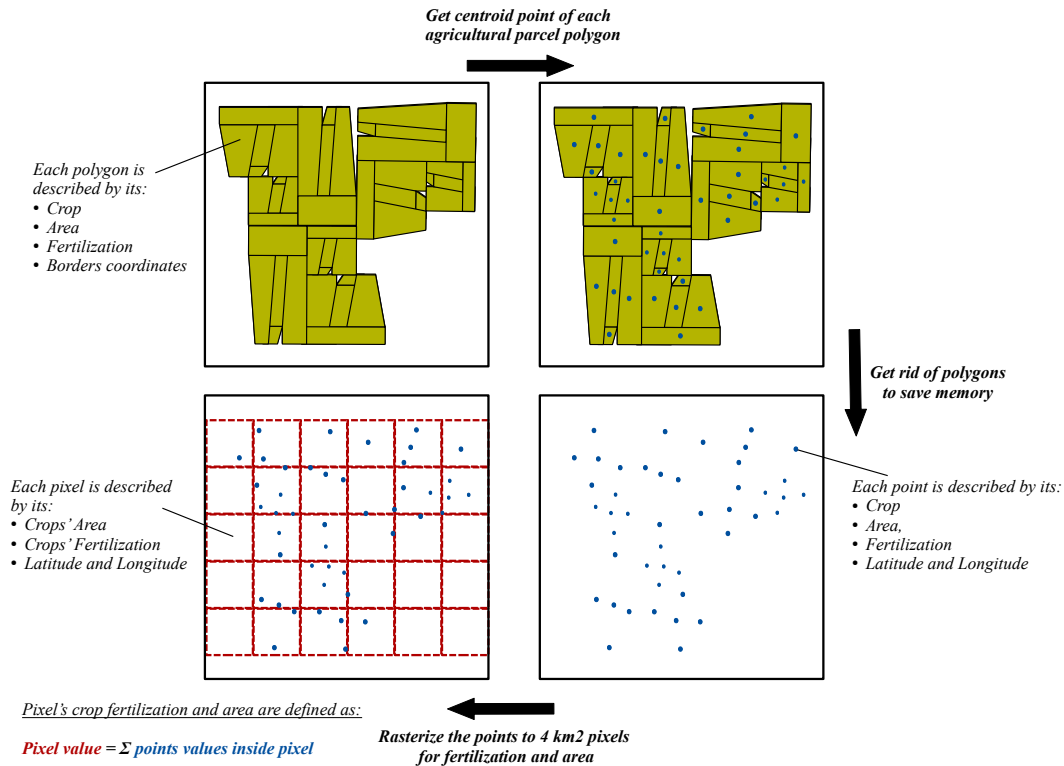


Figure 6.3: Procedure used to simplify to agricultural parcels polygons as reported in RPG and TOPO datasets into points and rasterized pixels. Rasterized datasets are used to plot the maps.

pixel they belong to. In my datasets, the vast majority of agricultural parcels has an area below 100 ha. I choose a pixel resolution of 4 km² (2 km x 2 km), or 400 ha, which is 4 times higher than the 100 ha threshold, as a security margin.

The obtained spatial distribution of all agricultural parcels (RPG + TOPO database) is displayed in Figure 6.4. Note that I use this aggregation procedure into pixels only to plot maps; for the attribution of excretions to surrounding agricultural parcels with an algorithm (Section 6.1.3), I use the unaggregated parcels points.

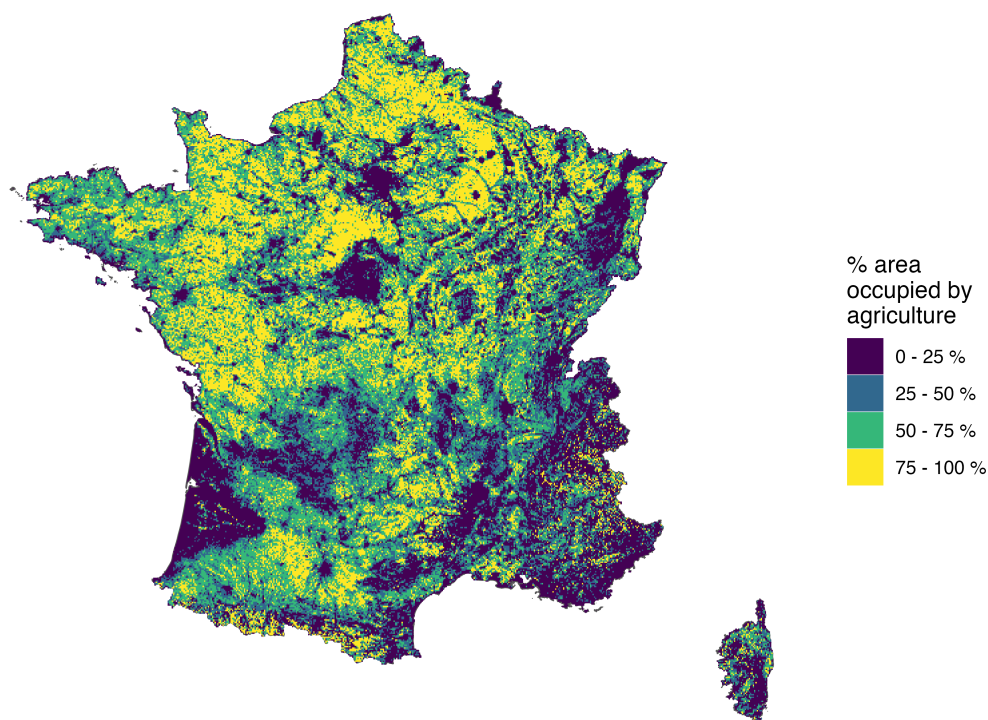


Figure 6.4: Spatial distribution of agricultural parcels (including permanent grasslands) in France. The main data source is the RPG database, enriched with the TOPO database for vineyards and orchards (Figure 6.2).

6.1.1.2 Fertilization on agricultural land. Resolution: regional

I use multiple sources to assign a fertilization level, in $\text{kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$, to each agricultural crop. My main interest is the synthetic N fertilization, but I also assign organic N fertilization to each plot.

Whenever I can, I use French cultivation practices surveys ("*Enquêtes Pratiques Culturales*") on fertilization practices (Agreste 2017b; Agreste 2019); if such surveys are not available for the considered crop, I use values from a report commissioned by the French Ministry of Agriculture (AND International 2022).

In these surveys, the reported organic fertilization is made of multiple forms: livestock excretions, the vast majority of the total, but also vegetal compost, wastewater treatment plant sludge, agro-industry sludge residuals, methanisation digestate... For context, a French collective expertise (Houot et al. 2014) on organic fertilizers estimated that around 2010, only about 5% of N organic fertilization in France came

from sources other than livestock manure⁶.

Finally, contrary to the previous Chapter 5, I do not focus on atmospheric deposition and legume biofixation, only on synthetic fertilizers and organic fertilizers (95% of which are from livestock manure), because handling of human excretions is much more similar to these 2 forms.

Major field crops: fertilization from cultivation practices survey In France, cultivation practices survey ("*Enquêtes Pratiques Culturelles*") are regularly conducted. Farmers are surveyed on many aspects of their agricultural practices, such as pesticide use, plowing, or previously cultivated crops. Of particular interest for my study are the surveys results about fertilization practices.

The latest survey for 17 major crops types and grasslands was carried out in 2017 (Agreste 2017b), detailed by French pre-2015 reform regions. I present the survey results for mean mineral and organic N fertilization on Figure 6.5. The attribution between 42 RPG crop denominations and these 17 cultivation practices survey categories is detailed in Appendix A.1, Table A.1.

Fertilization values are not available for every possible (crop; region) combinations. This is because some crops are barely grown in some regions, and thus were not surveyed, or the results were not statistically robust due to low sample size. For crops with no attributed fertilization rate in a particular region, I use the national crop mean.

Only for permanent and temporary grassland, was I not able to recover any open source regional data from the cultivation practice survey (though they exist, but require special demand). For temporary grasslands, I use the national value for all the regions. For permanent grassland, I was able to find regionalized fertilization data, but only for mineral N fertilization, reported in Billen et al. (2024): the values are generally between 10-30 kg N mineral per ha, except for Haute-Normandie which almost reaches 40 kg N mineral.

Similar major field crops Some crops reported in the RPG database are not surveyed in the French cultivation practices surveys, but are quite similar to the surveyed crops. I present the approximations I used in Appendix A.2, and its Tables A.2 and A.3. I also set a 0 kg·ha⁻¹ fertilization value for fallows, mountain pastures, and what is simply described as "miscellaneous" (*divers* in French, less than 1% of the reported agricultural land).

⁶in the text in French: "*Les Mafor (matières agricoles fertilisantes d'origine organique, nldr) autres que les effluents d'élevage ne représentent que 2, 5 et 3% du N, du P et du K utilisés en agriculture*". and also "*On estime qu'à l'échelle nationale, respectivement 39%, 70% et 82% de l'azote (N), du phosphore (P) et du potassium (K) utilisés en agriculture proviennent d'apports organiques exogènes*". So if we focus on organic fertilizers, the N share coming from sources other than animal manure is $2/39 = 5\%$ (and 7% for P and 4% for K).

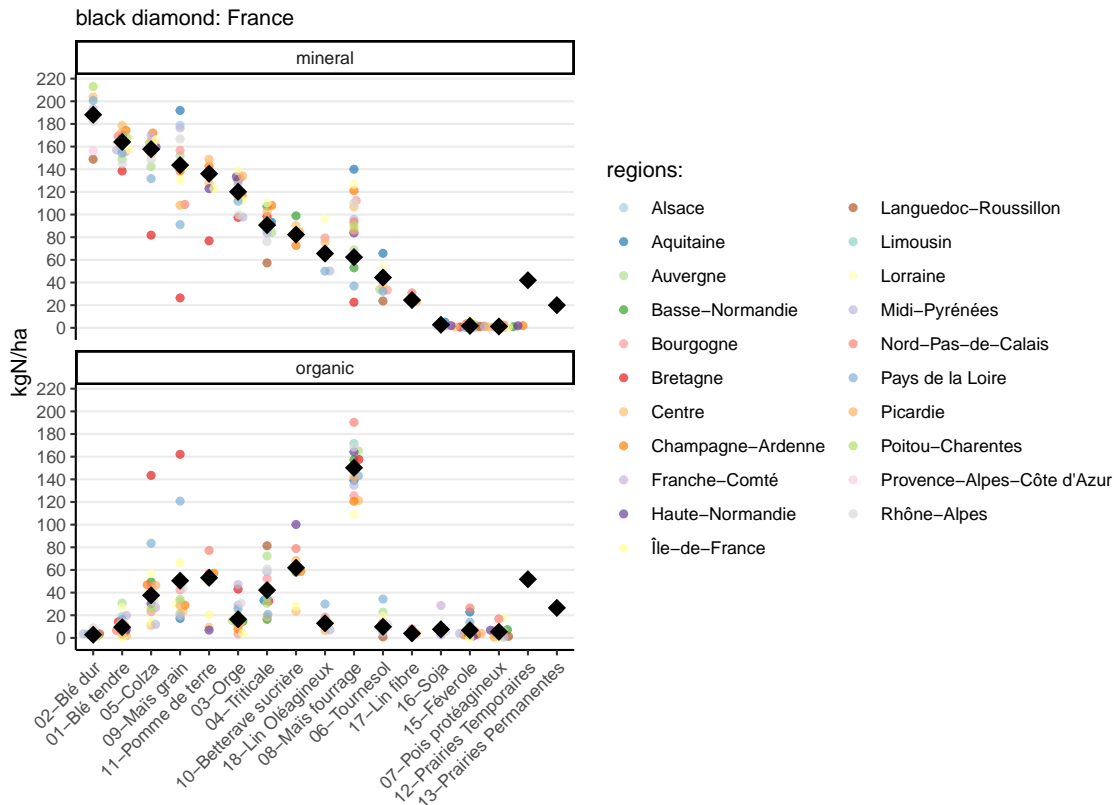


Figure 6.5: N Fertilization with mineral synthetic fertilizers (top) and organic fertilizers (bottom) for the different major field crops surveyed in the France cultivation practices survey (Agreste 2017b). Colored points: values for the 22 French regions (pre-2015 reform). Black diamonds: mean values for metropolitan France. For permanent and temporary grasslands (last 2 on the right), only national values were publicly reported.

Vineyards: fertilization from cultivation practices survey In addition to major field crops, a French cultivation practice survey was also carried out specially for vineyards in 2019 (Agreste 2019). Fertilization values are reported not by region but by 23 wine-growing basins (see Figure 6.6, top). To make it compatible with my procedure, I attribute one or several wine-growing basins to each French region (Table A.5). If one unique wine-growing basin is attributed to one unique region, I take its fertilization value for the whole region. If several wine-growing basins are attributed to one same region, I take their averaged value. The resulting fertilization rates by region are displayed in Figure 6.6, bottom.

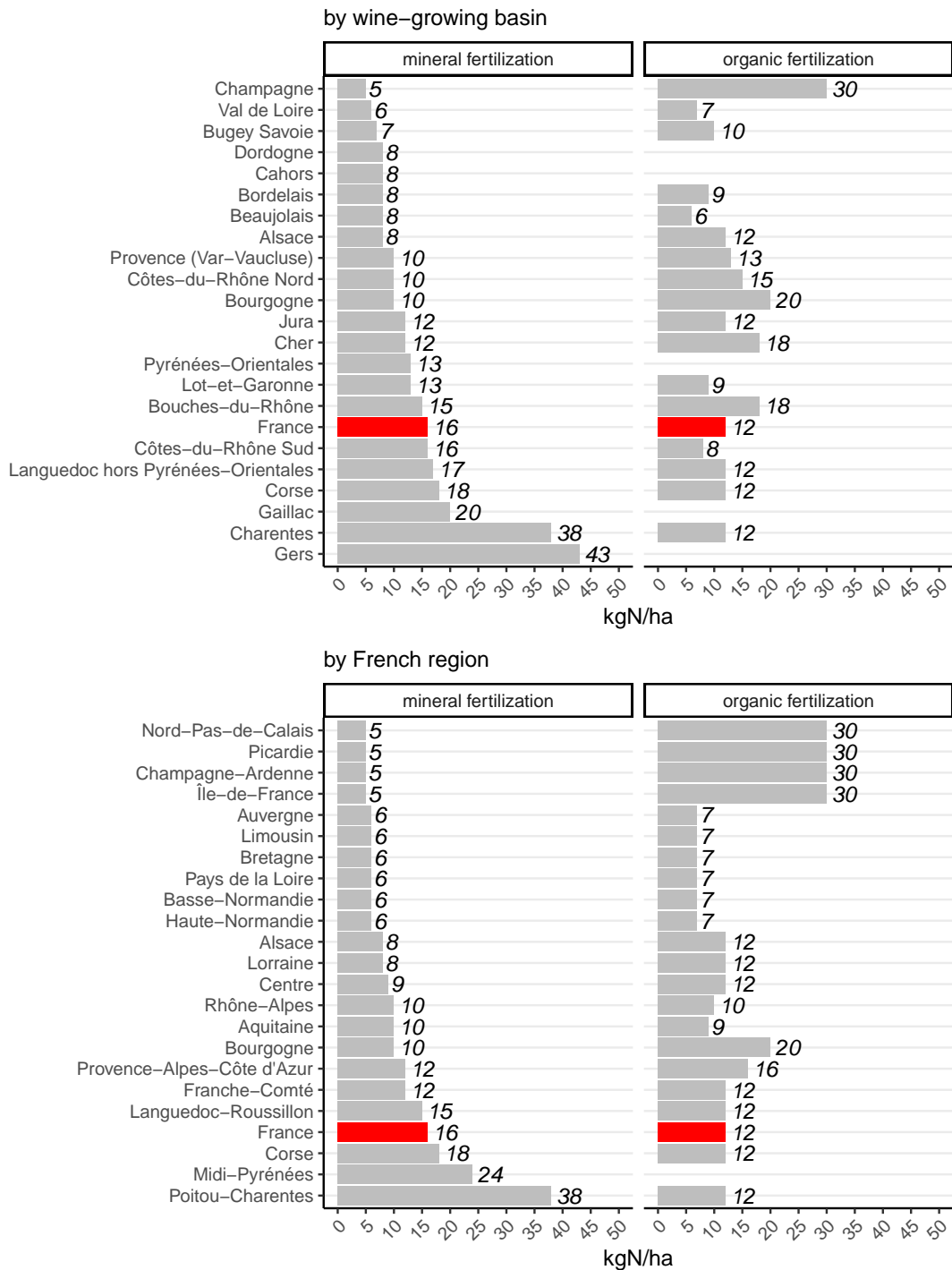


Figure 6.6: Top: N fertilization values in vineyards reported in French cultivation practice survey (Agreste 2019), by wine-growing basins; Bottom: values adapted to French regions, based on the matching in Table A.5. Left for N mineral fertilization, right for N organic fertilization.

Orchards: estimation of fertilization There was a French cultivation practice survey focused on orchards in 2015 (Agreste 2017a), but it only reported values for apricot, plums, cherries, peaches and apples. To overcome this limitation, I used a more complete report commissioned by the French Ministry of Agriculture (AND International 2022), which estimates in its methodology fertilization requirements for various crops, and in particular orchards⁷. Table 6.1 compares the few values surveyed by the French cultivation practices survey to the ones reported in AND International (2022); they are broadly similar.

These estimated requirements do not indicate how it is effectively distributed between mineral and organic fertilization. The French cultivation practice surveys indicate that for most major field crops (excluding leguminous crops), mineral fertilization represents often 75% or more of total N inputs; for vineyards, the distribution is more about 50/50% (Figure 6.7). I will use 66% of mineral fertilization for orchards as an order of magnitude.

Table 6.1: Comparison of orchards N fertilization values (in $\text{kgN}\cdot\text{ha}^{-1}$) in AND International (2022) report to the few values reported in the French cultivation practice survey (Agreste 2017a).

AND international report		Cultural practices survey	
label	value	label	value
plums	49	plums	37
apples	40	apples	39
other pome fruit and stone fruit	41	apricot	42
grapes	27	vineyards	28
other pome fruit and stone fruit	41	peaches	49
other pome fruit and stone fruit	41	cherries	35

Combining all this information, I estimate mineral and organic fertilization on different orchards types, featured in Figure 6.8, top.

However, while the incomplete RPG dataset reports the different orchards types, allowing detailed fertilization attribution, the more complete TOPO database that I use only reports a broad "*orchards*" category. For greater precision, I use the following procedure. With the incomplete areas reported in the RPG data, I compute the mean regional orchards fertilization, based on orchards types cross-referenced with fertilization values reported in Figure 6.8, top. Then I apply these mean regional values to all orchards fields belonging to that region in the TOPO database. This relies on the implicit assumption that the unreported orchards in the RPG data are quite similar to the reported ones.

⁷The report focuses on organic agriculture. For cereal field crops, it assigns lower fertilization values compared to conventional practices. However, for orchards, the fertilization values are often based on conventional practices, as there is minimal difference between the two production methods for this type of crop.

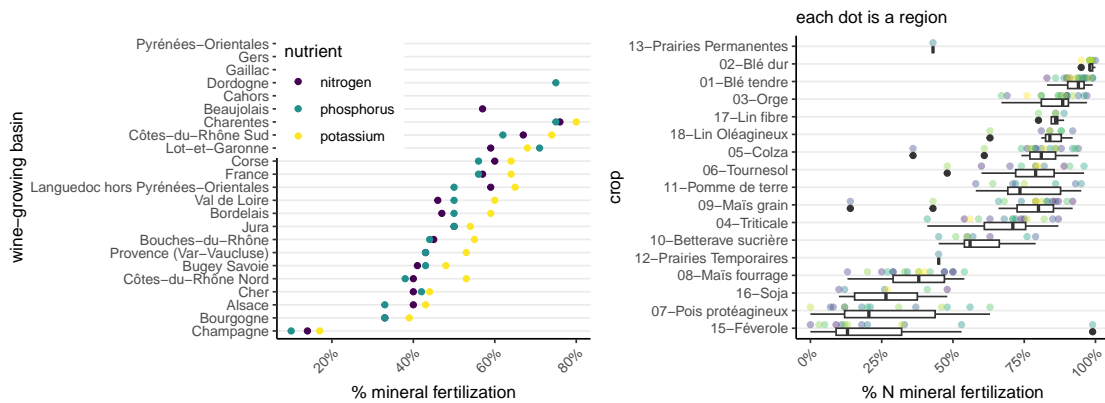


Figure 6.7: Percent of mineral fertilization in total fertilization. Left: for vineyards, in the different wine-growing basins, for N, P and K. Right: for major field crops, in the different regions, only for N.

My obtained regional values are displayed in Figure 6.8, bottom. The order of magnitude is $100\text{--}120 \text{ kgN}\cdot\text{ha}^{-1}$ for total fertilization, of which $2/3$ is mineral fertilization.

Vegetables and Floral crops The last broad category for fertilization attribution is Vegetables and Floral Crops. Table A.7 in Appendix A.5 details the attribution of the remaining 95 RPG items to this broad category. They are cross-referenced to fertilization values reported in the AND International (2022) report (see Figure 6.9). As for orchards, I estimate that 66% of the N fertilization is in the mineral form, and the rest in organic form. Vegetables growing is special in the sense it can have multiple harvest a year. This is partially captured by the AND International (2022) categories *Maraîchage sous abris* (covered market gardening) and *Maraîchage plein champs* (market gardening in open fields) in Figure 6.9, which have indeed high fertilization values.

Agro-ecological scenario For the agro-ecological fertilization scenario, I build on existing hypotheses made on scenarios at the European scale (Billen et al. 2024; Billen et al. 2021). These scenarios assume no change in the total agricultural area, or in the relative share between arable land, permanent grasslands and permanent crops. The scenarios are based on long term crop rotations, with a high share of leguminous crops to build the soil fertility.

I do not change the agricultural field spatial positions, implying that there is no change in France’s agricultural land spatial distribution.

I consider that permanent grasslands are not fertilized with human excretions. This is because my aim is to enhance short-distance synergies between human excretions and crops eaten by humans, and permanent grasslands are only grazed by

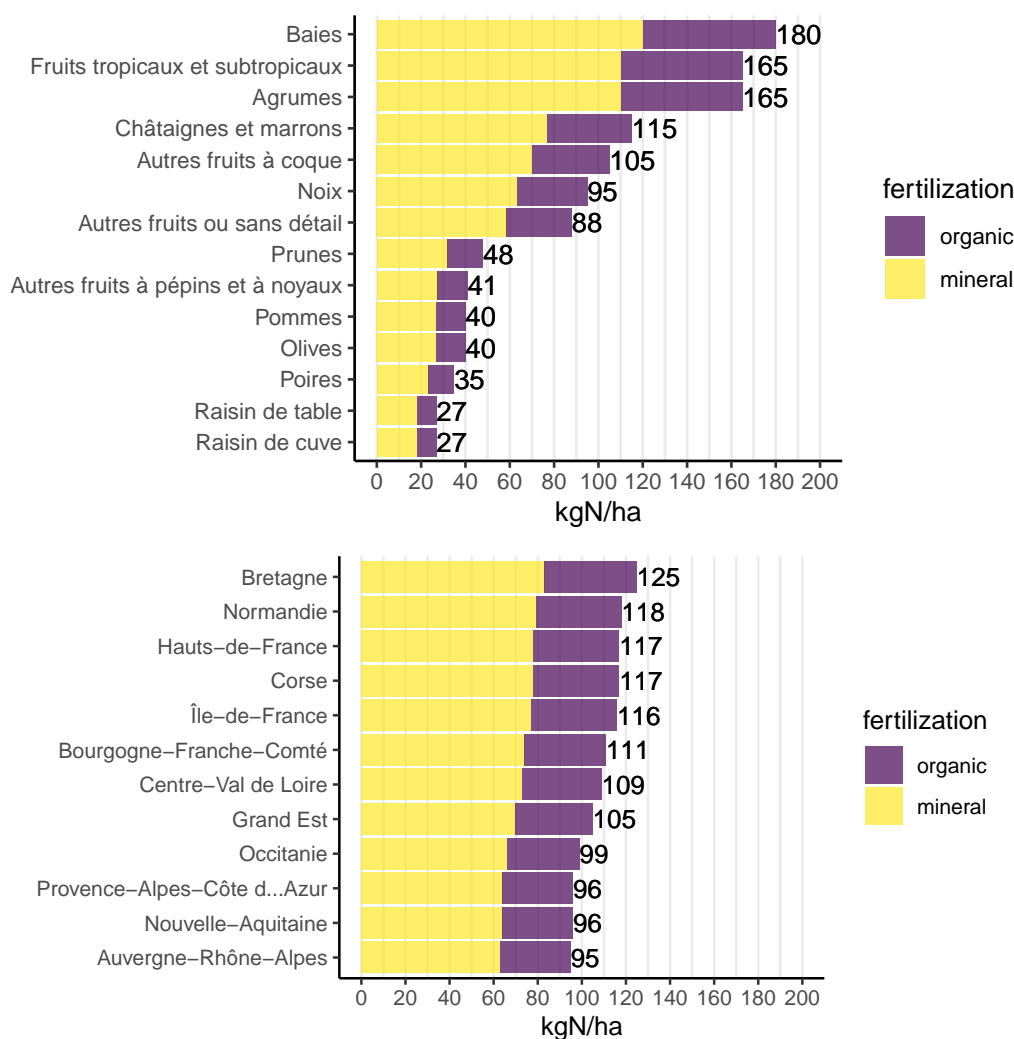


Figure 6.8: Top: orchards fertilization by crop type, based on AND International (2022) report. Bottom: estimated regional mean fertilization for orchards (see text).

ruminants. Moreover, permanent grasslands are semi-natural ecosystems, and in an agro-ecological scenario, it would be more coherent to not transform it into a high input parcel.

For orchards and vineyards, I consider that human excretions can be used up to the current mineral fertilization level. These levels are respectively $65 \text{ kgN}\cdot\text{ha}^{-1}$ and $16 \text{ kgN}\cdot\text{ha}^{-1}$ when considering national average (see Figures 6.8 bottom and 6.6 for the more specific values). These values are compatible with the low N inputs characterizing agroecological systems.

Likewise, on vegetables and floral crops, I estimate human excretions can be used up to the current mineral fertilization level⁸ (see Figure 6.9, national average at 90

⁸It would be interesting to conduct a sensitivity analysis in which vegetables are not fertilized with human excretions. As they are often eaten raw with minimal transformation, they are the most susceptible to contamination, and maybe regulation will encourage fertilization on crops like cereals. But note that vegetables are currently fertilized with animal manure, which poses similar issues regarding contamination.

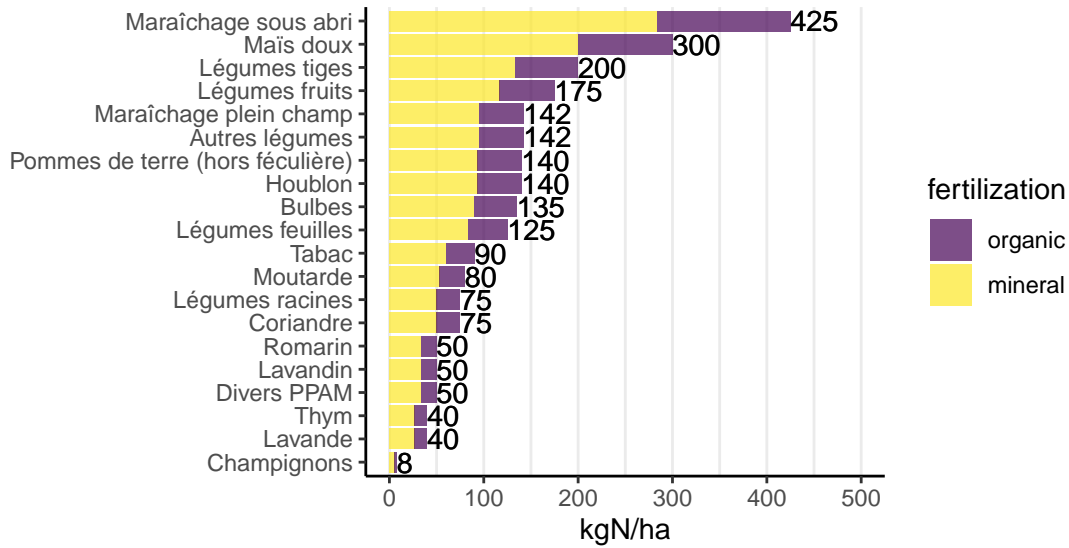


Figure 6.9: Fertilization values for vegetables and floral crops in the AND International (2022) report, with and hypothesis of 2/3 of mineral fertilization.

$\text{kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$). This represents a higher value than for vineyards and orchards. Indeed, even with agro-ecological practices, vegetables and market gardening are characterized by high inputs, namely because there can be multiple harvest a year.

For major crops on arable land, I need to consider that current N inputs are way higher than what is compatible with low input scenario. To do this, I rely on the scenario published by Billen et al. (2021) and focus on the simplified crop rotations in their Supplementary Materials (Table 6.2). I consider that, as it is the case currently, leguminous crops are not fertilized (see Figure 6.5). For non-leguminous crops, I need to fix a fertilization value with human excretions, that I arbitrarily fix to $50 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$, which could correspond to a spring N mineral boost brought to cereals with urine, in the context of an agro-ecological scenario relying more on organic fertilizers. This fertilization occurs only on non-legume crops, whose frequency in the rotation is between 5/8 and 2/3, resulting in a mean fertilization over the whole rotation around $31\text{-}33 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$ (see Table 6.2).

Table 6.2: Fertilization with human excretions in an agro-ecological scenario, based on crop rotations in Billen et al. (2021).

	Northern rotations	Southern rotations
legumes frequency	3/8	1/3
non-legumes frequency	5/8	2/3
fertilization with human excretions on non-legumes ($\text{kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$)	50	50
mean fertilization with human excretions over the whole rotation ($\text{kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$)	31	33

This $50 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$ rate seems reasonable considering the other values of fertilization in the scenario of Billen et al. (2021). They apply livestock manure on non legumes at a rate of $35 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$ ⁹. So the considered crops would be fertilized with human excretions at a rate about 50% higher than with animal manure. If we focus on all their inputs (symbiotic fixation, manure, atmospheric deposition) over a whole rotation, the rate is $109 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$ ¹⁰, so human excretions (around $31 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$ on the whole rotation) would bring this rate to $140 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$.

In their scenario, on a per capita basis, N inputs to croplands (excluding human excretions), amount to $23 \text{ kgN}\cdot\text{cap}^{-1}\cdot\text{year}^{-1}$; this is similar to the current inputs in Germany, Turkey, or Myanmar estimated in Figure 5.13 in Chapter 5. But these inputs would be 70% made of leguminous fixation, whereas it is currently less than 10% in France. The $3.5 \text{ kgN}\cdot\text{cap}^{-1}$ excreted in the scenario would represent 15% of these total inputs.

6.1.1.3 Fertilization consistency check

I aggregate my field-scale values to the regional and national scale, to compare with official data on fertilizers consumption and check the relevancy of my results. In France, UNIFA (2023) reports fertilizers delivery at the regional scale. Even though my main focus is on nitrogen, I will also use their data on phosphorus and potassium as a secondary consistency check. For mineral fertilizers consumption at the national scale, I use both FAOSTAT (2023a) and UNIFA (2023) data.

For organic N fertilization, I consider FAO data on all livestock manure applied to agricultural soils (FAOSTAT 2022f). Note that for both FAO and my data, N directly excreted by animals on grassland is not included in N organic fertilization. Livestock excretions are considered only if they are collected by the farmer, usually in the barn, and then spread on agricultural parcels.

In addition to livestock manure, my data based on the French cultivation practices also includes other sources of organic fertilization (industrial residues, municipal compost...), contrary to FAO data that only focuses on manure. I discuss in Section 6.1.1.2 that organic fertilization other than manure represents about 5% of total organic fertilization. So I estimate that my results and FAO official data on organic fertilization are quite comparable, but my results should be slightly higher by about 5%. Yet the discrepancy can be higher locally, especially in Northern

⁹In their supplementary spreadsheet, manure inputs on croplands are $23 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$ (396 ktN on 17.2 Mha of arable land). If we consider that manure is not spread on legumes, and that legumes represent 1/3 of the rotations, this translates into an effective rate of $35 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$.

¹⁰The total inputs are actually $118 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$ (2031 ktN over 17.1 Mha of arable land), but this already includes a partial (25%) human excretions application of 159 ktN, or a bit less than $10 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$. So the total excluding human excretions is $109 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$.

France which uses residuals of the sugar beet industries.

6.1.2 Spatial distribution of excretions

For the spatial distribution of people, I use population data published by INSEE (2019) at a 1 km² resolution. This source details the population by age categories. This allows me to couple the population data with INCA3 ingestion data by age (Dubuisson et al. 2019; data.gouv 2021), that I already detailed in Chapter 4. This results in a average ingestion and excretion of 4.5 kgN·cap⁻¹·year⁻¹.

For the agro-ecological scenario, I use data in Billen et al. (2021). They hypothesize a N food supply of 5 kgN·cap⁻¹·year⁻¹, of which 30% is lost or wasted, resulting in an effective ingestion and excretion value of 3.5 kgN·cap⁻¹·year⁻¹. Compared to the 4.5 kgN·cap⁻¹·year⁻¹, this implies a 0.78 ratio, which I apply to each pixel of 1 km². This implies that the age structure of each pixel is kept in the agro-ecological scenario compared to the current situation.

To plot the maps, and out of consistency with the aggregating procedure followed by agricultural parcels, I also aggregate human excretions at a 4 km² resolution. Contrary to agricultural parcels, there is no resolution issue in this case (Section 6.1.1.1 for the resolution issue with agricultural parcels). Note that in both cases, this aggregation is only used to visually represent the maps, but not for the algorithm used to attribute excretions to neighboring agricultural parcels, which I describe in the following.

6.1.3 How excretions are distributed to neighboring agricultural parcels

In the *current situation* case, I substitute mineral fertilization with human excreta (urine + feces), hypothesizing a 100% recovery rate. In the *agro-ecological* scenario, I fill the fertilization demand, as defined previously in Section 6.1.1.2 and Table 6.2, with human excreta, also with a 100% recovery rate.

Excretions are attributed to neighboring agricultural parcels with the following procedure, which was algorithmically implemented by Tanguy Fardet:

1. We sort the population pixels from the least to the most populated. We then begin with the pixel containing the least population, and proceed by increasing population.
2. We then select all the agricultural parcels within a 4 km radius of the selected pixel, and for each parcel we compute the distance to the pixel. Note that this is a straight-line distance, as the crow flies, and not an actual distance that would be covered by a truck constrained by the presence of a road.

3. Proceeding by increasing distance, we attribute N excretions to these parcels, until the N excretions of the pixel is depleted. If the parcels in the 4 km radius are not enough to deplete the pixel's excretions, we increase the radius by 50%, and start again step 3. If a radius of 300 km is reached, and the pixel's excretions are still not depleted, we stop, and the remaining excretions are not attributed to a parcel.
4. We move on to the next population pixel and start again at step 2, paying attention to not consider agricultural parcels which have already been attributed to a previous population pixel. When all pixels are treated, we stop.

This procedure gives priority to less densely populated areas over the more populous ones. So this favors less densely areas into having shorter distances. I agree that this is a disputable methodological choice that induces a bias. To see its influence, I re-run the algorithm, this time giving priority to the most populated areas in step 1, to compare the results in the 2 cases.

6.2 Results and Discussion

6.2.1 Consistency of fertilization values

6.2.1.1 Official data at national and regional levels

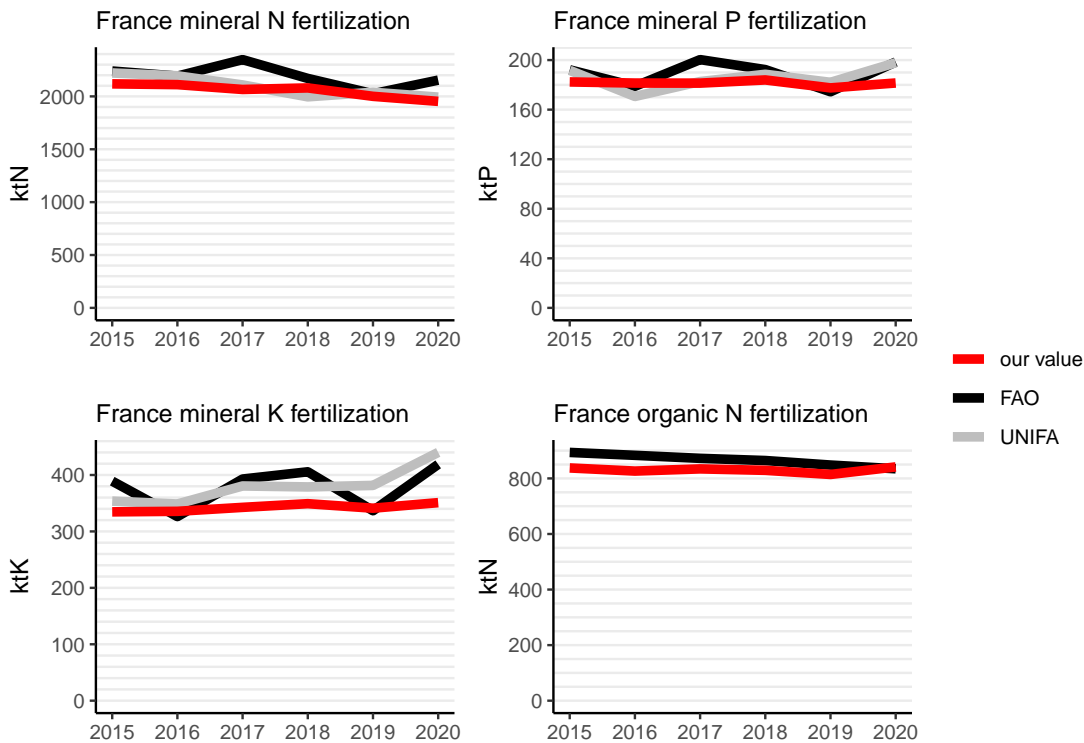


Figure 6.10: Comparison of results (red) regarding fertilizers use in France to official data from FAOSTAT (2022c) and UNIFA (2023).

The comparison of my estimate of fertilization at the national scale to official data is presented on Figure 6.10. For N and P, my estimates are well in line with FAO and UNIFA values across the 2015-2020 period, and typically represent 90-105% of these official values. For K, my estimate tend to be slightly lower than their official figures, typically within 85-100% of official values. My N organic fertilization values, virtually all of which is made of animal manure, is about 5% lower than official FAO estimation of N manure collected and spread on agricultural land; but considering that my data include all organic fertilizers and not only livestock manure, it should be 5% *higher* (see the discussion in Section 6.1.1.3), so the actual discrepancy is rather a 10% underestimation.

At the regional scale (Figure 6.11 for N, Figure 6.12 for P and K), I generally get the right order of magnitude, but there is a lot more variation. Part of this variation may be due to the fact that UNIFA reports deliveries, and fertilizers delivered in

one region might be used in another neighboring region. This can result in over- and under-estimations in the concerned regions.

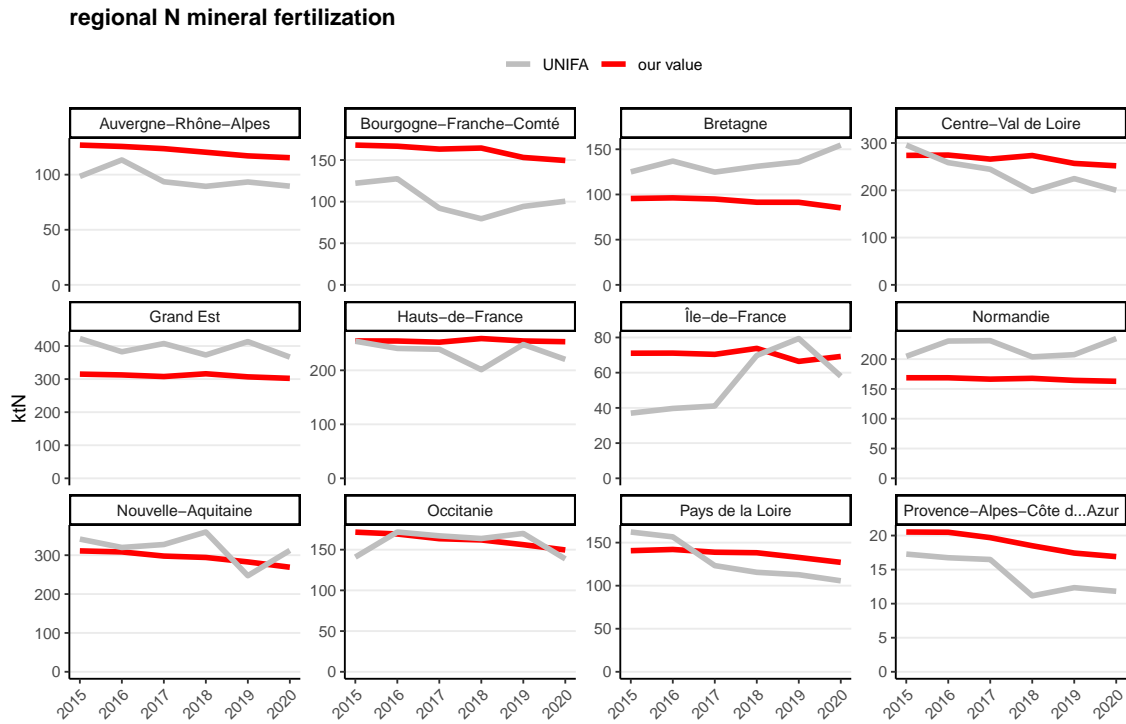


Figure 6.11: Comparison of results regarding N synthetic fertilization (red line) to official fertilizers delivery data from UNIFA (2023) (grey line), by French metropolitan region. Note that for UNIFA data, fertilizers delivered to one region may be spread in another neighboring region.



Figure 6.12: Comparison of our results regarding P and K synthetic fertilization (red line) to official fertilizers delivery data from UNIFA (2023) (grey line), by French metropolitan region. Note that fertilizers delivered to one region may be spread in another neighboring region.

6.2.1.2 Spatial distribution of fertilization

Besides national or regional value, we can also spatially analyze the data to assess the consistency of my results. Figure 6.13 presents my results regarding the spatial distribution of total N fertilization in France. Major cash crops, intensively fertilized, are located in the North of France, around Paris; this is coherent with the fact that it is the most fertilized region on my map (>10 ton of N per km^2).

total N fertilization

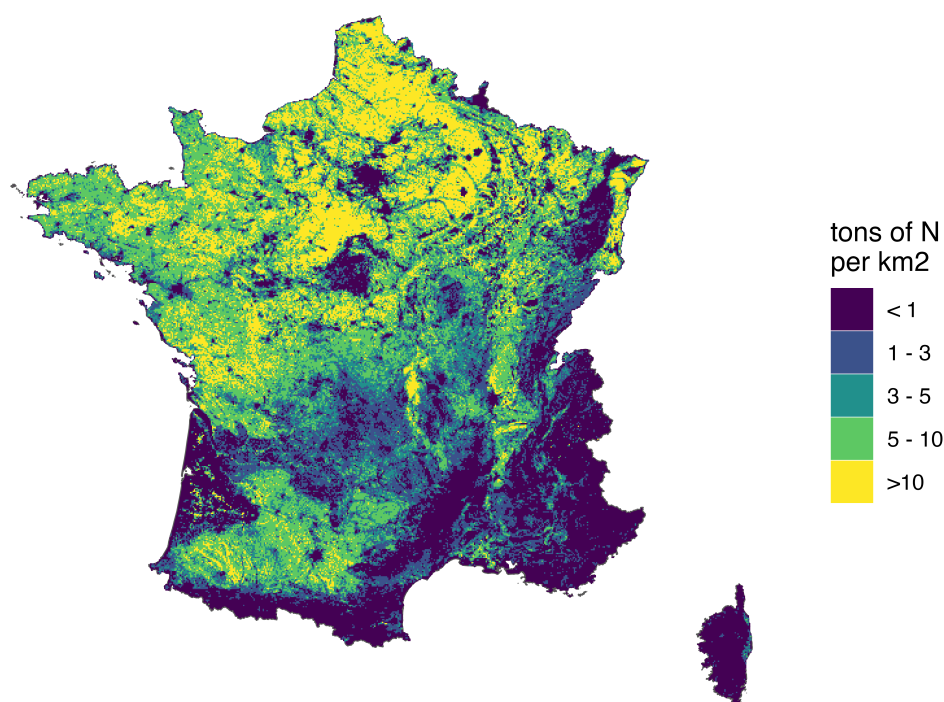


Figure 6.13: Total N fertilization in Metropolitan France. This includes mineral N and organic fertilization (but not legumes biofixation nor atmospheric deposition), which are broken down in Figure 6.14

Zones with low N fertilization (<1 ton of N per km^2 per year) indicate mountains (most of Corsica, Alps in South East, Vosges in North East, Pyrenees in South West), forests (the Landes in South West, Ardennes in North East, Sologne in the center, Cevennes in the South center); some large urban metropolitan areas with no fertilization are visible, particularly around Paris.

Most of the other areas are around 5-10 tons of N per km^2 per year. The major exception is the Massif Central, around 1-5 tons of N per km^2 . This is a region specialized in cattle production on extensive grassland, with little major crops, but nonetheless more croplands than mountainous or forests regions. Breaking

down fertilization between mineral and organic N fertilization (Figure 6.14) allows to better see the influence of livestock.

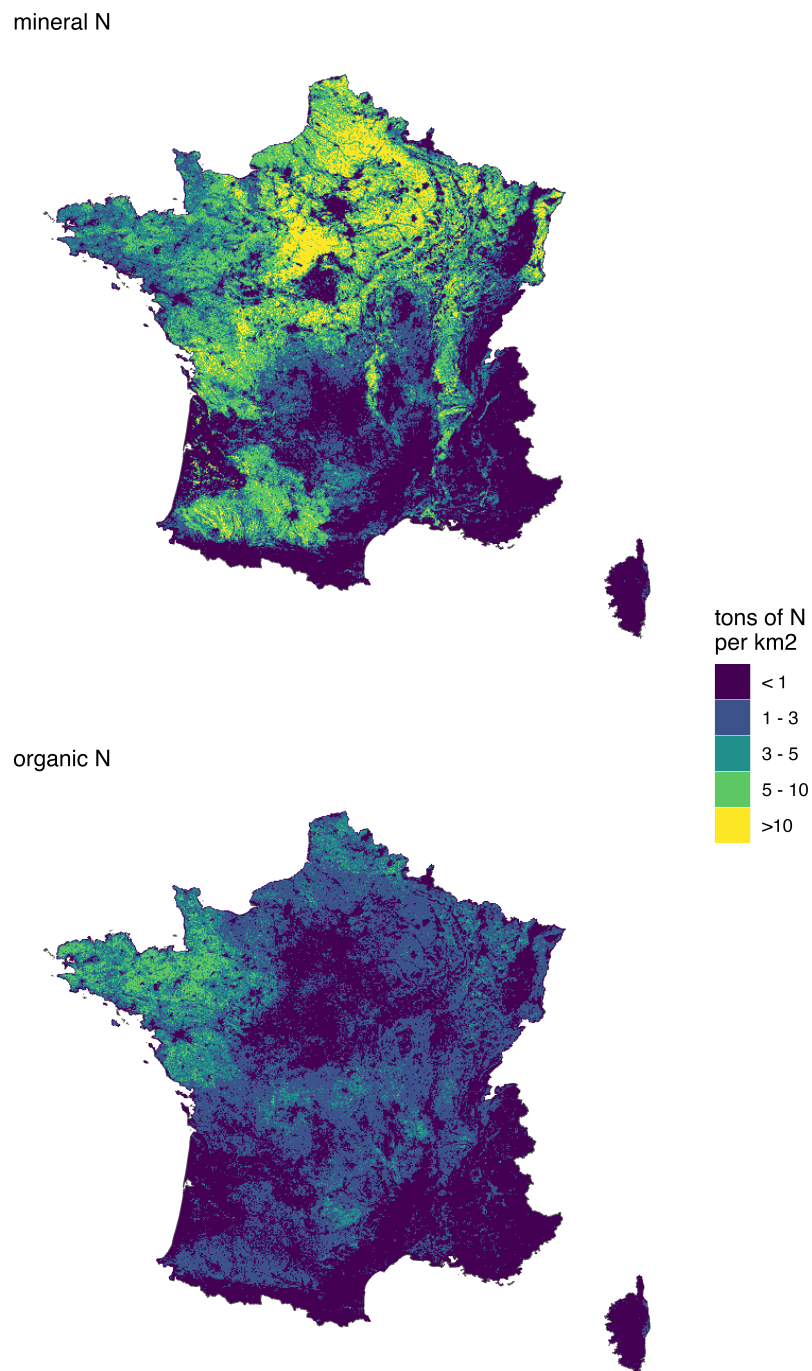


Figure 6.14: Spatial distribution of N mineral (top) and organic (bottom) fertilization in France.

Organic N fertilization is concentrated in Brittany and part of Normandy, which concentrates most of French monogastrics and many dairy cows. Note that ruminant

excretions in the center of France are not visible. This is because in our data, organic fertilization only accounts for manure collected by farmers and then spread on crops or grasslands, and does not considered situations where excretions are directly excreted by cattle on grasslands during pasture. As cattle breeding in the center of France is mostly extensive on permanent grasslands, many of their excretions are not visible on the map.

Mineral P and K fertilization (Figure 6.15) globally follows a pattern similar to mineral N fertilization, with one particularity for K, whose highest fertilization rates tend to be in the extreme North of France (region Hauts de France). This is coherent, since most of the K-demanding sugar beet and potatoes are grown in this region. In the cultivation practices survey (Agreste 2017b), mineral K fertilization is around 100-150 kgK·ha⁻¹ for potatoes and 50-100 kgK·ha⁻¹ for sugar beets, whereas other majors crops are consistently below 50 kgK·ha⁻¹, and often below 20-30 kgK·ha⁻¹ (results not shown here but visible on the website).

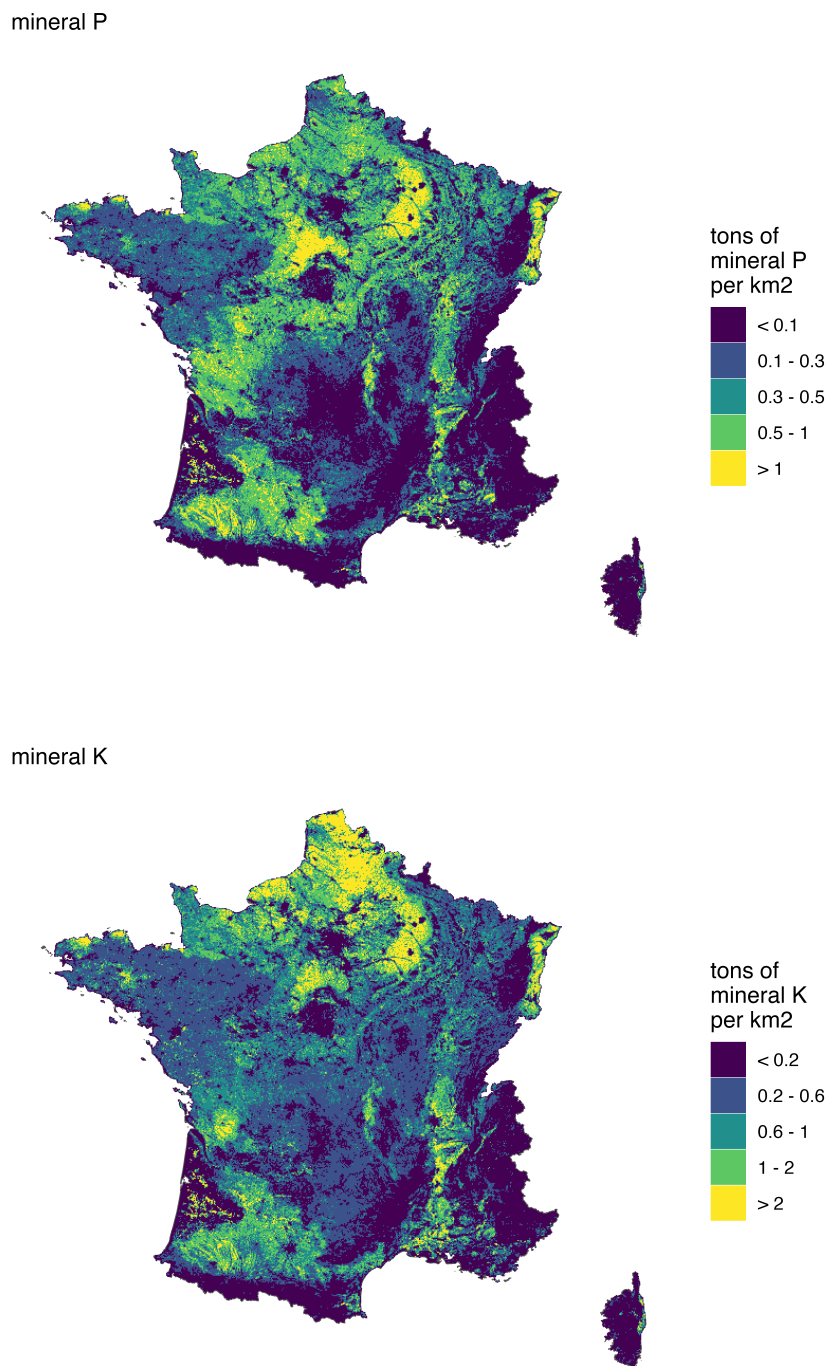


Figure 6.15: Spatial distribution of P (top) and K (bottom) mineral fertilization in France.

6.2.1.3 Interpretation of current fertilization practices

The results of the survey on major crops fertilization practices (Agreste 2017b) featured in Figure 6.5 are also informative. Leguminous crops, which can fix N

from the air themselves, are barely fertilized with nitrogen. Major cereals (soft and durum wheat, rapeseed, grain maize, potatoes) are fertilized with about 200 kgN·ha⁻¹·year⁻¹ (mineral + organic). There can be some variations between regions, but the main factor influencing the fertilization level is the kind of crop.

One exception is Brittany (red dot on Figure 6.5), especially visible for soft grain maize and rapeseed: mineral fertilization is way lower than other regions, but compensated by higher organic fertilization. This is coherent with the fact that these 2 crops are used in monogastric feeding, intensively bred in Brittany, and farmers have to handle their excretions, and use them as fertilizers.

Similarly, forage maize is much more organically fertilized than other crops, but this time this is true for all regions and not just Brittany. This is because forage maize is usually grown as part of mixed farming and fed to neighboring cattle, and their manure is in return used as fertilizer.

6.2.2 Distances to fertilize crops with excretions: current fertilization practices

When our procedure to replace current use of mineral fertilizers by human excretions is applied, human excretions are allocated to a total 6.1 Mha of agricultural land. This represents 23% of the agricultural land in Metropolitan France. 70% of excretions could be spread on an agricultural plot within less than 10 km. About 20-25% could be spread within 10-50 km; and 5-10% have to go further than 50 km. So, the vast majority of excretions could be used as fertilizers within a short distance. In the following subsections below, I analyze the factors influencing these transport distances.

6.2.2.1 Influence of geography

Figure 6.16 presents the excretions and associated crops for the different distances categories (<10 km; 10-50 km; and >50 km).

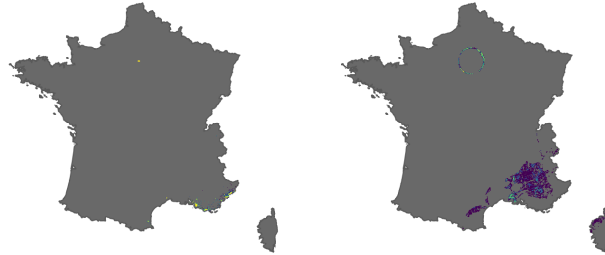
The case of Paris center and the Mediterranean coast: 5-10% of excretions Even without delving into fertilization, it is already clear that the Southeastern part of France seldom has agricultural land (Figure 6.4), and thus the demand to absorb human excretions used as fertilizers will be low. This problem is amplified by the fact that the Mediterranean coast is densely populated, with Marseille metropolitan area and it almost 2 million inhabitants, the second largest in France, and so the supply of excretions is quite high. Moreover there is no opportunity to spread it in the South, where there is the Mediterranean sea. And finally, the remaining Northward agricultural area is characterized by crops with low N inputs

Attribution of human excretions to surrounding fields with current fertilization practices and diets

transport >50 km (5-10% of excretions)

human excretions

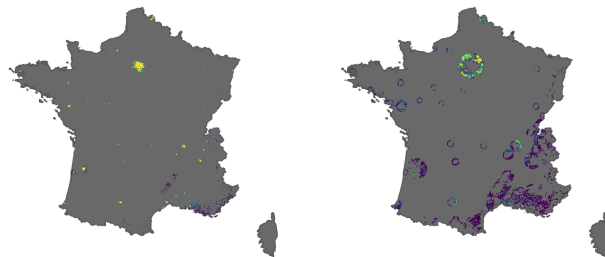
fertilization with excretions



transport 10 to 50 km (20-25% of excretions)

human excretions

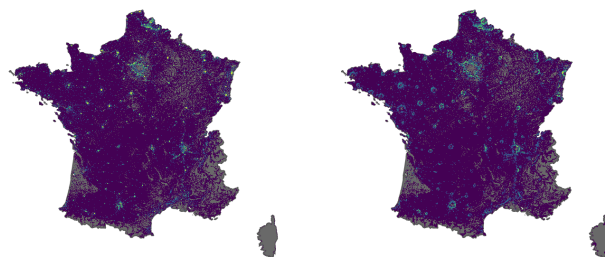
fertilization with excretions



transport <10 km (70% of excretion)

human excretions

fertilization with excretions



tons of N per km² < 1 1 - 3 3 - 5 5 - 10 > 10

Figure 6.16: Human excretions (left) and their associated fertilized agricultural parcels (right), broken down into 3 categories based on the distance between excretions and fertilized crops. This is based on current fertilization and diets, following our algorithm attributing excretions to neighboring parcels. Distances refer to straight-line distance, as the crow flies, and not an actual distance that would be covered by a truck constrained by the presence of a road. Gray areas are excretions and agricultural areas not concerned by the transport distance category.

such as orchards, vineyards or grasslands, and few cereals. As a result, many agricultural parcels in the Southeastern part are associated with a transport distance of more than 50 km, or sometimes even more than 100 km (Figure 6.16, top maps).

The other region with transport distances superior to 50 km is the Paris metropolitan area. This is obviously a densely populated area, with a high supply of excretions. However, contrary to the Mediterranean coast, it is surrounded by many croplands, especially cereals that are intensively fertilized. As a result, transport distances are just a bit higher than 50 km, but do not exceed the 100 km threshold, contrary to Southern France.

Synergies between medium-sized cities and their surrounding: 20-25% of excretions If we except the Paris metropolis and the Mediterranean coast, all the other French medium to large cities have their excretions that could be spread within just 10-50 km. This concerns cities that typically have a population of 0.1-1 million inhabitants. Actually, even for the Paris metropolis, the majority of human excretions can be spread within this distance, because it is surrounded by many intensively fertilized crops. The crowns drawn by the fields surrounding these cities are clearly visible on Figure 6.16 (the middle maps).

The vast majority of excretions, from rural areas and middle-sized cities: 70% of excretions The rest of human excretions can be spread within 10 km (Figure 6.16, bottom maps). Actually, about 65% of excretions can be spread within just 3 km. This concerns France rural areas, but also the urban center of medium-sized cities (Orléans, Le Mans, Tours, Nancy, Besançon...). To sum up, France benefits from having not a lot of very large metropolises (excepted Paris and Marseille) and an agricultural land pretty much distributed all over the Metropolitan area (excepted in the Southeastern part), with major crops like cereals that can absorb the supply of excretions.

6.2.2.2 Influence of population density

Figure 6.17a shows the distribution of distances in the current situation, broken down by population densities. Note that, all the excretions in a 1 km² pixel are attributed to the pixel center; so distances <500 m in the Figure are not meaningful given the spatial resolution of excretions, should simply be interpreted as "the agricultural parcels are within the excretions pixel of 1 km²".

For areas with less than 1,000 inhabitants·km⁻², which represents about 40% of the French population, virtually all of the excretions (>98%) can be spread within less than 10 km. Actually for areas with very low population density, <100

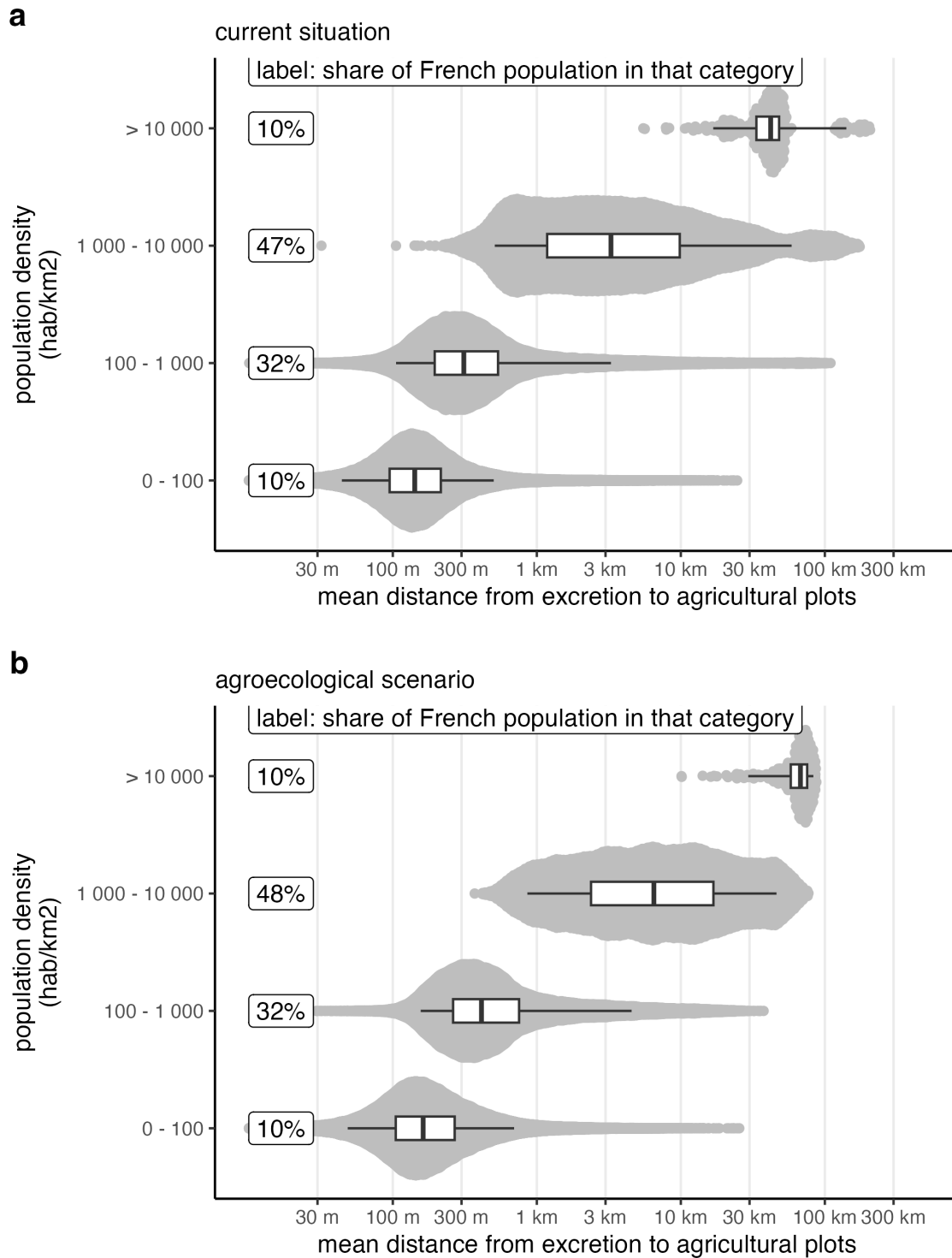


Figure 6.17: Influence of population density on required distance to evacuate human excretions to surrounding agricultural parcels, for current situation (top) and the agroecological scenario (bottom). Each dot is a 1 km² pixel of human excretions. On the x-axis, the distance is the mean distance from all the agricultural parcels attributed to the concerned pixel of human excretions. Distances refer to straight-line distance, as the crow flies, and not an actual distance that would be covered by a truck constrained by the presence of a road. Boxplots represent the following percentiles: 5, 25, 50, 75 and 95%.

inhabitants·km⁻², and gathering 10% of the French population, virtually all of the excretions can be spread within 1 km.

About 10% of French inhabitants live in areas with a high population density >10,000 hab·km⁻². For these cities, the mean distance to agricultural plots is almost always above 10 km, and often above 50 km, but generally stays below 100 km. However, sometimes, distances as far as 300 km are required. The median distance is about 40-50 km.

The most contrasted situation is for areas between 1,000 and 10,000 hab·km⁻², gathering almost half of the French population. The 95% percentiles of population distances contain values spanning 2 orders of magnitude. For them, the median distance is 3 km, and 75% of these excretions can be spread withing 10 km.

As I explained in the Methods Section 6.1.3, the algorithm used to attribute excretions to surrounding parcels is biased towards attributing closer parcels to the less densely populated areas. To assess the extent of that bias, I compare in Appendix B how the results are changed when we use an algorithm biased towards the most populated areas. This changes the median distance in each population density category and the tail of the distributions, but the general trends and order of magnitude are similar, and the results rather robust.

6.2.3 Distances to fertilize crops with excretions: agro ecological scenario

6.2.3.1 A decreased potential demand for excretions

Spatial distribution of demand In the current situation, the demand for fertilization with human excretions, presented in Figure 6.14, displayed a strong heterogeneity between regions.

On the contrary, in the situation of an agro-ecological scenario, the demand for human excretions is much more homogeneous (Figure 6.18). Due to our hypotheses regarding fertilization demand, there are 2 main land categories: agro-ecological rotations on arable land, with a demand between 1 to 3 tons of N per km², corresponding to the mean application rates of 30-35 kgN·ha⁻¹ (Table 6.2)¹¹; and permanent grasslands, on which we do not allow fertilization with human excretions. There are some rare clusters where demand can exceed 3 tons of N per km², because of a high density of croplands and the presence of permanent crops or vegetables and floral crops, which are more nitrogen-demanding.

¹¹30-35 kgN·ha⁻¹ corresponds to 3-3.5 tons of N per km², but only in the case where all of the 4 km² pixel is covered with agricultural parcels. As the covered area is often <100%, the category is usually 1-3 tons of N per km².

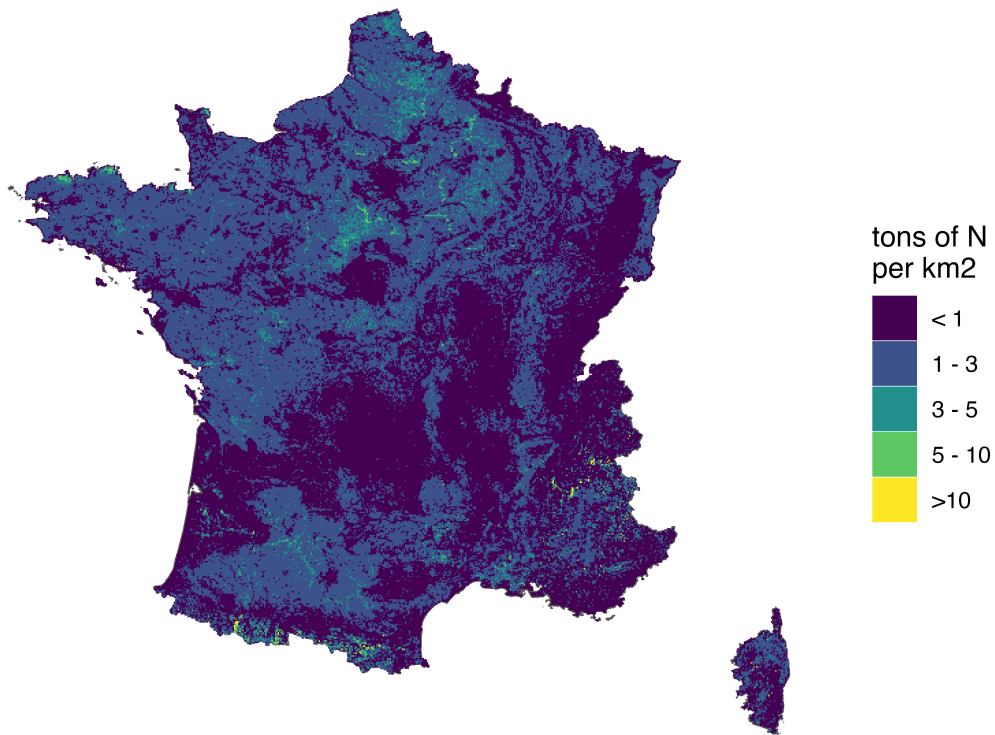


Figure 6.18: Demand for fertilization with human excretions in an agro-ecological scenarios, resulting from our hypotheses.

Matching of demand to excretions: national and regional levels The potential demand for fertilization with excretions in the agro-ecological scenario is about $680 \text{ ktN}\cdot\text{year}^{-1}$ at the national level, which is about 3 times less than the current mineral fertilization of about $2000 \text{ ktN}\cdot\text{year}^{-1}$. The vast majority of the demand is due to arable land and its agro-ecological rotations, and is still way higher than the $220 \text{ ktN}\cdot\text{year}^{-1}$ national excretions in this agro-ecological scenario (Figure 6.19, left).

However the diminished demand raises the question of the matching to local excretions. At a regional level, only Ile-de-France has an excretion amount higher than the potential demand, and thus needs to export about half of its excretions to neighboring regions (Figure 6.19, right).

Now that we know that there is no strong mismatch at the regional scale, we can dig deeper and study more precisely the distances from excretions to fields.

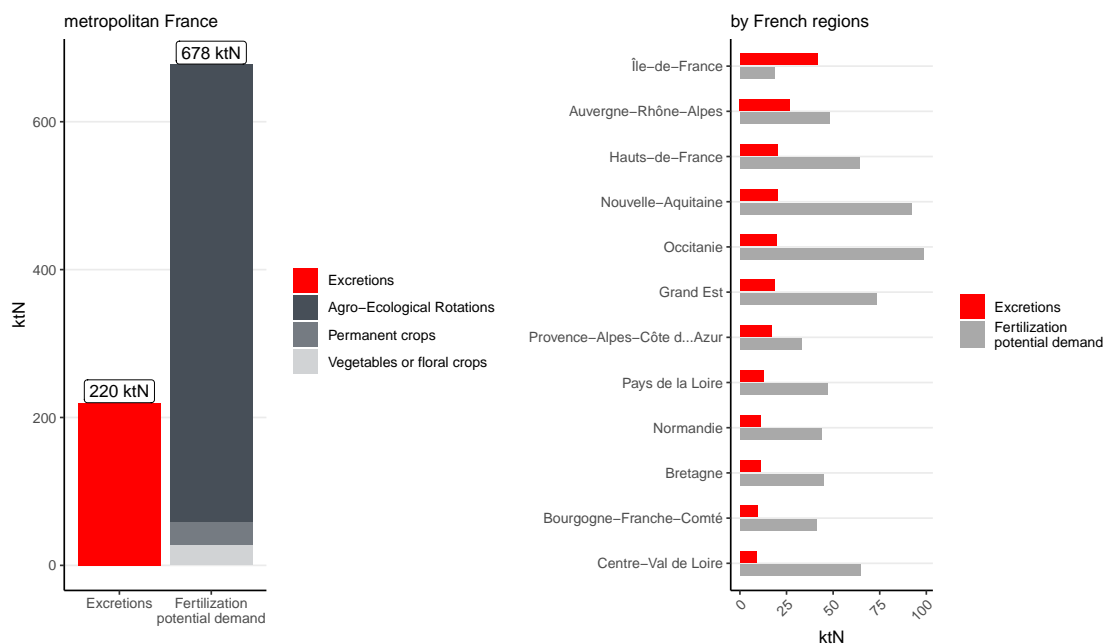


Figure 6.19: Comparison of excretions to potential demand for fertilization with excretions (considering $50 \text{ kgN}\cdot\text{ha}^{-1}$ on non-legumes crops) at the national level (left) and regional level (right).

6.2.3.2 Influence of geography

Figure 6.20 presents human excretions and their associated agricultural parcels, broken down by distance categories. In the agro-ecological scenario, the supply is slightly diminished, due to less protein in the diet: $3.5 \text{ kgN}\cdot\text{cap}^{-1}$ instead of $4.5 \text{ kgN}\cdot\text{cap}^{-1}$, a 22% decrease. But the decrease in fertilization is much more important, by a factor 3, from 2000 ktN to 680 ktN. So the change in fertilization is the major factor affecting the supply/demand, and thus the distances. Generally, distances are slightly increased in this agro-ecological scenario.

Large metropolises: 15% of excretions Compared to the current situation, more cities are concerned with distances exceeding 50 km: still Paris and Marseille, but also Bordeaux, Lyon, Nice and Toulon (Figure 6.20, top maps). The crown of parcels fertilized with Paris excretions is larger than in the current situation. This was expected, since it is currently composed of crops fertilized at levels of $100\text{-}200 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$, compared to only $30\text{-}35 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$ in the agro-ecological scenario.

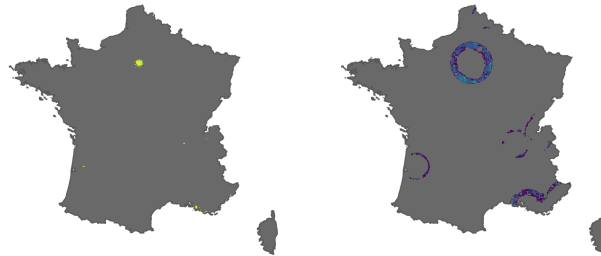
Medium-sized cities: 25% of excretions For distances between 10 to 50 km, I find a similar pattern than for the current situation: we clearly see the urban centers surrounded by crops in the middle maps of Figure 6.20. However more cities are

Attribution of human excretions to surrounding fields with agroecological scenario

transport >50 km (15% of excretions)

human excretions

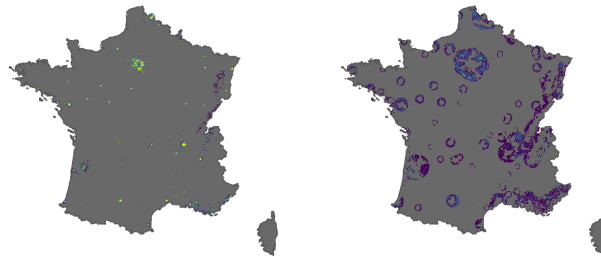
fertilization with excretions



transport 10 to 50 km (25% of excretions)

human excretions

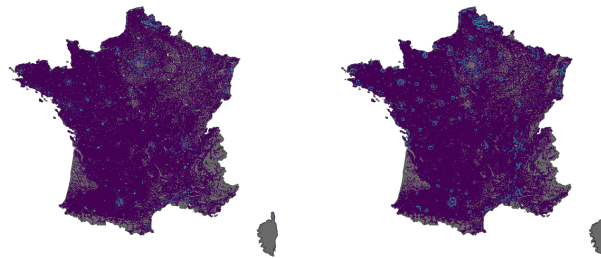
fertilization with excretions



transport <10 km (60% of excretion)

human excretions

fertilization with excretions



tons of N per km² ■ < 1 ■ 1 - 3 ■ 3 - 5 ■ 5 - 10 ■ > 10

Figure 6.20: Human excretions (left) and their associated fertilized agricultural parcels (right), broken down into 3 categories based on the distance between excretions and fertilized crops. This is based on agro-ecological scenario fertilization and diets (detailed in Section 6.1.1.2), following our algorithm attributing excretions to neighboring parcels. Distances refer to straight-line distance, as the crow flies, and not an actual distance that would be covered by a truck constrained by the presence of a road. Gray areas are excretions and agricultural parcels not concerned by the transport distance category.

concerned than in the current situation, because lower fertilization values require the excretions of medium size cities to be transported further.

The remaining population: 60% of excretions The rest of excretions, spread within a radius of 10 km, concerns 60% of excretions, which is slightly lower than the 70% for the current situation. Also, it concerns a smaller number of urban centers, and is more characteristic of rural areas, because in the current situation, high fertilization rates allowed to catch some excretions of urban centers within 10 km.

6.2.3.3 Influence of population density

Mean transport distances (weighted by the quantity of transported N) are slightly higher in the agro-ecological scenario compared to the current situation: 17 km vs 14 km. Breaking down the result by population density (Figure 6.17b) offers more detailed insights.

First, for population densities below 1,000 inhabitants·km⁻² (about 40% of population), virtually all their excretions can still be spread within 10 km.

Second, for population density between 1,000 and 10,000 inhabitants·km⁻², representing about half of the French population, the median distance is a bit higher (6 km instead of 3 km), but stays very modest. About three quarters of their excretions can be spread within 20 km (10 km in the current situation).

Finally, for the 10% living in areas with a density >10,000 inhabitants·km⁻², the distances are sensibly increased (median 70 km instead of 40 km) but never exceed 100 km, contrary to the current situation.

6.2.4 A distance/density typology that can help decisions

Both in the current scenario or in the agro-ecological one, clear typologies emerge from my analysis, which can inform the choices regarding logistics.

First, low-densities areas (<1,000 hab·km⁻²) with low transport distances (<10 km), gathering 40% of the French population. Generally, these areas will have space to store urine (except in small and dense urban centers in rural areas), and farmers won't be far. The challenge will maybe lie in optimizing the gathering in scattered settlements.

Second, high densities (>10,000 hab·km⁻²) with transport distances of a few tens of km, gathering about 10% of the population. Here the limited space for storage, associated with higher transport distances and the need to regularly evacuate high quantities of excretions might plead for concentrations techniques, to reduce the logistical hurdle.

Finally, the remaining 50% of population, between 1,000 and 10,000 hab·km⁻², which experiences very diverse situations regarding transport distances. The choices regarding recovery and transport methods will be more context-dependent, and treated on a case by case basis. Future works should particularly focus on that category.

Besides pure logistical convenience, Figure 6.1 can inform on the trade-offs regarding energy consumption in transport vs urine concentrations. Yet, several factors should be kept in mind when analyzing the graph. First, contrary to the Haber-Bosch process or WWTP denitrification, nitrogen concentration processes are generally not yet optimized, and further improvements and energy savings might still occur. Second, the graph features the energy cost associated to the transport of undiluted urine (8.7 gN/L), but in practice, the concentration will often be lower. For instance, diverting toilets still have a small flush, that can halve the N concentration, or urine collected in festivals tend to be more diluted, because of the volumes of beverages ingested. In any case, for now, the transport of raw urine on a distance of about 10 km consumes much less energy than almost all of the concentration processes, often by a factor at least 10.

6.2.5 Comparison with other similar works

Wielemaker et al. (2020) produced an interesting logistic optimization study to allocate human P excretions to agricultural parcels, within the municipality of Amsterdam. The interest of their work lies in that they defined collective storage point for urine, and calculated exact road distances for transport. However their results are difficult to compare. First, because they focused on phosphorus and not on nitrogen. Second, they stayed within the Amsterdam administrative boundary. So the 200 ktP excreted was way higher than the 75 ktP fertilization demand. They fulfilled only 40% of this demand, because beyond that threshold the number of storage points exponentially increased (from 500 at 40% to 2,500 at 100%) and wanted to focus on a stepwise transition. With these caveats in mind, more than three quarters of the transport distances were below 15 km.

Echevarria et al. (2021) used a slightly different framework than ours. They used a global dataset at a 5 arc minutes resolution (about 10 km x 10 km) for the year 2010. They then verified in each grid cell how much of the advised fertilization for crops could be covered with the supply of excretions. For France, they found that 45% of N excretions could be spread within the crops pixels without saturating the demand. That means that, according to these authors, at least 45% of human N excretions could be spread within 10 km. Some of the remaining 55% will also be within 10 km, but it is difficult to assess how much from their results, and so the

comparison with my 70% result is difficult.

Trimmer et al. (2018) computed the transport distances for 56 major world cities. Their data are quite old: crops distribution for the year 2000 and fertilization recommendation in the 90s. They also use a coarser resolution of 10 km x 10 km. However their results for the Paris metropolis are quite similar to mine: they find that 95% of the 10 million inhabitants excretions could be spread within 60 km, with a median distance of 35 km; I find that most excretions from this area can be spread within 10-50 km, with maximum distance just above the 50 km threshold.

A strength of my work is the spatial resolution. Most previous work at a national or global scale use grids of 10 km by 10 km, or 100 km². My population data is 100 time more precise, at 1 km². For agricultural parcels, I do not have a constant spatial extent, since I have data for all field crops. The largest ones are around 100 ha, or 1 km², and many are just 1-10 ha, or 0.01-0.1 km². In the case of Pakistan and Sweden, Akram et al. (2019) found that higher spatial resolutions decreased the estimated transport distances.

Finally, my distance results are simply "flying results". Actual transport distances, constrained by the roads network, will be higher. For comparison, found that in their case computing actual road transport increased distances by 7-21%. This suggests that my results would be barely changed if transport distances were actually computed. However one might expect that the results would be relatively more impacted for short distance transport than for long-distance.

6.3 Conclusion and perspectives

The main message of this chapter is that in both the current situation or an agro-ecological scenario with low N inputs, for more than half of excretions (respectively 70% and 60%), the distance to an agricultural parcel where it could be spread is less than 10 km.

About 20-25% of excretions can be spread within 10 to 50 km, and this usually concerns urban areas with a typical population of 0.1-1 million inhabitants. Finally, only about 5-15% of excretions require agricultural parcels situated further than 50 km.

The reader should remember that these results are specific to the particular hypotheses I made for this work, and which should be remembered. The distances calculated assume a 100% recovery rate of excretions, and lower recovery rates would result in less competition for agricultural land and thus smaller distances. On the other hand, in the *current* scenario, I assumed high fertilization rates with human excretions, sometimes $>150 \text{ kgN}\cdot\text{ha}^{-1}$, and lower fertilization rates would further increase the transport distances. I also only computed flying distances; actual transport distances by road would be a bit higher (maybe on the order of 10-20% based on Trimmer et al. (2018)).

In their life cycle analysis of stored urine used as fertilizer, Martin et al. (2023a) found little difference in the environmental impact between a 10 km vs a 25 km transport distance. The difference was more substantial for a 100 km distance, with a +50% energy consumption, and a +20% climate change impact (for lorries propelled by gasoline). But in all cases, the environmental impact of stored urine was better than the current system¹², except for the acidification impact, due to NH_3 volatilization, where it was broadly similar.

So, considering the typical distances I calculated and this life cycle analysis results, the issue is often not so much about the environmental performance of the concentration of urine-based fertilizers, but more about the logistical challenge. A farmer with 50 ha of major crops, fertilizing them with $50 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$ of urine at $5\text{gN}\cdot\text{L}^{-1}$ will need $500 \text{ m}^3\cdot\text{year}^{-1}$ of raw urine¹³. With freight lorries of 15 m^3 capacity, this means this farmer will need about 30 deliveries of urine a year to

¹²This nonetheless requires the logistics to be relatively efficient; if lorries are only half full, this will increase environmental impact.

¹³For context, in the late XIXth century, Mathey (2019) reports that Flemish farmers (with much smaller farms) already had tanks to store urine and feces with mean capacity of $120\text{-}140 \text{ m}^3$, so a storage 500 m^3 is clearly feasible. Moreover, feedbacks from field experiments indicates that "*The deposit must be available between February and May, requiring a 200 m^3 tank for approximately 0.3 to 1 t of nitrogen*" (L'Information agricole du Rhone 2024). My example of $50 \text{ kgN}/\text{ha}$ on 50 ha translates into 2.5 tons of N; so the mentioned 200 m^3 for 1 ton of N is coherent with my estimate of 500 m^3 .

be then stored in its tank, compared to only a few deliveries with current mineral fertilizers.

Likewise, on the other side of the supply chain, recovering and storing urine in rural households might be pretty straightforward; but for the 2.1 million inhabitants in intramural Paris, producing 1-1.5 liter of urine per day, resulting in 63,000 - 95,000 m³ per month, this would necessitate on the order of 6,000 to 10,000 lorries with a 10 m³ capacity to empty storage tanks in front of residential buildings, each month, or about 200-300 lorries each day ¹⁴. From a logistical viewpoint, in that case, it may be more interesting to concentrate urine rather than simply store it.

Hopefully, my results focusing on population density and distances to fertilize will help to inform the trade-offs behind these issues.

¹⁴According to Fabien Esculier (personal communication) there are around 400 garbage trucks circulating in Paris every day. This is consistent with a volume of waste of around 2L per person per day and a volume of urine of 1-1.5L.

General Conclusion

Summary of my results

My thesis began with some general context to the disruption of nitrogen and phosphorus flows, as well as how these nutrients contained in human excreta are handled; this is summarized in the following 2 sections.

Nitrogen and Phosphorus pollution

Nitrogen (N) and Phosphorus (P) are components of life's most essential molecules, and are crucial to agricultural systems productivity. But too much of these nutrients results in multiple pollutions impacting ecosystems and human health: eutrophication, contamination of groundwaters, atmospheric pollution, acidification of soils and contribution to climate change (Chapter 1).

This excess of nutrients has been exacerbated mainly by new reactive N and P inputs to agricultural systems. Up to the XXth century, new sources of nutrients (guanos, Chilean nitrates, bones acidulation...) barely made any difference at the global scale; but this changed with NH₃ fixed from atmospheric N₂ by the Haber-Bosh process, and phosphate rocks transformed into soluble PO₄³⁻ (Chapter 2). These new inputs rely on commodities not present on many countries' territories (gas or coal for N, phosphate rocks deposits for P). While there has been recent concerns of a peak P (i.e., the impossibility to further increase phosphate rocks production), I showed that up to know forecasts about this peak have always been postponed. From this, I argue that a more relevant aspect is the P vulnerability, to geopolitical or economic situations, a concept that can be extended to N.

In the past century, these new N and P inputs have drastically altered the N and P planetary cycles, with a roughly doubling of their flows intensity compared to the pre-industrial situation. This observation has triggered attempts to defined "N and P planetary boundaries", i.e. numerical values of the global N and P inputs viewed as "safe" in terms of pollution. With the N planetary boundary as a case study, I argue that it is not necessarily relevant to define such a boundary at the global level, as N impacts are very locally constrained, contrary to CO₂ and climate

change for instance.

As less than 20% of agricultural global N and P inputs are finally eaten by humans (this is the food system nutrient use efficiency), the majority does not reach the plate and ends up into losses and pollution, in waters for phosphorus, and in waters and air for nitrogen. Even the small part reaching the plate is eventually lost, as nutrient in human excreta (especially nitrogen) are barely reused as fertilizers at the global scale, and often end up simply discharged to the environment. Levers to decrease nutrients losses and pollution can be categorized into: increase the food system nutrient use efficiency (this includes: more vegetal diets, better crop fertilization, better handling of livestock manure, decreased food waste); do not over-eat N and P far beyond nutritional recommendations; and reuse nutrients in human excreta. This latter point is the main focus of this thesis.

Handling Human Excreta

In Chapter 3, I review past and present ways human excreta have been handled, and the consequence in terms of N and P recycling. It is difficult to estimate how much of the N and P in human excreta was recovered to fertilize crops in the distant past, where most people were farmers living just next to their agricultural field. We can hypothesize that a non negligible part was directly excreted on the field or mixed with animal manure. However, the linearity of the Western urban systems based on sewers have been extensively described, like London in the XIXth century. There was nonetheless some local exceptions, like Paris which reached a recovery of 50% N and 70% for P in excretions in the late XIXth, or the Flemish practices. By contrast, in China or Japan's Edo in the XIXth and part of the XXth century, reuse was widespread, even in urban areas.

But with the progressive adoption of sewers and wastewater treatment plants, reuse rates for N plummeted, while for P it can still be important. In wastewater treatment plants, P recovery can already be quite high (80-90%), but not necessarily in bio-available forms when it is precipitated in Fe or Al salts; there are also existing and developing processes to purify the recovered P. By contrast, there is currently no economically viable process to recover N in high amounts (beyond 30%) in wastewater treatment plants, because most processes rely on N effluents much more concentrated than wastewater effluents. However, most of the global population is not concerned by these processes, as less than half of humanity is currently connected to sewers.

Besides recovery of nutrients in wastewater treatment plants, I also present the approach of source separation of human excreta. The rationale is that excreta, and particularly urine, contain most of the N and P in wastewaters while constitut-

ing only around 1% of their volume. While there are numerous advantage to this option (less contaminated effluents, less pollution of water discharge, less resource consumption to produce fertilizers...), the challenges are more socio-technical, with a reorganization of the sanitation system, housing infrastructures, and farmers practices.

This general background, detailed in Part I of my thesis, has led me to define the following research question, to which each of the following 3 sections contributes to answer:

What is the current state regarding the fate of nitrogen and phosphorus in human excretions used as fertilizers, and what could be their agronomic potential ?

In France, N from human excreta is mostly lost, while P is partially reused as fertilizer

In Chapter 4, I described in details the current situation of the sanitation systems in rich industrialized countries largely connected to sewers and wastewater treatment plants, with a case study on France. For these countries, the reuse of N from human excreta to agriculture is most certainly not much higher than 10%, and the rest is lost. In France, about 50% is lost as inert atmospheric N_2 following energy-intensive nitrification/denitrification in wastewater treatment plants, and the remaining 40% is lost as reactive nitrogen in surface and groundwaters (NO_3^- , NH_4^+) and the atmosphere (NH_3 , N_2O). The current indicator currently followed by policy makers, the wastewater removal efficiency (70% for N in France), is blind to most of these losses, as it only looks at the share of nitrogen not discharged in waters, with the aim of limiting water pollution and eutrophication.

For phosphorus, reuse rates can come close to 50%, as in France, but are often below in most rich industrialized countries, from a few tens of percent to virtually zero for the countries which forbid the spreading of wastewater sludge. The main difference with nitrogen is that phosphorus removal in wastewater treatment plants relies on precipitation in sludge, allowing better recycling if removal efficiency is high (80% in France) and if sewage sludge are mostly spread on agricultural land (75% in France). But losses also happen before and outside wastewater treatment plants, in sewers and individual autonomous systems, further decreasing the reuse rate to 50% instead of $80\% \cdot 75\% = 60\%$.

These results are based on an extensive dataset I produced of the more than 20,000 wastewater treatment plants in France, reporting their nutrients inflow and outflows. The individual data from these facilities allow to visualize the influence

the European legislation on wastewater treatment plants removal efficiency through time. Increased efficiency concerned a few large facilities in areas classified as N or P sensitive, driving the national mean towards the 70% N and 80% P removal efficiencies; but there remains a multitude of wastewater treatment plants not concerned by the legislation whose discharges can have a local impact, especially in watercourses with small flow rate. Finally, the dataset also allows to visualize the reduction of a P pollution at the source, namely the progressive ban of P in dishwashers and washing machines detergents, which decreased P flows entering the wastewater treatment plants.

Future works could try to replicate my detailed analysis in other countries, when datasets of WWTP operational data are available. Also, some important uncertainties that should be reduced are the quantity of nutrients lost in sewers overflows and leaks, and the N content in sludge.

Quantification of the theoretical potential for N in human excreta to be used as fertilizer in food systems

My French case study is only representative of industrialized countries whose populations are highly connected to sewers and wastewater treatment plants. However only one third to 40% of the global population is currently connected to sewers, and by 2050 this will probably be not much more than 50%. For the rest, hundreds of millions of people in the world do not even have access to toilets, and the others rely on pit latrines and septic tanks. Assessing reuse rate of nutrients in human excreta requires to estimate their fate for these populations, and is stained with much uncertainties. For instance, published estimates of phosphorus reuse rate span from 10% to 50%. In Chapter 5, I give a tentative estimate for nitrogen, and my best guess is between 5% and 20% at the global scale, and more probably around 10-15%.

The current low reuse rate of nitrogen in human excreta at the global scale testifies of an unfulfilled agronomic potential, were it to be reused as fertilizer. I try to assess it in Chapter 5, for the current food systems of most world countries. N in human excreta expressed as a percentage of N inputs to croplands spans from 5% in high inputs agricultural exporters to 50% in low inputs agricultural importers, and most countries are between 10 to 30%. At the global scale, with no imports nor exports, the rate is 15-20%, and corresponds to the food system N use efficiency. I also give tentative estimates for a more frugal food systems (higher N use efficiency, less carnivorous diets, reduced food waste) and find upper plausible values of 30%, maybe 40% with considerable changes and mostly vegetarian diets.

Also, in most countries and at the global scale, quantities of N in human and non ruminant excretions are roughly similar. The global quantity of N in human excreta

is also roughly equivalent to the N in all livestock manure collected and spread on croplands. But, while manure is seen as a fertilizing resource to be recovered, human excreta is often simply viewed as a waste, to the point of destroying this N resource in wastewater treatment plants through denitrification.

In this Chapter I just compared the quantities of nutrients in human excreta to nutrient inputs to agricultural systems or to manure. But recovery of nutrients in human excreta will most probably be less than 100%. Future work should focus on plausible recovery potentials of nutrients in human excreta (which I did not assess in my thesis), allowing to expand my results beyond mere theoretical maximum potential.

Most of human excreta could be spread within a few kilometers in France

Actual recovery rate will depend on socio-technical factors, and in Chapter 6 I focused on one particular feature related to the logistics of handling human excreta. For France, I assessed the distance from excretions to crops, if all the human excretions were recovered and spread in an optimized manner. The distances I report are flying distances, and actual transport distances would be a bit higher. A side-product of this study is the production of a fine resolution dataset of N, P and K fertilization in France, at the agricultural parcel level, which might be of interest for other works.

With the current nitrogen fertilization rates, the mean distance would be 14 km, quite similar to the 17 km I found with low inputs fertilization practices for an agro-ecological scenario, with also more frugal protein intakes. In both cases, 60-70% of N in excretions could be spread within a short distance of 10 km; 20-25% of excretions could be spread within 10-50 km, mostly corresponding to medium-size cities (0.1-1 million inhabitants). Finally, 5-15% of excretions, all located in the populated regions of Paris and the Mediterranean coast, have to be spread further than 50 km, .

I also developed a typology based on population densities. For areas with less than 1,000 inhabitants·km⁻² (about 40% of the French population), the excretions can mostly be spread within a 3 km radius. For areas >10,000 inhabitants·km⁻² (10% of the population), the distance are mostly above 20-30 km, up to 100 km or more. The remaining half of the population experiences a variety of situations, from a few km to almost 100 km. These distances relate to deeply optimized logistics, and would be higher with less optimization; on the other hand, it hypothesizes a 100% recovery rate of N in human excreta, and lower recovery rates would decrease the distances.

These estimates can inform on trade-offs related urine-based fertilizers: is the environmental impact higher with raw urine or with concentrated urine-based fertilizers? A major parameter influencing the result in life-cycle analyses published on that matter is the transport distance: below a few tens of km, raw urine often scores better than concentrated urine. My results suggest that in France, in most of cases, concentration before transport would not be needed on environmental grounds, but would rather be a matter of logistical convenience (i.e., in densely populated areas) or of farmers preferences to handle smaller quantities.

Future related works could more specifically look at how to implement the logistics, by modeling actual transports along the road, and even more importantly how storage is handled, namely in terms of capacity and location of storage points.

Perspectives

Get insights from real-world projects

What can be the future perspectives for research on reusing human excreta in agriculture? To me, an important knowledge gap is how much of N and P in human excreta could actually be recovered in real-world conditions. Throughout my thesis, I always assumed a 100% recovery, and focused on maximum theoretical potential, but the real-world rate will certainly be below. Different logistics (storing urine in sealed containers vs composting with feces, using raw excreta vs highly transformed and concentrated urine...) will result in different losses along the chain. Likewise, if source-separation toilets are generalized, actual usage, dilution factors or separation rates will probably be different than anticipated. That is why I believe actual research-action programs are important on that matter, to get real world feedbacks. What is lacking the most is not theoretical knowledge on the benefits of nutrient recovery or its potential, but expertise and know-how on to best concrete implementation of such systems.

Future research on the topic might include (non exhaustive list): research into urine-diverting toilets, decentralized collection systems, and household-level nutrient recovery units, which could significantly improve the quality of recovered nutrients and reduce energy demands in wastewater treatment plants; research into developing efficient nutrient concentration techniques, which could facilitate storage and transport of recovered nutrients to agricultural regions; research into investigating the potential for integrating nutrient recovery with bioenergy production (e.g., biogas plants) and composting systems, which could promote a more holistic circular economy model.

Policy choices to increase nutrient recovery

However knowledge is not sufficient on its own, and it is most probable that large-scale source separation has little chance to be achieved without political support, just like wastewater treatment plants did not emerge out of nowhere, but as a result of law regulating discharge to waters. Likewise, high connection rate to sewers was achieved in the past by legal enforcement. If the aim is to recover nutrients from human excreta, and especially N, there are contrasted choices regarding the sanitation system: pursue in the current wastewater treatment plant system, with uncertain prospect concerning future technological ability to recover most of N in wastewaters, but with minimal organizational changes; or source separation, with major changes in infrastructure and logistics, but with high recovery rates that are known to be feasible on technical grounds.

Of course this dichotomy is simplified, and many other issues are at stakes: contamination of fertilizing materials, water and resource consumption, societal impacts of extractivism, resilience of the system... To properly compare the options, adequate metrics should be developed. Life-Cycle Analyses can inform on environmental performance of incremental changes, but will generally do not reflect the full consequences of undertaking major changes in the sanitation system. System thinking and scenarios can help better assess pros and cons.

Monitoring of excreta reuse should also be more regularly integrated to research and policy. It is possible to find international statistics on the share of livestock manure that is used to fertilize crops; there are numerous political plans to increase the recycling of urban biowastes (food waste, industrial wastes...); but policy makers and researchers are often blind to the fate of nutrients from human excreta. As I highlighted in my thesis, indicators related to the sanitation system only focus on pollution, with a 70% N removal efficiency in France that seems quite good; but there was no statistics on the poor 10% N recycling.

Integration into agricultural practices

Finally, recovering nutrients is not a goal in itself, but rather a means to decrease pollution, increase self-sufficiency, and fertilize agricultural parcels in a sustainable way.

Concerning self-sufficiency, this can constitute a key motive for policy-makers. The integration of nutrients derived from human excreta into agricultural practices offers significant potential for reducing dependence on finite resources such as natural gas and phosphate rock, which are critical for producing synthetic fertilizers. Phosphate rock reserves are geographically concentrated, raising concerns about geopolitical vulnerabilities. Similarly, nitrogen fertilizer production is heav-

ily reliant on natural gas through energy-intensive processes like the Haber-Bosch method, linking agricultural input costs to volatile energy markets. By recovering and recycling nutrients from human excreta, agriculture can decouple itself from these fossil resources, fostering greater resilience and independence.

Concerning fertilization, numerous works have been carried out in the past decades on crops fertilization with urine-based fertilizers, and more broadly with human excreta. An interesting and less studied perspective, beyond simple yield response and nutrient pollution, lies in how these new fertilizers could be synergistically integrated into crop rotations. This is particularly relevant for organic agriculture, which is nutrient limited. Should these fertilizers be added as a mineral nutrient boost during spring, leaving the rotation unchanged, or should it be used to add an additional cereal harvest to the rotation ? Should urine-based fertilizers be used in their mineral form, or rather incorporated in organic amendments like compost to respect organic farming principles ?

Contributions of my work

To conclude, I present some key insights from my work that could guide the development of public policies.

In Chapter 4, I quantified the major nutrient losses currently occurring within the sanitation system, ranging from individual autonomous systems to sewers, wastewater treatment plants (WWTPs), and unrecycled sludge. This analysis highlights critical points where immediate interventions could enhance nutrient recycling. More importantly, the chapter reveals that existing policy indicators, such as nutrient removal in WWTPs, are inadequate for monitoring nutrient recovery, pleading for the adoption of new indicators.

In Chapter 5, I demonstrated that, excluding fertilization linked to agricultural exports, the nitrogen contained in human excreta could account for approximately 10-40% of a country's agricultural fertilization needs, depending on the efficiency of its food system. This finding underscores the potential of nutrient recovery as a significant lever for achieving greater nutrient self-sufficiency and reducing reliance on imports—a message that could resonate strongly with policymakers.

Finally, Chapter 6 lays the groundwork for addressing the logistical challenges of implementing nutrient recovery in France, providing a preliminary framework for practical application. In addition, it shows that in France, most excretions could be spread just a few kilometers away.

I hope this thesis can help advancing nutrient recovery efforts and promoting more sustainable and less wasteful food and excreta management systems.

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Appendixes

Appendix A

RPG and TOPO items attribution

Attribution of fertilization to RPG and TOPO items

A.1 Major field crops and grasslands

The Table below presents which of RPG crop labels I associated with one of the 17 crops surveyed in the cultivation practices survey (Agreste 2017b).

Table A.1: Association of 42 RPG items to the 17 major field crops and grasslands surveyed in Agreste (2017b).

cultural practices survey label	RPG crop code	RPG crop label	RPG groups code	RPG groups label	my English label
Blé tendre	BTH	Blé tendre d'hiver	1	Blé tendre	Soft wheat
Blé tendre	BTP	Blé tendre de printemps	1	Blé tendre	Soft wheat
Blé dur	BDH	Blé dur d'hiver	4	Autres céréales	Durum wheat
Blé dur	BDP	Blé dur de printemps	4	Autres céréales	Durum wheat
Blé dur	BDT	Blé dur de printemps semé tardivement (après le 31/05)	4	Autres céréales	Durum wheat
Orge	ORH	Orge d'hiver	3	Orge	Barley
Orge	ORP	Orge de printemps	3	Orge	Barley
Orge	CHH	Autre céréale d'hiver de genre Hordeum	4	Autres céréales	Barley
Orge	CPH	Autre céréale de printemps de genre Hordeum	4	Autres céréales	Barley
Triticale	CHT	Autre céréale d'hiver de genre Triticum	4	Autres céréales	Triticale
Triticale	CPT	Autre céréale de printemps de genre Triticum	4	Autres céréales	Triticale
Triticale	TTH	Triticale d'hiver	4	Autres céréales	Triticale
Triticale	TTP	Triticale de printemps	4	Autres céréales	Triticale
Colza	CZH	Colza d'hiver	5	Colza	Rapeseed
Colza	CZP	Colza de printemps	5	Colza	Rapeseed
Tournesol	TRN	Tournesol	6	Tournesol	Sunflower

Tournesol	OEH	Autre oléagineux d'espèce Helianthus	7	Autres oléagineux	Sunflower
Pois protéagineux	PHI	Pois d'hiver	8	Protéagineux	Protein pea
Pois protéagineux	PPR	Pois de printemps semé avant le 31/05	8	Protéagineux	Protein pea
Pois protéagineux	PPT	Pois de printemps semé tardivement (après le 31/05)	8	Protéagineux	Protein pea
Maïs fourrage	MIE	Maïs ensilage	2	Maïs grain et ensilage	Forage maize
Maïs grain	MIS	Maïs	2	Maïs grain et ensilage	Grain maize
Maïs grain	CPZ	Autre céréale de printemps de genre Zea	4	Autres céréales	Grain maize
Betterave sucrière	BVF	Betterave fourragère	16	Fourrage	Fodder – Non Legume
Betterave sucrière	BTN	Betterave non fourragère / Bette	24	Autres cultures industrielles	Sugar beet
Pomme de terre	PTC	Pomme de terre de consommation	25	Légumes ou fleurs	Potato
Pomme de terre	PTF	Pomme de terre féculière	25	Légumes ou fleurs	Potato
Prairies Temporaires	PTR	Autre prairie temporaire de 5 ans ou moins	19	Prairies temporaires	Temporary grasslands
Prairies Temporaires	RGA	Ray-grass de 5 ans ou moins	19	Prairies temporaires	Temporary grasslands
Prairies Permanententes	PPH	Prairie permanente - herbe prédominante (ressources fourragères ligneuses absentes ou peu présentes)	18	Prairies permanententes	Permanent grasslands
Prairies Permanententes	PRL	Prairie en rotation longue (6 ans ou plus)	18	Prairies permanententes	Permanent grasslands
Féverole	FVL	Féverole semée avant le 31/05	8	Protéagineux	Faba bean
Féverole	FVT	Féverole semée tardivement (après le 31/05)	8	Protéagineux	Faba bean
Féverole	FF5	Féverole fourragère implantée pour la récolte 2015	16	Fourrage	Faba bean
Féverole	FF6	Féverole fourragère implantée pour la récolte 2016	16	Fourrage	Faba bean
Féverole	FF7	Féverole fourragère implantée pour la récolte 2017	16	Fourrage	Faba bean
Féverole	FF8	Féverole fourragère implantée pour la récolte 2018	16	Fourrage	Faba bean
Féverole	FFO	Autre féverole fourragère	16	Fourrage	Faba bean
Soja	SOJ	Soja	7	Autres oléagineux	Soybean
Lin fibre	LIF	Lin fibres	9	Plantes à fibres	Fiber flax

Lin Oléagineux	LIH	Lin non textile d'hiver	7	Autres oléagineux	Oilseed flax
Lin Oléagineux	LIP	Lin non textile de printemps	7	Autres oléagineux	Oilseed flax

A.2 Similar major field crops and grasslands

Some major field crops are not surveyed in the cultivation practices survey (Agreste 2017b), but are quite similar to one of the 17 crops surveyed. For these, Table A.2 presents the broad categories ("*my English label*") I associated with some particular surveyed crops. For some items like "fallow", "others", and "mountain pastures and heaths", I set their fertilization value to 0 kgN·ha⁻¹.

Table A.3 presents in more details the 116 RPG items I associated with my broad categories.

Table A.2: Broad categories of major field crops whose fertilization values I approximated by crops surveyed in the cultivation practices survey (Agreste 2017b). For Fallow, Others, and Mountain pastures and heaths, I simply set a 0 fertilization value. See Table A.2 for details about my matching of broad major field crops categories to RPG items.

my English label	Survey approximation French label	Survey approximation English label
Other cereals	Orge	Barley
Other oilseeds	Tournesol	Sunflower
Other fiber crops	Lin Fibre	Fiber Flax
Other protein crops	Féverole	Faba Bean
Other grain legumes	Soja	Soybean
Fodder- Non legume	Maïs fourrage	Forage Maize
Fodder- Legume	Féverole	Faba Bean
Fodder – Legume + Non Legume	Moyenne Maïs Fourrage et Féverolle	Forage Maize and Faba Bean averaged
Fallow	-	-
Others	-	-
Mountain pastures and heaths	-	-

Table A.3: Association of 116 RPG items to my broad categories, for the major field crops and grasslands not directly surveyed in Agreste (2017b)

my English label	RPG crop code	RPG crop label	RPG group code	RPG group label
Fallow	J5M	Jachère de 5 ans ou moins	11	Gel (surfaces gelées sans production)
Fallow	J6P	Jachère de 6 ans ou plus	11	Gel (surfaces gelées sans production)
Fallow	J6S	Jachère de 6 ans ou plus déclarée comme Surface d'intérêt écologique	11	Gel (surfaces gelées sans production)
Fallow	JNO	Jachère noire	11	Gel (surfaces gelées sans production)
Fodder – Legume	GES	Gesse	16	Fourrage
Fodder – Legume	JO5	Jarosse implantée pour la récolte 2015	16	Fourrage
Fodder – Legume	JO6	Jarosse implantée pour la récolte 2016	16	Fourrage
Fodder – Legume	JO7	Jarosse implantée pour la récolte 2017	16	Fourrage

Fodder – Legume	JO8	Jarosse implantée pour la récolte 2018	16	Fourrage
Fodder – Legume	JOD	Jarosse déshydratée	16	Fourrage
Fodder – Legume	JOS	Autre jarosse	16	Fourrage
Fodder – Legume	LEF	Lentille fourragère	16	Fourrage
Fodder – Legume	LFH	Autre lupin fourrager d’hiver	16	Fourrage
Fodder – Legume	LFP	Autre lupin fourrager de printemps	16	Fourrage
Fodder – Legume	LH5	Lupin fourrager d’hiver implanté pour la récolte 2015	16	Fourrage
Fodder – Legume	LH6	Lupin fourrager d’hiver implanté pour la récolte 2016	16	Fourrage
Fodder – Legume	LH7	Lupin fourrager d’hiver implanté pour la récolte 2017	16	Fourrage
Fodder – Legume	LH8	Lupin fourrager d’hiver implanté pour la récolte 2018	16	Fourrage
Fodder – Legume	LO7	Lotier implanté pour la récolte 2017	16	Fourrage
Fodder – Legume	LO8	Lotier implanté pour la récolte 2018	16	Fourrage
Fodder – Legume	LOT	Lotier	16	Fourrage
Fodder – Legume	LP5	Lupin fourrager de printemps implanté pour la récolte 2015	16	Fourrage
Fodder – Legume	LP6	Lupin fourrager de printemps implanté pour la récolte 2016	16	Fourrage
Fodder – Legume	LP7	Lupin fourrager de printemps implanté pour la récolte 2017	16	Fourrage
Fodder – Legume	LP8	Lupin fourrager de printemps implanté pour la récolte 2018	16	Fourrage
Fodder – Legume	LU5	Luzerne implantée pour la récolte 2015	16	Fourrage
Fodder – Legume	LU6	Luzerne implantée pour la récolte 2016	16	Fourrage
Fodder – Legume	LU7	Luzerne implantée pour la récolte 2017	16	Fourrage
Fodder – Legume	LU8	Luzerne implantée pour la récolte 2018	16	Fourrage
Fodder – Legume	LUD	Luzerne déshydratée	16	Fourrage
Fodder – Legume	LUZ	Autre luzerne	16	Fourrage
Fodder – Legume	ME5	Ménilot implanté pour la récolte 2015	16	Fourrage
Fodder – Legume	ME6	Ménilot implanté pour la récolte 2016	16	Fourrage
Fodder – Legume	ME7	Ménilot implanté pour la récolte 2017	16	Fourrage
Fodder – Legume	ME8	Ménilot implanté pour la récolte 2018	16	Fourrage
Fodder – Legume	MED	Ménilot déshydraté	16	Fourrage
Fodder – Legume	MEL	Autre ménilot	16	Fourrage
Fodder – Legume	MI7	Minette implanté pour la récolte 2017	16	Fourrage
Fodder – Legume	MI8	Minette implanté pour la récolte 2018	16	Fourrage
Fodder – Legume	MIN	Minette	16	Fourrage
Fodder – Legume	ML5	Mélange de légumineuses fourragères implantées pour la récolte 2015 (entre elles)	16	Fourrage

Fodder – Legume	ML6	Mélange de légumineuses fourragères implantées pour la récolte 2016 (entre elles)	16	Fourrage
Fodder – Legume	ML7	Mélange de légumineuses fourragères implantées pour la récolte 2017 (entre elles)	16	Fourrage
Fodder – Legume	ML8	Mélange de légumineuses fourragères implantées pour la récolte 2018 (entre elles)	16	Fourrage
Fodder – Legume	MLD	Mélange de légumineuses déshydratées (entre elles)	16	Fourrage
Fodder – Legume	MLF	Mélange de légumineuses fourragères (entre elles)	16	Fourrage
Fodder – Legume	PFH	Autre pois fourrager d'hiver	16	Fourrage
Fodder – Legume	PFP	Autre pois fourrager de printemps	16	Fourrage
Fodder – Legume	PH5	Pois fourrager d'hiver implanté pour la récolte 2015	16	Fourrage
Fodder – Legume	PH6	Pois fourrager d'hiver implanté pour la récolte 2016	16	Fourrage
Fodder – Legume	PH7	Pois fourrager d'hiver implanté pour la récolte 2017	16	Fourrage
Fodder – Legume	PH8	Pois fourrager d'hiver implanté pour la récolte 2018	16	Fourrage
Fodder – Legume	PP5	Pois fourrager de printemps implanté pour la récolte 2015	16	Fourrage
Fodder – Legume	PP6	Pois fourrager de printemps implanté pour la récolte 2016	16	Fourrage
Fodder – Legume	PP7	Pois fourrager de printemps implanté pour la récolte 2017	16	Fourrage
Fodder – Legume	PP8	Pois fourrager de printemps implanté pour la récolte 2018	16	Fourrage
Fodder – Legume	SA5	Sainfoin implanté pour la récolte 2015	16	Fourrage
Fodder – Legume	SA6	Sainfoin implanté pour la récolte 2016	16	Fourrage
Fodder – Legume	SA7	Sainfoin implanté pour la récolte 2017	16	Fourrage
Fodder – Legume	SA8	Sainfoin implanté pour la récolte 2018	16	Fourrage
Fodder – Legume	SAD	Sainfoin déshydraté	16	Fourrage
Fodder – Legume	SAI	Autre sainfoin	16	Fourrage
Fodder – Legume	SE5	Serradelle implantée pour la récolte 2015	16	Fourrage
Fodder – Legume	SE6	Serradelle implantée pour la récolte 2016	16	Fourrage
Fodder – Legume	SE7	Serradelle implantée pour la récolte 2017	16	Fourrage
Fodder – Legume	SE8	Serradelle implantée pour la récolte 2018	16	Fourrage
Fodder – Legume	SED	Serradelle déshydratée	16	Fourrage
Fodder – Legume	SER	Autre serradelle	16	Fourrage
Fodder – Legume	TR5	Trèfle implanté pour la récolte 2015	16	Fourrage
Fodder – Legume	TR6	Trèfle implanté pour la récolte 2016	16	Fourrage
Fodder – Legume	TR7	Trèfle implanté pour la récolte 2017	16	Fourrage
Fodder – Legume	TR8	Trèfle implanté pour la récolte 2018	16	Fourrage

Fodder – Legume	TRD	Trèfle déshydraté	16	Fourrage
Fodder – Legume	TRE	Autre trèfle	16	Fourrage
Fodder – Legume	VE5	Vesce implantée pour la récolte 2015	16	Fourrage
Fodder – Legume	VE6	Vesce implantée pour la récolte 2016	16	Fourrage
Fodder – Legume	VE7	Vesce implantée pour la récolte 2017	16	Fourrage
Fodder – Legume	VE8	Vesce implantée pour la récolte 2018	16	Fourrage
Fodder – Legume	VED	Vesce déshydratée	16	Fourrage
Fodder – Legume	VES	Autre vesce	16	Fourrage
Fodder – Legume	XFE	X-Felium de 5 ans ou moins	16	Fourrage
Fodder – Legume + Non Legume	CPL	Fourrage composé de céréales et/ou de protéagineux (en proportion < 50%) et/ou de légumineuses fourragères (en proportion < 50%)	16	Fourrage
Fodder – Legume + Non Legume	MC5	Mélange de légumineuses fourragères prépondérantes au semis implantées pour la récolte 2015 et de céréales		Fourrage
Fodder – Legume + Non Legume	MC6	Mélange de légumineuses fourragères prépondérantes au semis implantées pour la récolte 2016 et de céréales	16	Fourrage
Fodder – Legume + Non Legume	MC7	Mélange de légumineuses fourragères prépondérantes au semis implantées pour la récolte 2017 et de céréales	16	Fourrage
Fodder – Legume + Non Legume	MC8	Mélange de légumineuses fourragères prépondérantes implantées pour la récolte 2018 et de céréales et d'oléagineux	16	Fourrage
Fodder – Legume + Non Legume	MH5	Mélange de légumineuses fourragères prépondérantes au semis implantées pour la récolte 2015 et d'herbacées ou de graminées fourragères	16	Fourrage
Fodder – Legume + Non Legume	MH6	Mélange de légumineuses fourragères prépondérantes au semis implantées pour la récolte 2016 et d'herbacées ou de graminées fourragères	16	Fourrage
Fodder – Legume + Non Legume	MH7	Mélange de légumineuses fourragères prépondérantes au semis implantées pour la récolte 2017 et d'herbacées ou de graminées fourragères	16	Fourrage
Fodder – Legume + Non Legume	MLC	Mélange de légumineuses fourragères prépondérantes et de céréales et/ou d'oléagineux	16	Fourrage
Fodder – Legume + Non Legume	MLG	Mélange de légumineuses prépondérantes au semis et de graminées fourragères de 5 ans ou moins	16	Fourrage
Fodder – Non Legume	CAF	Carotte fourragère	16	Fourrage
Fodder – Non Legume	CHF	Chou fourrager	16	Fourrage
Fodder – Non Legume	DTY	Dactyle de 5 ans ou moins	16	Fourrage

Fodder – Non Legume	FAG	Autre fourrage annuel d'un autre genre	16	Fourrage
Fodder – Non Legume	FET	Fétuque de 5 ans ou moins	16	Fourrage
Fodder – Non Legume	FLO	Fléole de 5 ans ou moins	16	Fourrage
Fodder – Non Legume	FSG	Autre plante fourragère sarclée d'un autre genre	16	Fourrage
Fodder – Non Legume	GAI	Gaïlet	16	Fourrage
Fodder – Non Legume	GFP	Autre graminée fourragère pure de 5 ans ou moins	16	Fourrage
Fodder – Non Legume	MOH	Moha	16	Fourrage
Fodder – Non Legume	NVF	Navet fourrager	16	Fourrage
Fodder – Non Legume	PAT	Pâturin commun de 5 ans ou moins	16	Fourrage
Fodder – Non Legume	RDF	Radis fourrager	16	Fourrage
Mountain pastures and heaths	BOP	Bois pâturé	17	Estives et landes
Mountain pastures and heaths	SPH	Surface pastorale - herbe prédominante et ressources fourragères ligneuses présentes	17	Estives et landes
Mountain pastures and heaths	SPL	Surface pastorale - ressources fourragères ligneuses prédominantes	17	Estives et landes
Other cereals	AVH	Avoine d'hiver	4	Autres céréales
Other cereals	AVP	Avoine de printemps	4	Autres céréales
Other cereals	CAG	Autre céréale d'un autre genre	4	Autres céréales
Other cereals	CGF	Autre céréale de genre Fagopyrum	4	Autres céréales
Other cereals	CGH	Autre céréale de genre Phalaris	4	Autres céréales
Other cereals	CGO	Autre céréale de genre Sorghum	4	Autres céréales
Other cereals	CGP	Autre céréale de genre Panicum	4	Autres céréales
Other cereals	CGS	Autre céréale de genre Setaria	4	Autres céréales
Other cereals	CHA	Autre céréale d'hiver de genre Avena	4	Autres céréales
Other cereals	CHS	Autre céréale d'hiver de genre Secale	4	Autres céréales
Other cereals	CPA	Autre céréale de printemps de genre Avena	4	Autres céréales
Other cereals	CPS	Autre céréale de printemps de genre Secale	4	Autres céréales
Other cereals	EPE	Épeautre	4	Autres céréales
Other cereals	MCR	Mélange de céréales	4	Autres céréales
Other cereals	MLT	Millet	4	Autres céréales
Other cereals	SGH	Seigle d'hiver	4	Autres céréales
Other cereals	SGP	Seigle de printemps	4	Autres céréales
Other cereals	SOG	Sorgho	4	Autres céréales
Other cereals	SRS	Sarrasin	4	Autres céréales
Other cereals	RIZ	Riz	14	Riz
Other fiber crops	CHV	Chanvre	9	Plantes à fibres
Other fiber crops	CSE	Chanvre sans étiquette conforme	9	Plantes à fibres
Other grain legume	LEC	Lentille cultivée (non fourragère)	15	Légumineuses à grains
Other grain legume	PCH	Pois chiche	15	Légumineuses à grains
Other oilseeds	ARA	Arachide	7	Autres oléagineux
Other oilseeds	MOL	Mélange d'oléagineux	7	Autres oléagineux
Other oilseeds	NVE	Navette d'été	7	Autres oléagineux
Other oilseeds	NVH	Navette d'hiver	7	Autres oléagineux
Other oilseeds	OAG	Autre oléagineux d'un autre genre	7	Autres oléagineux
Other oilseeds	OEI	Œillette	7	Autres oléagineux
Other oilseeds	OHN	Autre oléagineux d'hiver d'espèce Brassica napus	7	Autres oléagineux

Other oilseeds	OHR	Autre oléagineux d'hiver d'espèce Brassica rapa	7	Autres oléagineux
Other oilseeds	OPN	Autre oléagineux de printemps d'espèce Brassica napus	7	Autres oléagineux
Other oilseeds	OPR	Autre oléagineux de printemps d'espèce Brassica rapa	7	Autres oléagineux
Other protein crops	FEV	Fève	8	Protéagineux
Other protein crops	LDH	Lupin doux d'hiver	8	Protéagineux
Other protein crops	LDP	Lupin doux de printemps semé avant le 31/05	8	Protéagineux
Other protein crops	LDT	Lupin doux de printemps semé tardivement (après le 31/05)	8	Protéagineux
Other protein crops	MPA	Autre mélange de plantes fixant l'azote	28	Divers
Other protein crops	MLS	Mélange de légumineuses non fourragères prépondérantes et de céréales et/ou d'oléagineux	8	Protéagineux
Other protein crops	MPC	Mélange de protéagineux (pois et/ou lupin et/ou féverole) prépondérants semés avant le 31/05 et de céréales	8	Protéagineux
Other protein crops	MPP	Mélange de protéagineux (pois et/ou lupin et/ou féverole)	8	Protéagineux
Other protein crops	MPT	Mélange de protéagineux semés tardivement (après le 31/05)	8	Protéagineux
Other protein crops	PAG	Autre protéagineux d'un autre genre	8	Protéagineux
Other sugar cane	CSA	Canne à sucre - autre	26	Canne à sucre
Other sugar cane	CSF	Canne à sucre - fermage	26	Canne à sucre
Other sugar cane	CSI	Canne à sucre - indivision	26	Canne à sucre
Other sugar cane	CSP	Canne à sucre - propriété ou faire valoir direct	26	Canne à sucre
Other sugar cane	CSR	Canne à sucre - réforme foncière	26	Canne à sucre
Others	ACA	Autre culture non précisée dans la liste (admissible)	28	Divers
Others	BFP	Bande admissible le long d'une forêt avec production	28	Divers
Others	BFS	Bande admissible le long d'une forêt sans production	28	Divers
Others	BOR	Bordure de champ	28	Divers
Others	BRO	Brome de 5 ans ou moins	28	Divers
Others	BTA	Bande tampon	28	Divers
Others	CAE	Châtaigneraie entretenue par des porcins ou des petits ruminants	28	Divers
Others	CEE	Chênaie entretenue par des porcins ou des petits ruminants	28	Divers
Others	CNE	Chênaie non entretenue par des porcins ou des petits ruminants	28	Divers
Others	CID	Cultures conduites en interrangs : 2 cultures représentant chacune plus de 25%	28	Divers
Others	CIT	Cultures conduites en interrangs : 3 cultures représentant chacune plus de 25%	28	Divers
Others	CUA	Culture sous abattis	28	Divers
Others	MCT	Miscanthus	28	Divers
Others	MRS	Marais salant	28	Divers
Others	NYG	Nyger	28	Divers
Others	PCL	Phacélie de 5 ans ou moins	28	Divers

Others	PEP	Pépinière	28	Divers
Others	ROS	Roselière	28	Divers
Others	SBO	Surface boisée sur une ancienne terre agricole	28	Divers
Others	SNA	Surface non agricole non visible sur l'orthophotographie	28	Divers
Others	SNE	Surface agricole temporairement non exploitée	28	Divers
Others	TBT	Tubercule tropical	28	Divers
Others	TCR	Taillis à courte rotation	28	Divers
Others	TRU	Truffière (chênaie de plants mycorhizés)	28	Divers
Others	VET	Vétiver	28	Divers
Others	ZZZ	Culture inconnue	28	Divers
Others	ACP	Autre culture pérenne	28	Divers

A.3 Vineyards

Table A.4: The 4 RPG items that I identified as vineyards. These items are removed from my RPG dataset. To identify vineyards fields, I use the TOPO database instead of the RPG dataset.

RPG crop code	RPG crop label	RPG group code	RPG group label
RVI	Restructuration du vignoble	21	Vignes
VRC	Vigne : raisins de cuve	21	Vignes
VRN	Vigne : raisins de cuve non en production	21	Vignes
VRT	Vigne : raisins de table	21	Vignes

Table A.5: Matching between French regions and wine-growing basins to adapt the results from French cultivation practice survey (Agreste 2019)

French regions (pre-2015 reform)	Associated wine-growing basins
Île-de-France	Champagne
Champagne-Ardenne	Champagne
Picardie	Champagne
Haute-Normandie	Val de Loire
Centre	Val de Loire; Cher
Basse-Normandie	Val de Loire
Bourgogne	Bourgogne
Nord-Pas-de-Calais	Champagne
Lorraine	Alsace
Alsace	Alsace
Franche-Comté	Jura
Pays de la Loire	Val de Loire
Bretagne	Val de Loire
Poitou-Charentes	Charentes
Aquitaine	Lot-et-Garonne; Dordogne; Bordelais
Midi-Pyrénées	Gers; Gaillac; Cahors
Limousin	Val de Loire
Rhône-Alpes	Bugey Savoie; Beaujolais; Côtes-du-Rhône Nord, Côtes-du-Rhône Sud
Auvergne	Val de Loire
Languedoc-Roussillon	Languedoc hors Pyrénées-Orientales; Pyrénées-Orientales
Provence-Alpes-Côte d'Azur	Provence (Var-Vaucluse); Bouches-du-Rhône
Corse	Corse

A.4 Orchards

Table A.6: The 28 RPG items that I identify as orchards. These items are removed from the RPG dataset. I rather use the orchards fields reported in the TOPO database. However I use RPG orchards items, coupled with AND International (2022) fertilization values, to determine averaged regional orchards fertilization values (Figure 6.8). This table shows the matching between AND International (2022) and RPG orchards labels.

AND International label	RPG crop code	RPG crop label	RPG group code	RPG group label
Agrumes	AGR	Agrume	20	Vergers
Autres fruits à coque	CAB	Caroube	22	Fruits à coque
Autres fruits à coque	NOS	Noisette	22	Fruits à coque
Autres fruits à coque	PIS	Pistache	22	Fruits à coque
Autres fruits à pépins et à noyaux	CBT	Cerise bigarreau pour transformation	20	Vergers
Autres fruits à pépins et à noyaux	PVT	Pêche Pavie pour transformation	20	Vergers
Autres fruits ou sans détail	VGD	Verger (DOM)	20	Vergers
Autres fruits ou sans détail	VRG	Verger	20	Vergers
Baies	PFR	Petit fruit rouge	20	Vergers
Baies	FRA	Fraise	25	Légumes ou fleurs
Châtaignes et marrons	CTG	Châtaigne	22	Fruits à coque
Fruits tropicaux et subtropicaux	ANA	Ananas	20	Vergers
Fruits tropicaux et subtropicaux	AVO	Avocat	20	Vergers
Fruits tropicaux et subtropicaux	BCA	Banane créole (fruit et légume) - autre	20	Vergers
Fruits tropicaux et subtropicaux	BCF	Banane créole (fruit et légume) - fermage	20	Vergers
Fruits tropicaux et subtropicaux	BCI	Banane créole (fruit et légume) - indivision	20	Vergers
Fruits tropicaux et subtropicaux	BCP	Banane créole (fruit et légume) - propriété ou faire valoir direct	20	Vergers
Fruits tropicaux et subtropicaux	BCR	Banane créole (fruit et légume) - réforme foncière	20	Vergers
Fruits tropicaux et subtropicaux	BEA	Banane export - autre	20	Vergers
Fruits tropicaux et subtropicaux	BEF	Banane export - fermage	20	Vergers
Fruits tropicaux et subtropicaux	BEI	Banane export - indivision	20	Vergers
Fruits tropicaux et subtropicaux	BEP	Banane export - propriété ou faire valoir direct	20	Vergers
Fruits tropicaux et subtropicaux	BER	Banane export - réforme foncière	20	Vergers
Fruits tropicaux et subtropicaux	CAC	Café / Cacao	20	Vergers
Noix	NOX	Noix	22	Fruits à coque
Olives	OLI	Oliveraie	23	Oliviers
Poires	PWT	Poire Williams pour transformation	20	Vergers
Prunes	PRU	Prune d'Ente pour transformation	20	Vergers

A.5 Vegetable and Floral crops

Table A.7: The 95 items RPG items that I identify as Vegetables and Floral crops, and their matching to AND International (2022) labels. This latter report is used to determine fertilization values.

AND International label	RPG crop code	RPG crop label	RPG group code	RPG group label
Autres légumes	AUB	Aubergine	25	Légumes ou fleurs
Autres légumes	BLT	Bleuet	25	Légumes ou fleurs
Autres légumes	BUR	Bugle rampante	25	Légumes ou fleurs
Autres légumes	CRA	Cresson alénois de 5 ans ou moins	25	Légumes ou fleurs
Autres légumes	CRN	Cornille	25	Légumes ou fleurs
Autres légumes	CRS	Cresson	25	Légumes ou fleurs
Autres légumes	DOL	Dolique	25	Légumes ou fleurs
Autres légumes	FLA	Autre légume ou fruit annuel	25	Légumes ou fleurs
Autres légumes	FLP	Autre légume ou fruit pérenne	25	Légumes ou fleurs
Autres légumes	GER	Géranium	25	Légumes ou fleurs
Autres légumes	HPC	Horticulture ornementale de plein champ	25	Légumes ou fleurs
Autres légumes	HSA	Horticulture ornementale sous abri	25	Légumes ou fleurs
Autres légumes	MRG	Marguerite	25	Légumes ou fleurs
Autres légumes	PAQ	Pâquerette	25	Légumes ou fleurs
Autres légumes	PAS	Pastèque	25	Légumes ou fleurs
Autres légumes	PMV	Primevère	25	Légumes ou fleurs
Autres légumes	POT	Potiron / Potimarron	25	Légumes ou fleurs
Autres légumes	PSE	Pensée	25	Légumes ou fleurs
Autres légumes	PVP	Poivron / Piment	25	Légumes ou fleurs
Autres légumes	ROQ	Roquette	25	Légumes ou fleurs
Autres légumes	RUT	Rutabaga	25	Légumes ou fleurs
Bulbes	AIL	Aïl	25	Légumes ou fleurs
Bulbes	OIG	Oignon / Échalote	25	Légumes ou fleurs
Coriandre	CRD	Coriandre	24	Autres cultures industrielles
Divers PPAM	ANE	Aneth	24	Autres cultures industrielles
Divers PPAM	ANG	Angélique	24	Autres cultures industrielles
Divers PPAM	ANI	Anis	24	Autres cultures industrielles
Divers PPAM	BAR	Bardane	24	Autres cultures industrielles
Divers PPAM	BAS	Basilic	24	Autres cultures industrielles
Divers PPAM	BRH	Bourrache de 5 ans ou moins	24	Autres cultures industrielles
Divers PPAM	CAV	Carvi	24	Autres cultures industrielles
Divers PPAM	CHR	Chardon Marie	24	Autres cultures industrielles
Divers PPAM	CIB	Ciboulette	24	Autres cultures industrielles
Divers PPAM	CML	Cameline	24	Autres cultures industrielles
Divers PPAM	CMM	Camomille	24	Autres cultures industrielles
Divers PPAM	CRF	Cerfeuil	24	Autres cultures industrielles
Divers PPAM	CUM	Cumin	24	Autres cultures industrielles
Divers PPAM	CUR	Curcuma	24	Autres cultures industrielles
Divers PPAM	EST	Estragon	24	Autres cultures industrielles
Divers PPAM	FNO	Fenouil	24	Autres cultures industrielles
Divers PPAM	FNU	Fenugrec	24	Autres cultures industrielles
Divers PPAM	MAV	Mauve	24	Autres cultures industrielles

Divers PPAM	MLI	Mélicse	24	Autres cultures industrielles
Divers PPAM	MLP	Millepertuis	24	Autres cultures industrielles
Divers PPAM	MRJ	Marjolaine / Origan	24	Autres cultures industrielles
Divers PPAM	MTH	Menthe	24	Autres cultures industrielles
Divers PPAM	ORT	Ortie	24	Autres cultures industrielles
Divers PPAM	OSE	Oseille	24	Autres cultures industrielles
Divers PPAM	PAR	Plante aromatique (autre que vanille)	24	Autres cultures industrielles
Divers PPAM	PMD	Plante médicinale	24	Autres cultures industrielles
Divers PPAM	PPA	Autre plante à parfum, aromatique et médicinale annuelle	24	Autres cultures industrielles
Divers PPAM	PPF	Plante à parfum (autre que géranium et vétiver)	24	Autres cultures industrielles
Divers PPAM	PPP	Autre plante à parfum, aromatique et médicinale pérenne	24	Autres cultures industrielles
Divers PPAM	PSL	Persil	24	Autres cultures industrielles
Divers PPAM	PSN	Psyllium noir de Provence	24	Autres cultures industrielles
Divers PPAM	PSY	Plantain psyllium	24	Autres cultures industrielles
Divers PPAM	SGE	Sauge	24	Autres cultures industrielles
Divers PPAM	SRI	Sarriette	24	Autres cultures industrielles
Divers PPAM	THY	Thym	24	Autres cultures industrielles
Divers PPAM	VAL	Valériane	24	Autres cultures industrielles
Divers PPAM	VNB	Vanille sous bois	24	Autres cultures industrielles
Divers PPAM	VNL	Vanille	24	Autres cultures industrielles
Divers PPAM	VNV	Vanille verte	24	Autres cultures industrielles
Divers PPAM	YLA	Ylang-ylang	24	Autres cultures industrielles
Divers PPAM	VER	Véronique	25	Légumes ou fleurs
Houblon	HBL	Houblon	24	Autres cultures industrielles
Lavande	LAV	Lavande / Lavandin	24	Autres cultures industrielles
Légumes feuilles	CES	Chicorée / Endive / Scarole	25	Légumes ou fleurs
Légumes feuilles	EPI	Épinard	25	Légumes ou fleurs
Légumes feuilles	LBF	Laitue / Batavia / Feuille de chêne	25	Légumes ou fleurs
Légumes feuilles	LSA	Légume sous abri	25	Légumes ou fleurs
Légumes feuilles	MAC	Mâche	25	Légumes ou fleurs
Légumes fruits	TOT	Tomate pour transformation	24	Autres cultures industrielles
Légumes fruits	CCN	Concombre / Cornichon	25	Légumes ou fleurs
Légumes fruits	CCT	Courgette / Citrouille	25	Légumes ou fleurs
Légumes fruits	CMB	Courge musquée / Butternut	25	Légumes ou fleurs
Légumes fruits	HAR	Haricot / Flageolet	25	Légumes ou fleurs
Légumes fruits	MLO	Melon	25	Légumes ou fleurs
Légumes fruits	PPO	Petits pois	25	Légumes ou fleurs
Légumes fruits	TOM	Tomate	25	Légumes ou fleurs
Légumes racines	CAR	Carotte	25	Légumes ou fleurs
Légumes racines	CEL	Céleri	25	Légumes ou fleurs
Légumes racines	NVT	Navet	25	Légumes ou fleurs
Légumes racines	PAN	Panais	25	Légumes ou fleurs
Légumes racines	RDI	Radis	25	Légumes ou fleurs
Légumes racines	SFI	Salsifis	25	Légumes ou fleurs
Légumes racines	TOP	Topinambour	25	Légumes ou fleurs
Légumes tiges	ART	Artichaut	25	Légumes ou fleurs
Légumes tiges	CHU	Chou	25	Légumes ou fleurs
Légumes tiges	POR	Poireau	25	Légumes ou fleurs
Maraîchage sous serre et abri	CSS	Culture sous serre hors sol	25	Légumes ou fleurs
Moutarde	MOT	Moutarde	24	Autres cultures industrielles

Romarin	ROM	Romarin	24	Autres cultures industrielles
Tabac	TAB	Tabac	24	Autres cultures industrielles

Appendix B

Influence of algorithm prioritizing more or less populated areas

Changing the algorithm priority (see Section 6.1.3), this time from most populated to least populated areas (Figure B.1 below), mainly affects the distribution tails in each population density category (compared to initial Figure 6.17 in Chapter 6). In the *current situation* scenario, median values are shifted by about 70%, but since the distributions span 4 orders of magnitude, this does not change the general trend that densely populated areas generally need way higher distances than less populated areas.

For areas with less than 1,000 inhabitants/km², the vast majority of distances are still less than 10 km. The main notable difference is that there is much more overlap between the 2 middle categories distributions (100-1,000 and 1,000-10,000 inhabitants/km²).

The total tons·km are barely changed between the 2 cases: 3.7 Mtons·km in the first case vs 3.9 Mtons·km in the second.

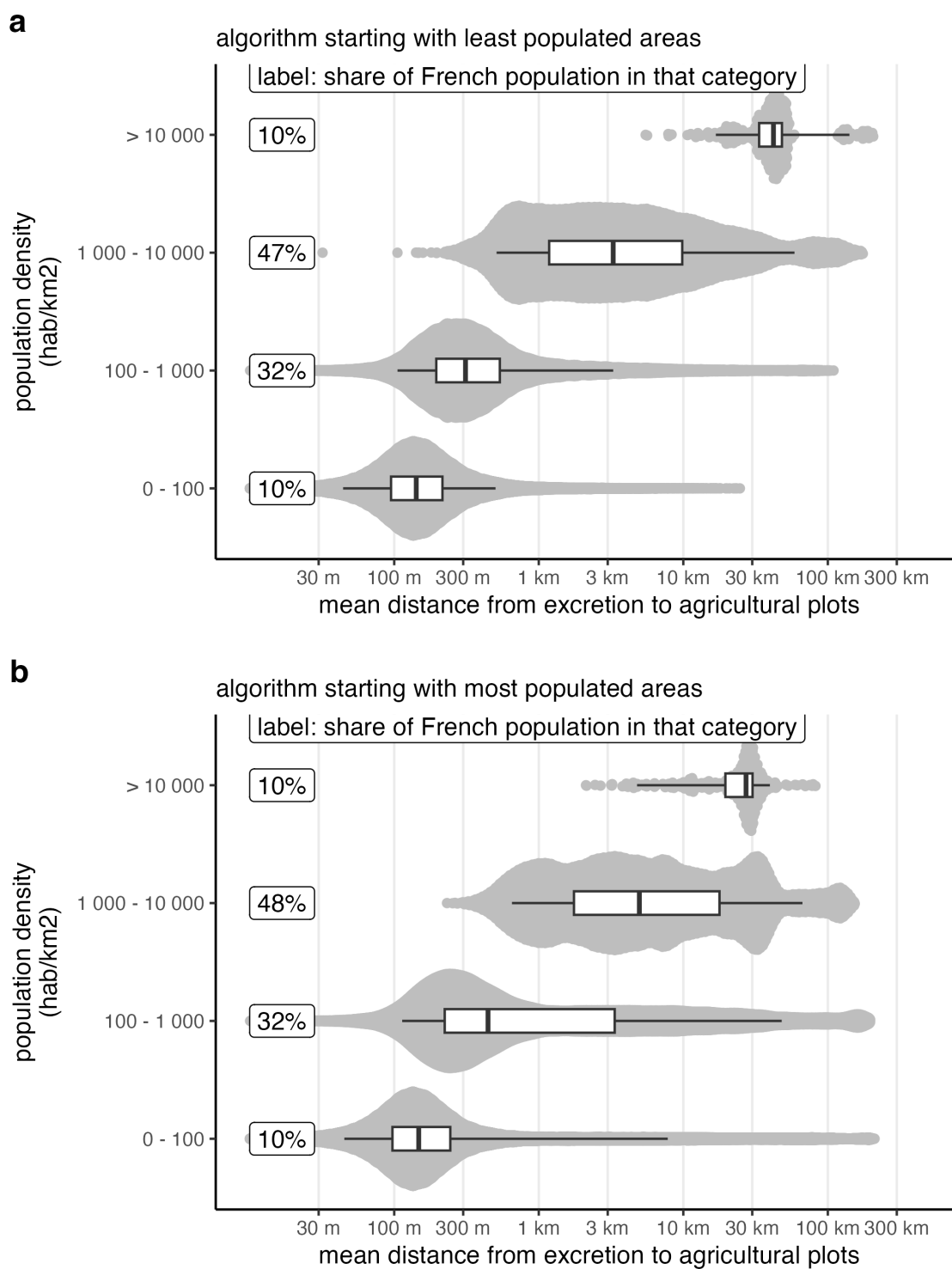


Figure B.1: Effect of changing the algorithm to evacuate human excretions to surrounding agricultural parcels for the *current situation* scenario. Top: starting allocation with least populated areas/ Bottom: starting allocation with most populated areas. Each dot is a 1 km² pixel of human excretions. On the x-axis, the distance is the mean distance from all the agricultural parcels attributed to the concerned pixel of human excretions. Distances refer to straight-line distance, as the crow flies, and not an actual distance that would be covered by a truck constrained by the presence of a road. Boxplots represent the following percentiles: 5, 25, 50, 75 and 95%.