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# Trace contaminants in the environmental assessment of organic waste recycling in agriculture: gaps between methods and knowledge

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## Abstract

Agricultural recycling of organic waste (OW) derived from urban, agricultural and agroindustrial sources is an essential sustainable development strategy. Yet repeated application of nutrient-laden OW in crop fields can also drastically boost contaminant levels in soil. This review focuses on the consideration of three categories of OW-borne contaminants, namely trace elements, organic contaminants and pathogens (including antibiotic resistance), in environmental assessments, chiefly involving life cycle assessment (LCA) and risk assessment (RA). The in-depth discussion also focuses on gaps between empirical knowledge and the models underlying these frameworks. Potential improvements to fill the identified gaps are proposed, including novel approaches and uses of existing approaches, while also featuring various levels of 'readiness'. Finally, a comprehensive theoretical framework to assess OW recycling scenarios, combining complementary approaches and models, is proposed and exemplified.

**Keywords:** antibiotic resistance; LCA; modelling; organic contaminants; pathogens; REACH; risk assessment; soil amendment; trace elements; USEtox

# 1 Introduction

## 1.1 Treatment and agricultural recycling of organic waste

There is a global consensus among scientists, economists, politicians and civil society on the need to recycle resources, engage in industrial symbioses whereby wastes are transformed into resources, while closing material loops in circular economies as a pathway to sustainable development (Costa et al., 2010; Frosch and Gallopoulos, 1989; Ghisellini et al., 2014; Gontard et al., 2018). The waste-agriculture nexus is particularly relevant in this context (Kurian and Ardakanian, 2015), especially from the bioeconomy perspective (EC, 2012; Loiseau et al., 2016). First, the ever-increasing waste production pattern has prompted the need for more sustainable waste management, from economic, environmental and human health standpoints (Morrissey and Browne, 2004; Singh et al., 2014) and secondly, the rising food demand is exerting additional pressure on agriculture and other food production systems that are expected to feed the growing population in more sustainable ways despite increased resource constraints (EC, 2011; Moreau et al., 2012). Both concerns could —and perhaps should!— be jointly addressed , but they are often studied separately. When addressed alone, waste management is generally viewed as a costly disposal activity, whereas agriculture consumes large quantities of finite resources while generating environmental burdens (e.g. emissions).

Agricultural recycling can allow for effective synergistic use of **organic waste** (OW), i.e. any organic biogenic waste (residue) derived from urban, agricultural, and agroindustrial sources, including crop residue, animal effluents, agroindustrial residue, landscaping residue, organic fraction of municipal solid waste, and sewage sludge (Avadí, 2020). These OWs contain nutrients that could become substitutes for mineral fertilisers and thus contribute to soil fertility. OWs also contain organic matter whose application on soils could contribute to increasing soil organic matter contents and help mitigate climate change —a mechanism that is currently being put forward to cope with this key concern (Minasny et al., 2017). These OWs are applied to agricultural soils in raw form or after processing for stabilisation, volume reduction, hygienisation, etc. Aerobic (composting) and

anaerobic digestion are the most obvious operational processes for OW treatment prior to soil application from a waste management perspective (see Figure 1 for OW, treatments and recycling pathways considered in this study). Source separation of the organic fraction of municipal waste followed by composting is considered to be an effective method for diverting organic material from landfills and incineration, while reducing the waste volume, eliminating pathogens and creating a stable product suitable for crop field application. The use of anaerobic digestion has also significantly increased in recent decades in several European countries, while representing an opportunity to convert OW into biogas and organic fertiliser (digestate). Both treatments have been found to produce stable fertilising materials by effectively retaining macronutrients and converting certain nutrients to phytoavailable forms (Houot et al., 2014). However, it is essential to take the organic, biologic and inorganic contaminants contained in waste and their environmental and human health impacts into account when studying the waste-agriculture nexus (Houot et al., 2014).

[Figure 1]

## **1.2 Contaminants in organic waste and effects of its agricultural recycling on soil**

Many studies have highlighted that repeated OW application in crop fields can drastically boost contaminant levels in soil, regardless of their origin (urban, agricultural, etc.), the type of OW (raw, digestate or compost) or the pedoclimatic conditions (Achiba et al., 2009; Börjesson et al., 2015; Formentini et al., 2015; Roig et al., 2012; Udom et al., 2004). Moreover, the contaminants borne by this raw or treated OW may have potential impacts on ecosystem functions and ultimately on human health.

Several national monitoring programmes have revealed that **trace elements**<sup>1</sup> are quantitatively the main type of contaminants spread on agricultural soils (Benoît et al., 2014; Senesi et al., 1999). Mean inputs typically range from 2 g ha<sup>-1</sup> year<sup>-1</sup> for Cd to 1 500 g ha<sup>-1</sup> year<sup>-1</sup> for Zn (Belon et al., 2012; Eckel et al., 2005; Luo et al., 2009; Nicholson et al., 2003). While atmospheric deposition is the major source of trace elements in the most industrialized countries, OW remains a major, if not the main

source of trace elements in agricultural soils. Zn and Cu input is typically —by several orders of magnitude— the highest among trace elements. This is particularly due to agricultural recycling of livestock effluents that exhibit high concentrations in Cu and Zn, two animal feed supplements. OW transformation by anaerobic digestion or composting usually tends to increase the total trace element concentration in OW due to organic matter mineralization and/or liquid phase elimination (Bożym and Siemiątkowski, 2018; Gusiatin and Kulikowska, 2014; Knoop et al., 2017). Accordingly, Kupper et al. (2014) recently calculated that fertilization with compost or digestates results in higher trace element loads than application of equivalent nutrient inputs through raw OW. It has also been found that dilution of the trace element concentration following anaerobic digestion or composting may occur when organic matter degradation during the thermophilic phase prompts the release of leachates containing some trace element complexes (Amir et al., 2005), or when substantial amounts of solid organic substrate containing lower trace element concentrations than in the initial raw OW are added to enable liquid OW composting (e.g. animal slurry and sewage sludge). Trace elements are usually: i) more concentrated in OW than in most soils, ii) not biodegradable, and iii) strongly bound to the soil solid-phase. Hence these substances tend to almost irreversibly to gradually accumulate in topsoil, with a residence time in soil typically much longer than 100 years (Benoît et al., 2014; Senesi et al., 1999). Cd, Cu, and Zn accumulation in agricultural soils is particularly worrisome since the estimated time needed to increase the soil concentration of these substances from the natural background level to regulatory limits is the shortest among all trace elements, while OW contributes substantially to this incremental pattern (Luo et al., 2009; Nicholson et al., 2003). Most of risk assessment methodologies point out trace elements as the type of contaminants that dominates the impacts on human health and, even more so, on aquatic and terrestrial ecotoxicity (Pettersen and Hertwich, 2008; Pizzol et al., 2011a, 2011b; Tarpani et al., 2020). The robustness of these methodologies has, however, been questioned as they do not account for the speciation and bioavailability which are essential factors in determining the toxicological and ecotoxicological impacts of trace elements (Plouffe et al., 2016, 2015a, 2015b; Sydow et al., 2020). It would therefore

be essential to develop robust risk assessment methodologies specifically focused on the input of trace elements from raw and transformed OW in agricultural soils to ensure the sustainability of agricultural recycling of OW.

**Organic contaminants** have been documented in a diverse range of OWs, and OW recycling is a possible gateway for these contaminants in agricultural soils (Hargreaves et al., 2008; Verlicchi and Zambello, 2015) and connected ecosystems (Balderacchi et al., 2013). An extensive review of the available data has nevertheless revealed that knowledge on this complex issue is highly heterogeneous depending on the origin and type of OW, as well as on the class of organic pollutant targeted (Houot et al., 2014). Contrary to trace elements, waste treatment involving composting or anaerobic digestion can partly reduce organic pollutant concentrations via dilution (mixtures with co-substrates), leaching (during composting), transformation (biotic or abiotic), volatilization or non-extractable residue formation. The extent of involvement of these processes largely depends on the type of treatment and on-site conditions. Yet these treatments seldom fully eliminate the contamination. In particular, transformation products and non-extractable residues are usually formed during OW decomposition and biodegradation (Benoît et al., 2014). Following OW spreading onto soil, organic compounds not removed during OW treatment can undergo physicochemical and biological processes that may alter their chemical forms and availability. These compounds may then pose a risk to humans upon their transfer to crops, water or air and to the environment via their impacts on terrestrial and aquatic organisms (Langdon et al., 2010; Thomaidi et al., 2016). Most available data concern organic contaminants in sewage sludge monitored for regulatory purposes, such as pesticides, polyaromatic hydrocarbons (PAH) and polychlorinated biphenyls (PCB). Data are also available on other yet to be regulated compounds, including nonylphenol ethoxylates, linear alkylbenzene sulfonate, polychlorinated dibenzodioxins, furans and bis(2-ethylhexyl) phthalate (Olofsson et al., 2012). Over the last 10 years, pharmaceuticals and personal care products (PPCP) have drawn greater attention due to their potential impacts on human health (Clarke and Smith, 2011; Sarmah et al., 2006). However, few field studies have focused on the fate of these PCPP in the agricultural OW recycling setting. OW

treatment influences the concentration and availability of organic contaminants in soil, their persistence and leaching but the impacts of these substances largely depend on their chemical class (Bourdat-Deschamps et al., 2017). Scant data is available on the transfer of many different organic contaminants and OW to plants, although some plant uptake and transport models have been designed for ionic chemicals, for instance (Trapp, 2004).

OWs can be vectors for the transmission of all categories of infectious agents, **pathogens**, ranging from viruses to parasites. Enteroviruses, adenoviruses, polyomaviruses, astroviruses, noroviruses and hepatitis A and E viruses have, for instance, been detected in raw and treated biosolids (Bofill-Mas et al., 2006; Chapron et al., 2000; Laverick et al., 2004; Sidhu and Toze, 2009). Biosolids from wastewater treatment plants have been shown to be major vectors of human enteric viruses (USDA - NASS, 2012) and other wastewater treatment plant-related pathogens, including standard waterborne pathogens, e.g. *Cryptosporidium*, *Salmonella* and *Pseudomonas aeruginosa* (Lagriffoul et al., 2009). Higher numbers of pathogens have also been reported in digestates and composts (Fradkin et al., 1989; Nell et al., 1983). Similarly, but to a lesser extent, livestock manure can also substantially contribute to the transmission of human infectious agents (so-called zoonotic agents). The main zoonotic microorganisms are *Salmonella enterica*, *Campylobacter jejuni*, *Listeria monocytogenes*, *Staphylococcus aureus*, the Shiga toxin-producing *Escherichia coli* and *Coxiella burnetii* (Q fever) and are involved in disease outbreaks. The microbiological safety of OW treatment products (i.e. digestate and compost) is controversial and hard to assess because of significant differences between the technologies used and the varied sanitary quality of the waste prior to treatment. It would be essential to gain insight into the number of pathogens in the initial OW —as reviewed for instance in Zhao and Liu (2019)— to help select appropriate treatments for the defined end uses. However, this would be difficult to achieve because of the high cost of such microbial source tracking analyses and the lack of background regarding the pathogens potentially found in these OW. Indicator bacteria such as *E. coli* and intestinal *Enterococci* are generally used to monitor the microbiological quality of digestate. Several knowledge gaps would need to be filled to be able to

predict the survival and fate of pathogens in digestate applied on agricultural soils. In fact, pathogens may survive in the environment for days to months, and their transport can occur naturally via wild uncontrollable animals or by insect or worm movements (Moore and Gross, 2010). Pathogen die-off in soils also depends on a number of factors, including the applied OW-bound nutrients, microbial species, and soil parameters such as moisture and pH (Girardin et al., 2005; Lepeuple et al., 2004; Pourcher et al., 2007). Indeed, no cultivable forms of *Salmonella* or *E. coli* were detected a month after manure and digestate application on soils, whereas the *Listeria* count in soil was only reduced by one order of magnitude 3 months after digestate application (Goberna et al., 2011). *E. coli* survival periods of more than 99 days in bovine manure amended pastures have nevertheless been reported for pathogenic serogroups (Bolton et al., 1999). These die-off datasets thus appear to depend upon the nature of the *E. coli* phylogroup considered. Microbial competition with the native soil microbiota is likely a key factor driving the survival of such pathogens in soils (Goberna et al., 2011), as also is predation. Microbial competition between native bacteria and pathogens generally occurs through antimicrobial production, and chelator-mediated nutrient deprivation (Buchanan and Bagi, 1999; Galia et al., 2017; Goberna et al., 2011; Haas and Défago, 2005; Sidhu et al., 2001). Unc and Goss (2004) emphasized that digestate application on soils can have negative environmental effects even when it has a low pathogen concentration, i.e. given its low solid content, any digestate-bound microbe could have higher mobility and thereby more readily colonize deeper soil layers and groundwater. Agricultural runoff may carry and transfer human pathogens from amended sites to water bodies (Barbarick and Ippolito, 2007; Bibby et al., 2011; Palmer et al., 2005).

Over 70% of antibiotics consumed by humans or animals are excreted un-metabolized (Kümmerer, 2009a, 2009b) and enter wastewater treatment plants or slurry lagoons —this can create suitable environments for resistance development and horizontal gene transfer of resistance between bacteria (O'Neill, 2016). Similarly, around 30–90% of antibiotics used for animal production end up in manure (Sarmah et al., 2006) which, when applied as fertilizer in fields, can potentially lead to antibiotic contamination of the soils (Fahrenfeld et al., 2014; Hou et al., 2015; Qiao et al., 2018; Sun

et al., 2018). **Antibiotic resistant bacteria (ARB), antibiotic resistance genes (ARG) and/or mobile genetic elements (MGE)** are also abundant in animal and human faecal materials that end up in agricultural and urban waste treatment systems. These systems are designed to reduce total faecal organism numbers in effluent before discharge into the environment, but they are not specifically designed for ARB, ARG and MGE removal. Wastewater treatment plants have been identified as potential hotspots for resistance development and ARG transmission between pathogenic and non-pathogenic bacteria. Indeed, bacteria from different sources (municipalities, hospitals and industries) are present at high density and in close contact during the purification process —this includes a diverse range of species across bacterial phyla. Many studies aimed at determining the extent of resistome changes throughout the treatment process have involved comparisons of influent and effluent contents. The conclusions of a meta-analysis (Harris et al., 2012) indicated that wastewater treatment led to a reduction in the total bacterial count but an increase in the ARB percentage, thereby highlighting that resistance-oriented selection pressure may prevail in wastewater treatment plants. Recently, Goulas et al. (2020) documented the extent of ARB, ARG and MGE reduction possible following wastewater processing in treatment plants, yet these markers were still found in both treated effluents and sewage sludge. Youngquist et al. (2016) compiled data on the effects of composting and anaerobic digestion on ARB and ARG persistence and reported that “some ARB and ARGs persist during mesophilic anaerobic digestion”, and that “thermophilic treatments are more effective at decreasing ARB and ARGs”. They observed more inconsistent results regarding composting as some studies even revealed increased ARG levels after treatment. Goulas et al. (2020) showed that treatments could decrease ARB amounts, and variations in the observed reduction rates could possibly be related to differences in initial ARB abundances between studies. They also reported a significant effect on ARG/MGE relative abundance during composting. Treated or not and irrespective of the treatment process, OW are a source of ARB, ARG and MGE when applied on soils. Most studies on OW-amended soils have focused on animal manure, generally investigating ARG and MGE persistence in soil and potential dissemination in vegetables. They were conducted at various

scales (microcosms to experimental fields) and targeted antibiotic resistance markers (Chee-Sanford et al., 2009; Ghosh and Lapara, 2007; Heuer et al., 2011; Ji et al., 2012; Munir and Xagorarakis, 2011; Muurinen et al., 2017). Manure (Graham et al., 2016; Heuer et al., 2011; Heuer and Smalla, 2007; Marti et al., 2013) or biosolid (Rahube et al., 2014) fertilization modifies the soil resistome, and increases in ARB and ARG levels in agricultural soils have been detected shortly after spreading and after repeated long-term applications. Antibiotic resistance may trickle down the food chain. Recent studies revealed that ARG were more abundant in vegetables harvested in soil fertilized with manure (Marti et al., 2013; Tien et al., 2017) or sewage sludge (Rahube et al., 2014) as compared to control soil. Human or animal consumption of these contaminated vegetables represents a potential route of ARG exposure (Zhu et al., 2017).

### **1.3 Scope and objectives of this review**

The advantages of using raw OW materials, compost or digestate as fertilizer and soil amendment need to be assessed alongside the potential environmental and toxicological impacts due to the presence of contaminants. This review focuses on three categories of contaminants —trace elements, organic contaminants and pathogens (including antibiotic resistance<sup>ii</sup>)— regarding their consideration in environmental assessments. In this review, ‘environmental assessment’ is broadly understood as *ex ante* (thus model-based) estimation of environmental impacts and risk indicators for decision support. This review thus aims to identify: 1) limitations in existing environmental assessment frameworks for agricultural recycling of OW, regarding the absence or inadequate consideration of some biophysical processes governing the fate of contaminants; and 2) suitable approaches for assessing contaminants involved in agricultural recycling of OW, such as complementary analytics, methods, and models. This review seeks to fulfil its objectives by highlighting the state of the art with regard to the consideration of contaminants in environmental assessment frameworks applied to OW treatment, products and their recycling in agriculture (section 2), followed by a discussion on gaps between empirical knowledge and the two most widely used environmental assessment frameworks, namely life cycle assessment (LCA) and risk assessment (RA),

including potential improvements (section 3). The state of the art is presented per framework and per type of contaminant. In the LCA context, the state of the art focuses on trace elements (section 2.3), because toxicity modelling in LCA does not yet consider pathogens, and terrestrial ecotoxicity models are not yet available for organic contaminants. The discussion on limitations of and potential improvements to assessment frameworks is also organized per type of contaminant (section 3), as approaches, methods and models are generally contaminant-specific for both frameworks.

## **2 Environmental assessment of organic waste treatment and agricultural recycling – state of the art: consideration of contaminants**

### **2.1 Environmental assessment frameworks**

Various environmental assessment frameworks have been applied to OW treatments and agricultural recycling of treated and raw OW. They are either procedural (focused on societal and decision-making aspects) or analytical (focused on technical aspects), according to the classification of Finnveden and Moberg (2005). Table 1 briefly defines the most common ones, indicates their usual application level (i.e. system size), and their main associated pros and cons. Among these frameworks, material and substance flow analysis (an accounting approach to flows and stocks of materials, often in a regional system) considers contaminants only in terms of flows and stocks. Other frameworks, such as environmental impact assessment (a project analysis toolbox), exergy (an indicator of useful energy) and emergy (a measure of quality differences among different energy forms) analyses, eco-efficiency analysis (ratio between socioeconomic benefits and environmental burdens) and strategic environmental assessment (policy analysis toolbox), often do not explicitly consider specific contaminants (Allesch and Brunner, 2015, 2014). Only **life cycle assessment (LCA)** and **risk assessment (RA)** consider contaminants explicitly in terms of their impacts on human and/or ecosystem health, via toxicity impact categories.

[Table 1]

LCA (sections 2.2 and 2.3) is an accounting framework geared towards documenting all resource consumption and emissions associated with the provision of goods or services, throughout the whole life cycle from raw material extraction to the construction, use, maintenance and end of life/final disposal of production means. LCA is formalized by an ISO standard (ISO, 2006a), and several guidelines are focused on its theory and practice (EC-JRC, 2010; EC, 2018). LCA estimates impacts under a range of impact categories, including human toxicity and environmental ecotoxicity. Human toxicity is often split into cancer and non-cancer effects, while ecotoxicity is further subdivided into freshwater aquatic, marine and terrestrial ecotoxicity. Toxicity assessment in LCA deals with huge numbers of substances in minimal detail, which results in high model and data uncertainty regarding environmental mechanisms and characterisation factors<sup>iii</sup> (Fantke et al., 2018; Rosenbaum et al., 2013).

RA (sections 2.4 and 2.5) is mainly a human health-related framework while also featuring ecological/environmental elements, such as the set of models used in ecological risk assessment (ERA) (Roast et al., 2007). In practice RA involves mandatory procedures, usually published by authorities, such as the different technical guidelines issued by the European Union and the United States Environmental Protection Agency. The two main branches of RA focus on ecological and human health risks. A variety of models and empirical tests are used in RA for identification and characterisation of (mainly toxicity-related) hazards, dose responses, exposure assessment and risk characterization of chemicals, as comprehensively reviewed in the literature (Roast et al., 2007; Thoeue et al., 2003; WHO, 2010). RA addresses a very limited number of substances in detail.

LCA and RA are complementary methods (Flemström et al., 2004; R. Rosenbaum, pers. comm.), i.e. the first is comprehensive but not detailed whereas the second is the opposite.

**Combined LCA and RA** for joint ecological and human health assessment is a promising option for agriculture and food production research (Jolliet et al., 2014; Notarnicola et al., 2017; Tukker et al., 2011), while providing a means to inform robust multicriteria decision-making (Huang et al., 2011; Linkov and Seager, 2011). Various studies have, for instance, attempted to integrate exposure RA

with LCA of wastewater treatments on the basis that both frameworks yield endpoint indicators on human health (Harder et al., 2016, 2015, 2014; Heimersson et al., 2014; Jolliet et al., 2014). The scope of such joint assessments could encompass OW treatment and agricultural use. Combined RA-LCA studies, despite their fundamental differences regarding aims and conceptual background (Olsen et al., 2001), often express chemical and pathogen exposure risk in terms of DALY (Harder et al., 2015, 2014), i.e. a common unit to express impacts on human health (Gao et al., 2015; Kobayashi et al., 2015).

Various studies have also advocated and demonstrated the efficacy of material flow analysis and substance flow analysis for tracking different trace contaminants (e.g. trace elements) through waste management systems, in addition to the modelling of downstream processes (e.g. agricultural use) (Laurent et al., 2014a, 2014b; Wassenaar et al., 2015). Combined substance flow analysis and LCA has also been put forward as a means for mapping pollutant flows in the urban environment (Arena and Di Gregorio, 2014; Azapagic et al., 2007), which could also be tailored to agricultural recycling scenarios.

Table 2 lists representative reviews on environmental, life cycle and risk assessments regarding OW treatment and the impacts of contaminants.

[Table 2]

Toxicity models used in LCA are usually based on RA, yet they differ in terms of metrics (e.g. health impacts vs. chemical concentrations), scope (all emissions from global life cycles vs. all local emissions including background concentrations), parameterization (e.g. thousands vs under 10 chemicals), modelling choices (average vs. worst case), compartments (e.g. sediment) and mechanisms (Table 3). These modelling principles, illustrated with examples of their application for OW-borne contaminant assessment, are outlined in the following subsections.

[Table 3]

## 2.2 Toxicity modelling in LCA

### 2.2.1 General principles

The LCA framework consists of four phases (ISO, 2006b, 2006a): 1) goal and scope, where the objectives and limits of the study are decided, including the system boundaries and other fundamental choices; 2) life cycle inventory (LCI), where all data regarding resource consumption and direct emissions from the studied system are compiled; 3) life cycle impact assessment (LCIA), where all LCI factors are multiplied by characterization factors so as to express them in a common unit per impact category, i.e. where the potential impacts are estimated; and 4) interpretation, where the quantitative results of both LCI and LCIA are interpreted and explained, and environmental hotspots of the process are identified.

LCA involves the use of toxicity models to assess the toxic effects of substances on human health and both aquatic and terrestrial ecosystems. Note that in most models terrestrial and marine ecotoxicity data considered in LCA are generally extrapolated from freshwater ecotoxicity data, thereby increasing the uncertainty associated with toxicity modelling. It has been pointed out that differences among LCIA methods regarding toxicity impacts are related to the characterization phase (e.g. the inclusion or not of specific substances, the type of multimedia model<sup>iv</sup>) (Geisler et al., 2005; Pizzol et al., 2011a, 2011b). None of these toxicity models take the combination of trace elements, organic contaminants and pathogens in OW into account, or all of the dynamics determining their fate following their soil application. Their uncertainty is high regarding characterization factors, which are usually based on total concentrations and are not regionalized. Moreover, many substances potentially present in OW —notably PAH, hydrocarbons and non-methane volatile organic compounds— lack characterization factors, which are only available across models for a limited set of substances out of the >30 000 most frequently used chemicals (Fantke (Ed.) et al., 2017; Huijbregts, 2000; Saouter et al., 2017). Moreover, these models disregard contaminant transformation, degradation and combination products after emission and within the period taken into account in the studies, i.e. usually a year and seldom longer. Lastly, toxicity models developed to

date do not assess pathogens, genetic material (e.g. antimicrobial resistant genes), or other forms of contamination, such as toxins and particulate matter, that negatively impact health.

The Uniform System for the Evaluation of Substances adapted for Life Cycle Assessment (USES-LCA, current version 2.0), is a global nested multimedia fate, exposure, and effects model to calculate characterization factors for terrestrial, freshwater and marine ecotoxicity, as well as for human toxicity, on both midpoint and endpoint levels (Van Zelm et al., 2009). It is based on empirical equations (Guinée et al., 2002; Huijbregts et al., 2000) and a class of risk assessment models for EU applications, i.e. the European Union System for the Evaluation of Substances (EUSES) toolkit, a harmonized quantitative risk assessment tool to calculate predicted environmental concentrations of chemical substances (Vermeire et al., 2005). It currently has a database of 3 396 (organic and inorganic) chemicals, with 10 emission compartments, including urban air, rural air, freshwater and agricultural soil. Based on three scales, i.e. regional (original USES-LCA only), continental and global, it calculates human-toxicological effect and damage factors per chemical with information related to intake route (inhalation and ingestion) and disease type (cancer and non-cancer). Moreover, it calculates endpoint ecotoxicological effect factors expressed in terms of changes in overall toxic pressure due to chemical concentration changes. These ecotoxicological effect factors consist of a slope factor and a chemical-specific toxic potency factor reflecting the average environmental toxicity of given chemicals (based on single species toxicity data).

The Tool for the Reduction and Assessment of Chemical and other environmental Impacts (TRACI, current version 2.0), is an LCIA method featuring toxicity models for human health cancer, non-cancer, and ecotoxicity (Bare, 2011). TRACI uses the CalTOX toxicity model for these impact categories (CalTOX is a collection of multimedia transport and transformation, exposure scenario and multiple-pathway exposure models incorporating uncertainty and population-distribution data (Mckone, 1993)). For toxicity, TRACI only characterizes chemicals included in the US EPA Toxics Release Inventory (<https://www.epa.gov/toxics-release-inventory-tri-program/tri-listed-chemicals>).

The EDIP programme and the Danish LCA Methodology Development and Consensus Creation Project, produced two LCIA methods, respectively EDIP97 and EDIP2003. The latter is an update of the former regarding the extent to which the characterization factor modelling takes the causality chain into account for all non-global impact categories: EDIP97 encompasses emissions, their fate and degradation, while EDIP2003 includes exposure, impacts and damage (Hauschild and Potting, 2005). Characterization factors in EDIP2003 are spatially resolved at the country level. EDIP includes human and ecotoxicity impact categories, whose characterization factors are based on 'some-fate' modelling rather than on multimedia 'full-fate' modelling. For human toxicity, EDIP97 characterization factors express "the volume of environmental compartment (air, water, soil) which can be polluted up to the human reference concentration or dose, the level not expected to cause effects on lifelong exposure" ( $\text{m}^3 \cdot \text{g}^{-1}$ ), while EDIP2003 characterization factors represent "the reciprocal of a fate-corrected human reference dose or concentration". Legacy models such as EcoSense and empirical equations are used to calculate characterization factors. Otherwise, ecotoxicity is assessed in aquatic and terrestrial ecosystems, as well as in wastewater treatment plants, through "simplified fate modelling based on a modular approach where redistribution between the environmental compartments and potential for biodegradation are represented as separate factors". Characterized substances include those compiled in the Danish Environmental Protection Agency List of Undesirable Substances and List of Effects.

IMPACT 2002+ is an LCIA method featuring damage factors, including human toxicity and ecotoxicity impact categories that take intake fractions, best estimates of dose-response slope factors, as well as severities, into account (Jolliet et al., 2003). IMPACT 2002 (without the '+', also known as Impact Assessment of Chemical Toxics) models risks and potential impacts per emission for several thousand chemicals. It proposes generic characterization factors for Western Europe and spatially explicit characterization factors for 50 catchments and air cells in Europe based on fate, exposure, dose-response and severity modelling (via models and empirical equations).

USEtox, current version 2.0 (Fantke (Ed.) et al., 2017), is a scientific consensus toxicity model and collection of human toxicity and aquatic freshwater ecotoxicity characterization factors developed under the auspices of the United Nations Environment Programme (UNEP) and the Society for Environmental Toxicology and Chemistry (SETAC) Life Cycle Initiative (Rosenbaum et al., 2008). The consensus process culminating in the development of USEtox included the developers of previous CalTOX, IMPACT 2002, USES-LCA, BETR, EDIP97, WATSON and EcoSense models. It was based on a quantitative comparison of these models on 45 organic substances, and on their underlying fate, exposure and effect modelling (Hauschild et al., 2008; Rosenbaum et al., 2008). The consensus-building process was carried out between 2003 and 2008 (Hauschild et al., 2008), partially based on previous toxicity assessment harmonization initiatives (De Koning et al., 2002; Molander et al., 2004). USEtox development and refinement is ongoing and, for instance, the model was recently implemented in the LC-IMPACT regionalized life cycle impact assessment method (Verones et al., 2020).

### **2.2.2 USEtox: the consensus fate, exposure and toxicity model**

USEtox features over 29 000 characterization factors for over 3 000 substances emitted in all ecosystem compartments, including trace elements (Ag, As, As, Ba, Be, Cd, Co, Cr, Cr, Cu, Hg, Mo, Ni, Pb, Sb, Sb, Se, Sn, Tl, V and Zn) and organic contaminants (e.g. alkylbenzene, PCB, nonylphenol and phthalates). Certain substance groups are still excluded, namely PAH, hydrocarbons, non-methane volatile organic compounds and particulate matter, because the model is best equipped for modelling “non-dissociating and non-amphiphilic organic substances” (Fantke (Ed.) et al., 2017). USEtox characterization factors are calculated on the basis of the cause-effect chain linking emissions to impacts through environmental fate, exposure and effects. It uses continental and global scales and relies on chemical property, ecotoxicity impact and physicochemical data, along with environmental multimedia and multi-pathway models to account for environmental fate and exposure processes, as well as impacts (damage). Compartments (air, soil, fresh water, etc.) are modelled as homogenous well-mixed boxes, and the inventory of a contaminant in each box depends

on the concurrent processes determining whether or not it will remain in the box, be transferred to another box (by dispersive and advective cross-media transfers), degraded (by oxidation, etc.) or removed (by leaching or burial). Fate modelling is thus based on a mass balance equation taking these processes into account. Exposure modelling transforms the amount of a chemical found in a given compartment into human intake via direct and indirect (via bioaccumulation in animal tissues) pathways, but this presently excludes intake by dermal contact and dust inhalation. Effect modelling is based on statistics for cancer and non-cancer effects (for human toxicity) and on species sensitivity distribution modelling (for freshwater ecotoxicity). USEtox does not consider speciation, aging and weathering (UNEP, 2019), complexation with organic matter (Weng et al., 2002) and other transformative dynamics of chemicals following emission, yet it does consider environmental conditions (temperature, rain, etc.) during the fate and exposure modelling process (Fantke (Ed.) et al., 2017).

**Human toxicity** characterization factors, expressed in comparative toxic units ( $\text{CTU}_h \cdot \text{kg}^{-1}$ ), represent the estimated increase in morbidity in the total human population per unit mass of an emitted chemical, expressed in  $\text{cases} \cdot \text{kg}^{-1}$  (assuming equal weighting between cancer and non-cancer effects).

**Aquatic ecotoxicity** characterization factors, expressed in comparative toxic units ( $\text{CTU}_e \cdot \text{kg}^{-1}$ ), estimate the potentially affected fraction of species (PAF, see Table 3) integrated over time and volume per unit mass of an emitted chemical ( $\text{PAF} \text{ m}^3 \cdot \text{day} \cdot \text{kg}^{-1}$ ). Freshwater aquatic ecotoxicity is defined as the comparative toxicity potential (CTP, i.e. equivalent to  $\text{CTU}_e$  defined above) of the total concentration of a chemical emitted to any compartment and reaching the freshwater body. USEtox is currently being expanded to include near-field exposure (Fantke et al., 2016; Jolliet et al., 2014) and terrestrial ecotoxicity of trace elements (Owsianiak et al., 2015, 2013; Verones et al., 2020), along with the development of a dynamic version of the model (the current steady-state version features an infinite time horizon) (Fantke et al., 2015). Discussions are also under way regarding developments in aquatic ecotoxicity of effluents, human toxicity of pesticide emissions and higher (warm-blooded) predator ecotoxicity of organic contaminant emissions (Fantke et al., 2018;

Rosenbaum et al., 2013), as well as the integration of REACH (section 2.4) toxicity data (Müller et al., 2017). The emphasis on trace elements (Fantke et al., 2018) and the lack of a terrestrial ecotoxicity model (UNEP, 2019) have been highlighted by the USEtox team. The current (standalone) USEtox model is summarized in Table 3.

### **2.3 Consideration of contaminants in LCA of organic waste treatments and agricultural recycling**

The impact categories/indicators taken into account in LCA studies on OW and OW treatment usually include the global warming potential (occasionally encompassing carbon storage), acidification potential, eutrophication potential, toxicity, and resource depletion (e.g. energy use), as well as other novel indicators such as water and land use footprints. LCA studies focused on OW treatments only consider the toxicity impacts of contaminants (trace elements, organic contaminants) in terms of their absolute quantities emitted over the study/simulation period, as well as their distribution among environmental compartments. OW commonly studied include solid residues from municipal solid waste, food waste and various types of source-separated waste management systems (Bernstad and la Cour Jansen, 2012; Cleary, 2009; Morris et al., 2013), as well as wastewater treatment sludge (Corominas et al., 2013; Lundin et al., 2004; Risch et al., 2015; Suh and Rousseaux, 2002; Teodosiu et al., 2016; Yoshida et al., 2013). For instance, a recent study (Avadí, 2020) compared the main OW treatments used in France for producing organic fertilizers and amendments, and identified the main impact contributors per treatment pathway. Contaminants were not explicitly assessed, as the agricultural recycling of OW treatment products was not a focus.

A few LCA studies on OW have addressed their potential 'recycling' pathways. These studies almost exclusively addressed the benefits of substituting chemical fertilizers by treated OW, or were focused on the contributions of their relative absolute emissions to various impact categories, or on trace element concentrations in treated OW (Bernstad and la Cour Jansen, 2012, 2011; Righi et al., 2013). Various initiatives and tools have been developed specifically to facilitate LCA studies on solid waste management systems and wastewater treatment plants. Among them, EASEWASTE and its successor

the EASETECH LCA model (<http://www.easetech.dk/>) is an assessment tool that is used to an increasing extent in solid waste management LCA studies (Jensen et al., 2016; Yoshida, 2014). For instance, in Boldrin et al. (2011), field application of treated OW is modelled as a function of the compost/digestate composition, crop rotation and soil properties, while taking advantage of the EASEWASTE use-on-land module. The latter is based on an agroecosystem model (Hansen et al., 1991) and field experiments. It includes nutrient uptake and degradation of organic persistent pollutants, various airborne emissions, leaching and runoff of pollutants (trace elements and four organic contaminants: bis(2-ethylhexyl) phthalate, nonylphenol ethoxylates, linear alkylbenzene sulfonate and PAH), as well as carbon sequestration (Hansen et al., 2006a, 2006b). EASEWASTE and EASETECH have been widely used in Danish and German environmental assessments of waste technologies. These include comparisons of different solid-waste treatment technologies (Kirkeby et al., 2006), specific technologies such as composting (Martínez-Blanco et al., 2013, 2009), agricultural recycling of treated OW (Hansen et al., 2006a), uncertainty quantification in LCA of waste management systems (Bisinella et al., 2015; Clavreul et al., 2012), LCA of treatment and recycling of specific feedstock such as biosolids and food waste (Righi et al., 2013; Yoshida, 2014), and LCA of source-separated organics (Morris et al., 2013). EASEWASTE/EASETECH includes the simplified toxicity model described in Hauschild and Wenzel (1998).

### **2.3.1 Organic waste treatment and soil application**

LCA has often been used for environmental assessment of **anaerobic digestion**, focusing on biogas production, substrate transformation (e.g. OW treatment), or both. LCA studies usually focus on the environmental impacts of the process (e.g. Rehl and Müller 2011), while often overlooking contaminants. Hospido et al. (2010) conducted one of the few LCA studies focused on the impact of contaminants associated with OW recycling in soils. The authors assessed the potential impacts of contaminants (trace elements and organic contaminants) on terrestrial ecosystems and their toxicity to humans following agricultural application of undigested sewage sludge and **digestate**. While the fate of pollutants following anaerobic digestion has been experimentally determined (Hospido et al.,

2010), their subsequent fate upon soil application was assessed through highly hypothetical, unvalidated concentration estimates in abstract environmental compartments that mobilize little knowledge of related biophysical processes (a constant feature in all LCA toxicity models. Pivato et al. (2015) empirically assessed the ecotoxicological risk associated with using digestate as a fertilizer using a matrix-based approach that integrated the behaviour of (and interaction among) pollutants in mixtures and within their real (soil) matrix. The authors derived USEtox-based ecotoxicity characterization factors for digestate as a whole. This was a worthwhile attempt to underscore the limits of conventionally independent assessment of individual contaminants, but the validity of such factors is of course limited to situations similar to the empirical reference.

LCA studies on **composting** are less common than on anaerobic digestion, and they often focus on comparisons of alternative composting system scales. A great deal of environmental assessment research has been devoted to investigating the benefits of compost recycling in agriculture (e.g. Martínez-Blanco et al. (2013)), but seldom its impacts due to contaminant transfer to soil. For instance, Saer et al. (2013) assessed nine standard LCA negative impact categories using TRACI, including impacts associated with feedstock collection, compost production and distribution, along with its use as a replacement for peat moss as soil conditioner. Quirós et al. (2014) qualified compost versus mineral fertilizer use based on standard LCA methods. An earlier study by Teglia et al. (2011a, 2011b) also characterized solid digestates for agricultural use after a composting post-treatment. Moreover, LCA has been used to assess different digestate post-treatment technologies, including composting, drying and physicochemical treatments (Rehl and Müller, 2011; Vázquez-Rowe et al., 2015). These authors identified composting as a suitable digestate treatment geared towards agricultural recycling, while discussing the advantages of nutrient recovery via agricultural recycling compared to alternative disposal pathways. Hermann et al. (2011) calculated the carbon and energy footprints of biodegradable waste material treatment, including home and industrial composting, anaerobic digestion and other treatments. None of these studies took contaminants into consideration, which implies that their conclusions were not fully informed.

Laurent et al. (2014a, b) critically reviewed 222 LCA studies on solid waste management systems, including investigations on the post-treatment use (i.e. recycling) of sewage sludge. At least 10% of the studies in this review addressed the issue of terrestrial use of biological treatment outputs. Another review (Zang et al., 2015) of 44 LCA studies on wastewater treatment plants showed that most authors used LCA to compare the relative environmental effects (negative and positive) of alternative treatment technologies and disposal routes. In these studies, impacts associated with trace elements and organic contaminants were only considered via toxicity LCIA methods (Zang et al., 2015), with their inherent limitations (see section 2.2.1). Different LCIA methods (ReCiPe, CML, etc.) and toxicity models (USEtox and USES-LCA) were used, thereby leading to marked discrepancies in toxic impact categories. A recent article (Tarpani et al., 2020) compared several sewage sludge recycling alternatives, while taking the ecotoxicity associated with trace elements and PPCP into account, using the ReCiPe life cycle impact assessment method with USES-LCA as toxicity model. The authors partially assessed speciation of trace elements (i.e. the exchangeable/acid soluble bioavailable fraction). They found that composting gave the lowest resource recovery rates, while anaerobic digestion had the highest freshwater ecotoxicity due to the high trace element, as compared to composted sludge spreading, incineration, pyrolysis and wet air oxidation. No dedicated LCA studies were found regarding agricultural recycling of livestock manure. Manure is included as a fertilizer in agricultural LCAs, but the impacts of its contaminants are seldom investigated, whereas several empirical studies have pointed out that manure is a major source of some trace elements in agricultural soils. Moreover, treated (e.g. stored, composted, digested) manure is more commonly included in LCA studies than raw manure (e.g. Cherubini et al., 2015).

### **2.3.2 Ecotoxicity of OW-borne trace elements**

Considering the major contributions of trace elements to ecotoxicity impact scores in LCA (Pizzol et al., 2011b), substantial improvements in LCA approaches have been achieved over the past two decades concerning trace element impact assessment. Before 2010, all LCA approaches only took the total concentration of trace elements in soil and freshwater into account when assessing their

ecotoxicological impact, which meant that there was a high risk of overestimation (Fairbrother et al., 2007). Indeed, there is considerable evidence that total concentrations are poor indicators of fate, bioavailability and toxicity of trace elements. Accordingly, the UNEP-SETAC working groups reached a consensus, i.e. the so-called Clearwater Consensus, to incorporate trace element speciation<sup>y</sup> and bioavailability for freshwater ecotoxicity assessment in the USEtox LCA methodology (Diamond et al., 2010). The proposed approach includes: a fate factor (FF) corresponding to the total trace element concentration in freshwater; a bioavailability factor (BF) representing the trace element fraction exhibiting a toxic interaction with freshwater organisms, estimated as the concentration of trace elements occurring as free ions and inorganic complexes (e.g.  $\text{CuCl}^+$ ,  $\text{NiCO}_3$ , and  $\text{ZnOH}^+$ ); and an effect factor (EF) corresponding to the PAF of species in freshwater, calculated as the hazardous concentration affecting 50% of species ( $\text{HC}_{50}$ , see Table 3).  $\text{HC}_{50}$  is calculated from the effective concentrations of trace elements at which 50% of a population displays a toxic effect ( $\text{EC}_{50}$ ). PAF is estimated by combining environmental concentrations (from interpolated measurements or model simulations) with field bioavailability estimates (Klepper et al., 1998). The Clearwater Consensus further suggested using the Windermere Humic Aqueous Model (WHAM) (Tipping, 1994), with freshwater chemical properties commonly available in databases (notably pH, dissolved organic carbon (DOC) and water hardness) as input parameters, to estimate the concentration of trace elements, in the form of free ion and inorganic complexes, necessary to calculate BF and EF. Gandhi and Huijbregts (2010) and Dong et al. (2014) developed this approach to calculate generic CTP values for 14 trace elements (i.e. Al(III), Ba, Be, Cd, Co, Cr(III), Cs, Cu, Fe(II), Fe(III), Mn(II), Ni, Pb, Sr, and Zn), and to define seven freshwater archetypes ranked according to their chemical properties (i.e. pH, DOC and water hardness). These new CTP values were similar or slightly higher than the default USEtox-derived CTPs, and in all cases the differences were within 2 orders of magnitude (Dong et al., 2014). Yet when Gandhi and Diamond (2011) applied the WHAM approach to two case studies for Cd, Co, Cu, Ni, Pb and Zn, they found that the contribution of trace element emissions to the overall freshwater ecotoxicity score was one to four orders of magnitude lower than the overall freshwater

ecotoxicity score calculated with default USEtox and USES-LCA. Gandhi et al. (2011) further showed that accounting for the geographic variability in freshwater chemistry was changing both region-specific CTP values for Cu, Ni, and Zn by up to three orders of magnitude and the ranking of CTP values for each trace elements between regions as compared to default USEtox and USES-LCA calculations. More recently, Hedberg et al. (2019) extended the WHAM approach to calculate a CTP value for Cr(VI) and to assess the impact of a temporal change in Cr speciation from Cr(VI) to Cr(III) on the CTP value. While not definitive, the overall results highlighted the need to account for the speciation and bioavailability of trace elements when assessing their freshwater ecotoxicity.

Terrestrial ecotoxicity modelling poses additional challenges with respect to freshwater ecotoxicity, as few toxicity data (especially chronic effects) are available for terrestrial species. Moreover, contrary to freshwater, the solid phase dominates the solution phase and is thereby crucial in the determination of trace element speciation and bioavailability. Recent studies have attempted to include the characterization of terrestrial ecotoxicity of trace elements in USEtox. The model proposed by Owsianiak et al. (2013) includes: a fate factor (FF), calculated as the total concentration of trace elements remaining in soil following their emission; an accessibility factor (ACF), defined as the reactive (i.e. available) fraction of the total trace element concentration in soil that mainly occurs in the solid phase; a bioavailability factor (BF), defined as the free ion fraction in soil solution of the reactive trace element exhibiting an interaction with soil organisms to generate a toxic effect; and a terrestrial ecotoxicity effect factor (EF), as defined for freshwater ecotoxicity. The solid-solution partitioning coefficients used to calculate FF, ACF and BF were estimated from soil physicochemical properties via multilinear regression. EF was estimated using the terrestrial biotic ligand model developed for plants, invertebrates and microorganisms (Thakali et al., 2006a, 2006b). This framework has been applied to estimate the CTP for Cu and Ni in 760 soils worldwide. CTP values for Cu and Ni were found to vary by 3.5 and 3 orders of magnitude over a geographical gradient. CTP values for Cu were mainly affected by the soil organic matter concentration and pH, while those for Ni were mainly impacted by the Mg concentration in soil solution. Owsianiak et al. (2015) further

implemented this framework to account for the different types of emission sources (geogenic versus anthropogenic) and aging. While emission sources significantly affected ACF, thereby suggesting the need to account for emission sources in LCIA, the effect of aging was inconclusive.

Plouffe et al. (2016, 2015a, 2015b) proposed an alternative way to calculate CTP for trace elements based on the Clearwater Consensus. Contrary to Owsianiak et al. (2013) who defined an ACF, the soil solid-phase is not specifically accounted for in this approach. BF is calculated from the total soil trace element concentration and WHAM, with soil properties commonly available in databases (i.e. pH, texture, organic matter and carbonate concentration, and cation exchange capacity) as input parameters. BF values are calculated as the total concentration of trace elements in soil solution or as the concentration of trace elements in soil solution in the form of free ions and inorganic complexes. Note that the Clearwater Consensus considers the latter as being the best indicator of trace element species bioavailable for organisms. The ability of the WHAM approach to fit experimental data was assessed for Zn on 80 soil samples and the findings were compared with the multilinear regression-based approach developed by Owsianiak et al. (2013). The results suggest that the WHAM and regression approaches predict BF values similarly based on the total Zn concentration in soil solution, and that the WHAM approach poorly predicts BF values based on the free and inorganic Zn complex concentration. The rankings of BF values calculated with WHAM and measured experimentally were moderately, but significantly, correlated, whereas those calculated with multilinear regressions and measured experimentally were not significantly correlated. Accordingly, the total trace element concentrations in soil solution simulated with WHAM suggested that this is the most reliable approach to calculate BF (Plouffe et al., 2015a). Note, however, that the extent of calculated BF values that were validated encompassed only 25% of the soil types included in the FAO Harmonized World Soil Database (FAO/IIASA, 2009), therefore underlying the need to further validate the WHAM approach.

Plouffe et al. (2016) also calculated CTP values by combining the aforementioned WHAM-based BF with an FF based on a modified USEtox framework using WHAM to estimate the solid-solution

partitioning coefficient, as well as with an EF based on the total Zn concentration in soil solution. Grid-specific CTP values were first calculated for 5 200 soil samples referenced in the FAO Harmonized World Soil Database, and a global CTP value was finally calculated by summing the product of CTP and population as a proxy of Zn emission in each grid. Interestingly, the resulting global CTP value based on the WHAM approach was 27- and 62-fold lower than the default terrestrial CTP values derived from IMPACT 2002 and USEtox, respectively. Plouffe et al. (2015b) implemented the IMPACT 2002+ method and showed that accounting for trace element speciation with the WHAM-based approach in the LCIA procedure decreased the Zn contribution to the total terrestrial ecotoxicity impact score from 26% in the default approach to less than 2% in the WHAM approach. While the WHAM approach was only validated for Zn, Aziz et al. (2018) and Santos et al. (2018) further implemented the WHAM approach for Cu and Ni terrestrial ecotoxicity assessment. They showed that the grid-specific and global CTP values, as well as the grid-specific and global impact score based on the WHAM approach, were about 3 orders of magnitude lower than the default IMPACT 2002- and USEtox-derived terrestrial CTP values and impact scores. Moreover, Santos et al. (2018) showed —with regard to soil Cu contamination in European vineyards— that increasing the spatial resolution of LCIA implementation decreased the CTP value and impact score variability, which suggests the need for a regionalized assessment of terrestrial ecotoxicity. Concomitantly, these overall results suggest that the contribution of trace elements to the terrestrial ecotoxicity impact score derived from current LCIA approaches is overestimated, thus highlighting the need to further account for trace element speciation and bioavailability. This conclusion was, however, recently challenged by the findings of Sydow et al. (2020) based on a comparison of the speciation-based method developed by Owsianiak et al. (2015, 2013) and the toxicity models implemented in the widely used IMPACT 2002+ and ReCiPe 2008 methods for ~13 000 life cycles of unit processes. They observed that CTP values and concomitant impact scores calculated with the speciation-based method were not systematically markedly lower than those calculated with IMPACT 2002+ or ReCiPe 2008 methods. Sydow et al. (2020) further found that, when accounting for their solid and

solution speciation in soils, trace elements still contributed to more than 90% of the total terrestrial ecotoxicity impact score (with less than 10% due to organic contaminants). These authors thus concluded that increasing the substance coverage of LCIA methods would be as beneficial as increasing their environmental relevance by considering trace element speciation in soils. This conclusion strongly differs from those of Plouffe et al. (2015b), Aziz et al. (2018), and Santos et al. (2018). In the light of the high contribution of trace elements on the total ecotoxicity impact score currently calculated by LCIA methods, there is therefore an urgent need to determine whether the time-consuming effort of developing new speciation-based LCA methods would improve LCIA methods by markedly changing the total ecotoxicity impact score calculation.

Leclerc and Laurent (2017) described a framework to inventory worldwide and some national emissions of trace elements (As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn) into agricultural soils due to livestock OW spreading. While some discrepancies were underlined, this framework generally appeared to be consistent with the findings of previous worldwide and national inventories. Leclerc and Laurent (2017) used USEtox 2.02 to further estimate the impacts of trace elements in OW-amended soils on human toxicity (cancer and non-cancer effects) and freshwater ecotoxicity. They showed that Hg had a dominant impact with regard to cancer effects, Zn for non-cancer effects and Cu for freshwater ecotoxicity. More interestingly, the high spatial variability in regional inventories and impact intensity pointed out the need for the development of country- or regional-specific impact assessment.

Tarpani et al. (2020) recently suggested a complementary way to account for the ecotoxicological impact of trace elements among other environmental impacts of sewage sludge treatment methods, namely to consider the concentration of the most available trace element fraction (i.e. chemically extracted in the exchangeable/acid soluble pool) as being potentially harmful.

Owsianiak et al. (2015) assessed the influence of the emission source on the terrestrial ecotoxicological impact of some trace elements (Cd, Co, Cu, Ni, Pb, and Zn). Among the emissions sources, an organic-related source associated with anthropogenic activities (i.e. historical soil contamination) was considered. This organic-related source theoretically encompassed a large range

of OW, such as biosolids, manure, compost and treated wastewater —it was considered to be characterized by trace elements bound mainly to organic matter in OW in comparison with trace elements occurring as oxides or sulfides respectively originating from combustion processes and mining activities. It was found that the emission source has a significant impact on the reactive pool of Cd, Co, Cu, and Zn, but not Ni and Pb. The organic-related source exhibited a reactive pool that was significantly less reactive than geogenic and airborne sources for Cd, less reactive than any other anthropogenic sources for Cu and, conversely, more reactive than airborne and geogenic sources for Zn. Accordingly, Owsianiak et al. (2015) recommended incorporating the emission source effect, including the OW-related emission source effect, in the CTP calculation to improve assessment of the terrestrial ecotoxicological impact of trace elements in LCA procedures.

Sydow et al. (2018) combined the national inventories performed by Leclerc and Laurent (2017) and the assessment of the terrestrial ecotoxicological impact of OW-related emission sources suggested by Owsianiak et al. (2015) to assess the terrestrial ecotoxicological impact of agricultural OW-bound trace element inputs after spreading on agricultural fields in Europe on a country scale. Trace element and country-specific impact scores were calculated from the product of the total mass of a trace element emitted from OW applied on agricultural land in a given country ( $m_{\text{total}}$ ) and the area-weighted comparative toxicity potential (CTP) for the trace element and the target country. The  $m_{\text{total}}$  value was calculated directly at the country scale from the product of the total OW applied and the concentration of each trace element in OW. The CTP value per country was first calculated at 1 km<sup>2</sup> grid resolution as the product of the grid-specific agricultural surface area and the grid-specific CTP. The CTP value was then area-weighted at the country scale. Grid-specific CTP values were finally calculated from a grid-specific soil properties database and the framework previously proposed by Owsianiak et al. (2013) for FF, ACF, BF, and EF calculation. To account for the fact that trace elements were added to soil from a specific emission source (i.e. OW), ACF was calculated from the mean ACF determined for the organic-related trace element source, as suggested by Owsianiak et al. (2015). Several major conclusions were drawn from this study. Firstly, the grid-specific CTP values per trace

element ranged from 1 to 4 orders of magnitude among the soils in Europe. Despite this marked variability in grid-specific CTP, the country-specific impact scores were closely correlated with  $m_{\text{total}}$  rather than with the area-weighted CTP. Sydow et al. (2018) attributed the relatively small effect of CTP on the impact scores to the area weighting of grid-specific CTP at the country scale, suggesting that OW, and hence trace elements, are homogeneously applied on agricultural surface areas. Interestingly, this study also pointed out that Cu and Zn dominated among trace elements with regard to the contribution to both  $m_{\text{total}}$  and area-weighted CTP, and hence to the impact scores.

### **2.3.3 Ecotoxicity of OW-borne organic contaminants**

Despite organic contaminants being included in LCA toxicity models, very few LCA studies on OW management and treatment include organic contaminants in their emission inventory, contrary to trace elements. Although the organic pollutant fate is known to often depend on the sorptive properties, USEtox fate modelling does not take interactions between organic matter and organic contaminants into account, and excludes certain substance groups (see section 2.2.2).

The fate of trace elements and organic contaminants following OW application on agricultural soils has been assessed by the CML LCIA method, and the findings revealed that trace element leaching from sludge had greater impacts than organic contaminants (e.g. PPCP). In this study, organic contaminant characterization factors not accounted for in the CML method and were estimated by EDIP97 and USES-LCA on the basis of very limited data (Muñoz et al., 2008). The paucity of data available for determining characterization factors for contaminants as hence a major shortcoming of toxicity methods in LCA, yet ongoing research is striving to overcome this issue. For instance, in some other studies, characterization factors (fate and toxicology) for emerging pollutants, their metabolites and transformation products have been approximated through analytical methods such as liquid chromatography combined with mass spectrometry (la Farré et al., 2008). Alfonsín et al. (2014) also used USEtox and USES-LCA to calculate new updated characterization factors for PPCP in a new wastewater treatment implementation context. They enhanced the calculations by including additional physicochemical properties and degradation rates (via the Estimation Program Interface

Suite v4.0), as well as human exposure data, human toxicological effect and ecotoxicological effect data (from experimental data and the ECOTOX and IRIS databases).

In reference to a 3-year field experiment where organic contaminants were found to have dissipated in the first months after OW spreading, Hansen et al. (2006) used the EASEWASTE tool (and its simplified toxicity model) to account for bis(2-ethylhexyl) phthalate, nonylphenol ethoxylates, linear alkylbenzene sulfonate and PAH (sum of 11 compounds) derived from composted or anaerobically digested source-separated municipal solid waste spread on soil. One of the goals in the study of Sablayrolles et al. (2010) was to improve the quantification of human toxicity when dried and composted sludge were spread in fields. The authors calculated human toxicity factors via plants (carrot, tomato) for PAH (sum), PCB (sum), and bis(2-ethylhexyl) phthalate based on postharvest concentrations, dry matter content, plant yield and average consumption using USES-LCA and empirical factors from a previous soil-biosolids-plant study (Sablayrolles, 2004). Hospido et al. (2010) focused on the potential impacts of some emerging pollutants when anaerobically digested sludge (four digestion scenarios according to the temperature and sludge retention time) was applied on soil. Their contribution to human and terrestrial toxicity was found to be small compared to that of trace elements. They used USES-LCA (via CML) and the characterization factors presented in Muñoz et al. (2008) and computed with experimental data and the EDIP97 and USES-LCA toxicity models. All of these studies carried out using USES-LCA to assess terrestrial ecotoxicity inevitably inherit the shortcomings of that model, including the lack of ability to extrapolate aquatic data to estimate terrestrial ecotoxicity. Harder et al. (2017) studied the USEtox-modelled effects on human toxicity and compared the results with regard to sludge spread on soil. In the context studied, 13 organic contaminants were found to be of greatest potential concern, including bis(2-ethylhexyl) phthalate, hexachlorobenzene, 1 PCB, 1 polybrominated diphenyl ether, 17 $\alpha$ -ethinyloestradiol, 7 PAH and one biocide (Mirex, CAS No. 2385-85-5). The authors underlined the different sources of uncertainty in the estimation of the aggregate burden of disease: the model adequacy (e.g. overestimation of uptake by plants compared to field measurements), the data gaps regarding contaminants

potentially present in sewage sludge but not monitored and the data gaps regarding chemical and biological properties for most of contaminants.

## 2.4 Toxicity modelling in RA

Risk assessment is another framework commonly used to study waste treatments, their outputs and OW recycling in general, from an environmental/ecological and/or human health perspective (Brooks et al., 2012; Jahne et al., 2015; Magid, 2012). RA relies on models to determine hazards (including toxicity) and exposure to released contaminants in an *ex ante* fashion. The term RA may also refer to *ex post* empirical research studies, but this review focuses exclusively on *ex ante* model-based RA (see 2.5.2). OW risk assessments have mainly been conducted to address wastewater treatments and sludge recycling. RA compares measured or estimated concentrations with legal limits (Huber et al., 2016). RA and toxicity studies on sewage sludge for agricultural use have addressed —via empirical research— the load, impacts on human health and occasionally the fate (i.e. the transport and degradation of a substance emitted into the environment) of trace elements, organic contaminants and pathogens (Aparicio et al., 2009; Meng et al., 2014; Moreira et al., 2008; Ning et al., 2015; Singh and Agrawal, 2008; Tobajas et al., 2015; Vaz-Moreira et al., 2008; Walter et al., 2006). Relevant empirical research may involve bioassays, including germination/growth and microorganism isolation tests. RA models focused on hazard, exposure and effect (risk characterization) of European relevance are identified in the European Union Technical Guidance Document on Risk Assessment (EU-TGD<sup>vi</sup>) (EC-JRC, 2003) and the European Union Registration, Evaluation, Authorization, and Restriction of Chemicals (REACH) legislation (EC 2007; Table 3), such as the quantitative structure-activity relationship (QSAR<sup>vii</sup>) for toxicity, as well as the European Centre for Ecotoxicology and Toxicology of Chemicals targeted risk assessment (ECETOC-TRA), Stoffenmanager (Marquart et al., 2008; Tielemans et al., 2008) and the Advanced REACH Tool (ART) for occupational exposure. Exposure models have been compared on a recent review by Riedmann et al. (2015). The findings revealed that each model had different perspectives, as shown in the weighting sets applied to the modelled physical phenomena, and that there is a trade-off between accuracy and precision the

model designs, and recommendations on their use ultimately depends on the availability and quality of the available data. The authors identified Stoffenmanager, which consists of system of linear equations for near and far-field exposure, as the most robust exposure model. Another key RA framework is that implemented by the United States Environmental Protection Agency (US EPA), which is also based on the exposure-effect paradigm (see Table 3 for the European REACH), and is highly modelling based (US EPA, 1992; USEPA, 1994). A heterogeneous international list of exposure models, often discussed among scientists at the European Joint Research Centre (JRC), is also presented in the supplementary material associated with Bopp et al. (2019). Contrary to LCA, where toxicity models are focused on a broad range of target contaminants (trace elements and organic contaminants), RA features a separate model per type of contaminant (trace elements, organic contaminants and pathogens). Models used in the OW treatment and agricultural recycling context, as well as those dealing with specific contaminant types, are presented in section 2.5.

## **2.5 Consideration of contaminants in organic waste treatment and agricultural recycling RA**

### **2.5.1 Organic waste treatment and soil application**

Risk assessment has mainly been used to study digestate recycling rather than anaerobic digestion facilities and their functioning (Brooks et al., 2012; Pivato et al., 2015; Zhao and Liu, 2019). The contaminants most commonly studied in relation to anaerobic digestion of OW are, in decreasing frequency: trace elements and pathogens, through approaches such as quantitative microbial risk assessment (QMRA) (section 2.5.4) (e.g. Brooks et al., 2012) and ecotoxicological tests (e.g. Pivato et al., 2015). We did not find any anaerobic digestion RA taking organic contaminants into account, but we did note a few examples of empirical determination and/or measurements of the effects of anaerobic digestion on organic contaminants present in sewage sludge (Bourdat-Deschamps et al., 2017; Carballa et al., 2007; Samaras et al., 2013).

Composting RA studies are also less common than on other waste treatment pathways. These studies address municipal solid waste, agricultural waste and sludge as substrates, and consider the

trace elements, pathogens and organic contaminants (Alvarenga et al., 2013; Déportes et al., 1995; Gusiatin and Kulikowska, 2014; Patureau et al., 2012). Gusiatin and Kulikowska (2014), while recognizing that it would be insufficient to only use the total trace element concentration in environmental assessments, proposed trace elements indicators for sludge compost RA (see 2.5.2). A non-peer reviewed meta-review (Magid, 2012), compiled conclusions from more than 150 RA and fertilization studies, where the fate of contaminants, in terms of PEC and PNEC (see Table 3), was determined through long-term field experiments, mathematical modelling compliant with EU-TGD, and worst-case input-based estimation according to REACH. These studies suggested that interactions among trace elements, organic contaminants and pathogens present in sludge are unlikely unless the contaminants share a mode of action, and that there is no conclusive evidence that sludge spreading on agricultural soils (within regulatory boundaries) is harmful for humans or animals. Several issues have been identified that require further investigation, namely the potential impacts of chlorinated paraffin and phthalates on human health, as well as the impacts of PPCP and endocrine-disruptors on soil quality and human health. These studies conclude that the possibility of health impacts from antibiotics and antibiotic-resistant bacteria in sludge is non-negligible, and that this issue requires further research.

For certain organic contaminant categories such as pharmaceuticals and PCPP, despite the growing number of publications, very few data are available on their environmental fate after OW recycling in agricultural fields (Houot et al., 2014).

Recently, Bourdat-Deschamps et al. (2017) studied the fate and exposure of several pharmaceutical residues in agricultural soils after repeated application of several types of OW, applied at usual doses for farmers and in different pedoclimatic settings. Predicted concentrations of pharmaceuticals in soil after several OW applications were higher than the measured concentrations due to degradation, strong sorption to soil constituents and/or leaching. Dissipation half-life times ( $DT_{50}^{viii}$ ) were assessed at around 1300-2500, 900 and <300 days for fluoroquinolones, carbamazepine and ibuprofen in temperate soils, and <150 days and 80 days for fluoroquinolones and doxycycline in tropical soils.

Based on the few data available on ecotoxicological effects on terrestrial organisms, potential risks were estimated using the risk quotient approach (section 2.5.3). The authors concluded that the environmental risk was low. However, the study clearly highlighted the lack of available ecotoxicological parameters for such contaminants. More generally, one difficulty encountered was to obtain relevant ecotoxicological endpoints for organic contaminants. This issue is still far from solved, even for compounds such as pesticides undergoing ecotoxicological and toxicological tests for registration purposes. Traoré et al. (2018) noticed that the availability of ecotoxicological parameters was lower for chronic than for acute toxicity endpoints, regardless of the targeted organisms. This distortion is due to the fact that simple cost- and time-effective tests are used for acute toxicity assessment. It is also clear that ecotoxicological parameter measurements are far more available for aquatic organisms (invertebrates) than for terrestrial invertebrates. Moreover, interpretation of results from terrestrial invertebrates requires a far more complex methodology because tests regularly involve non-equilibrium processes.

Brooks et al. (2012) performed QMRA (section 2.5.4) of scenarios featuring the application of sewage sludge and manure digestates and raw sewage sludge based on previously published experimental data and empirical equations to determine soil and crop contamination levels. They concluded that raw and digestate sewage sludge risks were associated with viruses, whereas manure risks concerned bacteria.

Komnitsas and Zaharaki (2014) reviewed recent ecological RA studies on compost application, performed under the US EPA framework. They presented a broad range of fate and risk factors and indices, while also focusing on human exposure. Thomaidi et al. (2016) performed environmental RA of raw and composted sewage sludge-amended soil, calculated aquatic and soil PEC, and estimated elevated risk quotients for synthetic phenolic compounds and siloxanes. A recent review of the environmental risk literature, including ERA, revealed that the recycling of sewage sludge in soils — raw or stabilized by composting or anaerobic digestion— generates risks from PAH (but not other organic contaminants) and trace element contamination risks (Liu, 2016).

RA on livestock effluent recycling in agriculture (treated or raw) has been almost exclusively focused on trace elements and organic contaminants. For instance, Río et al. (2011) proposed a dynamic environmental risk assessment multicompartmental model for assessing trace element-induced risks of long-term manure application. Several RA studies have investigated different aspects, including: PEC of selected antibiotics (e.g. tetracyclines, quinolones, sulfonamides) based on EU-TGD and European Medicines Agency (EMA) guidelines (Carballo et al., 2016; EMA, 2006; Li et al., 2015) (see section 2.4); effective internal doses of airborne pathogens (via empirical formulas (Jahne et al., 2015)); aerosol dispersion models (via empirical formulas (Heimersson et al., 2014)); and even qualitative scoring according to pedoclimatic conditions using a qualitative multi-attribute decision model for *ex ante* assessment of agricultural sustainability (Sadok et al., 2009). Prosser and Sibley (2015) assessed the human health risk based on reported empirical research findings on organic contaminant residues in edible tissues of plants grown in sewage sludge- or manure-amended or wastewater irrigated soils. They compared estimated daily intake to acceptable daily intake values (as defined by many different official international agencies). For all three amendment practices, hazard quotients <0.1 (low risk) applied to the majority of the reported residues. Yet the need for further investigation of risks related to antibiotic resistance associated with sludge and manure spreading has recently been pointed out in the light of the rising pharmaceutical contamination levels in the environment (Bondarczuk et al., 2016).

### **2.5.2 Ecological and human risks due to OW-borne trace elements**

Trace element RA has been developed to evaluate the impact of trace elements on ecosystems and human health through two main groups of approaches (Figure 2). The first involves dedicated experimental procedures and analytical determinations, while the second is based on quantitative and predictive modelling of the fate of trace elements in soils and their consequent transfer to surrounding environmental compartments, and to the human food chain.

[Figure 2]

The main equations used to compute the indicators associated with these approaches are summarized in Table 4.

[Table 4]

The experimental and analytical approach of RA for trace elements can be further divided in two sub-approaches, based on an *ex ante* RA of trace elements that could accumulate in soil upon OW application or an *ex post* RA of trace elements that were added in fields that have a long history of OW amendment (Figure 2). The *ex ante* approach is focused on the analytical characterization of trace element availability in OW and/or trace element ecotoxicity in freshly OW-amended soil samples (Komnitsas and Zaharaki, 2014). Trace element availability in OW is usually determined by the application of a sequential trace element extraction procedure to a range of OW types. The trace element concentration in each extracted fraction and their relative proportions enable calculation of three indicators of the potential availability level of trace elements that would be added to soil by the application of a given OW (Gusiatin and Kulikowska, 2014). The reduced partition index ( $I_R$ ) characterizes the binding intensity of each trace element in the OW solid phase, where a low  $I_R$  value indicates low binding and consequently a potentially high availability of trace elements in OW and in the amended soil. The risk assessment code (RAC) corresponds to the percentage of the total concentration of each trace element occurring in the most available fraction (e.g. the exchangeable fraction) and characterizes the trace element availability in OW. RAC is usually inversely correlated with the  $I_R$  and is typically interpreted according to the following scale: <1% (no risk), 1-10% (low risk), 11-30% (medium risk), 31-50% (high risk) and >50% (very high risk). Finally, the modified risk index (MRI) corresponds to the sum of the potential ecological risk of each individual trace element and combines a toxic-response factor, the measured availability, and a background or regulatory threshold for each trace element. Although MRI is highly impacted by the number of trace elements analysed and the background/regulatory thresholds considered, it is typically interpreted according to the following scale: <150 (low risk), 150-300 (moderate risk), 300-600 (considerable risk) and >600 (very high risk). This approach —based on the analytical characterization of trace element availability

in waste— has already been applied to OW, including agricultural waste and sewage sludge compost (Gusiatin and Kulikowska, 2014; Komnitsas and Zaharaki, 2014).

When there is not sufficient data to classify an OW based on its chemical composition, Pandard and Römbke (2013) suggested that *ex ante* characterization of its overall ecotoxicity (including the contribution of trace elements and other types of contaminants) could be performed through a battery of aquatic and terrestrial biotests on a dilution series of OW eluate or OW-amended artificial soil samples (Figure 2). Thresholds based on the level or percentage of OW dilution that induces a given reduction (e.g. 50%) of each ecotoxicological endpoint compared to the control were proposed to classify the OW as hazardous or not. This approach was specifically developed for OW meant for application on agricultural soils and deployed to assess a solid or eluted mixture of artificial soil with nine OW applied at 1- to 100-fold the recommended application rate (Huguier et al., 2015). The authors concluded that the use of terrestrial biotests (particularly plants and earthworms biotests) were the most sensitive and discriminant ecotoxicological endpoints and consequently the suggested minimal procedure to assess the ecotoxicological impacts in OW-amended soils. Similarly, a range of aquatic and terrestrial biotests was also used for the ecotoxicological RA of an agricultural digestate (Pivato et al., 2015). The dose-response curves derived from the measured ecotoxicological endpoints were used to determine a predictable non-effect digestate concentration in soil ( $PNEC_{Soil}$ , in mass of digestate per mass unit of soil) under which no unacceptable ecotoxicity on soil organisms would be expected. The authors also suggested to further reduce this  $PNEC_{Soil}$  by a dimensionless assessment factor, depending on the number of trophic levels assessed, the timespan of the exposure to OW-amended soil (short-term/acute vs. long-term/chronic), and on whether the ecotoxicological RA were performed on laboratory or field data. Renaud et al. (2019) recently proposed to refine the rendition of dose-response curves obtained from OW-amended soils by establishing a species sensitivity distribution for each tested OW. These species sensitivity distributions are based on ecotoxicological endpoints determined for several species (e.g. two endpoints for plants and four endpoints for soil invertebrates were used by Renaud et al. (2019)).

The suggested reference ecotoxicological endpoints were the effective concentrations causing 10 or 50% toxic effect ( $EC_{10}$  or  $EC_{50}$ ). A hazardous concentration (HC) was thereafter determined for each OW as a threshold value higher than 5 or 50% ( $HC_5$  or  $HC_{50}$ , respectively) of the community of soil organisms accounted for in the corresponding species sensitivity distribution. Interestingly, Renaud et al. (2019) showed that using an  $HC_5$  based on  $EC_{50}$  was more protective than an  $HC_{50}$  based on  $EC_{10}$  and thus seemed more suitable for ecotoxicological risk assessment. The  $PNEC_{add}$  value —which corresponds to the so-called hazardous concentration at x% ( $HC_x$ )— is hence protective for 1-x% of soil organisms considered in the database.  $PNEC_{add}$  is usually calculated for a value of  $x = 5$ .

The ecotoxicological impacts of OW can be also assessed *ex post* on soil sampled in fields that have a long history of OW amendment (Figure 2). For instance, Roig et al. (2012) sampled soils from a 16-year field experiment where unfertilized control plots and control plots receiving mineral fertilizers were compared to plots amended with anaerobically digested sewage sludge applied at several frequencies and application rates. The resulting physicochemical properties and trace element concentrations were determined in each soil sample. Ecotoxicological parameters (microbial activity in soil via respiration, phytotoxicity via germination and shoot growth tests, etc.) were further determined directly in soil samples or in a soil eluate. The overall ecotoxicological impact was finally assessed by comparing changes in soil physicochemical properties and ecotoxicological parameters between the OW-amended soil samples and control soil samples.

*Ex post* RA was also developed to assess the impacts of trace elements on human health (cancer and non-cancer effects; Figure 2) by considering multiple pathways of human (adults and children) exposure to amended soils, such as soil dust inhalation and the ingestion of soil, edible plant organs and livestock products (typically milk, meat and eggs) (Déportes et al., 1995). *Ex post* RA is used to measure trace element concentrations in relevant environmental compartments (i.e. waste, soil, air, water, plant and animal products) and then to calculate a health risk index (HRI) from the ratio between the average daily intake of a given trace element through one or many exposure pathways, along with the corresponding health benchmark value. For each trace element, health benchmark

values usually correspond to the reference dose for cancer and non-cancer effects following ingestion (expressed in trace element mass/body weight mass/day) and inhalation (expressed in trace element mass/m<sup>3</sup> of air) (Komnitsas and Zaharaki, 2014). Such health benchmark values have, for instance, been defined for a range of trace elements by US EPA. HRI can integrate the fact that human beings are only occasionally exposed to trace elements by taking the exposure frequency into account when calculating the target hazard quotient (THQ) for non-cancer effects, along with the target cancer risk (TCR). HRI and THQ values below one and TCR values below 10<sup>-4</sup> are considered to be within the health protection limit. THQ of a range of trace elements can be further summed to calculate the hazard index (HI), which takes the cumulative effect of ingested or inhaled trace elements into consideration. This kind of *ex post* RA of trace elements for human health was recently applied to the findings of a 10-year field experiment with soil amended with mineral fertilizers or OW and revealed that some trace elements (Br and Zn) posed a potential non-cancer risk to human health (Couto et al., 2018).

The approach based on experimental procedures and analytical determinations is sometime tedious, expensive and time-consuming, so a predictive approach (*ex ante* by design) was developed for trace element RA (Figure 2). This approach is based on quantitative modelling of the fate of trace elements following their emission into soil, and of their ecotoxicological and human health impacts. The primary step in this predictive approach involves mass-balance modelling of trace element inputs and outputs in soil at an annual time step. Trace element inputs in soil usually include atmospheric deposition, pesticide application and fertilization with mineral fertilizer and OW. The intermediate dynamic model for metals (IDMM) also accounts for the impact of OW inputs on soil properties other than the total trace element concentration in soil (Monteiro et al., 2010) (Figure 2). In IDMM, it is considered that OW is mixed with the top 5 cm soil layer and consequently alters the bulk density and organic matter concentration in this layer. It is further considered that 70% of the organic matter added by OW is mineralized at each time step. Trace element outputs from soil usually considered are uptake in plants and vertical leaching of trace elements in solution. In IDMM, the leaching of

trace elements associated with soil particles, as well as the erosion-induced loss of trace elements in soil solution and associated with soil particles, are also considered as outputs (Lofts et al., 2013b; Monteiro et al., 2010). The simplest models directly calculate trace element concentrations in plants (for the plant uptake output) or in soil solution (for the soluble leaching output) from the total trace element concentration in soil and the soil properties (i.e. pH, clay concentration and/or organic matter concentration) using multilinear regression equations (Río et al., 2011). The model of de Vries et al. (2004) estimates trace element leaching in soil solution using empirical multilinear regression equations to first calculate the available pool of trace elements in soil (called the reactive pool) from the total concentration of trace elements in soil and the soil properties (i.e. organic matter and clay concentration) and then the trace element concentration in soil solution from the calculated reactive pool and the soil properties. In IDMM, trace elements are more comprehensively distributed between four different pools: a first mineral pool of trace elements occluded in soil-bound primary and secondary minerals; these can weather over time and give rise to a second pool of available/reactive trace elements bound to the soil solid-phase; the latter is in equilibrium with a third pool of free ionic trace elements in soil solution; and finally this pool is in equilibrium with a fourth pool of trace elements bound to inorganic and organic ligands in soil solution (Lofts et al., 2013b; Lofts and Walker, 2016). The third and fourth pools thus jointly constitute the total pool of trace elements occurring in soil solution. The free ionic concentration of trace elements in soil solution is calculated from the concentration of trace elements bound to the soil solid-phase using multilinear regression based on the soil properties (i.e. pH and the clay and organic matter concentration), while the total concentration of trace elements in soil solution is simulated from the free ionic concentrations and the chemical composition of the soil solution (i.e. pH and the concentration of major cations and anions and DOC) with WHAM. Monteiro et al. (2010) and Lofts and Walker (2016) further implemented IDMM to enable the available pool of trace elements bound to the soil solid-phase to kinetically decrease over time to become strongly fixed to the soil solid-phase following aging. This aged pool was also able to kinetically resupply the mineral pool. The rate

of trace element weathering from the occluded pool to the available pool is set at a given value per trace element, while the rate of trace element aging depends on the soil pH. Simulations of trace element accumulation in soil and transport from soil to the surrounding water resources are usually performed over several decades, thereby questioning the relevance of uncertainty in the model parameter estimations. In IDMM, up to three soil layers are considered, corresponding to the top 5 cm layer, the 5-25 cm layer and the layer from 25 cm to the artificial drainage depth (typically 1 m). In the most advanced IDMM version, a surface water model was implemented to manage the supply of drained and eroded trace elements (Lofts and Walker, 2016). The range of trace elements for which each mass-balance model is parameterized usually depends on the model complexity. The simplest models, such as that proposed by Río et al. (2011), are parameterized for five trace elements (i.e. Cd, Cu, Ni, Pb and Zn), while more complex models such as those proposed by de Vries et al. (2004) or IDMM are completely parameterized for one or two trace elements only (i.e. Cu and/or Zn).

Based on mass-balance modelling outputs, ecotoxicological impacts are predicted for a given time horizon by calculating the PEC/PNEC ratio that is considered safe if it is lower to 1 (de Vries et al., 2004; Monteiro et al., 2010) (Figure 2). The PEC value corresponds to the total trace element concentration in soil predicted by the mass-balance model for a given time horizon. PNEC is usually considered to be the sum of the natural pedogeochemical background concentration of trace elements in an uncontaminated soil layer and the  $PNEC_{add}$  specifically calculated for trace elements added to the soil by anthropogenic activities (i.e. contamination).  $PNEC_{add}$  is derived from a selected ecotoxic endpoint given for a range of soil organisms (usually plants, invertebrates and microbial processes) in ecotoxicity databases, e.g. preferentially  $EC_x$  (usually  $EC_{10}$  is considered) or, when no reliable  $EC_x$  is available, the highest no observed effect concentration (NOEC, see Table 3) or lowest observed effect concentration (LOEC) (de Vries et al., 2004; Oorts, 2018). Several corrections are further applied to account for the fact that experimental conditions (e.g. freshly spiked soil samples in laboratory conditions rather than from long-term field experiment) and soil types used to derive

EC<sub>x</sub>, NOEC and LOEC do not comprehensively mimic realistic field conditions, or encompass the whole range of soil properties that may govern trace element ecotoxicity. The EC<sub>x</sub>, NOEC, and LOEC are multiplied by a lab-field (L/F) factor varying between 1 for Cd to 4 for Ni at high soil pH and Pb (de Vries et al., 2004; Oorts, 2018). Jensen et al. (2018) further suggested differentiating two L/F factors to account for the specific case of repetitive long-term OW applications. This proposal was based on the assumption that, if trace elements added to a soil over a period of more than a year are highly aged (and consequently less available) relative to freshly spiked soil under laboratory conditions, OW-bound trace elements applied within the ongoing year would be less aged (and thus more available). A second correction is then applied to EC<sub>x</sub>, NOEC, and LOEC to account for the specific physicochemical properties of studied soils. This soil factor is either empirically set at a given value depending on soil type, e.g. 1.1 for sandy soil to 1.5 river clay soils (de Vries et al., 2004), or semi-mechanistically calculated from the effective cation exchange capacity, organic matter concentration, clay concentration, pH, and/or the background concentration determined for each studied soil (Oorts, 2018). The PNEC<sub>add</sub> value is finally calculated as the value corresponding the x<sup>th</sup> (usually x=5) percentile of the corrected EC<sub>x</sub>, NOEC, or LOEC distribution, and therefore corresponds to the so-called hazardous concentration at x% (HC<sub>x</sub>).

Based on the mass-balance modelling outputs, the risk-based decision tool for OW management in agriculture and farming activities (FARMERS) was developed to assess trace element impacts on human health (Río et al., 2011) (Figure 2). The exposure module considers that human beings will mainly be exposed to trace elements via milk and meat consumption, while dermal contact, ingestion, and soil particle inhalation are considered as negligible exposure routes (Franco et al., 2006). The exposure module therefore first calculates the transfer and consequent concentration of trace elements in cattle meat and milk by accounting for the contribution of the ingestion of forage and soil particles from the area receiving OW and the ingestion of water and feed additives that may contain additional trace elements. The calculated concentrations are then used in the risk characterization module to estimate the cancer and non-cancer risks. The non-cancer risk is

estimated by calculating the HRI (as defined above) for each exposure route or for the sum of multiple exposure routes and by summing the HRI for each trace element in a global non-cancer risk, i.e. HI (as defined above), whose maximum permissible limit must be provided to run a RA simulation. The cancer-risk is estimated by calculating the TCR (as defined above). Accordingly, FARMERS basically calculates the cancer and non-cancer risks for an OW recycling scenario in a given agricultural area and may highlight human health risks by integrating a geographical information system (GIS). In addition, FARMERS can optimize conversely parameters based on targeted HI and TCR criteria to calculate the maximum permissible concentration of each trace element in soils, or the maximum OW application rate to remain within an acceptable health risk context (Río et al., 2011).

### **2.5.3 Ecological and human risks of OW-borne organic contaminants**

It is not possible to perform comprehensive experiments to assess waste-bound organic compound exposure and hazards due to the high diversity of organic compounds, OW and agricultural conditions (soils, crops). Several RA strategies have been developed to deal with this, ranging from elementary ranking approaches to complex dynamic models.

**Screening/ranking approaches** aim at assessing the likelihood of organic contaminants posing risks to surface water, groundwater, human health or of accumulating in the food chain following waste application on agricultural soils, while prioritizing compounds of most concern (Clarke et al., 2016; Duarte-Davidson and Jones, 1996; Wilson et al., 1996). These approaches are mainly based on organic compound physicochemical properties, such as water solubility, vapour pressure, Henry constant, octanol-water partition coefficient, adsorption coefficient and degradation half-life, or on empirical (e.g. multi-parameter exponential/logarithmic) models based on these properties (Duarte-Davidson and Jones, 1996; Wilson et al., 1996). A more complex ranking approach was developed by Clarke et al. (2016) from the probabilistic Environmental Potential Risk Indicator for Pesticide indicator of Trevisan et al. (2009) to estimate human exposure to organic contaminants bound in biosolids destined for grassland application. The approach considers four major compartments:

concentration in top soil, surface runoff and groundwater, and the chemical intake level (or human exposure). Human health risk is estimated using the intake toxicity ratio, which corresponds to the ratio of the measure of the effects (concentration lethal to 50% of the population,  $LC_{50}$ ) to the estimated exposure (i.e. concentration in runoff or groundwater). The environmental fate of each contaminant is assessed using parameters such as those described previously (adsorption coefficient, half-life) to estimate the concentration in soil after application, potential subsequent runoff from the OW-treated field into surface water, leaching into groundwater, water consumption and overall toxicity. Beyond the usefulness regarding individual contaminants, the data are used to rank the human health risk of contaminated water consumption (Clarke et al., 2016). Screening is a useful tool for determining the possible fate of organic compounds introduced in the soil system via OW and for prioritizing compounds for experimental validation (Wilson et al., 1996). Other ranking methods not focused on OW could also be used to prioritize organic contaminants, e.g. methods based on quantitative structure-activity relationships (Sangion and Gramatica, 2016). The screening/ranking approach has major limitations as it does not allow quantitative environmental risk assessment of waste-bound organic contaminants. Moreover, since the processes determining their fate are not considered, interactions between contaminants and waste and subsequent modifications in interactions are also not taken into account. Sludge application increases the soil organic matter content and introduces complex components in soils, which change the soil properties. Organic contaminants may adsorb on (organic matter-rich) sludge particulates, and subsequently strongly bind the soil solid phase, thus decreasing the leaching potential, volatilization and/or plant uptake (Duarte-Davidson and Jones, 1996; Wilson et al., 1996). In other cases, some organic materials can enhance organic contaminant leaching by increasing the contaminant infiltration rate through the formation of soluble complexes and/or sorption on colloidal material (Wilson et al., 1996).

The **risk quotient (or hazard quotient) approach**, as recommended by EU-TGD (section 2.4) and EMEA (section 2.5.1), is one of the most highly used approaches to assess the environmental risk related to OW-bound organic contaminants after application on soils (Bourdat-Deschamps et al.,

2017; Chen et al., 2011; González et al., 2010; Hernando et al., 2006; Kloepper-Sams et al., 1996; Langdon et al., 2010; Liu et al., 2014; Martín et al., 2012; Martin, 2015; Thomaidi et al., 2016). The risk quotient corresponds to the PEC/PNEC ratio (Table 3). In most case studies, risk quotients are calculated for terrestrial risk assessment using measured or calculated (according to EU-TGD)  $PEC_{soil}$  values corresponding to the level one year after one OW dose application (EC-JRC, 2003; García-Santiago et al., 2016; Verlicchi and Zambello, 2015). Some authors have also assessed the risk for aquatic organisms from  $PEC_{water}$  and  $PEC_{sediment}$  (Hernando et al., 2006; Langdon et al., 2010).

According to EU-TGD, risk assessment in soil is based on short-term toxicity impact data on terrestrial organisms, such as plants, earthworms and/or soil microorganisms. If no terrestrial toxicity data are available,  $PNEC_{soil}$  are sometimes determined from  $PNEC_{water}$  (González et al., 2010; Martín et al., 2012; Thomaidi et al., 2016). PNEC for the aquatic compartment can be estimated from the half maximal effective concentration ( $EC_{50}$ ), from  $LC_{50}$  values obtained via acute toxicity tests or from NOEC values obtained in the long term (on algae, daphnia and fish), as well as by application of assessment factors to account for extrapolation from intra- and inter-species variability in sensitivity (EC-JRC, 2003; EMEA, 2006). For final risk assessment, a common criterion, as proposed by Sánchez-Bayo et al. (2002) and Hernando et al. (2006), is generally applied: low risk if  $0.01 < \text{risk quotient} < 0.1$ , medium risk if  $0.1 \leq \text{risk quotient} \leq 1$ , high risk if  $\text{risk quotient} \geq 1$ . To calculate the mixture toxicity risk quotient of several compounds, Thomaidi et al. (2016) chose to sum up their risk quotient according to Escher et al. (2011). This approach is also consistent with the findings of Backhaus and Faust (2012) who obtained evidence that concentration addition is a precautionous first tier mixture risk assessment model, and that summing up PEC/PNEC ratios might serve as a justifiable concentration addition approximation. The risk quotient approach should only be seen as a preliminary way to estimate the ecological threat for terrestrial and/or aquatic organisms. A number of uncertainty sources is often included, such as the use of aquatic toxicity due to the lack of terrestrial experimental toxicity data for most organic compounds. In addition, most risk quotients are calculated from PNEC determined from acute data rather than from long-term exposure data (e.g.

chronic toxicity tests or bioaccumulation studies) (Bourdat-Deschamps et al., 2017; Hernando et al., 2006; Martín et al., 2015). Finally, as degradation patterns observed for OW-borne organic compounds may differ from those that are spiked in soils, it may be misleading to use these degradation rates at this hazard assessment level (Langdon et al., 2010). Note, however, that risk quotient values are likely to be overestimates of the actual risk due to conservative assumptions used throughout the assessment process (i.e. PEC calculation is based on the maximum organic compound concentration in OW without taking sorption, degradation, etc., into account).

Various **other basic approaches** have been proposed. For instance, to assess whether PAH in textile dyeing sludge would have adverse biological effects, Ning et al. (2014) compared measured PAH concentrations in sludge to the effects range-low (ERL) and effects range-median (ERM) values, as developed by Long et al. (1995). ERL and ERM define the chemical concentration ranges that will rarely (<ERL), occasionally (>ERL and <ERM) and frequently (>ERM) have adverse biological effects. ERL is defined as the lower 10<sup>th</sup> percentile of the effects data range per chemical, while ERM is the 50<sup>th</sup> percentile of the effects data range (Long et al., 1995). Since ERL and ERM values based on sediment quality assessments may overestimate or underestimate the risk, Ning et al. (2014) also compared PAH concentrations to ecological soil screening levels (USEPA, 2007). These are contaminant concentrations in soil that are protective of ecological receptors that commonly come in contact with and/or consume biota that live in or on soil. Ecological soil screening levels are derived separately for four groups of ecological receptors: plants, soil invertebrates, birds and mammals. Chaney et al. (1996) used the pathway risk assessment approach to assess risks related to PCB in municipal sewage sludge applied on soil. Risk assessment was conducted to protect highly exposed individuals who use biosolid products on their lawns, gardens or farms, and who ingest foods produced on biosolid-amended soils as a high portion of their diet over a 70-year period of their lives. Several scenarios of exposure to PCB through biosolids, such as 'biosolids to human', 'biosolids to soil to animal to human', and 'biosolids to soil to plant to animal to human' were defined and studied. Several parameters were calculated for each scenario, such as the allowed daily

ingestion or the reference soil concentration that cannot be exceeded at the selected risk level. It should be underlined that the highly exposed individuals approach is very conservative because it considers that humans (adults or children) are continuously exposed to the same amount of biosolid-bound PCB (Chaney et al., 1996). Chari and Halden (2012) developed an empirical equation to estimate the aqueous-phase concentration of hydrophobic organic contaminants that could leach from biosolids and potentially reach groundwater and/or surface water. The equation estimates the pore-water concentration of a compound at equilibrium in the solid and liquid phases (Langdon et al., 2010). Pore-water concentrations indicate the migration potential of compounds applied on soils through biosolids (Chari and Halden, 2012). The pore-water concentrations are then compared to aquatic toxicity values for risk assessment. Note that the mass fraction of compounds present in pore water is very small compared to the amount of chemical sorbed onto particles, and that equilibrium concentrations in pore water will be substantially diluted during rainfall events thereby lowering the risk of harmful exposure (Chari and Halden, 2012; ECHA, 2016). Hazard assessment based on equilibrium pore-water concentrations should therefore be interpreted as a worst-case scenario. Brambilla et al. (2016) evaluated the potential carryover of persistent organic contaminants from amended soil-to-milk of extensive farmed sheep. The prediction model was based on farming practices, soil intake, organic pollutant toxicokinetics and dairy product intake in children. The PEC in topsoil relevant for intake assessment in exposed animals was based on the 'agronomic evaluation' of fate and behaviour of the considered persistent organic contaminants in soil in the treated area. Inputs of the considered contaminants from OW in topsoil were thus estimated by accounting for the median organic carbon content of agricultural topsoils and the annual maximum inputs of organic carbon from waste application. The annual progressive input of persistent organic contaminants from waste was then computed by dividing the level of waste contamination descriptors with various computed timeframes for the different fertilizer typologies. This was based on the following assumption: levels of considered hydrophobic contaminants in soil were correlated with the organic carbon content, while taking into account inputs from sources other than biosolids and losses due to

degradation. The impact of predicted environmental concentration in topsoil on food safety/security was computed using a consolidated physiological toxicokinetics model based on the soil uptake of the considered contaminants, their oral bioavailability, distribution, metabolism and excretion rate in sheep milk. In line with the screening/ranking and risk quotient approaches, the simple approaches described above do not account for the various processes involved in the fate and impacts of organic compounds in OW applied on soils such as interactions between compounds and waste.

In the REACH context, risk assessment distinguishes between releases before and after a biological sewage treatment plant. Releases 'before' are those from the use-process as such, with or without specific measures to prevent losses and/or to treat emissions onsite. Biological treatment —i.e. biodegradation and associated mechanisms that remove substances from the waterway (adsorption and sedimentation, volatilization, etc.)— is modelled by static and dynamic models such as SimpleTreat (Kah and Brown, 2011; Struijs, 2014; Struijs et al., 2016), WWTREAT (Cowan et al., 1993), Water9 (USEPA, 1994), TOXCHEM (Melcer et al., 1994), or activated sludge models such as WEST (Plósz et al., 2012). Various reviews have been published on contaminant fate models in wastewater treatment plants, describing their dynamicity and listing the pollutant families modelled (Clouzot et al., 2013; Plósz et al., 2013; Pomiès et al., 2013). The outputs of these models are concentrations/fluxes discharged into water which are then used by EUSES for impact assessment at local and regional scales based on mechanistic models (ECHA, 2016). At both scales, the exposure calculation covers soil, air, groundwater, fresh and marine water and sediment, secondary poisoning (fish-eating and worm-eating predators) and humans via the environment (through food consumption, e.g. fish, crops, meat and milk, and drinking water [oral route], and via the inhalation of air [inhalation route]). Once the expected exposure is estimated, exposure levels are used to characterize the risks by comparing them with the hazard assessment outcome. Quantitative risk characterization can involve comparing the exposure concentration in each compartment with the relevant PNEC (so-called risk characterization ratios) (ECHA, 2016). This approach is similar to the risk quotient approach, but the PEC calculation is based on a more complex method. To assess exposure

following the release of pesticide co-formulants into agricultural soil and into edge of field water bodies via spray drift and runoff/drainage, the European Crop Protection Association Local Environment Tool (ECPA LET<sup>ix</sup>) can be used instead of EUSES, since the latter is not able to take direct and indirect releases into edge of field water bodies into account. While widely used, EUSES has the following limits: it does not cover substances such as ionizing chemicals, trace elements, nanomaterials or substances which undergo transformation within the environment, and it only takes direct releases into soil at the local scale into account. New release mechanisms should therefore be included into the model, while the fate and transport mechanisms such as bioaccumulation in fish and secondary poisoning, human exposure via the environment need to be updated (ECHA, 2016). Exposure dynamic models such as SimpleTreat 3.1 (Legind et al., 2011) do not take any organic compound transformation in plants into account (Polesel et al., 2015). These models, while very useful for preliminary risk assessment, do not consider OW and organic compound interactions or compound degradation products.

#### **2.5.4 Ecological and human risks of OW-borne pathogens and antibiotic resistance**

Risk assessments include the following four basic steps: hazard identification, exposure assessment, dose-response assessment, and risk characterization (National Research Council, 1983). The **quantitative microbial risk assessment (QMRA)** approach has been added to the risk analysis process (Havelaar et al., 2008): it contains the last four basic steps (illustrated in Figure 3), as well as risk management and risk communication (National Research Council, 2009). QMRA combines information, data and mathematical models to describe the exposure to and spread of pathogens and to illustrate the nature of the adverse outcomes. A description of the different types of models used in QMRA that are applied to water safety management is available in (WHO, 2016). Large datasets and information are thus needed to calculate the microbiological risk, by defining the pathogen-specific dose-response assessment, which requires fitting of a dose-response model (e.g. exponential, Beta-Poisson) (Rose et al., 2008). The infective dose, taking all potential exposure routes into account, must also be calculated based on the quantity of medium involved in the exposure and

the associated pathogen concentration (Soller and Olivieri, 2003). QMRA relies on important inputs such as pathogen loads, decay rates, transport, inactivation, as well as on dose response parameters (Brooks et al., 2012; Haas et al., 2014). A typical example of the QMRA model output is the probability of infection or illness associated with the application of OW treatment end-products on agricultural soils. Pathogen fates following raw sludge, digested sewage sludge and manure applications on agricultural soils have been assessed by QMRA (Brooks et al., 2012). Infectious risks by pathogen groups confirmed the greater bacterial risk from manure, whereas viral risks were exclusive to biosolids. The greatest single risk in various pathogen exposure scenarios involving fomite (e.g. truck or tractor handles), soil, crop or aerosol exposure was found to be associated with *C. jejuni* and enteric viruses. Figure 3 depicts the QMRA processes associated with each parameter or step regarding land application of residual waste.

[Figure 3]

Current microbial RA models need to be fitted with high quality datasets. Furthermore, their use should be associated with uncertainty values reflecting the dataset quality per simulation or investigation according to the *Codex Alimentarius* (FAO-WHO, 2014) or other guidelines (e.g. FAO-WHO, 2003; Vose, 2008). QMRA is limited by the pathogen-specific dose-response data quality, thereby calling for appropriate treatment of QMRA output uncertainty (Beaudeau et al., 2015). Westrell et al. (2004) proposed the first published hazard analysis and critical control points (HACCP<sup>x</sup>, see section 3.4.1) approach for the anaerobic digestion process (no HACCP/QMRA composting frameworks were found in the literature). As a first step, exposure scenarios were defined by Westrell et al. (2004), and ranked in order of severity. These scenarios were based on standard operating conditions. As pathogen die-off is hard to assess during the process, pathogen concentrations were considered to not have changed from those measured in the initial waste. Inhalation exposure was considered to be the most hazardous. The ingestion exposure frequency was also estimated. Workers at the digestion plant were considered the most likely to ingest particles (1-5 g/year), followed by those involved with the spreading of the digestates as fertilizer. The

consumption of raw vegetables grown on digestate-amended soils was shown to have a low pathogen exposure risk, but could lead to some infections. A major difficulty with this first HACCP proposal was the great diversity (parasites, bacteria and viruses) and variations in numbers of pathogens in the OW. Specifying the microbial pathogens that could occur in such OW is thus a key yet hard to determine factor. However, their presence appears to be highly stochastic. This is likely because of the overall lack of knowledge on the origin and nature of OW involved in such processes. Nevertheless, this first scheme suggested that gastrointestinal viruses represented the highest risk. According to this study, all workers taking part in OW handling would be exposed to these viruses, and could thereby regularly suffer from a related infection. However, these workers did not seem to have used suitable recommended protection materials to reduce exposure risks. This specific situation is not representative of actual practices. Enterohemorrhagic *E. coli* was found to be the second most likely pathogen that could infect these 'unprotected' workers. This first HACCP framework for OW treatment identified major critical points that would need to be considered in any HACCP framework. In anaerobic digestion, pathogen inactivation is directly dependent on the four main transformation steps, which drastically modify the physicochemical conditions in the digester. A major critical HACCP to be assessed would thus be the proper functioning of each microbial functional group. Any deviation in these groups and in their synergistic functioning could result in failure of the digestion process, and would constitute a strong warning of the risk of production of hygienically inadequate treatment products. Another key critical point is the duration of the digestion processes. A mesophilic anaerobic digestion process must run at a temperature of 34-42°C for at least 50 days, while a thermophilic process has to run at 50-65°C for at least 30 days (JORF, 2017). pH is also a major critical point and must be between 7 and 8.5 to ensure proper functioning of the digestion process. Regarding the composting process, critical HACPP is the number of days (which should be >6 according the requirements in most countries [e.g. Hogg, 2002]) at a minimum temperature of 65°C. Furthermore, as the level of pathogenic bacteria remaining at the end of the

composting process is dependent on the pH value (should be >8), this critical parameter should be monitored.

No models for biological contaminants have been developed so far to assess the impact of OW recycling on human health. However, as mentioned by Ashbolt et al. (2013), “environmentally derived antibiotic-resistant bacteria may adversely affect human health (e.g. reduced efficacy in clinical antibiotic use, more serious or prolonged infection) either by direct exposure of patients to antibiotic-resistant pathogen(s) or by exposure of patients to resistance determinants and subsequent horizontal gene transfer [...] to bacterial pathogen(s) on or within a human host”. A quantitative model would need to be developed to assess the risk of transmission of resistance to humans (i.e. infection by antibiotic-resistant bacteria/pathogens). The overall risk for ARB and ARG can be estimated through outputs resulting from the combination and integration of all **microbiological risk analysis** (MRA) steps, namely hazard identification, hazard characterization and exposure assessment, based on the WHO framework used for food safety purposes (WHO, 2010), which is not commonly used in RA. This MRA approach for assessing the risk of antibiotic resistance transmission from the environment to humans has been well described by Manaia (2017). MRA applications performed to estimate the adverse health effects of resistant faecal bacteria on human populations exposed to recreational waters (Limayem and Martin, 2014) or of resistant *E. coli* in lettuce attributable to irrigation water and gene transfer (Njage and Buys, 2017) are exemplary examples.

**Quantitative risk assessment** (QRA) of antibiotic resistance associated with the application of OW treatment end-products on agricultural soils could require: i) determination of the level and dynamics of prioritized ARG in the raw OW, in treated OW and then in soil after treated OW spreading, ii) assessment of flows of these ARG between ARG-bearing non-pathogenic species to pathogens and the extent of occurrence in the various environmental compartments, and iii) determination of the extent to which abiotic and biotic parameters control horizontal gene transfer of these ARG and/or the selection and growth of ARG-bearing pathogens. Antibiotic resistance may

be measured within an environmental matrix by detecting and quantifying ARB and/or ARG using culture-dependent and -independent methods. The former rely on bacterial extraction on selective (i.e. single or multiple antibiotic addition) or non-selective media, with or without further species and ARG identification. The latter involve total DNA extraction from the environmental sample and quantitative polymerase chain reaction (PCR) targeting ARG, or metagenomic analysis. The culture-independent approach is being used to an increasing extent as it is less time consuming and provides a broader view of ARG diversity. Single-genome and metagenome analysis have revealed the tremendous amount and diversity of ARG and their widespread distribution across bacterial phyla and environments, including OW, treated products and soil. Changes in ARG abundance over time have been studied using quantitative PCR targeting a small fraction of ARG related to specific antibiotic families (i.e. frequently *sul*, *tet*, *bla* and *erm* associated with resistance to sulfamide, tetracycline, beta-lactam and macrolide, respectively), or hundreds of antibiotic families using high-throughput qPCR. These approaches might, however, overestimate antibiotic resistance as bacterial hosts might carry an ARG without having a resistant phenotype as expression is a matter of host and gene location on the genome. Furthermore, it is likely that not all ARG are equivalent regarding risks to human health. A classification could be based: i) on those conferring resistance to last resort antibiotics and/or highly virulent pathogens, and ii) those easily transferable and/or associated and frequently co-transferred with high ARG counts (Martínez et al., 2015; Oh et al., 2018). Zhang et al. (2019) recently used an empirical approach to categorize the risk of ARG to human health based on three factors, namely anthropogenic enrichment, mobility and host pathogenicity. They ranked 4 050 known ARG in four categories and prioritized 3% (i.e. 132) of them in Rank I (the most at risk of dissemination amongst pathogens) and 0.3% in Rank II (high potential emergence of new resistance in pathogens). The authors observed that “Rank I was strongly correlated with the potential exposure of clinical antibiotics, while Rank II was associated with industrialized lifestyles”.

### **3 Gaps in environmental assessment methods and empirical knowledge regarding contaminants in organic waste treatment and agricultural recycling**

### 3.1 Empirical knowledge on contaminants in organic waste

A major limitation in the environmental assessment of contaminants is that the development of assessment methods lags behind advances in knowledge in analytical research fields. The effects of contaminant **speciation** have not yet been accounted for in assessment methods. For instance, Schaubroeck et al. (2015) investigated the impacts of sludge-borne trace elements during agricultural recycling via USEtox and USES-LCA. By assuming that trace elements are exclusively present in ionic form, the authors estimated a highly theoretical human toxicity effect. Recent empirical research reported findings on a wide range of effects of agricultural reuse of digestate and compost. As typical examples, Hargreaves et al. (2008) performed an analysis on the findings of 30 case studies on municipal solid waste composting, while Brändli et al. (2007) characterized a high variety of organic contaminants in compost and digestate. Both studies overlooked the dynamics of these pollutants after soil applications. Du and Liu (2012) highlighted the potential ecological risks and public health threats of antibiotic pollution in soil–vegetable systems, and reviewed research on the fate and ecological risks of antibiotics. Empirical research has been focused especially on the following issues: changes in soil physical and biological properties, and trace element and persistent organic contaminant loading (Hargreaves et al., 2008); contaminant biodegradation and bioavailability (Smith, 2009a, 2009b); pathogen risks of manure and biosolids (Brooks et al., 2012); characteristics and risks of biosolid use (Uggetti et al., 2012); accumulation and risk of pharmaceuticals and PPCP (Chen et al., 2014); and ecotoxicological impacts (Pivato et al., 2015). A substantial corpus of knowledge has thereby been collected to date, even concerning pollutants that are emerging like pharmaceuticals, notably present in urban sewage sludge (Mailler et al., 2014). A recent review of experimental results (Insam et al., 2015) discusses the effects of the digestion process on digestate properties. It provides an exhaustive overview of existing findings concerning all digestate-borne substances and the pathways through which soil fertility might be affected following digested OW spreading as compared to raw OW. The authors do not provide clear evidence on the fate and risk of contaminants, although they suggest that a reduction in trace element solubility and availability

upon digestion would affect the subsequent fate of these compounds. The review also demonstrates the disparity of knowledge available on various compounds, as well as the difficulty of drawing general conclusions in the absence, in many cases, of a sound understanding of the mechanisms involved. The review concluded that, in comparison to raw OW, the pros of recycling digestate outweigh the cons. Nkoa (2014) stresses the importance of the application conditions and timing, non-respect of which could induce various types of hazard. Surprisingly, this review overlooks organic contaminants. Bonetta et al. (2014) performed an exhaustive experimental study, but again surprisingly overlooked organic contaminants and, moreover, their experimental setup did not account for contaminant mobility in soil beyond plant uptake. Teglia et al. (2011a; 2011b) found that post-treatment of digestate with composting would reduce risks and largely enhance the agronomic value of feedstock compared to conventional composting. Table 5 lists some of the most representative reviews of empirical research-based knowledge on the fate of contaminants in OW following soil application. These reviews showcase the frontiers in empirical research, including the importance of the condition of contaminants (trace element speciation and organic contaminant metabolites) on their fate. A critical view of the comprehensiveness of contaminant consideration in environmental assessment is presented in the following subsections.

[Table 5]

### **3.2 Trace elements in LCA and RA: limits and solutions**

Fantke et al. (2018) proposed a harmonized framework for the improvement of ecotoxicity assessment in LCIA. They recognized that trace elements deserved special attention in the light of both the enhanced knowledge on the environmental fate of trace elements and the major impact of trace elements on ecotoxicity results in LCIA. Moreover, the need for a terrestrial ecotoxicity model has been pointed out (Aziz et al., 2018; Fantke, 2019; Fantke et al., 2018). The following aspects of biophysical processes deserve more specific attention: i) chemical properties of OW, ii) physicochemical processes that drive trace element availability in soil, and iii) the ways soil organisms influence trace element bioavailability in soil. The following subsections discuss the relevance of

these aspects and propose modelling approaches that could be suitable for their inclusion in LCA and RA.

### **3.2.1 Chemical properties of organic waste altering trace element availability in soil: trace element speciation in organic waste**

As underlined in section 2.3.2, one approach has already been shown to improve ecotoxicity assessment by accounting for the trace element origin (Owsianiak et al. 2015). For instance, the ACF of trace elements from atmospheric emissions was differentiated from that from organic sources, encompassing a broad range of OW such as biosolids, manure, compost and treated wastewaters. Trace element speciation and availability in all of these OW have so far been viewed as homogeneous in this approach, with trace elements mainly considered to be bound to organic matter. Yet recent research has shown that trace element speciation and availability in OW (which may be related to the ACF) can vary substantially according to the trace elements, OW solid or liquid state and OW treatment.

Two methods are mainly used to study trace elements in OW, namely chemical extraction and X-ray absorption spectroscopy (XAS). Chemical extraction is widely used, as it is inexpensive, easy to apply and highly reproducible (Bacon et al., 2005). Concerns regarding the non-selectivity of reagents (Doelsch et al., 2008, 2006) or readsorption of elements following release are frequently reported (Martin et al., 1987). However, this method is well suited for assessing available and exchangeable fractions of a substantial number of trace elements in various OW (Doelsch et al., 2010). For example, Flyhammar (1998) studied Zn, Cu, Pb, Cr, Ni and Cd in municipal solid waste and sewage sludge and highlighted that the exchangeable fraction (ammonium acetate extraction) accounted for 1–20 % of the total trace element concentration. XAS is used to a much lesser extent than chemical extraction as synchrotron beamtime is limited. XAS is nevertheless a powerful tool that enables determination of trace element speciation in complex samples. XAS is notably able to distinguish trace element species such as metal sulfides that otherwise would be overlooked by chemical extraction (Legros et al., 2017). Recent studies highlighted that trace element speciation in OW is trace elements-dependent. For instance, Zn is mainly related to OW inorganic phases (Zn sulfide,

amorphous Zn phosphate and Zn sorbed to iron hydroxide), whereas Cu is mainly associated with organic matter (Donner et al., 2011; Legros et al., 2010). But this general pattern is affected by the physicochemical storage conditions (i.e. dry matter and oxidation-reduction potential) and by the OW treatment (i.e. composting, anaerobic digestion). Liquid OW in suboxic to anoxic redox conditions tend to have a majority of reduced trace element species. For example, Zn and Cu occur mainly or entirely as sulfides, with Cu in the form of reduced Cu(I) in swine slurry (Formentini et al., 2017; Legros et al., 2010). Only oxidized trace element species are present in solid OW under oxic redox conditions —Zn phosphate, Zn sorbed to iron hydroxide and Cu(II) complexed to organic matter are the major species in biosolids (Donner et al., 2011). Anaerobic digestion has been found to always favour nanosized Zn sulfide (nano-ZnS) formation (>70% of zinc in digestate) (Le Bars et al., 2018; Legros et al., 2017; Lombi et al., 2012). However, nano-ZnS becomes a minor species (<10% of zinc) after 1–3 months of OW composting. In compost, Zn is consequently mostly present as amorphous Zn phosphate, with Zn sorbed to ferrihydrite (Le Bars et al., 2018; Lombi et al., 2012). The relationship between trace element speciation in OW and trace element availability in OW-amended soils was further investigated in short- and long-term studies. Tella et al. (2016) studied single *ex situ* soil application of six OW mainly stabilized under oxic conditions. They put forward the hypothesis that the increased Zn availability measured with the diffusive gradient in thin films (DGT) technique in OW-amended soil could be explained by the desorption of Zn sorbed to iron hydroxide in OW. In a long-term field experiment (22 pig slurry applications), Formentini et al. (2017) observed that ZnS, which had accounted for 100% of Zn speciation in swine slurry, was not detected in the amended soil despite a nearly 2-fold increase in the soil Zn concentration due to swine slurry spreading. Instead, the initial Zn speciation (i.e. Zn bound to kaolinite and Fe oxyhydroxide) in the un-amended soil remained predominant in the amended soil along with organic matter-bound Zn accumulation. The authors hypothesized that the ZnS initially present in the swine slurry was weakly stable under the soil oxic conditions, thereby explaining the radical change in Zn speciation after soil application. This hypothesis was reinforced by the findings of Le Bars et al. (2018) who

demonstrated that OW-borne ZnS was present as nanosized ZnS, which is much less chemically stable than the macroscopic counterparts. Contrary to Zn, Cu speciation in OW was mainly dominated by organic matter-bound Cu (Donner et al. 2011; Tella et al. 2016; Legros et al. 2017). In a study of single *ex situ* soil application of six OW, Tella et al. (2016) found that the increase in Cu availability measured by DGT in the OW-amended soil was linearly correlated with the mineralization potential of the OW-bound organic carbon. Accordingly, the authors suggested that the release of available Cu in OW-amended soils could be driven by the mineralization of organic matter from OW. Trace element speciation in OW is the main driver controlling the fate of this element in amended soil (Hodomihou et al., 2020). Clearly, trace element speciation must be taken into account to gain an overall understanding of the environmental fate of trace elements. Chemical extraction has already been used in RA and LCA (e.g. Tarpani et al. (2020)) but this approach is not well suited for determining trace element speciation in OW, as we discussed above. It would, however, not be feasible to routinely determine trace element speciation in OW via XAS. An alternative option could be to develop a qualitative typology of OW based on basic OW characteristics (e.g. OW nature and treatment) related to physicochemical parameters such as the pH and redox potential which drive trace element speciation in OW. It would also be essential to consider an indicator based on the mineralization potential of OW organic matter in this typology. Considering the broad diversity of OW treatments, the mineralization potential of organic matter over time is very heterogeneous between OW (Thuriès et al. 2002). An indicator of residual organic carbon remaining in amended soils ( $I_{ROC}$ ) based on some OW biochemical properties and the short-term mineralization of organic carbon has been developed for the main types of OW applied to soils (Bouthier et al., 2009; Lashermes et al., 2010, 2009). This typology could be further used to develop a correcting factor to the availability of trace elements estimated with the generic methodology used for assessing environmental risk.

### **3.2.2 Soil physicochemical processes driving the trace element fate in soil**

OW application on soil induces a net increase in the soil trace element concentration and can lead to trace element export, in soluble form, via water drainage depending of the type of trace elements.

Yet this trace element export has a relatively low net impact. Total trace element export by drainage always represents a few percentage of the OW trace element inputs (Cornu et al., 2001; Formentini et al., 2015; Hodomihou et al., 2020; Legros et al., 2013; Lekfeldt et al., 2017; Richards et al., 1998).

#### ***Colloidal transfer for trace element leaching prediction***

Colloidal transfer is suspected to facilitate trace element transport through the soil to groundwater.

Colloids are generally characterized by dispersed particles with average diameters of 1 nm to 10 µm.

According to Kretzschmar et al. (1999), four key conditions are required for colloid-facilitated contaminant transport to become environmentally significant: i) mobile colloidal particles must be present in sufficiently high concentrations, ii) the particles must be transported over long distances through uncontaminated zones of the porous medium, iii) the contaminants must sorb strongly to the mobile particles and desorb slowly, and iv) the contaminant must be toxic at low concentration so that even trace concentrations in groundwater cannot be tolerated.

Contaminants that meet the latter two criteria notably include trace elements. For example, nanosized metal oxide (including ferric, manganese, aluminium, titanium, magnesium and cerium oxides) provide high surface area and specific affinity for trace element adsorption (Hua et al., 2012).

Regarding the first criterion, many studies have shown the presence of colloids in soils. For example, clays minerals that make up the colloid fraction of soils, sediments, and water. There is also evidence that colloids are present in OW. For example, nano-ZnS is a major Zn species in raw liquid OW (Kim et al., 2014; Le Bars et al., 2018), and organic colloids have been observed in biosolids (Ghezzi et al., 2014).

Regarding the second criterion, soil colloids can transfer trace elements to groundwater. According to Kretzschmar and Schäfer (2005), under certain conditions, colloid facilitated transport can become the major transport mechanism of strongly sorbing trace elements in soils and aquifers. This was

demonstrated for Pb transport in water-saturated soil columns (Grolimund et al., 1996), for Cu, Zn and Pb in biosolid amended soils (Karathanasis et al., 2005), and more recently for Cu, Cr and Pb in a cattle slurry amended soil (Lekfeldt et al., 2017).

In almost all fate modelling approaches for environmental risk assessment, trace element leaching is conceptualized with two compartments —the immobile solid matrix phase and the mobile aqueous solution phase. A third compartment is considered in the conceptual model when colloidal transport is considered (Mccarthy and Zachara, 1989). Mallmann et al. (2017) suggested accounting for colloid-facilitated transport of trace elements, increasing the range of transport processes considered in HYDRUS 1D simulation of Cu and Zn transport in amended soil. However, models that consider such processes, such as the HYDRUS-1D C-Ride module, require many additional parameters, such as those characterizing trace element sorption on colloids, which have to be measured experimentally. Whether colloid facilitated transport of trace elements should be considered in fate modelling and in environmental risk assessment depends on the added value vs additional uncertainty and complexity trade-off.

### ***Variability in dissolved organic matter binding properties***

The most advanced approaches used to calculate BF in LCA (sections 2.2.2 and 2.3) and several RA approaches (section 2.4) estimate soil organism exposure to trace elements through the determination of trace element speciation in soil solution. These approaches all directly or indirectly consider that the binding properties of dissolved organic matter (DOM) to trace elements are a major driver of trace element speciation in solution, in line with the findings of specialists in this field (e.g. Weng et al., 2002). Moreover, between-soil variations in trace element binding to DOM are considered to be solely dependent on the DOM concentration, not the DOM binding properties (i.e. the density of binding sites and their binding affinity towards trace elements). This has led scientists conducting ERA to use geochemical models, e.g. WHAM, with the benchmark fulvic acid binding properties representing those of DOM (e.g. Lofts et al., 2013b; Tipping et al., 2003). Yet research focused on DOM binding properties in the past decade have shown that the binding properties of

DOM to trace elements can vary considerably in soil and freshwater (Chen et al., 2018; Li et al., 2017; Weng et al., 2002). This has firstly been attributed to the substantial proportion of DOM corresponding to non-humic substances with binding properties that differ from that of benchmark fulvic acid (Baken et al., 2011; Groenenberg et al., 2010; Ren et al., 2015), and secondly to the variability in DOM properties such as its aromaticity, which seems to partly account for DOM binding properties (Amery et al., 2008; De Schampheleere and Janssen, 2004). Djae et al. (2017) studied more than 50 soil samples exhibiting very contrasted physicochemical properties and confirmed that WHAM parameterized without accounting for DOM binding property variability skewed the prediction of  $\text{Cu}^{2+}$  activity in soil solution by up to three orders of magnitude, while also showing that the prediction of Cu toxicity to microbial functions, invertebrates and plants in soils was substantially skewed as well. Laurent et al. (2020) recently studied dozens of field samples of soils that had been amended or not with OW over a decade and showed that adequate WHAM prediction of Cu (but not Zn) speciation in soil solution required DOM binding property optimization. When considering the chemical properties of trace elements, it is thus crucial to account for the variability in the binding properties DOM to trace elements exhibiting a high affinity for organic matter (e.g. Cu and Pb), except with regard to trace elements exhibiting a much lower affinity for organic matter (e.g. Cd, Ni and Zn).

The point is to know how, in practice, to account for DOM binding property variability in RA and LCA approaches. The only recommendation the specialized literature to date is to perform additional analytical determinations. Among these, specific UV-absorbance and fluorescence analysis of soil solution samples seem to be the only analytical determinations strategy that could be routinely applied to a high number of samples while, in addition, revealing some correlations with DOM binding properties that could be optimized in geochemical models (Amery et al., 2008; Baken et al., 2011; Zhang et al., 2020). Although specific UV-absorbance was found to be a better candidate from a practical standpoint and Amery et al. (2008) showed that specific UV-absorbance was a good proxy when assessing a soil receiving a single low OW application, Baken et al. (2011) and Laurent et al.

(2020) suggested that specific UV-absorbance would be a much less relevant proxy for DOM binding properties in freshwater and soil receiving high repetitive OW inputs. When developing a method for RA of Pb bioavailability in a lake, Zhang et al. (2020) also underlined the importance of accounting for the variability in the binding properties of DOM to Pb and that fluorescence analysis could be helpful in this respect. These authors thus developed a multilinear regression model based on fluorescence and usual water chemical parameters to predict Pb binding to DOM in the target lake at a regional scale. In the light of this example, we therefore stress the need to develop such regression models to estimate trace element binding to DOM in soil. These regression models could then be applied in site-specific RA, providing that fluorescence analyses could be done on each specific investigated site. Such analytical determinations would, however, not be feasible in LCA context. A second step would thus be necessary and could, for instance, consist of developing an empirical regression model to predict the fluorescence properties of DOM with conventionally determined soil physicochemical parameters, as previously carried out in LCA to predict DOC concentrations (Owsianiak et al. 2013). Such regression models have yet to be developed and would therefore deserve further attention.

### ***Kinetics of soil physicochemical processes***

In all RA and LCA approaches, physicochemical processes that drive trace element availability in soil are assumed to occur under equilibrium conditions, and thermodynamic constants are used to determine the solid-solution partitioning and the proportion of each trace element chemical species (Degryse et al., 2009a; Weng et al., 2001). There is, however, a growing body of evidence that when trace elements interact with a soil organism the physicochemical process kinetics can become a major driver of trace element availability in soil (Almås et al., 2004; Bade et al., 2012; Bravin et al., 2010a; Nolan et al., 2005). Although the free trace element species is still considered to be the main (if not sole) trace element species taken up by soil organisms under kinetically-driven conditions, the expected depletion of free trace element activity in solution at the soil-organism interface means that trace element uptake would no longer be driven by the trace element activity. It is instead driven by the ability of soil (i.e. trace elements bound to the solid phase and to ligands in solution) to

resupply free trace element species in solution, which depends on the diffusion limitations and dissociation kinetics (Degryse et al., 2012). Accordingly, new soil tests were developed to determine the extent to which soil could resupply trace elements in solution under kinetic limitations. Among these, DGT seems particularly promising (Davison and Zhang, 2012; Degryse et al., 2009b; Zhang and Davison, 2015). Kinetic considerations could therefore be integrated in RA and LCA approaches by developing an empirical regression model to predict DGT measurements with conventionally determined soil physicochemical parameters (Owsianiak et al., 2013). Such empirical regression models have already been designed in specific case studies (Bravin et al., 2010b; Guan et al., 2018), but would now be developed on a broader range of soils. Note that estimation of trace element availability in soil via a tool like DGT would require modification of the mathematical expression of exposure in RA and LCA approaches. In LCA, for instance, the exposure factor —the ACF x BF product— is currently expressed as the mass ratio of free to total trace elements in soil. Estimating the exposure factor with DGT would lead to expression of the ACF x BF product as the mass ratio of all DGT-available soil-borne trace elements. Consequently, EF should also be expressed as a function of DGT-available trace elements. Ecotoxicological endpoints based on DGT-available trace elements have already been determined, but only for one trace element (i.e. Cu) and one toxic endpoint (i.e. tomato shoot growth) (Zhao et al., 2006). Empirically deriving new ecotoxicological endpoints for a meaningful range of trace elements, soil organisms, soil types and toxic endpoints is a huge tedious task that has already been consistently carried out for numerous ecotoxicological endpoints based on total and free trace element concentrations in soil (Hamels et al., 2014; Lofts et al., 2013a; Thakali et al., 2006b). Toxic endpoints based on DGT-available trace elements could thus be derived from toxic endpoints based on total or free trace element concentrations by using the empirical regression models suggested above to predict DGT measurements from conventionally determined soil physicochemical parameters.

### ***Effects of repetitive long-term organic waste applications***

Organic waste is usually applied on soils annually over the long-term and seldom only once. Repetitive long-term OW application has three major consequences on trace element availability in soil that have yet to be accounted for in RA and LCA approaches. The first is related to the aging process, which usually tends to decrease trace element availability in soils over time. This aging process is, however, likely counteracted by fresh trace element inputs following each new OW application. Accordingly, Jensen et al. (2018) suggested an RA approach in which the availability of newly added trace elements would be higher in comparison to the availability of trace elements added over a year earlier. This factor should also be incorporated in the LCA approach by deriving an ACF through the integration of individual ACF values calculated at each time step (i.e. each OW application) over the whole LCA time scale (typically infinite). The second consequence is related to the specific pattern of trace element availability in soils contaminated by long-term OW application in comparison to trace element availability in soils contaminated by other trace element sources. Smolders et al. (2012) showed that trace element availability in soils at 22 sites where OW had been applied on soils for 1–112 years was on average 5.9-fold lower than in the corresponding unamended soils spiked with Cu salt and pH adjusted in the laboratory so as to have a total Cu concentration and pH similar to the levels found in OW-amended soils. Such a protective factor could thus be applied to the Cu availability value calculated in RA and LCA approaches. Although this kind of protective factor has not been derived for any other trace elements, a similar protective effect was observed for Cd by Kukier et al. (2010), who showed that the increase in plant Cd uptake due to Cd spiking under laboratory conditions was lower in soils amended with OW for 20 years than in the corresponding unamended soils with adjusted pH. The derivation of protective factors for a range of trace elements would thus deserve further investigation for potential incorporation in RA and LCA approaches. The third consequence of the repetitive and long-term OW application on soil is the fact that, in addition to trace elements, OW also supplies many other types of organic and inorganic matter. This regular supply of additional materials usually ultimately leads to the modification of some important physicochemical properties in soils, such as pH and the organic matter concentration and binding

properties (Nobile et al., 2018; Senesi et al., 2007; Smolders et al., 2012). These soil properties are known to highly influence trace element availability in soils (Degryse et al., 2009a; Sauvé et al., 2000; Weng et al., 2001), so their modification is expected to gradually alter the trace element availability in OW-amended soils over time. Laurent et al. (2020) recently obtained clear evidence of this effect. In three field trials, they showed that the increase in pH and in the DOM concentration, aromaticity, and binding properties induced by OW application over a decade mitigated Cu and Zn availability in amended soils despite a concomitant increase in the soil contamination level. This is consistent with the findings of Alvarenga et al. (2017), who showed that sewage sludge application promoted a significant increase in the plant concentration of Cu, Ni and Zn, which otherwise did not occur in association with compost application. This could be explained by the fact that sewage sludge application induced a decrease in soil pH, which was not observed following compost application. To be able to account for temporal changes in pH and organic matter properties in OW-amended soils in RA and LCA approaches, it would be necessary to be able to predict pH and organic matter patterns over time with routine models. Apart from predicting temporal changes in soil organic matter concentrations, such models have yet to be developed and future research should consequently be focused on overcoming this issue.

### **3.2.3 Influence of soil organisms on trace element availability and bioavailability**

Living organisms at risk of soil contamination are not passive trace element samplers, but instead may have a biological effect by substantially altering the soil physicochemical parameters and actively regulating trace element interactions with biological surfaces and by consequence trace element uptake. We identified four aspects for which RA and LCA approaches could better account for this influence of soil organisms on trace element availability in soils and consequently on trace element bioavailability and toxicity to soil organisms.

The most advanced RA and LCA approaches use the biotic ligand model to link trace element speciation in solution to trace element toxic effects on aquatic and soil organisms (Niyogi and Wood, 2004; Thakali et al., 2006b, 2006a). Despite the obvious assets of the biotic ligand model, there is

high variability in the binding constant numbers and values thus defined in comparable experiments (Ardestani et al., 2014), thereby questioning whether the biotic ligand model parameterized in this way would be sufficiently generic for operational application in RA and LCA approaches. The recent development of a more complex biotic ligand model formalism for predicting the toxic effects of trace element mixtures has further fuelled the debate on this issue. To deal with this complexity, Tipping and Lofts (Tipping and Lofts, 2015, 2013) suggested using WHAM to predict not only trace element speciation in solution but also trace element binding and toxicity to aquatic and soil organisms using the humic acid profile defined in the WHAM database as a surrogate of biological surfaces. Coupling the WHAM default parameterization with a toxicity function ( $F_{\text{TOX}}$ ), the WHAM- $F_{\text{TOX}}$  approach was found to successfully predict the toxic effect of trace element mixtures for a range of aquatic and soil organisms (Balistreri and Mebane, 2014; Guigues et al., 2016; He and Van Gestel, 2015; Qiu et al., 2016; Yen Le et al., 2015). This WHAM- $F_{\text{TOX}}$  approach therefore seems promising for ecotoxicological assessment in RA and LCA approaches.

Soil organisms are known to interact with the surrounding soil, e.g. the so-called rhizosphere for plants and the drilosphere (bioturbated soil) for earthworms. Plants excrete protons or hydroxyls, thereby changing the rhizosphere pH by up to several orders of magnitude compared to the bulk-soil pH (Hinsinger et al., 2003). Plants also exude a broad range of small molecular weight organic molecules, some of which are able to bind trace elements while the rest are known to increase microbial activity in the rhizosphere (Jones et al., 2009). Such plant-induced changes in the physicochemical and biological properties of the soil are known to alter the trace element availability in the rhizosphere compared to the bulk soil, and consequently the trace element bioavailability and toxicity to plants (Bravin et al., 2012; Michaud et al., 2007; Wenzel et al., 2011). Earthworms have also been found to be able to alter the pH, DOM concentration, microbial populations and consequently the trace element availability in their drilosphere (Sizmur and Hodson, 2009).

Ecotoxicological tests are able to account for the effects of the biological response of soil organisms on the soil physicochemical and biological properties, but estimates of soil organism trace element

exposure do not. This is because the latter estimates are based on measurements of trace element availability and speciation in bulk soil rather than in the volume of soil impacted by the presence of soil organisms. To the best of our knowledge, no models are available that could be operational in current RA and LCA approaches. Some biotests, such as the RHIZOtest for plants (Bravin et al., 2010b), have nevertheless been developed to specifically account for this influence of soil organisms in the assessment of trace element bioavailability to soil organisms. Such biotests could first be used to assess the extent to which this influence of soil organisms could skew current soil ecotoxicological assessments, before designing a way to use these biotests in RA and LCA approaches.

Conventional ecotoxicology methods to derive ecotoxicological endpoints used in RA and LCA approaches are based on dose-response curves regarding soil organism exposure to single trace elements or mixtures. In these experimental procedures, only the trace element exposure level is expected to change. This context differs markedly from that of soil organism exposure to OW-amended soils, as OW brings a mixture of organic and inorganic contaminants but also alters the soil physicochemical environment overall (see above). Some statistically significant relationships may be noted between some soil parameters and the measured toxic effect when soil organisms are exposed to increasing OW application rates, but it is very difficult to identify the soil parameters to which the measured toxic effect could be mechanistically related. It would therefore be highly challenging to design mechanistic models able to predict the toxicity of given trace elements to organisms in OW-amended soils. This issue could first be addressed by no longer relating the measured toxic effect to the trace element availability (or that of any other soil contaminant), but rather more empirically relating it to the soil OW application rate. Application of this strategy in RA and LCIA has been supported by the findings of Pandard and Römbke (2013), Huguier et al. (2015), Pivato et al. (2015) and Renaud et al. (2019). This issue could alternatively be addressed by relating trace element exposure to trace element bioaccumulation in soil organisms, since this bioaccumulation is considered as a prerequisite for any toxic effect. Such biotests are already being used to assess trace element bioaccumulation in plants and soil macroorganisms such as earthworms

(Beaumelle et al., 2016; Bravin et al., 2010b). For microorganisms, quantitative measurement of trace element-resistant genes is currently a promising approach for relating biological responses to the exposure to specific soilborne trace elements (Miranda et al., 2018; Roosa et al., 2014). The potential benefits of such biotests should be first assessed in the specific context of OW-amended soils before being proposed as new ecotoxicological endpoints in RA and LCA approaches.

Among the range of trace elements added to soil by OW application, some trace elements (i.e. B, Cu, Mo, Ni, and Zn) can have a beneficial effect on soil organisms by serving as micronutrients at a low concentration before generating eventually a toxic effect at a higher concentration. This beneficial effect of trace elements has yet to be accounted for in RA and LCA approaches. The UNEP-SETAC Ecotoxicity Task Force addressed this issue and decided for the moment not to consider such beneficial effects in ecotoxicity characterization in the USEtox framework (Fantke et al., 2018), but suggested that future research should focus on this issue.

### **3.3 Organic contaminants in LCA and RA: limits and solutions**

As for trace elements, the fate of OW-borne organic contaminants highly depends on the OW composition and speciation, as well as on the nature of contaminant binding in soils upon which OW is applied. The nature of this binding will affect: the persistence of soilborne contaminants, their mobility from amended soil into water and their bioavailability to plants and animals. In contrast to trace elements, organic contaminants are degradable but the possibility of degradation is very variable. Their biotic or abiotic breakdown entails the transformation of known parent molecules into sometimes unknown, and often scarcely studied, transformation products. Certain categories of organic contaminants, moreover, form strong bonds with organic particles in OW or in the soil, giving rise to 'bound residues' (non-extractable residues) which cannot be quantified by the chemical extraction methods used to measure contaminant levels. Total organic contaminant levels in OW are consequently not always known. Organic pollutants are often divided into two major groups according to their apparent half-life in soil: 1) persistent contaminants (apparent half-lives ranging from one to several years: PCB, PAH, dioxins, flame-retardants, etc.) and 2) non-persistent

contaminants, whose dissipation is more rapid (half-lives ranging from a few days to a few months: phthalates, bisphenol A, detergents, etc.). This distinction is usually accounted for in a first approach through dissipation half-life time parameters but also involves the consideration of non-extractable residue formation in process-based models describing the fate of organic contaminants.

Improvements to existing RA approaches, although being limited by empirical data collection costs, is possible through a combination of different modelling approaches. *In silico* approaches based on QSAR knowledge can be used to classify organic contaminants according to their potential behaviour and effects. For instance, TyPol (typology of pollutants) is a suitable classification method (Servien et al., 2014). It is based on statistical analyses combining several environmental (i.e. sorption coefficient, degradation half-life, Henry constant) and ecotoxicological (i.e. bioconcentration factor, EC<sub>50</sub>, NOEC) parameters, as well as structural molecular descriptors (i.e. number of atoms in the molecule, molecular surface, dipole moment, energy of orbitals). Molecular descriptors are calculated using an *in silico* approach. Environmental and ecotoxicological parameters are extracted from available databases and the literature (Servien et al., 2014). TyPol has so far mainly focused on pesticides and few transformation products. Recent improvements in TyPol have addressed the implementation of pesticide ecotoxicological endpoints (Traoré et al., 2018), prediction of the environmental fate of putative or recently detected transformation products (Benoit et al., 2017; Storck et al., 2016), and the expansion of the database to encompass pharmaceuticals and personal care products and some of their transformation products. This approach is generic but not sufficient for RA since it cannot account for the environmental specificity (pedoclimatic conditions, OW characteristics and agricultural practices), or for the organic contaminant concentrations, forms and bioavailability. However, the method may initially help rank organic contaminants in different clusters, each with relatively similar behaviour or impact. Therefore, it could help select representative molecules from each cluster so as to be able to conduct more comprehensive studies based on analytical studies or biophysical modelling, as described below.

### **3.3.1 Interactions between organic matter and contaminants**

To the best of our knowledge, few models have been developed to assess the fate of organic contaminants during applied OW decomposition in soil, despite the recently demonstrated importance of soil organic matter for soil microbiota that degrade organic contaminants (Neumann et al., 2014). The COP-Soil model describes the fate of organic contaminants during compost decomposition in soil (i.e. mineralization, adsorption/desorption, and formation/remobilization of non-extractable residues) combined with the organic carbon dynamics (Geng et al., 2015). The model uses one module for organic matter and one for organic contaminants, both of which are linked by several common assumptions. The organic C module is based on that proposed by Zhang et al. (2012) which describes organic matter transformation during composting and is adapted to the soil environment. Organic C in compost is divided into several pools: available organic carbon, soluble in neutral detergent, hemicellulose-like, cellulose-like and lignin-like. The organic contaminant module is inspired by the COP-Compost model that was developed to simulate the coevolution of organic contaminants and organic matter during composting (Lashermes et al., 2013), and is based on the same four pools and the same equations. Organic contaminants are distributed among mineralized, water-soluble, adsorbed and non-extractable residue fractions. The model was tested against experimental data and its performance was found to be acceptable (Geng et al., 2015). The model does not consider organic contaminant volatilization because it is assumed that this mainly occurs earlier in the process. Finally, the model does not consider the formation of degradation products nor include leaching or runoff processes. Brimo et al. (2018) modified the approach of Geng et al. (2015) to model the fate of compost-bound PAH in amended soil. Goulas (2006) used the same model to simulate the fate of sulfonamide antibiotics in soil amended by manure and by sewage sludge compost. The model consists of three modules: 1) a biodegradation/adsorption module, where adsorption is based on a bi-phasic first-order kinetic model and where PAH biodegradation can produce CO<sub>2</sub>, metabolites and non-extractable residues (the influence of the temperature and water content on biological processes are also taken into account (Brimo et al., 2016)), 2) an organic carbon module which describes organic matter transformation during composting and compost

decomposition in soil (Geng et al., 2015; Lashermes et al., 2013), and 3) a PAH release module which describes the release of compost-bound PAH during the decomposition of compost applied to the soil. The model assumes that PAH is distributed among the organic carbon fractions proportionally to the relative mass of each fraction. One fraction of PAH is taken up in the soil solution, the other is strongly sorbed on the surface of the soil-compost mixture. Following calibration of some parameters, the model gave acceptable predictions of PAH transformation in soils. The most significant features of this model are that it considers the organic matter characteristics of the applied compost, the production of non-extractable residues and the biodegradation route (Brimo et al., 2018b). The model was then mainstreamed in the Virtual Soil Platform (Lafolie et al., 2014) to predict the long-term fate of phenanthren. The performance of the model was tested against field data and found to be acceptable. The model was further used to test different long-term climate change or increased PAH bioavailability scenarios (Brimo et al., 2018a). This model could hence be a useful tool to test various organic contaminants/OW soil application scenarios. In the future, a module incorporating plant functions and compost fertilization will be mainstreamed in the Virtual Soil Platform (Brimo et al., 2018a).

Using the same Virtual Soil Platform, biogeochemical processes occurring during organic contaminant and dissolved organic matter transport in deep soil horizons were included in another model (PoDOC) for the purpose of assessing agricultural soils receiving urban waste compost as organic amendments (Chabauty, 2015). In the PoDOC model, two types of dissolved organic matter have been considered: native dissolved organic matter present in the illuviated<sup>xi</sup> horizon and dissolved organic matter percolating from the surface horizon. The model considers associations between percolating dissolved organic matter and organic contaminants such as pesticides and pharmaceuticals (ibuprofen and sulfamethoxazole) and can simulate reductions in pollutant transport through cosorption processes (Totsche et al., 1997) or accelerations in pollutant transport through cotransport processes (Keren and Communar, 2009).

### **3.3.2 Extending and complementing hazard and exposure models used in LCA and RA**

### ***Plant and animal uptake models***

A major public concern regarding agricultural application of treated wastewater and biosolids is the plant uptake of OW-bound contaminants because it may pose potential human health risks (Christou et al., 2017; Prosser and Sibley, 2015; Wu et al., 2015).

The Biosolids-Amended Soil: Level 4 (BASL4) model calculates the fate of chemicals entering soil in association with contaminated biosolid amendment (Hughes et al., 2005; Hughes and Mackay, 2011). The model simulates the fate of organic chemicals present in biosolids applied to a two-layer soil and addresses the processes of chemical degradation, volatilization, leaching, diffusion and sorption to decaying organic matter (fast- and slow-degrading organic matter). Uptake in invertebrates (worms), small mammals (shrews) and vegetation is simulated according to three complexity levels, namely simple equilibrium partitioning, steady state bioaccumulation and dynamic bioaccumulation. For simple equilibrium partitioning, biota are assumed to achieve the same fugacity as the soil (the assumption is that the plant roots and foliage achieve equilibrium with the soil pore water at all times). For steady state bioaccumulation, the mechanistic bioaccumulation model includes the possible effects of concentration increases by biomagnification and concentration decreases by metabolic conversion, reproductive loss and growth dilution. For dynamic bioaccumulation, a dynamic mechanistic model simulates the time course of uptake and clearance. Amendment is considered as 100% (dry) organic matter, and the chemical and organic matter masses are directly increased upon the addition of chemical or biosolid amendments. The soil properties and chemical distributions between phases are then adjusted accordingly. The increase in the soil water retention capacity with increased organic matter is ignored. Layer depths change because of changes in the quantity of organic matter present, but these changes are considered sufficiently small that diffusion distances remain constant. Leaching and diffusion rates thus remain unchanged throughout the simulation. The model does not simulate runoff. Concentrations in edible tissue predicted by the model can then be used to calculate estimated daily human consumer intake values. Finally, the ratio of the estimated daily intake value to the acceptable daily intake value allows calculation of a risk

coefficient and a risk coefficient  $\geq 0.1$  is considered as an indicator of a potential hazard to human health (Prosser et al., 2014). The model is designed to run for a simulation period of a single growing season because, in its present version, there is no option for entering changing temperatures and other seasonal effects. Multiple runs of the model in which the user manually determines any losses that may occur during the winter months, however, may determine year-to-year accumulation. Alternatively, users may choose to run the model for many months to obtain an estimate of long-term accumulation (Hughes et al., 2005; Hughes and Mackay, 2011). The model assumes that chemicals in the soil are mobile and therefore constantly available to plants, but this assumption is problematic for chemicals that strongly sorb to soil solids. In addition, the model does not take growth dilution or biotransformation into account, and as such are likely to give higher than realistic plant concentrations (Wu et al., 2015). It does not account for sorption of chemicals to root surfaces where equilibrium with the surrounding soil may be achieved, it cannot account for the non-linearity of degradation, and there is considerable uncertainty about parameter values that control partitioning, degradation and transport.

Legind et al. (2012) coupled SimpleTreat 3.1 (Legind et al., 2011) with a model for water and solute transport in soil, which includes a discrete cascade approach for the water balance (tipping buckets model) (Trapp and Matthies, 1998), to simulate the uptake and fate of organic contaminants from biosolid amended soils. The model consists in five soil layers and four plant compartments (root, stem, leaf, and fruit/seed). In each period, the water and substance balance in the five soil layers is solved iteratively while considering precipitation, soil surface evaporation, infiltration, leaching and transpiration (i.e. water uptake from soil by growing plants). Chemical contaminant input via organic amendment is calculated from measured contaminant concentrations in OW and simulated as pulse inputs by the model (Prosser et al., 2014; Trapp and Eggen, 2013). The dissolved concentration is calculated from the chemical distribution coefficient in soil. Chemical uptake in plants occurs via the water taken up by the roots at various depths. It is assumed that the roots grow to where water can be accessed. A similar approach was used to simulate the fate of three ionisable trace chemicals from

human consumption/excretion to the accumulation in soil and uptake in plants following field amendment with sewage sludge or irrigation with river water receiving treated wastewater (Legind et al., 2012; Polesel et al., 2015; Trapp and Eggen, 2013). A comparison with previously published results showed that the model predictions were close to experimental data regarding elimination in sewage treatment plants, concentrations in sewage sludge and bioconcentration factors in plant tissues, for which high variability was nevertheless noted (uncertainty of the combined models). One shortcoming of the plant uptake model is that organic contaminant transformation in plants is not taken into account (Polesel et al., 2015).

### ***Multi-compartment transport models***

A wide range of exposure estimation models can be used to simulate the fate and distribution of substances among the different environmental compartments. These models vary in their complexity and purposes (ECHA, 2016; Kester et al., 2005). However, the models tend to be based on simplified assumptions, and field application of the models must consider heterogeneity issues. Land application of organic contaminants is also often assumed to be random rather than uniform in the models. Several methods can be used to account for this heterogeneity. One way is to run the model under the range of conditions found in the field, with the most conservative (i.e. the lowest thresholds from an RA standpoint) results selected for the target organic contaminant. This generates a conservative model result that may be taken into account when making risk management decisions (Kester et al., 2005). The models most commonly used to assess the environmental fate of pesticides, especially their transfer to groundwater, surface water and sediment, are those used in the European pesticides registration framework: MACRO (Larsbo et al., 2005), PEARL (van den Berg et al., 2016), PELMO (Klein et al., 1997), PRZM (Carousel et al., 2005) for groundwater (FOCUS, 2014, 2000), and STEP1-2 and SWASH—which includes a substance characteristics database (SPIN), spray drift calculations, soil drainage (MACRO), run-off (PRZM) and surface water (including sediment) fate (TOXSWA) models—for surface water and sediment (ECHA, 2016; FOCUS, 2001). These models can be used for any organic contaminants, but they are not able

to represent OW and its interaction with contaminants, and agricultural practices (tillage, organic matter management, etc.) are not fully represented (Mottes et al., 2014). RZWQM is a process-based agricultural management model that includes water, chemical, and heat transport modules; a plant growth module; an evapotranspiration module; an equilibrium chemistry process module; an organic matter-N cycling module; a pesticide fate module; and a management practices module with crop rotation, tillage, irrigation, fertilizer, pesticide and manure applications (Ma et al., 1998; Malone et al., 2004). RZWQM was successfully used to simulate the fate of N in manure and poultry litter amended soil (Feng et al., 2015; Ma et al., 1998), so it might be suitable for simulating the environmental fate of organic contaminants present in manure/OW applied on soil. Lammoglia et al. (2017) combined the STICS (Brisson et al., 2003) crop model and the MACRO (Larsbo et al., 2005) pesticide fate model to simulate the fate of pesticides in complex cropping systems. STICS simulates the decomposition of OW, including farmyard manure, compost and sewage sludge. Enhanced coupling of STICS-MACRO could allow simulation of the fate of organic contaminants present in various OW applied on soil. Finally, inverse modelling showed that the HYDRUS-1D model (Šimůnek et al., 2012) and a two-site sorption model were able to predict transport parameters of phthalate esters from biosolid amended soil in corn cropfields using an inverse solution approach. However, they did not consider the interaction of biosolids with organic contaminants (Sayyad et al., 2017). Different soil structures due to tillage and compost are accounted for in HYDRUS-2D for the purpose of simulating water, Cu and Cd transport in amended soils (Filipović et al., 2016a). A similar approach was used to simulate the impact of compost application on isoproturon transport, although this substance was not added to soil via compost. After a calibration step, the HYDRUS-2D model was found to accurately simulate water and isoproturon fluxes in agricultural soil following repeated urban waste compost application (Filipović et al., 2016b, 2014). The fate of organic contaminants could also be better predicted by accounting for soil heterogeneity at the soil profile scale generated by OW amendment (Vieublé-Gonod et al., 2009). Yet another means to improve the prediction of risk in soil following amendment with OW containing organic contaminants would be to differentiate

non-extractable residues of biogenic and abiotic origin. Indeed, microorganisms incorporate carbon and in some cases carbon from organic contaminants, leading to the production of biogenic non-extractable residues (Kästner et al., 2014). Contrary to non-extractable residues formed from parent organic contaminants or metabolites, biogenic non-extractable residues do not present environmental risks (Kästner et al., 2014). Environmental risks related to the formation of non-extractable residues from organic contaminants are thus often overestimated (Nowak et al., 2013, 2011).

In summary, to improve risk assessment regarding organic contaminants in OW applied on soils, it would be essential to develop/improve process-based models that consider, in the long term: interactions between organic contaminants, the different processes involved in the fate of organic contaminants, such as sorption, degradation, generation of transformation products, volatilization, leaching, uptake, etc., and modifications in soil properties following OW application. Models such as those described here should also be able to represent a diverse range of OW (in terms of quantity and quality). The variability of the parameters/processes described above should also be considered. In addition, most of the models are 1-D models, but there is a need to extend risk assessment to larger scales, e.g. field to watershed, region, country, etc. Finally, impact assessment should be improved by taking chronic data, mixtures, etc., into account.

### **3.4 Pathogens and antibiotic resistance in RA: limits and solutions**

The methods used in RA for monitoring fate, exposure and infectious outcome (i.e. damage) (Table 3 and 2.4) have limitations with regard to the consideration of microbiological hazards during OW treatment and recycling, as they do not fully capture the complexity of the mechanisms and processes involved. LCA is not yet designed to consider pathogens, nor is its development in that direction foreseen in the near future (e.g. not mentioned in Fantke et al. (2018)). Combining LCA and RA has been suggested for situations where the environmental performance and pathogen-driven health risks are of joint interest (Jolliet et al., 2014).

### 3.4.1 Microbial fate during organic waste treatment

The main concerns regarding the microbiological hazards of anaerobic digestion processes are the survival and spread of zoonotic and human pathogens, as well as the antibiotic resistance dynamics, including ARG transfer from non-pathogenic organisms (including commensals) to pathogenic bacteria (Zhao and Liu, 2019). Moreover, OWs could also spread plant pathogens, but these considerations are beyond the scope of this review. The efficacy of OW sanitization depends on several parameters, with the most critical ones being the initial amount of pathogens, ARB, ARG and MGE in the OWs to be recycled and sanitized (Strauch, 1991), in addition to their respective infectious doses and die-off rates in relation with the environmental constraints that will develop in the bioreactor. Microorganism die-off rates during anaerobic digestion mainly depend on the time of exposure to unfavourable temperatures, pH or toxic substances such as hydrogen sulfide (Ottoson et al., 2008; Sahlström, 2003). Other parameters, such as the O<sub>2</sub> concentration, the presence of ammonium and short-chain fatty acids (e. g. acetate), mainly change the ratios in terms of numbers of cells per microbial taxa, but are not likely to increase their death rate. Biotic factors affecting microbial pathogens, such as predation through bacteria ingestion by microfauna or parasitism, or bacteriophage attacks on bacteria during anaerobic digestion, have not been documented. Predation is more likely to occur during the treated OW storage period, at the end of the process. Storage has therefore been suggested as a means for improving the microbiological quality of these end-products (Sahlström, 2003). Digestion processes are based on microbial ecological successions and synergies between key functional groups. By definition this process thus induces significant changes in OW microbiomes (Razaviarani and Buchanan, 2014). A highly diverse range of microbial pathogens, ARB, ARG and MGE can become involved in these digestion processes via cross-contaminations or faecal carriage by humans and livestock. The hydrolysis step during these processes seems to be compatible with the functional abilities of some bacterial pathogens such as *Pseudomonas aeruginosa* and *Clostridium perfringens*. However, the die-off rates of human and zoonotic viruses are directly related to their susceptibility to the abiotic constraints accumulating from the beginning of the

process. Predation or antagonism between bacteria might also have an impact on such reduction rates, but these interactions have yet to be documented. Die-off rates of a panel of pathogens have been reported over the years for the anaerobic digestion process (see Sahlström, 2003, for a review), which has led to the development of HACCP and QMRA scenarios (see section 2.5.4). The high variation in reported die-off values led Westrell et al. (2004) to assume that pathogens occur in similar numbers in raw and digested OW when they developed their QMRA and HACCP scenarios. Note that digestate spreading with immediate burial is recommended in France (JORF, 2017). This practice is likely to increase microbial contaminant exposure to soil components, including bacterial grazers. However, digestate microbiota, especially pathogenic taxa, can affect the soil biota (Teglia et al., 2011a). Few ecotoxicological studies have been conducted to assess the ecological impact of digestate application on soil. Moreover, ERA studies (US EPA, 1998) have mainly been focused on specific chemicals contained in the digestate and their associated ecotoxicity. Risk assessment in ERA could be enhanced by including both chemical and biological contaminants in ecotoxicological assays. The coalescence of communities from digestates and soils should also be investigated to evaluate resistance/resilience to invading taxa.

During organic waste composting, *Actinobacteria*, *Bacteroidetes*, *Proteobacteria* and *Firmicutes* species are the key microbial players (Bru-Adan et al., 2009). High *Proteobacteria* population levels could then be observed in mature compost. The main genera identified are *Pseudomonas* (28%), *Serratia* (20%), *Klebsiella* (11%) and *Enterobacter* (5%) (Boulter et al., 2002). Several microbial species of human health concern could thus play a role in the composting process (Reynnells et al., 2014). Microbial RA of composted OW could be assessed by monitoring: i) host-specific human/animal pathogens (or indicators of their occurrence) found in the initial organic matter but unlikely to develop during the process, and ii) cells of pathogens such as *Listeria*, *Clostridium* and *Klebsiella* that could be enriched by this process.

Several QMRA models have been suggested, but predicting the behaviour of zoonotic pathogens in OW is still a challenge. Including HACCP in RA model simulations is thus recommended. Westrell et

al. (2004) proposed the first published HACCP approach for anaerobic digestion, involving identification of critical points to control health hazards combined with a preventative management and quality assurance approach throughout the production and distribution steps. This approach generates safety criteria for each control point. Corrective measures are required if this critical limit is reached (Bryan, 1990; Orriss and Whitehead, 2000). As a first step in any HACCP framework, exposure scenarios must be defined and ranked in order of severity (Westrell et al., 2004). These exposure routes can involve: i) direct contact with the pathogen during the digestion process or while handling OW treatment products —the pathogens could enter the body through wounds, ii) inhalation of contaminated air, and iii) ingestion of contaminated matter, OW treatment products or amended soil. However, as for QMRA, thorough knowledge of the type of pathogens and doses that can lead to infection according to these routes is required to be able to accurately assess the relative incidence of the observed hazards. Yet their presence appears to be highly stochastic, likely because of our scant knowledge of the origin and nature of OW involved in such processes. The HACCP framework of Westrell et al. (2004) led to the identification of major critical points requiring consideration in anaerobic digestion processes. The fate of microbial pathogens is highly dependent on the efficacy of the main biochemical transformation processes involved (temperature, pH, duration). Inferences regarding the performance of these processes could also be made by monitoring bacterial groups likely to have die-off values related to those of the pathogens. Generally, indicator bacteria such as *E. coli* and intestinal Enterococci are used to monitor the microbiological quality of digested OW (JORF, 2017). Their numbers are considered satisfactory if they do not exceed 3 log/g (JORF, 2017).

System models based on Bayesian networks<sup>xii</sup> are now emerging as an alternative to QMRA models to overcome uncertainty associated with the underlying datasets (Aguilera et al., 2011; Barton et al., 2012; Beaudequin et al., 2015). Bayesian networks have been used to make RA predictions from incomplete datasets or small samples (Fenton and Neil, 2012; Kontkanen et al., 1997). Furthermore, Bayesian networks, by their nature, can be used for 'forward inference' (whereby the system

variables are specified and the impact on the outcome can be observed) and for ‘backwards reasoning’ (whereby the effect or outcome is specified and subsequent changes in the causal variables can be computed). Bayesian networks can hence reveal variables that are key drivers of an outcome or ,conversely, the sensitivity of an effect to the system variables (Ben-Gal, 2008; Coupé et al., 2000; Pollino and Hart, 2006). A Bayesian network framework can be used to generate scenarios and reduce microbiological risks by performing simulations with modified variables. This can help identify key control areas in a process where microbiological risks could be reduced, and to propose interventions (Albert et al., 2008). However, representing the distribution of a variable in a Bayesian network as continuous or discrete series remains a challenge. Moreover, quantification of conditional probability tables underlying each variable in a Bayesian network is often difficult. These distributions can be assessed on the basis of observed data, published papers or reports, statistical models or simulations (Pollino et al., 2007). Bayesian network predictions could be bolstered by linkages with DALY models. It could be interesting to apply this approach to subpopulations such as workers in contact with OW and OW treatment products. These estimates could then be used in Bayesian network models to infer key drivers influencing life expectancy and quality.

### **3.4.2 Role of antibiotics and non-antibiotic chemical stressors on antibiotic resistance selection and antibiotic resistance dynamics after treated OW spreading on soil**

In treatment systems, stressors such as antibiotics are often in mixtures and at high enough levels to increase the likelihood of antibiotic resistance. It was shown that antibiotics can exert selection pressure at far below inhibitory concentrations (Gullberg et al., 2011; Lundström et al., 2016). Bengtsson-Palme and Larsson (2016) attempted to determine minimal selective antibiotic concentrations in complex systems and they predicted no-effect concentrations for resistance selection for 111 antibiotics based on available minimum inhibitory concentrations for clinically relevant bacteria. These authors suggested that “emission limits for antibiotics must be set individually for each compound, and that different antibiotics have very different potential to be selective”. They also mentioned that antibiotic concentration limits must account for antibiotic effects on mutagenesis, transfer of genetic material between bacteria, chromosomal DNA

mobilization and biofilm formation as subinhibitory antibiotic concentrations were shown to affect those mechanisms. In addition, the concomitant presence of non-antibiotic chemical stressors that have antimicrobial properties might contribute to the spread and selection of antibiotic resistance. Among them, trace elements (Baker-Austin et al., 2006; Seiler and Berendonk, 2012), biocides (Capita et al., 2013; Pal et al., 2015), PAH and PCB (Chen et al., 2017; Gorovtsov et al., 2018) could co-select for ARG. This phenomenon exemplifies the so-called ‘cocktail effect’ (see section 3.5). The co-selection of resistance is based on: co-resistance (a resistance gene towards a chemical is selected because it is genetically linked to another resistance gene under positive selection; this occurs when genes are co-located on the same genetic element, i.e. plasmid, genomic island, integron, transposon); cross-resistance (a resistance gene can confer resistance to various chemicals); and cross-regulation (different resistance genes conferring resistance to various chemicals are controlled by common/identical regulatory systems) (Bengtsson-Palme et al., 2016; Dickinson et al., 2019; Pal et al., 2017; Poole et al., 2019; Rutgersson et al., 2020; Yu et al., 2017).

In a meta-analysis, Goulas et al. (2020) addressed the effect of chemicals (effects of antibiotic administration to animals and spiking OW with antibiotics and/or metals) on antibiotic resistance markers after composting or anaerobic digestion of sludge or animal waste. They reported a “non-significant reduction of ARG/MGE after composting —regardless of whether the animals had received antibiotics or not— and no significant effect of antibiotic use on anaerobic digestion. When OW was spiked with antibiotics and/or metals before treatment, the ARG/MGE relative abundance was still reduced (but not significantly) compared to the absence of spiking (significant) after composting, and the ARG/MGE reduction was significant in both cases after anaerobic digestion”. They related these observations to the “variability originating from the diversity in the duration of the exposure and/or in the concentrations of spiking molecules, which could affect the bacterial response to contamination, as well as the variation in the latency of measurement affecting the [antibiotic resistance] measurement. Another bias could explain the heterogeneity in results, which is linked to the range of ARG/MGE chosen by authors”.

Soil acts as a resistance reservoir and high-throughput sequencing technology has been used to conduct metagenomic analyses of soil bacterial communities and the findings have helped describe ARG, their origins and potential to be transferred between species (Nesme and Simonet, 2015). Human activities such as spreading OW on soil might alter their intrinsic resistome, thereby introducing allochthonous ARB, ARG and exogenous chemical stressors. Resistome changes are determined: i) by the survival and growth ability of ARG-bearing bacteria, ii) potential gene exchanges between ARG-bearing bacteria and the autochthonous soil population (i.e. probability of contact, phylogenetic relationships), and iii) by the nature, speciation, bioavailability and bioaccessibility of input chemicals as well as those already present (i.e. trace metals and pesticides in agricultural soils). Few data are currently available regarding the specific survival of ARG-bearing bacteria in terms of competition, predation or adaptation to soil environmental conditions (van Veen et al., 1997), or regarding horizontal gene transfer efficiency *in situ*. Moreover, further investigations are needed to determine whether ARG are carried by potential pathogens or co-located on mobile elements. The co-selection of resistance in agricultural soils mostly involves data related to the role of trace elements. These elements accumulate and can persist in soil for long periods because they are non-degradable and hard to remove. Their presence in animal waste consecutive to their use for growth enhancement and disease control, as well as in biosolids, inorganic fertilizers and pesticides (Hölzel et al., 2012; Mortvedt, 1996; Rangasamy et al., 2017; Singh and Agrawal, 2008), could exert selection pressure on antibiotic resistance in soils (Singer et al., 2016; Zhu et al., 2013). Studies by Berg et al. (2010, 2005) using both culture-based and culture-independent approaches demonstrated that Cu exposure in agricultural soils led to the selection of Cu-resistant bacteria and indirect selection of ARB. Various studies have shown a positive correlation between the presence of trace elements and the abundance of some ARG in agricultural soils receiving manure amendments. For instance, significant positive correlations were noted between some *sul* genes and various trace elements, i.e. Cu, Zn and Hg, in manure and soil samples collected from multiple feedlots in Shanghai (Ji et al., 2012). In addition, these authors indicated that the presence of ARG was relatively

independent of their respective antibiotic inducers. Similarly, Hu et al. (2017) found that changes in ARG abundance were independent of the antibiotics present and were affected by long-term Ni contamination in agricultural soils. Based on a nationwide sampling campaign across China (Zhou et al., 2017) showed significant co-selection between trace elements and antibiotic resistance genes among Cu, Hg and *sul* genes. Arsenic in swine manure has also been shown to increase ARG and MGE abundance (Zhu et al., 2013). The impact of applying ARG-containing fertilizers on already trace element-contaminated soils (e.g. vineyard soils known for their high levels of Cu due to fungicide applications) should be taken into account, especially because trace elements may influence ARG mobility (Hu et al., 2017, 2016). Unlike in RA, in LCA these factors could only be taken into account in the inventory phase, since antibiotic resistance and other pathogen-related mechanisms are not accounted for in fate, bioavailability and effect modelling. Studies on bacterial communities in the gut of earthworms exposed to As and sulfamethoxazole contamination revealed synergistic interactions of both contaminants, leading to an increased incidence of ARG and MGE and changes in the bacterial community structures correlated with ARG profiles (Wang et al., 2019). These results suggested that the gut of soil microfauna is a neglected hotspot of antibiotic resistance. Furthermore, these authors highlighted an ARG/MGE co-occurrence pattern, indicating that horizontal gene transfer via MGE may occur in the earthworm gut (Ding et al., 2019). However, correlation-based analyses were carried out in most studies, but this may not reveal causal evidence of patterns in complicated systems. Laboratory experiments should be conducted under controlled conditions in micro- or mesocosms so as to validate the role of trace elements in antibiotic resistance dynamics and to quantify the role of the various co-selection mechanisms. Furthermore, as these studies were mainly based on total trace metal concentrations, further studies are needed to also take speciation, bioavailability and bioaccessibility at the microscale into consideration.

### **3.5 Contaminant mixtures in LCA and RA: limits and solutions**

Few examples were found in the literature of RA of OW treatments and recycling focused on the possible effects of physicochemical interactions among trace elements, organic contaminants and

pathogens (i.e. co-toxicity), as well as on their joint fate after OW spreading. In-depth discussions on the so-called 'cocktail effect' of the environmental emission of mixed contaminants are ongoing, while the limitations of current RA approaches to address this effect have been analysed and some authors calling for a change of paradigms to take this issue into full account (Backhaus and Faust, 2012; Bopp et al., 2016; Svingen and Vinggaard, 2016). The cocktail effect is not considered in LCA as, by design, it deals with thousands of substances in a cumulative fashion. Under the current paradigms, substance mixtures would have to be treated as new inventory items, and toxicity data would be needed for the mixtures to be considered in LCA.

Bopp et al. (2016) reviewed 22 case studies of RA applied to chemical mixtures and found that the main shortcomings of current frameworks that would have to be overcome to perform mixture risk assessments include: i) the difficulty of determining the exact composition of unintentional mixtures (e.g. in wastewater and sludge), ii) the lack and high uncertainty of exposure and toxicity data, as well as of information on the mode of action of mixtures. The authors highlighted that "Data gaps seem to be the major hurdle when it comes to deal with risk assessment of chemical mixtures [...]" (Bopp et al., 2016). Pesticides are among the most commonly studied mixtures. The study concluded that the Information Platform for Chemical Monitoring data (IPChem) should be further populated to assist future chemical mixture RA (further explored in Bopp et al. (2019), as outlined below). They provided recommendations for future case studies: i) compare different populations and vulnerable groups, ii) include new chemicals and substance groups, iii) address interactions at low exposure concentrations, iv) develop criteria for assessing interactions, v) investigate approaches for substance grouping (e.g. per effect or mode of action), and vi) examine the relevance of assessing mixtures across regulatory regimes. Bopp et al. (2019) discussed a number of further challenges and potential solutions associated with RA of chemical mixtures, disaggregated by hazard, exposure, risk and risk management. One challenge highlighted concerned the simultaneous vs. sequential exposure to multiple chemicals, which entails the mechanics and timing of exposure and the cumulative effect of concentrations that may individually be below the level at which the effect is observed. The authors,

based on previously published findings, proposed a combined adverse outcome pathway/aggregate exposure pathway<sup>xiii</sup> framework to “put into context exposure models, biokinetic and dynamic modelling, the results from monitoring, biomonitoring, and *in vitro* testing”. This framework is designed to integrate a variety of models (multimedia fate models, *in vitro* fate mathematical models, physiologically based kinetic models, etc). There is published evidence that this framework is being built and applied (e.g. Clewell et al., 2020; Price et al., 2020).

#### **4 Potential environmental assessment enhancement and the prospect of a comprehensive framework for agricultural organic waste recycling assessment**

In the light of the literature on LCA and RA applied to OW treatments and their recycling, a life cycle based approach covering both waste processing and recycling is clearly lacking. LCA toxicity methods do not account for the transformation/degradation/combination of trace elements, organic contaminants and pathogens, or their fate following the spreading of untreated or treated OW on cultivated soils. These methods are hampered by high uncertainty regarding human and ecotoxicity characterization factors, which are usually based on total concentrations and not regionalized, apart from a few very recent regionalized LCA methods (Bulle et al., 2019; Verones et al., 2020). Moreover, available characterization factors exclude a plethora of substances that may be present in OW. RA methods fail to consider a number of biophysical processes that determine the fate of emitted contaminants, including trace elements, organic contaminants and pathogens. Moreover, methods such as QMRA rely on datasets featuring high data uncertainty, and combinations of contaminants are not considered in RA. Both frameworks could benefit from additional models and approaches (including those used in other disciplines) to integrate overlooked biophysical processes and thus bridge the assessment gaps. Existing frameworks and models need to be combined to be able to accurately assess the environmental and human health impacts of regional strategies for recycling OW in agriculture. Indeed, no single framework/model currently encompasses all pathways via which contaminant-bearing OW could affect the environment and living organisms. Moreover, the overall dynamics of given contaminants after OW spreading are not taken into account.

A combination of environmental and risk assessments may have to be applied to recycling scenarios which need to be defined and, ideally, co-constructed with impacted stakeholders. Conventional LCA, which considers the fate of contaminants applied to agricultural soils, might be insufficient for comprehensive environmental assessment. LCA deals only with systems in equilibrium (steady state), while excluding the dynamics of substances during a given study period (generally a year), and also it deals with absolute quantities (flows, stocks). LCA should therefore be combined with tools that account for all relevant flows and stocks of all materials and specific substances to ensure comprehensive and exhaustive assessments. Material and substance flow analyses (Allesch and Brunner, 2015; Bouman et al., 2000) are the natural candidates for such integration because, together with LCA, both tools are included in industrial ecology toolboxes and share key features. Material flow analysis encompasses all flows while substance flow analysis takes substances of interest into account in a given system (e.g. a region). Combining both of these tools with LCA would require additional models to estimate not only direct emissions due to OW spreading on soil, but also the fate of contaminants of interest (e.g. cocktail effect, available vs. total concentrations, speciation, etc.). This modelling should rely on both empirical knowledge (e.g. soil experiments) and existing models. Table 6 summarizes a few novel or complementary approaches that could be used to enhance LCA and RA, including their levels of 'readiness' (i.e. the extent to which such approaches could be applied or integrated today). The body of empirical and modelling knowledge on contaminants is vast, but the extension/complementation of LCA and RA would be a complex task, considering the data requirements at different levels as well as model complexity and suitability for a diverse range of situations. Additional toxicity experiments focused on terrestrial ecotoxicity could enrich the body of characterization factors used in LCA to assess the toxicity impacts of target contaminants.

[Table 6]

A comprehensive framework (Figure 4) could also include the set of approaches and models compiled in Table 6, depending on the goal and scope of the intended assessment. This theoretical

framework could help guide the integration of models and approaches to be able to achieve comprehensive assessments of OW recycling in soils.

[Figure 4]

For instance, in region X, animal effluents and sewage sludge constitute a surplus that would have to be dealt with at a cost. In parallel, region X features vast periurban agriculture areas requiring fertilization at a cost (mineral fertilizers are imported from outside the region). An assessment is sought by stakeholders, and required by legislation, regarding potential impacts on the environment and human health associated with agricultural recycling of a portion of the surplus OW. The team entrusted with performing the analyses starts by designing and proposing stakeholders various industrial ecology-inspired possibilities for OW recycling in crop fields, i.e. a so-called 'plausible promise' (Wassenaar et al., 2014). This systemic change strategy —to be elaborated in detailed scenarios with stakeholders via participatory approaches (e.g. as described in Queste and Wassenaar (2019))— is underpinned by the idea that the recommended recycling would bring overall improvements to the regional system (namely cheaper OW management and fertilization without worsening current impacts on the environment and human health, despite changes in agricultural practices and logistics). The team must demonstrate the environmental soundness of the plausible promise by comparing the 'conventional' (i.e. impact addressed by current LCA and RA methods) environmental impacts of both of the proposed scenarios with the current situation, while exploring and quantifying additional environmental and human health impacts associated with the increased transfer of contaminants to agricultural soils. This is done first by performing an MFA (see table A) to establish the flows and stocks of 'matter' (Wassenaar, 2015) in the system, and then estimating those that would occur under the proposed scenarios. Given the specificities of the region and the available OW, the team determines that further insight into the fate of specific substances of interest is needed and they perform an SFA for this purpose (Wassenaar et al., 2015). Based on the MFA and SFA findings, the team performs LCA of a sample of the current and prospective systems (OW generation, treatment and utilization), as well as RA targeting the substances of interest in the

context of OW treatment and utilization. The team discovers that LCA —despite estimating a battery of conventional impact categories— is not well equipped to consider the fate of certain target substances due to limitations in the underlying terrestrial ecotoxicity and human toxicity models. The team thus decides to mobilize additional models to more accurately simulate the fate and bioavailability of the target substances. These models include: i) the approach described in Owsianiak et al. (2013) to determine the terrestrial ecotoxicity of trace elements, informed by experiments conducted on soil and OW (e.g. chemical extractions), ii) the COP-Soil model to determine interactions between organic contaminants and organic matter, and iii) QMRA to predict potential exposure to pathogens in tandem with an assessment of the extent of survival ARG-bearing bacteria regarding their adaptation to the soil environmental conditions. The team also deploys: i) STICS (Brisson et al., 2003) to model the proposed agricultural systems in which a considerable share of the crop nutrient needs are supplied by organic fertilizers via the application of treated OW —the idea is to improve the quality of mineral fertilizer substitution estimations based on static indicators such as mineral fertilizer equivalents (Brockmann et al., 2018; COMIFER, 2013), and ii) the AMG soil organic carbon model (Clivot et al., 2019) to predict the effects of repetitive long-term OW applications on soil organic carbon. The team finally uses the overall available experimental and modelling results to assess the environmental soundness of the proposed scenarios, in consultation with the stakeholders via participatory approaches. Finally, results derived from a sample of systems are extrapolated to the regional scale, as described by Avadí et al. (2017).

## **5 Conclusion**

The present review demonstrates that some aspects of organic waste treatment and recycling and some contaminant families have received far more attention than others. For contaminants, as trace elements have been studied to a greater extent than other contaminants, this largely relates to their much longer research history, which in turn could be explained by the respective analytical capacities. Organic contaminant and (to a lesser extent) pathogen research is slowly catching up, but environmental assessment frameworks are slow to follow. Combined OW treatment and recycling is

less studied in combination, due to the complexity of these processes and the multidisciplinary scope. Common approaches for ecosystem and human impact assessment still fail to consider all relevant pathways and mechanisms involved when organic waste-borne contaminants are applied on soil. A number of refinements have thus been proposed to at least partially bridge these divides. Such refinements —consisting of models and approaches at different ‘readiness’ and application complexity levels— are discussed regarding their suitability to fill gaps in conventional LCA and RA practice. In principle, most elements required for comprehensive consideration of trace contaminants in the environmental assessment of agricultural recycling of OW are available in one form or another, and their application would depend on the assessment needs and resources, as certain approaches may be resource (e.g. data, modelling skills) intensive. A comprehensive framework combining several complementary approaches has been proposed, which could help generate comprehensive environmental assessments of agricultural recycling of organic waste. The overall framework is a theoretical instrument, mainly designed to fuel discussion while providing directions for further research.

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## Tables

**Table 1. Main frameworks for environmental assessment of waste treatments and their products**

<b>Procedural frameworks</b>	<b>Focus/level</b>	<b>Pros</b>	<b>Cons</b>
<ul style="list-style-type: none"> <li>• Environmental impact assessment (EIA): Multi-tool framework aimed at explicitly considering environmental and social impacts associated with new project developments. Often required by legislation in public projects.</li> </ul>	Micro (site)	Possibility of learning from previous projects and reducing mitigation costs of unforeseen impacts, improves long-term viability <sup>a</sup>	Exclusive focus on biophysical issues <sup>a</sup> , some at the local scale and some at the global scale
<ul style="list-style-type: none"> <li>• Strategic environmental assessment: Multi-tool framework similar to EIA but oriented towards policy instrument evaluation, often in situations of high uncertainty.</li> </ul>	Meso, macro (policy)	Identify potentially unsustainable development alternatives at an early stage <sup>a</sup>	Perceptions of increased costs (work, time, resources); absence of a single step-by-step approach <sup>a</sup>
<ul style="list-style-type: none"> <li>• Multi-criteria decision analysis: A collection of decision support methods aimed at comparing alternatives based on a set of decision criteria. Suitable for conflicting decision situations.</li> </ul>	Micro to macro (project, policy)	Evaluation of alternatives against multiple, often opposing criteria <sup>b</sup>	Subjective weighting scores, lack of a coherent universal framework <sup>b</sup>
<b>Analytical frameworks</b>	<b>Focus/Level</b>		
<ul style="list-style-type: none"> <li>• Material flow assessment/analysis/accounting (MFA): Systematic accounting of flows and stocks of materials and energy prevailing within an economic system, often a whole region or country. <ul style="list-style-type: none"> <li>○ Substance flow analysis (SFA): MFA-type assessment focused on the fate of specific substances at regional or national levels.</li> <li>○ Material input per service unit (MIPS): Estimation of the environmental pressure associated with products and services expressed as a life cycle-wise ratio of natural resource consumption to benefit provided.</li> </ul> </li> </ul>	Macro (policy, plan)	Multiple applications: industrial ecology, environmental management, resource management	No impacts are calculated, only environmental pressure indicators are provided
	Micro, macro (production systems, regions)	Similar to MFA, focused on details pertaining to specific substances of interest	Similar to MFA
	Micro (product, service)	Simple and intuitive cost (in terms of environmental pressure) to benefit ratios	Similar to MFA
<ul style="list-style-type: none"> <li>• Energy/exergy/emergy analysis: Group of methods aimed at accounting for energy flows in the studied system, usually a process or product system. Exergy refers to energy of a certain quality (useful to produce work). <ul style="list-style-type: none"> <li>○ Energy return on investment: A ratio of industrial energy embedded in a product vs. the energetic content of the product, representing energy efficiency.</li> </ul> </li> </ul>	Micro (process, product, service)	Multiple resource consumption issues are expressed in common intuitive units	Reliance on energy-related impact categories at the expense of other relevant environmental impact categories
	Micro (process, product, service)	Similar to MIPS, very relevant in energy intensive systems	Similar to energy analyses

<ul style="list-style-type: none"> <li>• Risk assessment (RA): Assessment toolset aimed at evaluating human health, environmental and safety-related risks associated with projects or product systems (chemicals, hazardous substances, and industrial facilities). An emerging research area is HERA: Health and ecological risk assessment, also known as Environmental risk assessment (i.e. health + ecological)<sup>e</sup>. <ul style="list-style-type: none"> <li>○ Ecological risk assessment (ERA): a process that evaluates the likelihood that adverse ecological effects may occur or are occurring to ecosystems exposed to one or more stressors (risk factors) <sup>c</sup>.</li> <li>○ Human health risk assessment: a process intended to estimate the risk to a given target organism, system or (sub)population, including the identification of attendant uncertainties, following exposure to a particular chemical, biological or physical agent, taking into account the inherent characteristics of the agent of concern as well as those of the specific target system <sup>d</sup>.</li> </ul> </li> </ul>	<p>Micro (site, chemicals)</p> <p>Organism to socioecological system</p> <p>Organism to socioecological system</p>	<p>Highly regulated framework, mainly focused on human health; allows for absolute quantitative comparisons; considers casual mechanisms affecting health</p> <p>Provides a system-oriented perspective for holistic risk evaluation and management <sup>c</sup></p> <p>Similar to ERA</p>	<p>Lack of full environmental impact assessment, often focuses on toxicity; high data requirements; emphasis on expert knowledge</p> <p>Lack of model integration, which complicates the selection and application of the most suitable models for each assessment scale, subjective weighting scores <sup>c</sup></p> <p>Lack of model integration and modelling challenges, especially regarding hazard characterization (description of inherent properties of the agent) and exposure assessment (evaluation of the concentration or amount of a particular agent that reaches a target population)</p>
<ul style="list-style-type: none"> <li>• Eco-efficiency analysis: Concept aligned with the growing environmental concerns in economic sectors, which can be defined as a management philosophy encouraging business to search for more environmentally-sound alternatives producing similar economic benefits.</li> </ul>	<p>Micro (product, service)</p>	<p>Developed and used by companies (e.g. BASF), balances economic and environmental concerns of alternatives</p>	<p>Lack of full environmental impact assessment</p>
<ul style="list-style-type: none"> <li>• Life cycle assessment (LCA): Life-cycle tool aimed at accounting for the environmental impacts, expressed in a number of impact categories, associated with the provision of a good or service over its whole life cycle. Various existing 'footprints' are related to LCA, but focused on single issues/indicator categories: <ul style="list-style-type: none"> <li>○ Carbon footprint (CFP): Can be considered as an LCA subset focused on the global warming potential.</li> </ul> </li> </ul>	<p>Micro (process, product, service), macro, meso (footprints, experimental regional LCA)</p> <p>Similar to LCA</p>	<p>Highly standardized framework, immense body of knowledge (theory and application), ample coverage of impact categories; prevents burden transfer among life cycle stages or impact categories; allows comparative assessments of multiple pathways with equivalent functions</p> <p>Similar to LCA</p>	<p>Data intensive, controversial design/methodological details and schools of thought (e.g. system boundaries, allocation of impacts among co-products, uncertainty management, descriptive vs prospective, inclusion of market data, carbon modelling, etc.)</p> <p>Focused on a single impact category</p>

<ul style="list-style-type: none"> <li>○ Ecological footprint (EF): Accounts for land use associated with the provision of a product. It can be complemented by human appropriation of net primary production (HANPP), which studies the proportion of original primary production that remains on a spatially-specifically defined land area with given specific land use practices.</li> </ul>	Similar to LCA	Similar to LCA	Similar to CFP
<ul style="list-style-type: none"> <li>○ Water footprint (WF): Accounts for freshwater resource appropriation (including fresh, rain and polluted water volumes affected) associated with the provision of a product, in a spatiotemporally explicit fashion.</li> </ul>	Similar to LCA	Similar to LCA, regionalizable	Focused on a single impact category, multiple competing approaches

Adapted from Jeswani et al. (2010) and Avadí (2014), except for: <sup>a</sup> <http://www.environmental-mainstreaming.org/>, <sup>b</sup> Velasquez and Hester (2013), <sup>c</sup> Chen et al. (2013), <sup>d</sup> WHO (2010), <sup>e</sup> SETAC (2018)

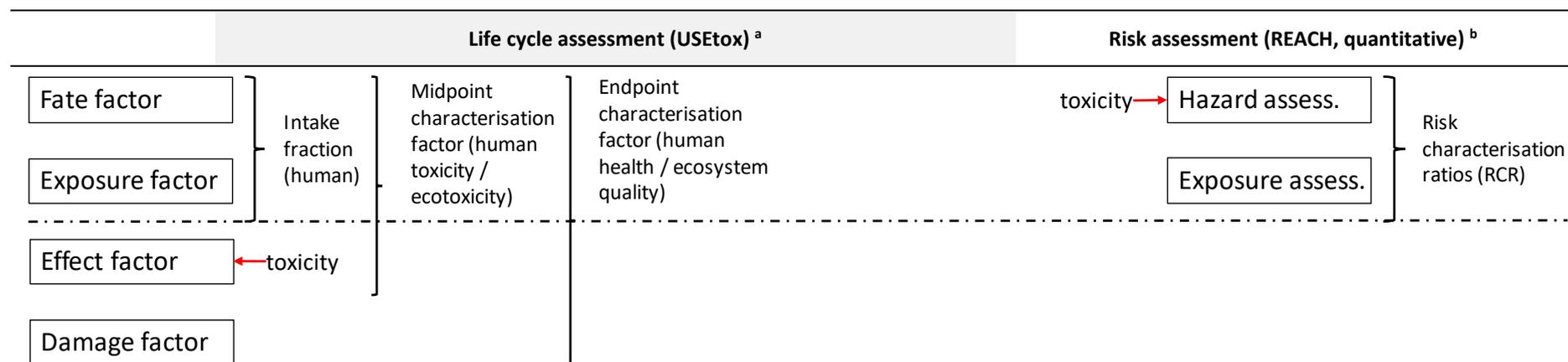
**Table 2. Recent reviews on environmental and risk assessment of organic waste treatments, including the consideration of contaminants**

<b>Publication: No. of studies reviewed</b>	<b>Assessment frameworks considered</b>	<b>Waste streams and treatments</b>	<b>Nutrients, contaminants and impacts considered</b>	<b>Main findings and recommendations</b>
Bernstad and la Cour Jansen (2012): 25	Life cycle assessment	Food waste, various treatments	Environmental impacts	<ul style="list-style-type: none"> <li>Assumptions on the content of toxic compounds in treated waste vary largely</li> <li>Mass-flows of carbon, nutrients and trace elements are often not respected in life cycle assessments due to cut-offs and the use of published values rather than transfer coefficients throughout the treatment chain</li> </ul>
Bopp et al. (2016): 21	Human and ecological risk assessment	Surface and groundwater, wastewater treatment	Pharmaceuticals and personal care products, VOCs, PPPs, trace elements, pesticides, etc.; chemical mixtures	<ul style="list-style-type: none"> <li>Several factors might lead to an underestimation or overestimation of the potential risk, e.g. uncertainty in reference values used, incompleteness of monitoring data, etc.</li> <li>It would be relevant to improve data sharing regarding toxicity and exposure information</li> </ul>
Gallagher et al. (2015): 10 studies, 5 issue papers	Cumulative risk assessment	Various industrial streams and impoundments, including wastewater treatment sludge	Single chemicals and chemical mixtures	<ul style="list-style-type: none"> <li>An iterative approach is essential; a tiered approach is particularly useful</li> <li>Vulnerable populations need to be considered</li> <li>Early involvement of multiple stakeholders facilitates the process</li> <li>New methods are required for multiple exposure routes, pathways, and chemicals</li> <li>The spatial scale affects the methods used, data needs, and types of risk management questions that can be addressed</li> </ul>
Harder et al. (2015): 30 <sup>a</sup>	Combined life cycle and risk assessment	Water and wastewater treatment	Pathogens	<ul style="list-style-type: none"> <li>Environmental assessment case studies based on elements taken from RA and LCA can be designed in many different ways (combination, integration, combined use, or hybridization)</li> <li>There are a number of implications and pitfalls at the model structure level, which analysts should be aware of (e.g. the potential asymmetry between the handling of local risks and impacts in the immediate sphere of interest to the decision maker and elsewhere)</li> </ul>
House and Way (2014): 88	Human and ecological risk assessment	Industry, agroindustry, agriculture, wastewater treatment	Sewage and septic tank sludge, manure, waste plant matter, food waste, abattoir waste, molasses, compost, paper waste	<ul style="list-style-type: none"> <li>For many organic materials little organic chemical data are available</li> <li>Widely used veterinary medicines, such as antiparasitic treatments, are likely to be of greatest concern in cattle due to the greater active ingredient quantities required for treatment</li> </ul>

				<ul style="list-style-type: none"> <li>Monitoring the concentrations of veterinary chemicals in organic materials, and the carbon contents of the soils to which they are being applied would greatly reduce uncertainty in environmental risk assessment</li> </ul>
Laurent et al. (2014a; 2014b): 222	Life cycle assessment	Solid waste management systems (non-organic, biowaste, sludge), various treatments	Environmental impacts (non-toxic, toxic, resources, energy)	<ul style="list-style-type: none"> <li>There is generally little decisive agreement among studies: the strong dependence of each system on its local context hampers a consistent generalization of life cycle impact assessment results</li> <li>A number of recommendations are provided to improve future life cycle assessments of waste management systems</li> </ul>
Zang et al. (2015): 44	Life cycle assessment	Wastewater treatment plants, activated sludge	Environmental impacts, trace elements, pharmaceuticals and personal care products	<ul style="list-style-type: none"> <li>Life cycle assessments of wastewater treatments should include spatial differentiated characterization methods, considering the emission location, spatial dimensions (transfer between environmental compartments) and pollutant properties</li> </ul>

<sup>a</sup> Most studies were on contaminated site remediation; we refer here to three studies on water and wastewater treatment

**Table 3. Models for human toxicity and ecotoxicity in LCA (USEtox) and RA (REACH)**



Factors	Human	Ecosystem	Human	Ecosystem
Fate (LCA) Hazard (RA)	<ul style="list-style-type: none"> <li>Measurements and models, to determine emission rates and intermedia transfers of chemicals using a mass balance differential equation for each compartment (indoor air, urban air, continental rural air, continental freshwater, continental seawater, continental agricultural soil, continental natural soil, and crop residues) due to various processes (deposition, volatilization, degradation). The initial transfer rate matrix is then inverted to determine the time-integrated, steady state cumulative transfer rates (the fate of a chemical in the long term, across all compartments) <sup>c</sup>.</li> <li>Data required includes partition coefficients (air-water, octanol-air and octanol-water)</li> <li>The mass balance equation system consists of a matrix of transfer rates among compartments <sup>c</sup>.</li> </ul>		<ul style="list-style-type: none"> <li>Measurements, to determine the derived no-effect level (DNEL).</li> <li>Measurements and models to determine toxicity.</li> </ul>	<ul style="list-style-type: none"> <li>Measurements to determine the predicted environmental concentration (PEC) and predicted no-effect-concentration (PNEC).</li> <li>Measurements and models to determine the toxicity.</li> </ul>
Exposure (LCA and RA)	<ul style="list-style-type: none"> <li>Human exposure factors are ratios between the intake by the population</li> </ul>	<ul style="list-style-type: none"> <li>The environmental exposure factor for freshwater ecotoxicity is the</li> </ul>	<ul style="list-style-type: none"> <li>Measurements and models (when no</li> </ul>	<ul style="list-style-type: none"> <li>Measurements and models (when no</li> </ul>

	<p>of a polluted medium (direct: air, water; indirect: food) via an exposure pathway; and the bulk density and volume of the medium <sup>c</sup>:</p> <ul style="list-style-type: none"> <li>○ General direct exposure pathways = direct intake of a polluted medium * population / bulk density of the medium * volume of the medium</li> <li>○ General indirect exposure pathways = biotransfer and accumulation of a contaminant in a substrate * individual ingestion rate of a polluted substrate * population/bulk density of the medium * volume of the medium</li> <li>● Indirect exposure pathways include bioaccumulation factors, a plant uptake model and a pesticide model (inspired from the dynamiCROP <sup>e</sup> model, which accounts for exposure via crop residues and crop to soil transfers).</li> </ul>	<p>fraction of a chemical dissolved in freshwater, depending on the water-suspended solids and water-dissolved organic carbon partition coefficients, suspended matter concentration, bioconcentration factor in fish and concentration of biota in water:</p> <ul style="list-style-type: none"> <li>○ Exposure factor for freshwater ecotoxicity = dissolved mass of chemical in freshwater / total mass of chemical in freshwater</li> <li>○ Partition coefficients are determined with the WHAM7 model <sup>f</sup>; bioaccumulation factor for fish are from databases.</li> <li>● For terrestrial ecotoxicity, a preliminary USEtox model separates exposure into accessibility and bioavailability factors: <ul style="list-style-type: none"> <li>○ Accessibility (reactive fraction in soil) and Bioavailability (free-ion fraction of the reactive fraction in soil) are determined from empirical regression models <sup>g</sup>.</li> </ul> </li> </ul>	<p>measured data exists), to determine inhalation, dermal and oral exposure by workers (occupational), consumers and humans in general via the environment.</p> <ul style="list-style-type: none"> <li>● Models for occupational exposure: <ul style="list-style-type: none"> <li>○ Tier 1: ECETOC-TRA <sup>h</sup>, EMKG-Expo-Too</li> <li>○ Tiers 2-3: Stoffenmanager <sup>i</sup>, RISKOFDERM, Advanced REACH Tool (ART)</li> </ul> </li> <li>● Models for environmental exposure: chemical assessment and reporting tool (CHESAR), which contains ECETOC and EUSES tools, and which integrates functions to calculate PEC values; probabilistic material flow modelling (PMFA) for establishing exposure scenarios.</li> </ul>	<p>measured data exists), to determine exposure of life forms in pelagic water, sediment, aquatic food chain, sewage treatment and air compartments.</p>
Effect (LCA) Risk characterisation (RA)	<ul style="list-style-type: none"> <li>● Effect factor: change in the lifetime disease probability due to a change in lifetime intake of a chemical) = <math>0.5/ED_{50}</math></li> <li>● Effect data rarely exist for humans, and are often extrapolated based on relative body weight from animal data. <math>ED_{50}</math> are listed in various databases.</li> </ul>	<ul style="list-style-type: none"> <li>● Effect factor: change in the potentially affected fraction of species (PAF) due to a change in concentration of a chemical, for freshwater aquatic or terrestrial ecosystems = <math>0.5/HC_{50}</math></li> </ul>	Risk characterisation ratio (RCR) = exposure/DNEL	RCR = PEC/PNEC
Damage (LCA)	<ul style="list-style-type: none"> <li>● The damage factor is the effect factor expressed as disability-adjusted life years (DALY) per number of cases of a</li> </ul>	<ul style="list-style-type: none"> <li>● The damage factor is the effect factor expressed as the potentially disappeared fraction of species</li> </ul>	N/A	N/A

specific disease caused by exposure to a contaminant.	(PDF), achieved by a factor of 0.5 (i.e. PDF/PAF = 0.5) <sup>i</sup> .
o Different factors are applied to cancer effects and non-cancer effects, with the former being >4 times higher than the latter.	

Indicator definitions:

- DALY: years of healthy life lost to premature mortality in the population and loss of quality of life (disability) due to health issues (Fantke (Ed.) et al., 2017). It is a World Health Organization metric. DALYs measure the health condition in a population, accounting for the sum of years of life lost (YLL) due to premature death and years lost due to disability (YLD): DALY = YLL + YLD. One DALY represents one lost year of 'healthy' life. Calculation of YLL requires information on the number of people who died from a disease and their life expectancy at age of death. YLD incorporates the number of incident cases, symptom duration, and disability weight (symptom severity) (Solberg et al., 2017).
- DNEL: is the maximum permissible level of exposure to a substance, as mandated by the REACH legislation.
- ED<sub>50</sub>: the daily dose per kg body weight that causes a disease probability of 50% on a species, per exposure route.
- HC<sub>50</sub>: the geometric mean of chronic EC<sub>50</sub>s for freshwater or soil species.
- NOEC: the no observed effect concentration is the dose, determined by toxicity tests, which will not cause harm on the tested species. It is an expression of the No effect concentration (NEC).
- PAF: the fraction of a species exposed to a concentration above their NOEC.
- PEC: the concentration of a substance in the environment, as determined by exposure models such as EUSES (Vermeire et al., 2005).
- PNEC: the maximum concentration of a substance at which an ecosystem may be exposed without adverse effects.

Sources: <sup>a</sup> Fantke et al. (2017), <sup>b</sup> REACHnano (2015), <sup>c</sup> Rosenbaum et al. (2007), <sup>d</sup> Gadaleta et al. (2016), <sup>e</sup> <http://dynamicrop.org/model.php>, <sup>f</sup> <https://www.ceh.ac.uk/services/windermere-humic-aqueous-model-wham>, <sup>g</sup> Owsianiak et al. (2015), <sup>h</sup> <http://www.ecetoc.org/tra>, <sup>i</sup> <http://nano.stoffenmanager.nl>, <sup>j</sup> Jolliet et al. (2003)

**Table 4. Computation of risk assessment indicators for trace elements**

Indicators	Main equations	Remarks
<b>Indicators calculated on waste</b>		
Reduced partition index	$IR = \frac{\sum_{i=1}^k (i^2 \cdot F_i)}{k^2}$	$i$ is the number of the extraction step from 1 for the first and weakest extractant to $k$ for the last and strongest extractant $F_i$ is the percentage of the total trace element recovered after the extraction step $i$
Risk assessment code	$RAC = \frac{[TE]_{available}}{[TE]_{total}} \cdot 100$	$[trace\ elements]_{available}$ is the concentration (mg kg <sup>-1</sup> dry soil) of a given available trace element recovered after the first extraction step(s) $[trace\ elements]_{total}$ is the total concentration (mg kg <sup>-1</sup> dry soil) of a given trace element
Modified risk index	$MRI = \sum_{i=1}^n (T_{r,i}) \cdot \frac{[TE]_{total,i} \cdot (A\beta + B)}{[TE]_{ref,i}}$	$T_{r,i}$ is the toxic-response factor for trace element $i$ (e.g. 1 for Zn, 5 for Cu, Ni, and Pb, and 30 for Cd according to Håkanson (1980)) $[trace\ elements]_{total,i}$ is the total concentration (mg kg <sup>-1</sup> dry soil) of trace element $i$ $A$ is the percentage of total trace element recovered after the first extraction step $\beta$ is the toxic index of trace elements recovered after the first extraction step; The $\beta$ value is defined as a function of the $RAC$ value (see 1.2) as follows: 1.0 for $1 < RAC \leq 10$ , 1.2 for $11 \leq RAC \leq 30$ , 1.4 for $31 \leq RAC \leq 50$ , and 1.6 for $RAC > 50$ (Zhu et al., 2012) $B$ is the sum of percentages of total trace element recovered in all extraction steps except the first one $[trace\ elements]_{ref,i}$ is the background or regulatory threshold concentration (mg kg <sup>-1</sup> dry soil) used a reference for trace element $i$
<b>Indicators calculated on contaminated soils</b>		
Health risk index	$HRI = \frac{[TE]_{food} \cdot D_{food}}{[TE]_{ref} \cdot BW}$	$[trace\ elements]_{food}$ is the concentration (mg kg <sup>-1</sup> dry food) of a given trace element in food $D_{food}$ is the daily intake of food (kg dry food day <sup>-1</sup> ) $[trace\ elements]_{ref}$ is the reference dose of trace element ingested with food (mg kg <sup>-1</sup> dry food day <sup>-1</sup> ) $BW$ is the average body weight of an exposed human being (adult or child)

Target hazard  
quotient

$$THQ = HRI \cdot \frac{EF_{nc}}{ME}$$

$EF_{nc}$  is the exposure frequency (day) to non-carcinogenic effects of a given trace element for 1 year

$ME$  is the maximal exposure (365 days) to carcinogenic or non-carcinogenic effects of a given trace element for 1 year

Hazard index

$$HI = \sum_{i=1}^n THQ_i$$

$THQ_i$  is the target hazard quotient for each trace element  $i$

Target cancer  
risk

$$TCR = \frac{[TE]_{food} \cdot D_{food} \cdot CPS \cdot EF_c}{BW \cdot ME}$$

$CPS$  is the carcinogenic potency slope (kg body weight day  $mg^{-1}$  trace element)

$EF_c$  is the exposure frequency (day) to carcinogenic effects of a given trace element for 1 year

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**Table 5. Recent reviews of empirical research-based knowledge on the fate of contaminants contained in organic waste following soil application**

<b>Publication (reviewed studies)</b>	<b>Waste streams and treatments</b>	<b>Nutrients, contaminants and impacts considered</b>	<b>Main findings and recommendations</b>
Bondarczuk et al. (2016): dozens	Sewage sludge	Antibiotics, antibiotic-resistant bacteria and antibiotic resistance genes Links: trace elements-pathogens, via maintenance of antibiotic resistance, organic contaminants - pathogens, via antibiotics	<ul style="list-style-type: none"> <li>• There is increased pharmaceutical pollution in the environment</li> <li>• Significant correlations have been observed between the presence of trace elements and antibiotics in sludge</li> <li>• Spreading contributes to the dissemination and further development of antibiotic resistance</li> <li>• Need for further investigations on human and ecological risk assessment and the fate of antibiotic resistant bacteria and genes</li> </ul>
Hargreaves et al. (2008): 30	Biowaste compost	Nutrients, trace elements, persistent organic contaminants	<ul style="list-style-type: none"> <li>• Safe use in agriculture of municipal solid waste compost depends on the production of good quality compost, specifically, mature and sufficiently low in trace elements and salt content</li> <li>• The best method of reducing the trace element content and improving the quality of this type of compost is early source separation</li> <li>• Further bioavailability research would be warranted</li> </ul>
Insam et al. (2015): dozens	Manure, anaerobic digestion	N, P, K, trace elements, organic contaminants, antibiotics, pathogens	<ul style="list-style-type: none"> <li>• Despite concerns of environmental impacts, odour emissions, and pathogen spreading associated with the production and agricultural recycling of digestate, its use is preferable to that of raw manure, from agricultural and environmental standpoints</li> </ul>
Nkoa (2014): dozens	Anaerobic digestates (unspecified)	Environmental impacts (atmospheric, water and soil pollution; chemical and biological)	<ul style="list-style-type: none"> <li>• Efficacy of digestates as fertilizer lies between that of livestock manure and mineral fertilizers</li> <li>• Digestate has a higher potential than livestock manure regarding ammonia and nitrous oxide emissions, and may contribute to soil contamination by trace elements</li> </ul>

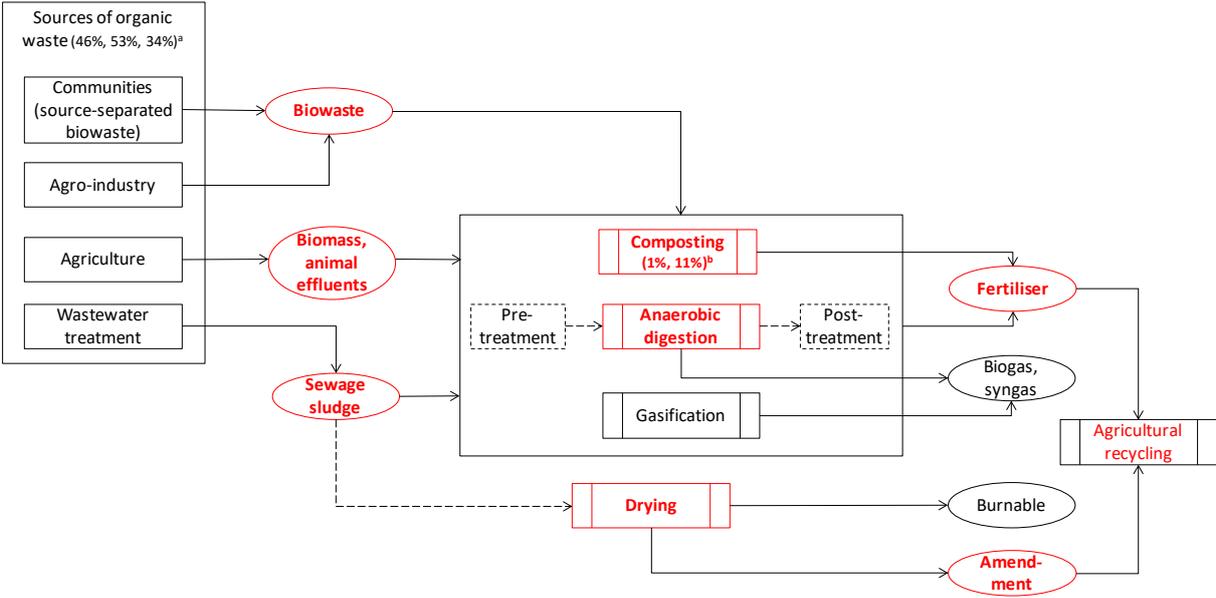
**Table 6. Key aspects of biophysical processes overlooked in RA and LCA, and suggested models to address them**

Key aspects	Proposed models and approaches	Readiness of approaches	References
<b>Trace elements</b>			
Trace element speciation in organic waste	Creation of a qualitative typology of OWs based on their basic characteristics related to physicochemical parameters driving trace element speciation in OW, such as pH and redox potential.	Relatively easy to implement, but would require a research project to develop a classification method similar to TyPol (Servien et al., 2014).	(Tella et al., 2016)
Colloidal transfer of contaminants	Models for simulating the colloid facilitated transport of trace elements in fate modelling (e.g. HYDRUS 1D).	Fully functional model.	(Šimůnek et al., 2012)
Multiple soil layers for the prediction of trace element fates in soil	Empirical or semi-mechanistic models to determine the fate of trace elements per soil layer over a given period.	Would require a review and validation of existing models.	(Jacques et al., 2008)
Characterization of the binding properties of dissolved organic matter	Empirical regressions based on soil physicochemical parameters to predict specific dissolved organic matter binding properties.	Fully functional approach, yet requiring a literature review to identify models for specific trace elements. Implemented in LC-IMPACT (Verones et al., 2020).	(Owsianiak et al., 2013)
Changes in trace element availability over a time course and consequences in terms of ecotoxicological endpoints	Data acquisition with DGT. Ecotoxicological endpoints based on DGT-available trace elements.	Fully functional approach, requiring empirical analyses and exploration of toxicity databases to identify suitable ecotoxicological endpoints.	(Zhao et al., 2006) (Thakali et al., 2006b)
Influence of soil organisms on trace element (bio)availability	Data acquisition with a biotest (e.g. RHIZOtest). Models to predict bioavailability (e.g. WHAM-F <sub>TOX</sub> ).	Fully functional approach, requiring empirical analyses and model calibration to specific conditions.	(Bravin et al., 2010b) (Tipping and Lofts, 2015)
<b>Organic pollutants</b>			

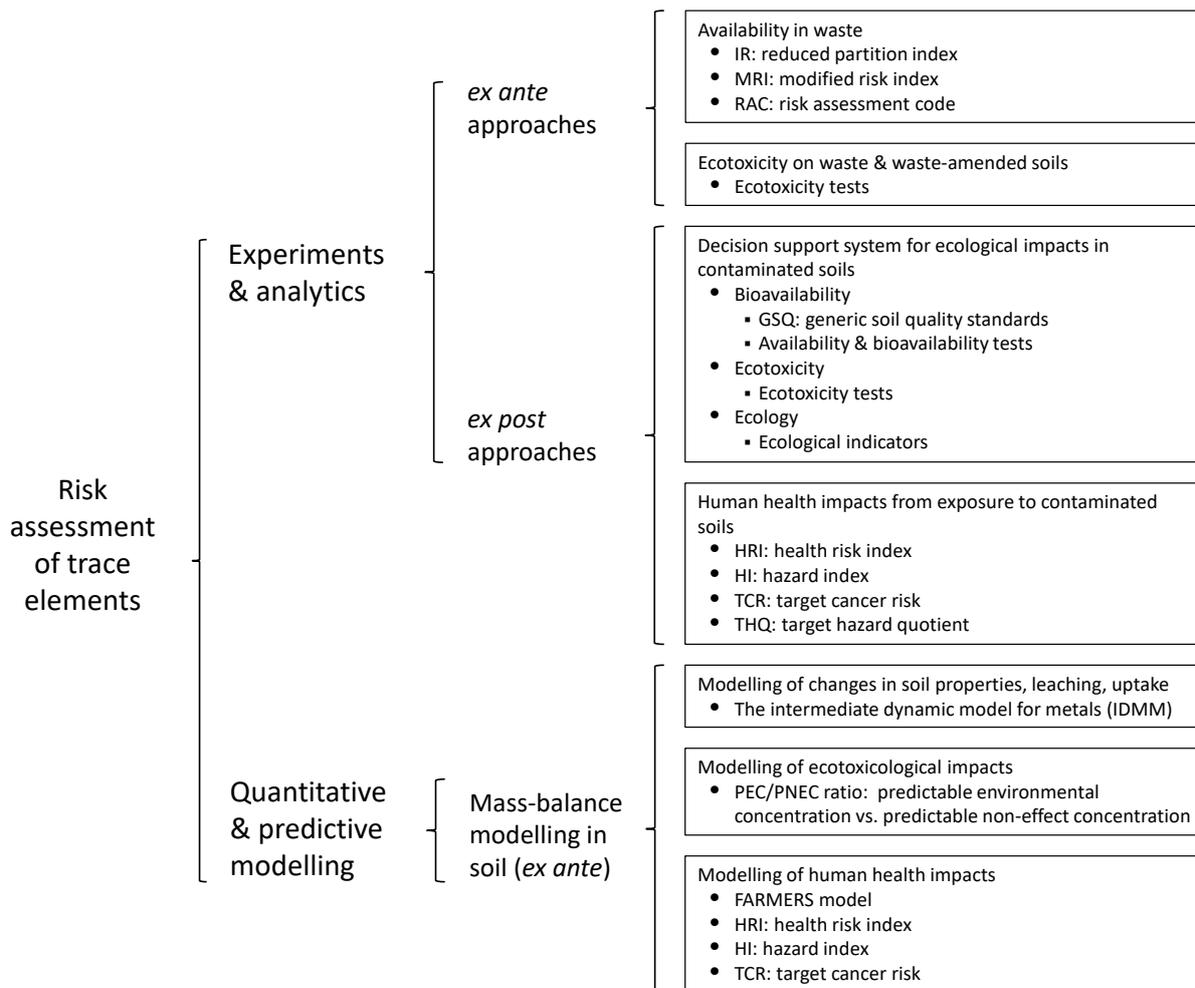
Fate of organic contaminants post spreading (mineralization, bound residues)	Plant uptake models (e.g. SimpleTreat 3) coupled with models for water and solute transport in soil.	Fully functional approach, requiring some model coupling and calibration. Data intensive.	(Legind et al., 2012) (Polesel et al., 2015)
Interactions between organic contaminants and organic matter after spreading	Models describing organic contaminants and organic matter interactions (e.g. COP-Soil).	Fully functional approach, requiring some model calibration. Data intensive.	(Geng et al., 2015) (Goulas, 2006) (Brimo et al., 2018a, 2018b)
Heterogeneity in the fate of organic contaminants spread on soils due to: interactions between organic contaminants and OW, various processes involved in the fate of organic compounds (sorption, degradation, production of transformation products, volatilization, leaching, uptake, etc.), the modification of soil properties following OW application, and the diversity of quantities and qualities among OW	Models used in the context of European pesticides registration.	Fully functional approaches, requiring some model selection and calibration. Data intensive.	(ECHA, 2016)
	Agricultural management models featuring water, chemicals, and heat transport modules; a plant growth module; an evapotranspiration module; an equilibrium chemistry process module; an organic matter-N cycling module; a pesticide fate module; and a management practices module including crop rotations, tillage, irrigation, fertilizer, pesticide and manure applications (e.g. RZWQM, STICS).	Fully functional approaches, requiring some model selection and calibration. Data intensive.	(Ma et al., 1998) (Malone et al., 2004) (Brisson et al., 2003)
	Sorption models (e.g. HYDRUS-2D).	Fully functional model.	(Šimůnek et al., 2012) (Filipović et al., 2016a, 2016b, 2014)
<b>Pathogens and antibiotic resistance</b>			
Direct exposure to antibiotic-resistant pathogens or to	WHO framework for food safety. Microbiological risk analysis (MRA).	Fully functional approach.	(Manai, 2017; WHO, 2010)

resistance determinants (and subsequent horizontal gene transfer to bacterial pathogens on or within a human host)			
Antibiotic resistance	Explore the gut of soil microfauna as a hotspot of antibiotic resistance; apply metagenomic analyses of soil bacterial communities to describe ARG, their origins, and their potential to be transferred between species.  Explore specific survival of ARG-carrying bacteria regarding competition, predation, and adaptation to soil environmental conditions.	Quite well described approach, requiring empirical analyses and selection of specific models to fit observational data.  Quite well described approach, requiring empirical analyses and selection of specific models to fit observational data. Data seems scarce to date.	(Ding et al., 2019) (Nesme and Simonet, 2015)  (van Veen et al., 1997)
Uncertainty management, quality of dose-response datasets	Bayesian networks.	Fully functional approach.	(Fenton and Neil, 2012)
Behaviour of a zoonotic pathogens in OW	Combined HACCP and QMRA.	Fully functional approach.	(Westrell, 2004; Westrell et al., 2004)
<b>Other aspects</b>			
Mixture effects	No specific model, but further population of the Information Platform for Chemical Monitoring data (IPChem) and a number of areas of further study have been suggested as a way forward	Immature approach.	(Bopp et al., 2016)
Effect of repetitive and long-term applications of organic waste	Models and databases to predict/fit pH and organic matter patterns over time.	Various semi-mechanistic models exist, which are rather data intensive, thus requiring empirical analyses and model calibration to local conditions.	(Clivot et al., 2019; Schädel et al., 2019)

# Figures



**Figure 1. Main global organic waste sources and disposal routes. Treatments and products studied here are highlighted. Less common/optional routes and treatments are dashed. Statistical sources: Hoornweg and Bhada-Tata (2012); UNEP (2015). Notes: <sup>a</sup> Contribution of organic waste to total waste composition for the world, low income and high income countries, respectively. <sup>b</sup> Contribution of composting to total waste management for low income and high income countries, respectively**



**Figure 2. Overall framework of the approaches used for risk assessment of ecotoxicological and human health impacts of trace elements (applicability for organic waste), indicators are detailed in Table 4**

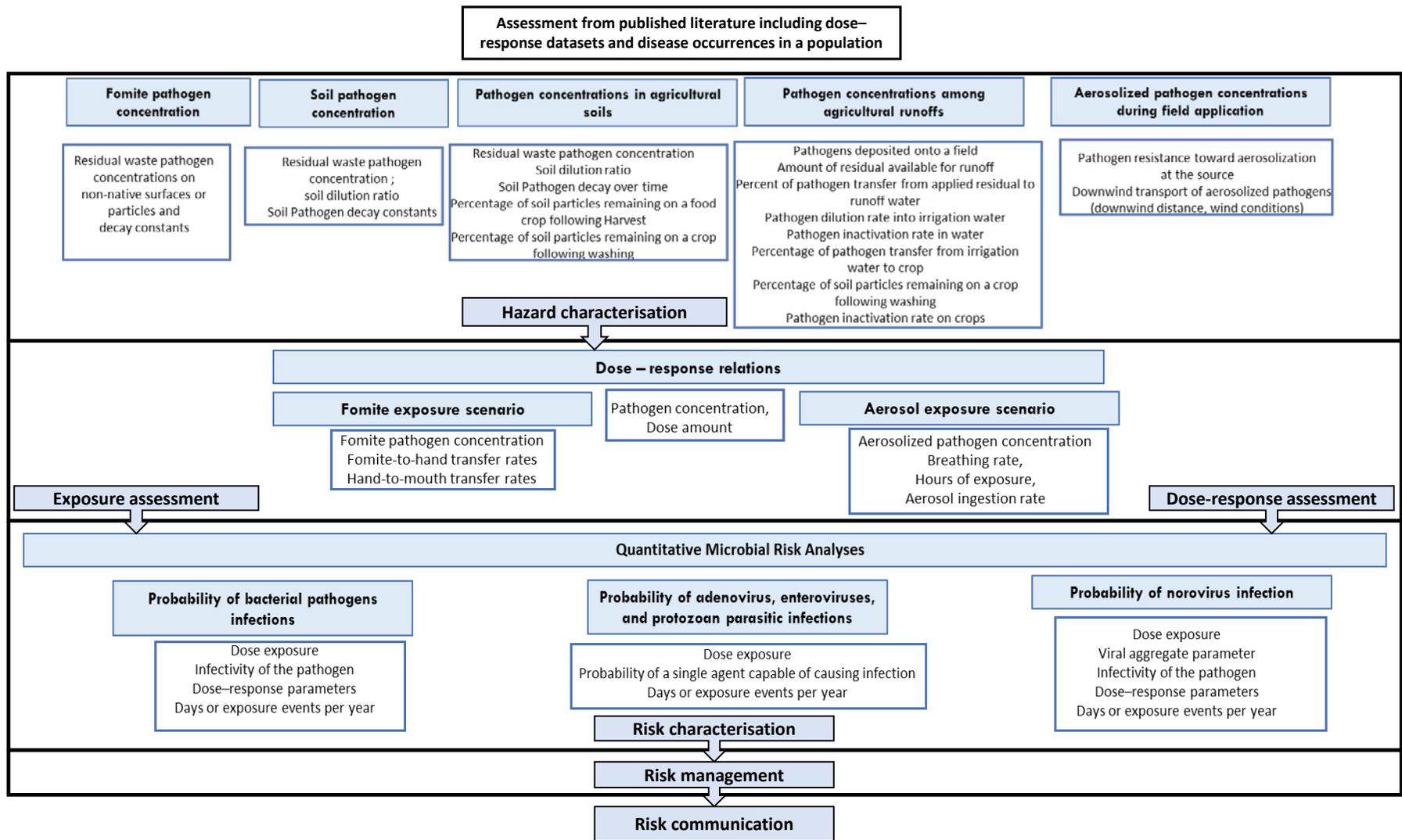


Figure 3. Schematic depiction of quantitative microbial risk assessment (QMRA) processes associated with each parameter or step in the land application of residual waste (manure and Class B biosolids). Exposure process during scenarios involving fomite, soil, crop, and aerosol exposure are included

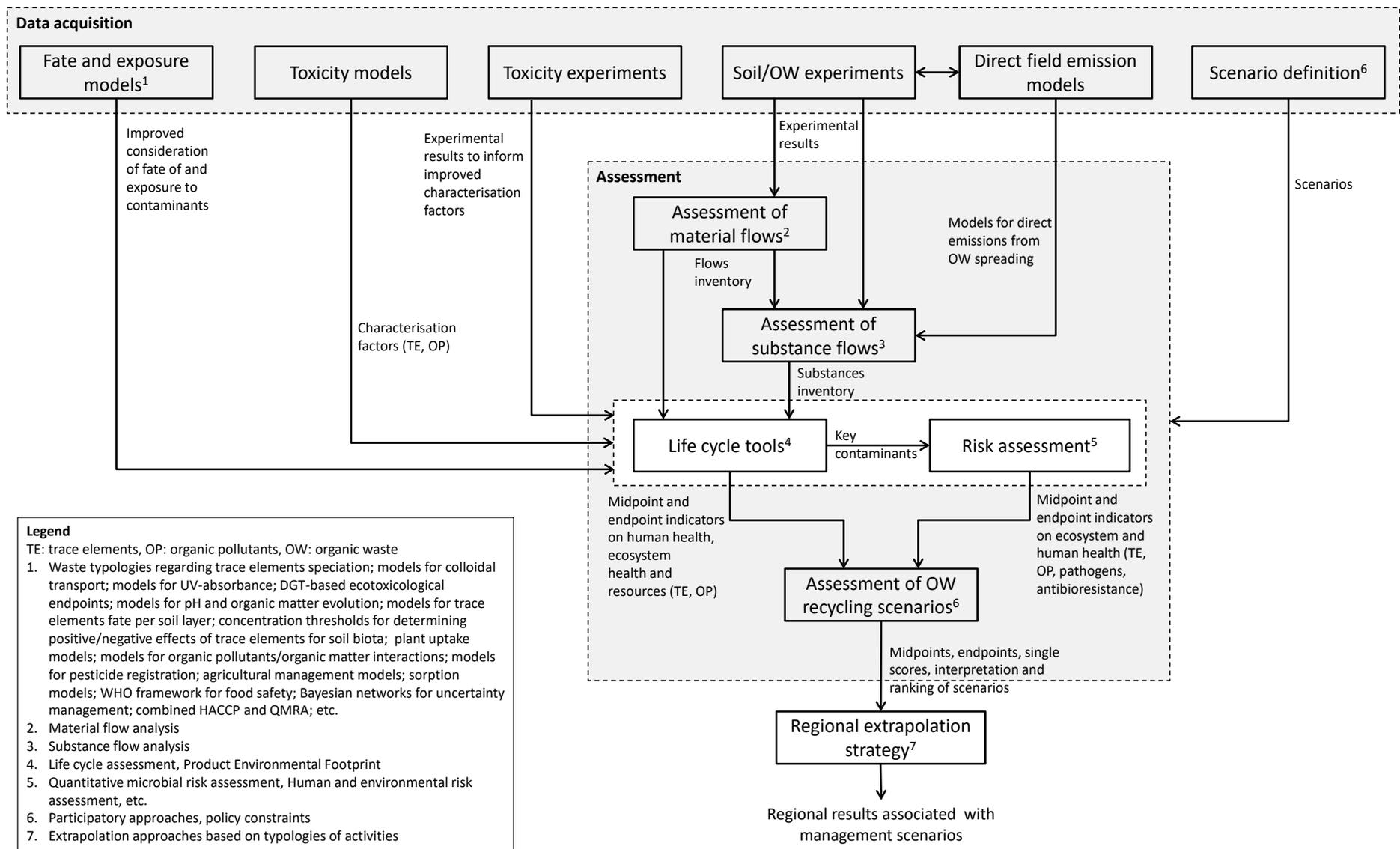


Figure 4. Diagram of a comprehensive framework for environmental assessment of agricultural recycling of organic waste

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## Footnotes

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- <sup>i</sup> Trace elements usually refer to chemical elements in soil that occur at lower than 100 mg kg<sup>-1</sup> concentration. The term ‘trace elements’ encompasses metals, metalloids, and non-metals and was herein preferred over the many other unsatisfactory synonyms commonly found in the literature (e.g. heavy metals, potentially toxic metals, etc.) (Duffus, 2002).
- <sup>ii</sup> Other contaminants present in organic waste, such as toxins (i.e. toxic substances produced by living entities) and particulate matter, were excluded from this review, mainly because their study is beyond the coauthors’ scope of expertise. In LCA, particulate matter formation is treated as an impact category separate from toxicity (Huijbregts et al., 2016; Humbert et al., 2011). There is abundant literature on toxin (e.g. Böhnel and Lube 2000; Ibelings and Chorus 2007; Then 2010; Brown et al. 2011) and particulate matter (e.g. US EPA 2005; Levy 2016) consideration in risk assessment. Toxins are not taken into account in LCA.
- <sup>iii</sup> Characterization factor: a factor applied to convert a given flow of a given substance, assigned to an impact category, into a ‘potential impact’ expressed in the common unit of all flows contributing to that impact category (ISO, 2006a). Generally, this common unit corresponds to that of one of the substances potentially contributing to the category at hand, such as CO<sub>2</sub> for all greenhouse gases.
- <sup>iv</sup> Multimedia model: a type of model used in environmental chemistry that expresses the diffusion of a substance between environmental media/compartments based on the chemical properties of the concerned substance (e.g. partition coefficients, etc.).
- <sup>v</sup> “Speciation takes into account chemical and physical properties such as the element isotopic composition, oxidation state, coordination and molecular structure” (Reeder et al., 2006).
- <sup>vi</sup> EU-TGD has been deprecated and replaced, since 2007, by the REACH legislation (EC, 2007), but many approaches and models were recycled in REACH.
- <sup>vii</sup> Quantitative structure-activity relationship (QSAR): “computer-based methods able to quickly predict and assess the toxicity of large numbers of chemicals using a mathematical algorithm implemented in a software programme” (REACHnano, 2015).
- <sup>viii</sup> Dissipation half-life times (DT<sub>50</sub>): rate of degradation of chemicals, a period after which half of the initial load has degraded. ‘Dissipation’ refers to “a composite of processes describing volatilization, wash-off,

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leaching, hydrolysis, chemical and biological degradation, and other individual processes” (Fantke and Juraske, 2013)

- ix ECPA LET (<http://www.ecpa.eu/industry-resources/reach-registration-evaluation-authorisation-and-restriction-chemicals>)
- x According with the FAO/WHO Codex Alimentarius (<http://www.fao.org/3/y1579e/y1579e03.htm>), HACCP is “a system which identifies, evaluates, and controls hazards which are significant for food safety”.
- xi Illuviation: “accumulation of dissolved or suspended soil materials in one area or horizon as a result of eluviation from another” (<https://www.merriam-webster.com/dictionary/illuviation>)
- xii A Bayesian network is a flexible graphical model with variables represented by nodes and connected by directed arcs indicating dependencies and strength of the causal links (Nielsen and Verner Jensen, 2007). Each variable is assigned a probability distribution conditional to its parent variables. Uncertainty in a Bayesian network model is thus represented by a probability distribution. By construction, Bayesian networks can characterize and describe complex relationships among variables associated with a complex outcome (Donald et al., 2009).
- xiii An adverse outcome pathway “describes a logical sequence of causally linked events at different levels of biological organization, which follows exposure to a chemical and leads to an adverse health effect in humans or wildlife” (online OECD resource <https://doi.org/10.1787/2415170X>); an aggregate exposure pathway is a similar concept, proposed as a framework to organize dispersed exposure data (Tan et al., 2018).