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# Anthropogenic global shifts in biospheric N and P concentrations and ratios and their impacts on biodiversity, ecosystem productivity, food security, and human health

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1 **Anthropogenic global shifts in biospheric N and P concentrations and ratios and**  
2 **their impacts on biodiversity, ecosystem productivity, food security, and human**  
3 **health**

4  
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43 **Abstract**

44 The availability of carbon (C) from high levels of atmospheric carbon dioxide (CO<sub>2</sub>) and anthropogenic  
45 release of nitrogen (N) is increasing, but these increases are not paralleled by increases in levels of  
46 phosphorus (P). The current unstoppable changes in the stoichiometries of C and N relative to P have no

47 historical precedent. We describe changes in P and N fluxes over the last five decades that have led to  
48 asymmetrical increases in P and N inputs to the biosphere. We identified widespread and rapid changes in  
49 N:P ratios in air, soil, water, and organisms and important consequences to the structure, function, and  
50 biodiversity of ecosystems. A mass-balance approach found that the combined limited availability of P and  
51 N was likely to reduce C storage by natural ecosystems during the remainder of the 21st Century, and  
52 projected crop yields of the Millennium Ecosystem Assessment indicated an increase in nutrient  
53 deficiency in developing regions if access to P fertilizer is limited. Imbalances of the N:P ratio would likely  
54 negatively affect human health, food security, and global economic and geopolitical stability, with  
55 feedbacks and synergistic effects on drivers of global environmental change, such as increasing levels of  
56 CO<sub>2</sub>, climatic warming, and increasing pollution. We summarize potential solutions for avoiding the  
57 negative impacts of global imbalances of N:P ratios on the environment, biodiversity, climate change,  
58 food security, and human health.

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62 **Keywords**

63 Biospheric N and P concentrations, water, soil and plant N:P ratios, anthropogenic global shifts,  
64 biodiversity, ecosystem productivity, food security, human health

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69 **1. Introduction**

70 The availability of carbon (C) from high levels of atmospheric carbon dioxide (CO<sub>2</sub>) and anthropogenic  
71 inputs of nitrogen (N) on ecosystems are increasing. These increases are, however, not paralleled by those  
72 of phosphorus (P), and current inexorable changes in the stoichiometry of C and N relative to P have no  
73 historical precedent (Peñuelas et al., 2013). The shifts in organisms' N:P ratio resulting from different  
74 environmental conditions are strongly related with shifts in ecosystems structure and function (Sterner &  
75 Elser, 2002; Loladze & Elser, 2011; Peñuelas et al., 2013). Imbalances between these two nutrients, N and  
76 P in natural, semi-natural, and managed ecosystems (Liu et al., 2010; Sardans & Peñuelas, 2012; Carnicer  
77 et al., 2015; Ulm et al., 2016; Peñuelas et al., 2013, Delgado-Baquerizo et al., 2017; Hu et al., 2018).  
78 reduce C capture and global food provision and security (Van der Velde et al., 2014; Lu & Tian, 2017;  
79 Peñuelas et al., 2017a; Wang et al., 2018a; Kahsay, 2019). These effects may be further exacerbated in  
80 cropland in the future by limited access to reserves of mineable P (Cordell et al., 2011; MacDonald et al.,  
81 2011; Li et al., 2016; Mew, 2016; Weikard, 2016; Lun et al., 2018).

82 Changes in the global P cycle, status and resources, together with associated economic impacts,  
83 were first debated at least a century ago (Liu et al., 2017). More recent studies have recognized that  
84 increases in N:P ratios with rising anthropogenic release have consequences for P and N cycling in soil and  
85 water, biodiversity, and ecosystem function (Elser et al., 2010a, b; Peñuelas et al., 2012; Peñuelas et al.,  
86 2013). The link between increasing imbalances in biospheric N:P ratios and their impacts on global  
87 ecology and socioeconomics is supported by evidence from many studies that have identified clear  
88 relationships between drivers of global change and anthropogenic N and P releases and with shifts in  
89 ecosystem N:P ratios. These studies have also demonstrated feedbacks and synergies of shifts in the N:P  
90 ratios in soil, water, and organisms with increases in atmospheric CO<sub>2</sub> concentrations, climate change,  
91 species invasions, ecosystem eutrophication, and changes in soil use (Sardans & Peñuelas, 2012; Sardans  
92 et al., 2012a, 2013, 2016a, 2017a; Zhang et al., 2013; Ferretti et al., 2014; Gargallo-Garriga et al., 2014; He  
93 & Dijkstra, 2014; Deng et al., 2015; Yuan & Chen, 2015; Chen et al., 2016; Delgado-Baquerizo et al., 2016;  
94 Jiao et al., 2016; Zhu et al., 2016; Kruk & Podbielska, 2018; Schmitz et al., 2019; Yuan et al., 2018; Peng et  
95 al., 2019).

96 We reviewed our current understanding and identified gaps in our knowledge of the effects of  
97 global change on ecosystem N and P ratios and associated impacts on ecosystem function, food security,  
98 and socioeconomics. Specifically, we addressed (i) the shifts in N:P ratios mediated by anthropogenic  
99 drivers of global change, (ii) the impacts of shifts in N:P ratios of human inputs on organisms,  
100 communities, and ecosystems, (iii) the impacts of N and P ratios on food security and human health, and

101 (iv) political, economic, and technological strategies to mitigate the negative impacts of unbalanced N:P  
102 ratios.

103

## 104 **2. Shifts in N:P ratios mediated by anthropogenic drivers of global change**

105 Further evidences accumulated in the last six years after Peñuelas et al (2013) robustly confirm the  
106 inexorable changes in the stoichiometry of C and N relative to P, which have no historical precedent (Fig.  
107 1). Furthermore, the increasing emissions of NO<sub>x</sub> and NH<sub>3</sub> to the atmosphere lead to large imbalances in  
108 the ratios of total atmospheric N:P deposition, with higher ratios for total atmospheric N:P than standard  
109 averages for soil, water, and organisms (Fig. 2).

110 Activities involved in food production, such as the application of fertilizer, cultivation of N<sub>2</sub>-fixing  
111 species of crop plants, livestock husbandry, and the release of N and P to the atmosphere from the  
112 combustion of fossil fuels, which are re-deposited on the surface, are key historical and contemporary  
113 contributors of bio-active N and P and drivers of these nutrient imbalances (Peñuelas et al., 2012; 2013;  
114 Yuan et al., 2018). For example, the N:P ratios of atmospheric total depositions are higher than the  
115 average N:P ratios of waters, soils, and organisms (Fig. 3).

116

### 117 *2.1. Effects of drivers of global change on N:P ratios of water, soil, and plants*

118 Many recent studies have reported increases in the N:P ratio in the soil, water and plants of terrestrial  
119 and aquatic ecosystems (Crowley et al., 2012; Lepori & Keck, 2012; Hessen et al., 2013; Yu et al., 2018; Xu  
120 et al. 2019; Jirousek et al., 2011; Blanes et al., 2013; Huang et al., 2016a; Zivkovic et al., 2019) in response  
121 to high levels of atmospheric N deposition (Table 1).

122 Some studies, however, have not clearly detected changing patterns in soil-plant C:N:P  
123 stoichiometry along natural gradients of N deposition (Stevens et al., 2011). The decrease in N deposition  
124 in some areas of North America and Europe in recent decades has substantially decreased N:P ratios in  
125 lakes (Gerson et al., 2016; Isles et al., 2018). Atmospheric P deposition is also increasing due to the rising  
126 levels of anthropogenic emissions of P to the atmosphere (3.5 Tg P y<sup>-1</sup>), which have led to current net  
127 continental and oceanic rates of P deposition of 2.7 and 0.8 Tg P y<sup>-1</sup>, respectively (Wang et al., 2015a). This  
128 deposition has been particularly intense in areas of the world with emerging economies, such as eastern  
129 Asia, which may account for the low N:P ratios reported in some freshwater systems in Japan (Miyazako  
130 et al., 2015).

131 The P cycle and N:P ratios are affected by many drivers of global change other than anthropogenic  
132 emissions of N and P (Table 1). Higher concentrations of atmospheric CO<sub>2</sub> are correlated with decreases in  
133 plant N and P concentrations and increases in the ratios of C:N and C:P (Peñuelas & Matamala 1990;

134 Peñuelas & Estiarte 1997; Sardans et al., 2012b; Deng et al., 2015), but the effects on plant N:P ratios are  
135 less clear. For example, recent meta-analyses have found that rising CO<sub>2</sub> concentrations have led to  
136 decreases in N:P ratios in different plant tissues (Deng et al., 2015) and woody plants but not herbaceous  
137 plants or mosses (Yue et al., 2017). Yuan and Chen (2015) in a meta-analysis of 315 studies with non-  
138 differentiation of plant organs observed an overall decrease in N:P ratios in controlled field conditions  
139 under elevated levels of CO<sub>2</sub>. However, another review of 215 studies (Sardans et al. 2017b), mostly under  
140 controlled field conditions, revealed that increased atmospheric concentrations of CO<sub>2</sub> led to decreased  
141 N:P ratios in roots, but not in leaves. Moreover, King et al. (2015) reported increased N:P ratio in one  
142 phytoplankton species, decreased N:P ratio in three other species, and no change in N:P ratio in other  
143 three species under high levels of CO<sub>2</sub>, thus suggesting that the effects of CO<sub>2</sub> enhancement on  
144 stoichiometry appear to be species-dependent. It is thus likely that the ongoing increases in atmospheric  
145 CO<sub>2</sub> concentrations are reducing N:P ratios in plants, which would be apparently consistent with the GRH  
146 for plants under favorable growth conditions (Sterner and Elser, 2002). The hypothesis that atmospheric  
147 increases in CO<sub>2</sub> stimulate higher plant uptakes of P than N (Deng et al., 2015) thus remains to be  
148 unequivocally demonstrated but begins to have some observational and experimental support (Table 1).

149 Less information is available regarding the relationships of the rise in atmospheric CO<sub>2</sub>  
150 concentration with N and P concentrations and N:P ratio in soil. Huang et al. (2014) observed that a rise in  
151 atmospheric CO<sub>2</sub> concentration did not change total soil P concentrations but increased P-available to  
152 plants and decreased more recalcitrant soil-P. Increased CO<sub>2</sub> concentrations can indirectly decrease soil N  
153 and P concentrations by several mechanisms including higher plant N and P demands, higher N and P  
154 resorption rates and higher exudates production and N and P uptake (Jin et al. 2015; Liu et al. 2018; Van  
155 Vuuren et al. 2018). However, the potential impact of CO<sub>2</sub> enhancement of soil N:P ratios also remains  
156 inconclusive.

157 The changes in N and P concentrations and N:P ratios in soil-plant systems in response to warming  
158 vary with biome and soil type (Sardans et al., 2008b, 2017b; Yue et al., 2017). They also suggest that low  
159 soil N and P concentrations tend to be associated with higher temperatures along natural long-term  
160 climatic gradients, but the reverse occurs for phenotypic responses of species to N in short-term field  
161 studies with climatic manipulation (Yuan et al. 2017). Several studies have indeed reported decreases in  
162 aboveground plant N:P ratios under warming that were attributed to the greater allocation of P to stems  
163 and/or to greater plant growth capacity (Dudareva et al. 2018; Wang et al. 2018d,2019b). The effects of  
164 warmer temperatures on plant and soil C:N:P ratios along natural gradients are not easy to distinguish  
165 from those of precipitation, radiation or atmospheric N deposition, which frequently correlate with the  
166 geographical temperature gradient (Jiao et al. 2016).

167 The projected total land surface occupied by warm semi-arid surfaces may become 38% larger in  
168 2100 compared to the present (Rajaud and de Noblet-Ducoudré, 2017; Huang et al. 2016, 2017). The  
169 effects of aridity (combination of high temperatures with low precipitation) on plant N:P ratios along  
170 natural long-term climatic gradients also differ from the effects in field studies with climatic manipulation  
171 (Yuan et al., 2017; Luo et al., 2018a, b; 2019). Increases in canopy N and P concentrations and decreases  
172 in plant C:P and N:P ratios have been recorded along transects of increasing aridity. Future increases in  
173 aridity are also likely to lead to lower N:P ratios in atmospheric depositions (Zarch et al., 2017; Lin et al.,  
174 2018). In contrast, plant N and P concentrations have tended to decrease and N:P ratios have tended to  
175 increase (He & Dijkstra, 2014; Yuan & Chen 2015) in short-term manipulation studies where water  
176 availability decreased (Jiao et al., 2016; Luo et al., 2018b) (Fig. 4), despite between-site variations in foliar  
177 N and P concentrations (Sardans & Peñuelas, 2007, 2013a,b; Sardans et al., 2008a, b, 2017b; Luo et al.,  
178 2018b). These increases in foliar N:P ratios in response to experimental drought are generally because low  
179 soil-water contents limit P uptake more than N uptake (Sardans & Peñuelas, 2013a; Urbina et al., 2015;  
180 Sardans et al., 2017b; Luo et al., 2018a, b). Plants notably respond to sudden conditions of drought and  
181 warming in manipulated field experiments with increased allocations of N, P, and potassium (K) to roots,  
182 leading to lower root N:P ratios associated with higher primary metabolism linked to growth, protein  
183 synthesis, and pathways of energy transfer (Gargallo-Garriga, et al., 2014;2015). In contrast, shoots have  
184 lower concentrations of N and P and higher N:P ratios linked to the activation of anti-stress metabolic  
185 pathways (Gargallo-Garriga et al., 2014;2015).

186 Contrasting responses of soil nutrients to short- and long-term drought conditions have also been  
187 reported, where soil N and P concentrations tended to decrease with aridity in natural (long-term)  
188 gradients but tended to increase in some biomes and soil types under conditions of short-term drought  
189 (Yuan et al., 2017) (Fig. 4). Delgado-Baquerizo et al. (2013) observed a negative effect of aridity on the  
190 concentration of soil organic C and total N, but a positive effect on the concentration of inorganic P in  
191 semi-arid and arid areas. In these conditions, P and N shift from soil to plants, so plant communities  
192 adapted to long-term drought conditions retain higher levels of N and P (Luo et al., 2018a, b). These  
193 effects are consistent with observations of lower ratios of N:P in water from deeper soil layers and  
194 indicate P limitation in soil under arid climatic conditions (Sardans & Peñuelas, 2014). Long evolutionary  
195 processes likely drive the conservative use of nutrients in droughted environments.

196 Our understanding of the impacts of extreme climatic events on plant-soil stoichiometry is  
197 limited. For example, Wang et al. (2016) observed that rapid production of litter in coastal wetland during  
198 typhoons led to larger and faster releases of N and P, characterized by low N:P ratios, but the associated  
199 potential impacts on soil microbial communities and trophic chains were unclear. The projected increases

200 in extreme climatic events indicate that quantifying the impacts on N and P cycles and their ratios is  
201 essential.

202 Invasion by non-native plants is an emerging driver of global environmental change (Seabloom et  
203 al., 2015), where establishment depends on differences in the uptake and use efficiency of nutrients  
204 between native and invasive species (Daehler, 2003, González et al., 2010; Peñuelas et al., 2010; Sardans  
205 et al., 2017a). The impacts of invasive species on N and P cycles and stoichiometry on the plant-soil  
206 system may vary between nutrient-rich and nutrient-poor ecosystems (González et al., 2010; Matzek,  
207 2011; Sardans et al., 2017a). For example, successful invasive species have higher capacities to take up  
208 and efficiently use nutrients that are limited (Aragon et al., 2014; Wang et al., 2015b; 2018a; Ulm et al.,  
209 2016; Sardans et al., 2017a), so the concentrations of N and P in photosynthetic tissues tend to be higher  
210 in invasive than native species. Total soil N concentrations and availabilities of N and P correlated with  
211 higher mineralization capacity are higher for invasive species, particularly in nutrient-poor environments  
212 (Sardans et al., 2017a). A higher capacity for N and P resorption in invasive species may account for these  
213 differences in concentrations and ratios of N and P (Sardans et al., 2017a and references therein). The  
214 possible effects of anthropogenic changes in soil and water N:P ratios on competitive relationships  
215 between native and invasive species have received little attention, but changes in soil elemental  
216 composition and stoichiometry have been linked with the success of alien species (Sardans et al., 2017a).  
217 Further research is clearly required to improve our understanding of the relationships between successful  
218 species invasion and ecosystem N and P cycles and stoichiometry, including the role of the interaction  
219 with other drivers of global environmental change. For example, increased flooding intensity in coastal  
220 wetlands due to sea-level rise drives the effects of invasive plant species on N and P cycling (Wang et al.,  
221 2015b, 2016b, 2018b).

222 Anthropogenic land-use changes are heterogeneous, but they tend to be associated with changes  
223 in soil N and P concentrations and N:P ratios (Wang et al., 2014; Zhao et al., 2015a; Liu et al., 2018; Zhou  
224 et al., 2018a, b; Urbina et al., 2019). For example, invasion by shrubs on grassland previously grazed by  
225 livestock is frequently associated with changes in soil-plant N and P concentrations and N:P ratios (Bui &  
226 Henderson, 2013; Urbina et al., 2019). These changes go in parallel to a transition from rapid nutrient  
227 cycling, with high concentrations of N and P in the plant-soil system, to slower N and P cycling, with lower  
228 concentrations of N and P in the system, and higher accumulations of N and P stocks in the higher  
229 aboveground shrub biomass (Zhou et al., 2018a, b; Urbina et al., 2019) that has a larger capacity to obtain  
230 nutrients from deep soil layers (Blaser et al., 2014). These trends, however, vary with the traits of the  
231 shrub species (Knapp et al., 2008; Eldridge et al., 2011; Zhou et al., 2018b). Shifts in soil N:P ratios during

232 processes of habitat transition may vary with soil layer, but soil N:P ratios tend to increase in the upper  
233 layers (Feng & Bao, 2018; Zhou et al., 2018a, b).

234 If croplands replace tropical forests, which have high rates of biological N fixation, the rates may  
235 decrease as a result of this anthropogenic land-cover change. These likely effects of land use change have  
236 not been investigated, even though they may have strong impacts on both N and P, on N because of  
237 increased leaching and biological N fixation, and on P because of erosion and replacing a community  
238 adapted to retain P by others that are not.

239 So, in summary, the current global trend is generally towards increasing N:P ratios in water, soil  
240 and plants, but with many exceptions. For example, widespread P enrichment of crop soil has led to  
241 declines in N:P ratios in several parts of the world (Peñuelas et al., 2009; Wang et al., 2015b; Delgadillo-  
242 Vargas et al., 2016, Wironen et al., 2018). The differences in immobilization, leaching, and volatilization  
243 between the two elements leads to higher soil retention of P than N (Peñuelas et al., 2012; 2013). This trend  
244 in P retention tends to be more pronounced where the density of livestock, particularly pigs and/or poultry  
245 is high (Arbuckle & Downing, 2001; Gomez-Garrido et al., 2014, Hentz et al., 2016; Peñuelas et al., 2009;  
246 Wironen et al., 2018), because the manure waste generated is characterized by very low N:P ratios (Humer  
247 et al., 2015; Oster et al., 2018). In conclusion, whereas in cropland soils and surrounding habitats such as  
248 lakes and ponds directly receiving non treated or diffuse wastes and leachates, N:P ratio has decreased in  
249 last decades, in the majority of other continental and coastal areas N:P tends to rise as a result of a greater  
250 spread capacity of N than P.

251

## 252 *2.2 Spatial heterogeneity in anthropogenic N and P imbalances: River basins as case studies*

253 The study of N and P concentrations and N:P ratios in rivers and basins allows the analysis of the effects of  
254 multiple human activities on nutrient budgets (Zhang et al., 2019) across a range of land uses (Sardans et  
255 al., 2012a; Zhang et al., 2019a; Romero et al., 2019) (Fig. 5). Environments where N is transported by  
256 aquatic systems, such as in the lower stretches of rivers and estuaries (Zhang et al., 1999; Capriulo et al.,  
257 2002; Turner et al., 2003; Chai et al., 2006; Yin & Harrison, 2007; Harrison et al., 2008; Li et al., 2010) and  
258 along coasts (Yin et al., 2004; Turner et al., 2006; Wei & Huang, 2010; Lipizer et al., 2011; Chen et al.,  
259 2014), or by deposition, such as in remote lakes (Arbuckle & Downing, 2001; Hessen et al., 2009; Liess et  
260 al., 2009) and forest and grassland ecosystems (Fenn et al., 1998; Franzing et al., 2010; Prietzel & Stetter,  
261 2010; Veresoglou et al., 2014; Du et al., 2016; Wang et al., 2017; Schmitz et al., 2019), tend to be enriched  
262 more rapidly by N than P, thereby increasing the N:P ratios (Fig. 5). This trend has been exacerbated by  
263 the progressive replacement of P-rich with N-rich detergents (Sardans et al., 2012b and references  
264 therein). The exceptions occur in areas with growing diffuse livestock densities (Frost et al., 2009; Zhang  
265 et al., 2015) and in countries with emerging economies and demography, such as Turkey, Mexico, and

266 India where the loads of non-treated wastes with great charges of human and animal dejections to rivers  
267 are increasing (Bizsel and Uslu, 2000; Ruiz-Fernández et al., 2007; Sardans et al. 2012b; Ramesh et al.,  
268 2015) (Fig. 5). These trends are recent, but the ongoing construction and use of wastewater treatment  
269 plants (Tong et al., 2019) has led to emergent re-oligotrophication of water and improved management of  
270 fertilization (Kara et al., 2012). Wastewater treatment plants generally retain approximately 60% of N and  
271 80% of P, so treated water released to the aquatic system has low N and P concentrations and high N:P  
272 ratios (Ibañez & Peñuelas, 2019) (Fig. 5). The number of wastewater treatment plants will likely increase,  
273 so assessing the potential impacts of re-oligotrophication will be important. For example, anoxic  
274 conditions may change to more aerobic conditions, and increases in water N:P ratios associated with low  
275 N and P concentrations may increase the abundance of aerobic species with low growth rates (Elser &  
276 Sterner, 2002; Sardans, Rivas-Ubach & Peñuelas, 2012b).

277 N and P concentrations and ratios at regional scales generally tend to differ between agricultural  
278 areas with no or low levels of livestock and areas with higher densities of livestock. The ratios of N:P  
279 inputs tend to be higher in areas with low livestock densities that are treated with inorganic fertilizer,  
280 (Sardans et al., 2012b; Dupas et al., 2015; Sun et al., 2017; Romero et al., 2019). Instead, leachates tend to  
281 be rich in P, with low N:P ratios (Szögi et al., 2015) in areas with high densities of livestock, particularly  
282 monogastric (nonruminant) livestock, such as poultry and pigs, so large amounts of P are released through  
283 estuaries to oceans, as observed in some Indian rivers (Ramesh et al., 2015), associated with deposition  
284 with low N:P ratios (Wang et al., 2018c) (Fig. 5).

285

286

### 287 **3. Impacts of shifts in the N:P ratios of human inputs on organisms, communities, and ecosystems**

#### 288 *3.1 Cascading effects*

289 The cascades of effects due to anthropogenic shifts in N:P ratios are similar in aquatic systems (lakes,  
290 estuaries, streams) and terrestrial ecosystems, where water and planktonic N:P ratios tend to increase in  
291 response to atmospheric deposition, leading to lower growth rates, complexity of community structure,  
292 and trophic diversity (Fig. 6, Table S1). Exceptions to these trends, however, have been recorded for  
293 aquatic systems, such as a decrease in N:P ratios in Japan due to the increasing deposition of P from dust  
294 dispersed from countries in southeastern Asia (Miyazako et al., 2015), and for European and North  
295 American lakes in areas with recent reductions in N deposition (Gerson et al., 2016; Isles et al., 2018).  
296 Although most studies of urban and crop wastes and leachate loads to rivers and estuaries (83.3%) have  
297 found increasing N:P ratios associated with increasing N:P ratios from human inputs, other studies

298 (13.7%) tended to find decreasing ratios in areas with high livestock densities (Arbuckle & Downing, 2001;  
299 Jonhson et al., 2006) (Fig. 6, Table S1).

300 Increasing evidence has established links between phylogeny and the elemental compositions of  
301 microbes, plants, and animals, including N and P concentrations and N:P ratios (Sardans et al., 2015;  
302 González et al., 2017, 2018; Bartrons et al., 2018; Godwin & Cotner, 2018; Peñuelas et al., 2019a).  
303 Anthropogenic increases in environmental and organismic N:P ratios in aquatic and terrestrial systems are  
304 generally associated with cascades of effects that benefit organisms with lower growth rates and lead to  
305 shifts in species community composition and function (Carrillo et al., 2001; Arnold et al., 2004; Wassen et  
306 al., 2005; Shurin et al., 2006; Ballantyne et al., 2008; Schindler et al., 2008; Wardle et al., 2008; Apple et  
307 al., 2009; Hall, 2009; Bishop et al., 2010; Cernusak et al., 2010; Chen et al., 2010; Elser et al., 2010a;  
308 Laliberté et al., 2010; Sasaki et al., 2010). Increases in plant N:P ratios can upregulate secondary  
309 metabolism and downregulate primary metabolism linked to growth and energy transfer, whereas  
310 decreases in N:P ratios have the opposite effect, especially when both N and P are not limiting (Peñuelas  
311 & Sardans, 2009; Rivas-Ubach et al., 2012; Gargallo-Garriga et al., 2014).

312 Changes in N and/or P availability and associated shifts in N:P ratios drive changes in species  
313 competition and dominance in communities of terrestrial plants (Sardans et al., 2004; Zhang et al.,  
314 2019b), animals (Jochum et al., 2017), microbes (Fanin et al., 2013; Zechmeister-Bolstenstren et al., 2015;  
315 Delgado-Baquerizo et al., 2017; Shao et al., 2017; Ren et al., 2017), and plankton (Elser et al., 2009a, b; He  
316 et al., 2013; Plum et al., 2015; Grosse et al., 2017; Moorthi et al., 2017). Changes in media (water or soil)  
317 N:P ratios affect the structure of terrestrial (Fanin et al., 2013; Scharler et al., 2015; Zechmeister-  
318 Bolstenstren et al., 2015) and aquatic (Sitters et al., 2015) food webs, but associated impacts on  
319 community diversity are unclear. For example, some studies have reported increases in N:P ratios due to  
320 N deposition or land-use change associated with reduced diversity of microbes (Zhang et al., 2018b),  
321 plants (Güsewell et al., 2005; DeMalach, 2018), and animals (Wei et al., 2012; Vogels et al., 2017), but  
322 other studies have found increases in microbial (Ren et al., 2016, 2017; Aanderud et al., 2018) and plant  
323 (Wassen et al., 2005; Laliberté et al., 2010; Pekin et al., 2012; Yang et al., 2018) diversity. The diversity of  
324 plant species has been associated with an optimum plant N:P mass ratio near 20 (Sasaki et al., 2010), but  
325 the tendency for biodiversity to depend on concentrations of N and P in soil hinders the establishment of  
326 a generalized hypothesis for the relationship between N:P ratios and diversity for all components of  
327 terrestrial communities (DeMalach, 2018).

328 Uncertainty of the effects of N:P ratios on community diversity derives from studies in which  
329 higher plant-community diversity has been correlated with higher N:P ratios and lower variation of plant  
330 N:P ratios. Higher plant-community diversity may be driven by optimizing nutrient uptake (Abbas et al.,



331 2013), but other studies have found higher variation in N:P ratios among sympatric species (Alexander et  
332 al., 2015; Urbina et al., 2015; 2017), indicating that these species tend to maintain different elemental  
333 stoichiometries to avoid direct competition. For example, greater partitioning of resources among niches  
334 (in this case, N and P) has been demonstrated in sympatric species of diatoms under field conditions,  
335 where the expression of genes in the N and P metabolic pathways varied (Alexander et al., 2015).

336 Links between N:P ratios and species diversity are clearer in marine and freshwater ecosystems,  
337 particularly lakes. For example, the typically negative relationships between N:P ratios and the diversities  
338 of zoo- and phytoplankton (He et al., 2013) are associated with the shortened pathways and lower  
339 transfer rates of matter and energy along trophic webs under P limitation (Elser, 2010a). Nutrient  
340 limitation and high N:P ratios are consistently associated with shifts from fast- to slow-growing species in  
341 all types of media (Peñuelas et al., 2013; Busch et al., 2018), and soil microbial and decomposer faunal  
342 compositions are consistently associated with soil and litter N:P ratios (Leflaive et al., 2008; Barantal et al.,  
343 2014; Lee et al., 2015; Su et al., 2015; Eo & Park, 2016; Delgado-Baquerizo et al., 2017; Lee et al., 2017;  
344 Ren et al., 2017).

345 Impacts of changes from N to P limitation on the relationships between bacteria and hosts (and  
346 vice versa) are strong due to the short life cycles of bacteria. Host selection in the cyanobacterium  
347 *Synechococcus* is more discriminant under N than P limitation, leading to changes in the co-evolution of  
348 microbial communities associated with hosts that depend on intermediate N:P ratios (Larsen et al., 2019).  
349 Similarly, changes in key ecosystem processes indirectly involved in community species composition, such  
350 as the transfer of energy and elements through trophic levels and nutrient cycling, have been correlated  
351 with changes in organismic N:P ratios (Vanni et al., 2002; Agren, 2004; Arnold et al., 2004; Zhang et al.,  
352 2004; Güsewell & Verhoeven, 2006; Güsewell & Gessner, 2009; Peñuelas et al., 2013 and references  
353 therein). The directions of effects on community diversity and ecosystem structure in terrestrial and  
354 marine ecosystems due to shifts in N:P ratios, however, are inconsistent (DeMalach, 2018), so an  
355 understanding of the response mechanisms and generalities in ecosystems, particularly terrestrial  
356 ecosystems, is lacking.

357 Recent studies of the C:N:P ratios in mammalian dung have found strong impacts on plant  
358 diversity (Váldez-Correcher et al., 2019), indicating that top-down effects of changes to ecosystem  
359 community structure may be driven by N:P ratios and nutrient cycling. More research, however, is needed  
360 to support this hypothesis. Several drivers of global change, such as N deposition and increasing aridity,  
361 together with imbalances in anthropogenic N:P ratios, are generally shifting ecosystem N:P ratios that in  
362 turn affect species community composition and diversity. Soil, water, and organismic N:P ratios have thus  
363 been associated with basic traits of ecosystem structure and function, such as growth, photosynthetic

364 activity, investment in reproduction, structure of trophic webs, life-history strategy, and species diversity  
365 (Sardans et al., 2012b; Peñuelas et al., 2013; Carnicer et al., 2015, Peñuelas, et al., 2017 and references  
366 therein).

367

### 368 *3.2 N:P ratios and the capacity of terrestrial ecosystems to capture C*

369 N:P ratios in ecosystems with the largest capacity to accumulate large amounts of C, such as forests and  
370 major estuaries, have tended to increase, including tropical forests that are usually P limited (Sardans et  
371 al., 2012a; Peñuelas et al., 2013; Du et al., 2016). These increases in N:P ratios may limit the capacity of  
372 terrestrial ecosystems, mainly tropical forests, to store C (Goll et al., 2017; Peñuelas et al., 2017a; Wang et  
373 al., 2019). The availability of key nutrients, such as K and P, are predicted to decrease the sensitivity of  
374 ecosystems to increasing CO<sub>2</sub> emissions and warming (Fernandez-Martinez et al., 2014; Peñuelas et al.,  
375 2017a; Wang et al., 2019). For example, climate-system models have predicted that limited P availability  
376 and corresponding imbalances in N:P ratios will decrease the capacity of terrestrial ecosystems to remove  
377 CO<sub>2</sub> (Peñuelas et al., 2013, 2017; Goll et al., 2017; Sun et al., 2017; Wang et al., 2019). Similarly, other  
378 studies report that recent climatic warming has increasingly decreased the capacity of the biosphere to  
379 store C (Fernandez-Martínez et al., 2019), and only forests with nutrient-rich soil had higher net primary  
380 production (NPP) in response to increases in gross primary productivity (Fernández-Martinez et al., 2014).  
381 Recent improvements to models, such as including N and P cycles in C-cycling models, have predicted that  
382 the capacity of the biosphere to store C will decrease when N:P ratios become unbalanced (Wang et al.,  
383 2018). Recent studies of the feedbacks and interactive effects of shifts in N:P ratios on climate change  
384 mediated by effects on the capacity of ecosystems to store and release CO<sub>2</sub>, where N and P cycles have  
385 been incorporated into general C and climatic models (Peñuelas et al., 2013; Goll et al., 2017; Wang et al.,  
386 2017a), challenge current understanding of the impacts of the interactive effects of global change. Closing  
387 this knowledge gap is a priority for future studies. These models have questioned whether changes in P  
388 and N availability and N:P ratios may alter the capacity of the biosphere to fix C from anthropogenic CO<sub>2</sub>  
389 emissions. Simulated changes in NPP and increases in vegetation and soil-C storage in response to rising  
390 CO<sub>2</sub> levels and longer growing seasons in the Northern Hemisphere have likely been overestimated  
391 (Hungate et al., 2003; Peñuelas et al., 2017a). Recent progress in implementing mechanistic N and P  
392 schemes in models of the terrestrial C cycle, however, underscores the importance of nutrient feedbacks,  
393 with reductions in productivity of up to 50% in the 21st Century (Goll et al., 2012). No consensus, though,  
394 has yet been reached on future spatial patterns, the degree of nutrient limitation (Zaehle & Dalmonech,  
395 2012), and associated interactions with the coupled system of climate and the C cycle, despite these  
396 advances.

397           Increases in NPP with more N and P must be balanced with increased decomposition with greater  
398 N and P supply. Increasing N:P ratios may actually lead to lower decomposition rates and hence greater C  
399 storage. If, however, there is less NPP feeding C pools, the net effect could be less storage. The  
400 stoichiometric constraints on microbial decomposition would play a key role in these changes in C storage  
401 and turnover. The relationship between litter N:P ratio and litter decomposition is not simple. Some  
402 studies have observed that litter decomposition is mostly related to lignin and/or secondary compounds  
403 concentrations, and only weakly dependent on litter N:P ratio both in tropical forests (Hattenschwiler and  
404 Jorgensen (2010) and high latitude ecosystems (Aerts et al., 2012). Other studies have observed that litter  
405 decomposition rates were positively (Zang et al. 2018) or negatively (Wang et al. 2016a) related to N:P  
406 ratios. These relationships between litter decomposition rates and N:P ratio strongly depend of the level  
407 of concentrations of N and P (Güsewell & Gessner, 2009). Litter with N:P > 22 has P-limited decomposition  
408 (Güsewell & Freeman (2005). In the frame of growth rate hypothesis, lower N:P ratios should increase  
409 microbial growth rate and thus favor fast litter decomposition but only when both N and P are in high  
410 concentration; instead, a positive relationship or no relationship between N:P ratio and growth rate of  
411 microorganisms occur under low N and P concentrations.

412           Declining health (high mortality and defoliation) has been recorded in forests with long-term and  
413 persistently high atmospheric loads of N (Carnicer et al., 2015), imbalances in soil nutrients, and  
414 increasing P limitation (Veresoglou et al., 2014; Schmitz et al., 2019). The capacity of temperate forests to  
415 store P increases with age (Sardans & Peñuelas, 2015), and proportional allocation among organs is linked  
416 to growth-trait strategies. For example, more N is allocated to leaves than roots in slower growing species  
417 (Sardans & Peñuelas, 2013b). The N:P ratios of plant organs may be involved in the phenomenon of  
418 masting, which intensifies at extreme low and high values of N:P (Fernandez-Martinez et al 2019).  
419 Anthropogenic nutrient imbalances and the declining health of temperate forests in the Northern  
420 Hemisphere (Veresoglou et al., 2014; Schmitz et al., 2019) may thus affect the capacity of forest  
421 ecosystem services, such as C storage. Such impacts on ecosystem function and service delivery remain to  
422 be quantified.

423

#### 424 **4. Impacts of shifts in N, P, and N:P ratios on food security and human health**

##### 425 *4.1. Food security*

426 Agriculture may face a potential long-term scarcity of P (McDonald et al., 2011; Obersteiner et al., 2013),  
427 likely due to the exhaustion of mineable P reserves (Cordell & White, 2011) and lack of financial access to  
428 P fertilizers in poorer countries due to high and fluctuating market prices (Obersteiner et al., 2013). The  
429 scarcity of P has long been debated, but ongoing increases in global reserves of mineable P have obscured

430 the potential risk of physical long-term P scarcity (Cordell & White, 2011), although the limited access of  
431 many countries still poses a risk to global food security (Fig. 7). The emergence of the global biospheric  
432 imbalanced N:P ratio has increased the complexity of the implications of P scarcity (Peñuelas et al., 2013;  
433 Lu & Tian, 2017), including risks to food production in agroecosystems (van der Velde et al., 2014; Lu &  
434 Tian, 2017). Most P reserves are in only three countries, with Morocco estimated to contain 85% of the  
435 global share, followed by China with 6% and the USA with 3% (MacDonald et al., 2011), exacerbating the  
436 global problem of supplying P fertilizers.

437         Recent reports about environmental problems related to P availability and imbalances in N:P  
438 ratios, and the P trilemma among rich, poor, and P supplier countries (Obersteiner et al., 2013) have  
439 attempted to address issues and solutions for P availability (Fig. 7). Some issues for avoiding the impacts of  
440 potential P scarcity on global food security for an increasing human population are important  
441 (Obersteiner et al., 2013; Rosemarin & Ekane, 2016), including increased demand and prices for P  
442 fertilizers that will likely render them inaccessible to poor and food-insecure countries (Obersteiner et al.,  
443 2013; Kahsay, 2019). Projections of demands for P fertilizers estimate a doubling of current levels by 2050  
444 (Mogollon et al., 2018b), consistent with short-term predictions (Matsubae et al., 2011; Jedekhauser et  
445 al., 2018; Withers et al., 2018a, b).

446         The predicted growth in P demand may be exacerbated by additional demands, such as for  
447 fertilizing grassland for livestock production, estimated at about 4–12 Tg P y<sup>-1</sup> globally (Mogollon et al.,  
448 2018b), and for fish farms, especially in eastern Asia (Vass et al., 2015). P reserves under these scenarios  
449 are expected to become depleted within the next 40–400 years, depending on the method of projection  
450 (Elser & Bennett, 2011; Cordell et al., 2012; Peñuelas et al., 2013; Cordell & White, 2011,2015). The  
451 prospect of exhausting P reserves is a particular concern for P-poor cropland in sub-Saharan Africa, South  
452 America, India, Australia, and Russia, especially where farmer income and the capacity of crop production  
453 are low (McDonald et al., 2011; Cordell et al., 2013; Rao et al., 2015; Sanyal et al., 2015), such as in sub-  
454 Saharan Africa, where low P content and high N:P ratios in some areas are alarming (Sileshi et al., 2017).

455         Geopolitical tensions associated with P scarcity (Obersteiner et al., 2013) are likely to increase  
456 between economically rich and poor P consumers, food-insecure P consumers, and P-producing countries  
457 (Obersteiner et al., 2013; Matsubae et al., 2011). These tensions indicate the increasing imbalances in N:P  
458 ratios due to socioeconomic and asymmetric (access to N vs P) differences in anthropogenic inputs of  
459 biologically active N and P to the biosphere (Peñuelas et al., 2013). Imbalances in total emitted  
460 anthropogenic N:P ratios to the biosphere increased exponentially during 1961–2013, with multiple  
461 detrimental effects. For example, P limitation has increased in several crops, predominantly in Africa and  
462 Asia, which may affect future responses to N fertilization (Lu & Tian, 2017). The accumulated addition of P

463 for 2000–2050 has been estimated at 1232 Tg P across the four Millennium Ecosystem Scenarios  
464 (Peñuelas et al., 2013), so the P deficit for cereal crops may increase exponentially, especially in large  
465 areas of Africa and Russia (Peñuelas et al., 2013; van der Velde et al., 2014).

466 In addition to the problems of P scarcity, P cycling has become a global concern, due to the very  
467 low solubility of P and its propensity to be adsorbed on some soil components and to precipitate to form  
468 diverse salt species, depending on the pH and mineral components of the soil (Srinivasarao et al., 2007;  
469 Dumas et al., 2011; Arai & Livi, 2013). Long-term continuous inputs of P fertilizer in cropland have led to  
470 estimates that 50% of total globally applied P fertilizer during 2002–2009 has accumulated in the soil (Xi  
471 et al., 2016; Lun et al., 2018). No chemical forms of P are directly available for uptake by crop plants, so  
472 efforts to improve P-use efficiency constitute a key global challenge (Sattari et al., 2012; Li et al., 2015,  
473 2016; Liu et al., 2016; Bai et al., 2016; Withers et al., 2018a, b).

474 The three-fold global increase in livestock production for human consumption over the last five  
475 decades has been a key driver of scarcity, environmental distribution, and decrease in the efficiency of P  
476 use (Liu et al., 2017). Globally, 70% of livestock comprises monogastric animals, such as poultry and pigs,  
477 which cannot absorb P from phytates and produce manure with very high P concentrations and low N:P  
478 ratios that lead to very low P-use efficiency (Prasad et al., 2015; Oster et al., 2018; Wang et al., 2018e).  
479 Land used for the intensive production of monogastric animals and that is fertilized with their manure  
480 exacerbates environmental imbalances in N:P ratios (Peñuelas et al., 2009; McDonald et al., 2011; Sileshi  
481 et al., 2017). A change in human diet to one with a larger proportion of plant-based food may be an  
482 effective tool to improve P-use efficiency (Reijnders, 2014; Withers et al., 2015). Studies have indicated  
483 that food security may be assured by improving P recycling by the application of a range of technologies  
484 and improved and efficient management of N and P fertilization to avoid imbalances in N:P ratios and  
485 subsequent associated cascades of environmental and economic problems (Cordell et al., 2012;  
486 Rosemarin & Ekane, 2016; Weikard, 2016; Rahman et al., 2019).

487

#### 488 *4.2 Human health*

489 Changes in N, P, and N:P ratios cascade up the trophic chain, potentially to humans from food production,  
490 when the effects of over-fertilization and imbalances in N:P ratios in crops may become apparent  
491 (Peñuelas et al., 2017b; Peñuelas et al., 2019b). N fertilization has historically been excessive in rich  
492 countries and has led to the over-production of food, and the low use of fertilizers has staved off  
493 malnutrition in poor countries (Smil, 2002). Men born in rich countries in the 1980s were an average of  
494 1.5 cm taller than men born in the 1960s, whereas the height of males born in the same decades in poor  
495 countries did not differ (Peñuelas et al., 2017b). Differences in per capita N, P, and N:P intake explained

496 these differences in the height of men born in rich countries better than did socioeconomic and sanitary  
497 variables, such as gross domestic product, the human development index and birth weight according with  
498 FAO, OCDE and WHO integrated data analyses (Peñuelas et al., 2017b). Some malign neoplasms,  
499 particularly of the colon and lung, contain higher concentrations of P and lower N:P ratios than do healthy  
500 organs and surrounding tissue (Elser et al., 2007a, b). High N and P intakes from an increased  
501 consumption of animal-based foods in some developed countries would therefore likely lead to higher  
502 heights, albeit with a higher risk of mortality from cancer.

503 The intensification of crop management and use of fertilizers (especially N) have changed the  
504 composition of food intake per capita. Peñuelas et al (2019b) reported that the global intensification of N  
505 fertilization may increase the allergenic proteins concentrations in wheat increasing the mean annual per  
506 capita intake of these proteins at global scale thus rising the risk of higher prevalence of some illness such  
507 as coeliac pathology. Using wheat as an example, global N fertilization increased from 9.84 to 93.8 kg N  
508  $\text{ha}^{-1} \text{y}^{-1}$  during 1961–2010 (Curtis, 2019), similar to the overall rate of increase (10.5%  $\text{y}^{-1}$ ) across all types  
509 of farmland (from 11.3 to 107.6 Tg N  $\text{y}^{-1}$ ) (Lu & Tian, 2017). The increases in N availability have led to  
510 increased concentrations of gluten (Klikocka et al., 2016; Litke et al., 2018; Zheng et al., 2018) and the  
511 gliadins in gluten (Daniel & Triboi, 2000; Kinderd et al., 2008; Guardia et al., 2018). These gliadins are  
512 responsible for triggering (Petersen et al., 2015; Morrell & Melby, 2017; Dubois et al., 2018) and  
513 maintaining (Hischenhuber et al., 2006; Akobeng & Thomas, 2008; Gil-Humanes et al., 2014) celiac  
514 disease. Indeed, the higher availability of N has been associated with higher expression of gliadin genes  
515 (Shewry et al., 2001).

516 Evidence suggests that P is accumulating in some cropland soils (Yuan et al., 2018) (Fig. 7), which  
517 increases uptake by crop plants that may increase P concentrations in food and therefore dietary intake.  
518 Some studies have reported high levels of P uptake by crops (Selles et al., 1999; Zhang et al., 2007;  
519 Fernandez et al., 2017; Gomez et al., 2019) and non-crop plants (Xu & Timmer, 1998; Ostertag, 2010; Da  
520 Ros et al., 2018) under high soil P concentrations. However, the potential relationship between the global  
521 accumulations of P in crop soil and P concentrations in the food produced and subsequent consequences  
522 on human health are currently unknown. Future research on effects of dietary increases in P intake is  
523 warranted since health problems, such as bone health, risk of cancer, and heart failure, have been linked  
524 to the increased use of P additives in foods (Dhingra et al., 2010; Wulaningsih et al., 2013; Takeda et al.,  
525 2014), albeit with inconsistent effects when P intake is excessive (Cooke, 2017). Sufficient evidences of a  
526 shift in food composition at elemental and molecular level produced by changes in N and P crop  
527 management are available. Human health can be affected, which opens a new potential perspective in  
528 medical studies

**530 6. Strategies to limit and mitigate the negative impacts of P scarcity and imbalances in N:P ratios**

531 Several policy and management mitigative strategies have been proposed to meet the challenges that the  
532 negative effects of P availability pose to food security, environmental health, and geopolitical and  
533 economic stability among countries (Dumas et al., 2011; Obersteiner et al., 2013; Cordell & White, 2015;  
534 Metson et al., 2015; Hukari et al., 2016; Withers et al., 2017). Key global approaches to ensuring  
535 sustainable P management and the avoidance of future P scarcity and limitation include stabilizing P  
536 prices, balancing the requirements of P supply and demand, limiting eutrophication, optimizing P cycling,  
537 remobilizing and recovering P stores in cropland soil, designing and implementing novel biotechnologies  
538 for crop and livestock production, and moving toward plant-based diets (MacDonald et al., 2011; Neset &  
539 Cordell, 2011; Schröder et al., 2011; Suh & Yee, 2011; Cordell et al., 2013; Cordell & White, 2015; Withers  
540 et al., 2015; Bai et al., 2016; Lukowiak et al., 2016; Metson et al., 2016; Wu et al., 2016; Roy, 2017;  
541 Jedelhauser & Binder, 2018; Jedelhauser et al., 2018; Withers et al., 2018a, b; Kasprzyk & Gajewska,  
542 2019).

543 The consensus indicates that increasing the use and cycling efficiencies of P will be the most  
544 effective approaches to prevent P scarcity for food production and reduce environmental problems  
545 involving P (Suh & Yee, 2011; Hanserud et al., 2016; Weikard, 2016; Melia et al., 2017; Withers et al.,  
546 2018a, b; Rahman et al., 2019). The direct recovery of P from all types of waste may yield large  
547 proportions of previously used P, reducing the need to exploit and release novel sources of bioactive P  
548 into the P cycle (Withers et al., 2018b), where secondary fertilizers are produced using recovered P  
549 (Hanserud et al., 2016; Talboys et al., 2016; Weikard, 2016; Jedelhauser & Binder, 2018). The efficiency of  
550 P recovery in some countries such as Finland and Denmark has reached 67.5 and 53.7%, respectively, but  
551 only 0.5% in the USA, a high P consumer (Rahman et al., 2019). A recovery of 37% of recyclable P in the  
552 USA would meet the P demand for corn crops (Metson et al., 2016).

553 Methods to increase plant accessibility to P sources have been proposed (Cordell et al., 2011;  
554 Adhya et al., 2015; Li et al., 2015; Rowe et al., 2016; Roy 2017; Withers et al 2015; Withers et al., 2018a)  
555 as approaches to increase P-use efficiency. At least 50% of the P fertilizer applied to cropland accumulates  
556 in the soil (van Dijk et al., 2016; Fun et al., 2018; Lun et al., 2018). For example, cropland soil in Brazil was  
557 estimated to store 30 Tg P in 2016 (Withers et al., 2018) (Fig. 7). Exploitation of these stocks may mitigate  
558 future scarcity of P fertilizer or inflated prices, where possible approaches include breeding novel  
559 microbial genotypes and crop varieties that could re-mobilize and re-use stored P (Adhya et al., 2015;  
560 Rowe et al., 2016; Vandamme et al. 2016).

561 The use of novel management techniques and biotechnologies provide opportunities to improve  
562 P-use efficiency (Adhya et al., 2015; Vandemme et al., 2016; Rowe et al., 2016; Zheng et al., 2019). In  
563 addition to the development and use of novel strains of microbes with a high capacity for remobilizing  
564 stored P from crop soil (Adhya et al., 2015; Zheng et al., 2019), other technological improvements, such as  
565 novel crop genotypes (Vandemme et al., 2016; Rowe et al., 2016), may be used to improve P-use  
566 efficiency (Fig. 7). Improved P-use efficiencies in soil and plants have also been achieved using  
567 combinations of novel and technologically improved traditional management techniques (Wang et al.,  
568 2016c; Zheng et al., 2019), such as the application of biochar integrated with approaches of organic  
569 agricultural management (Chintala et al., 2014) and crop rotation (Lukowiak et al., 2016).

570 The recovery of P from human urine and feces may meet 22% of the total P demand (Mihelic et  
571 al., 2011), but its success may be hindered by technological and politicoeconomic constraints.  
572 Precipitation with iron and aluminum salts is the simplest method to recover P from waste and water, but  
573 the resulting product has limited bio-availability and is a pollutant (Melia et al., 2017). The precipitation of  
574 P from wastewater as struvite is more promising (Melia et al., 2017), because the bio-availability of P in  
575 struvite as a fertilizer is high (Talboys et al., 2016), and transport costs between treatment plants and  
576 farmers is low (Jedelhause & Binder, 2018). Recovery capacity, however, is limited (approximately 25%)  
577 unless expensive chemical methods of extraction are applied (Melia et al., 2017). P recovery may be  
578 highest from the combustion of solid waste that produces energy and P-rich ash for use as fertilizer  
579 (Thitanuwat et al., 2016). Research into the efficient recovery of P from wastes is ongoing and yielding  
580 substantial advances (Roy, 2017; Kasprzyk & Gajewska, 2019).

581 Stimuli for recycling P tend to be controlled by legislative regulations and instruments at the  
582 national or regional administrative level, sometimes supported by subsidies (Withers et al., 2015; Hukari  
583 et al., 2016). Legislation is usually not harmonized or coordinated among national agencies, so the  
584 likelihood of the large-scale production of secondary P fertilizer from processes of P recovery is low and  
585 requires multinational adoption of cutting-edge technologies (Withers et al., 2015; Hukari et al., 2016;  
586 Oster et al., 2018). Increases in the costs of P extraction and transport, however, may increase the  
587 economic feasibility of secondary P fertilizers (Mew, 2016).

588 Reduction of livestock production has been suggested as the most effective approach to reduce  
589 global P demand and ensure global food security (MacDonald et al., 2011; Schröder et al., 2011; Withers  
590 et al., 2018b). The three-fold increase in livestock production in the last five decades (Liu et al., 2017) has  
591 led to decreased P-use efficiency of inorganically fertilized forage crops and P surpluses from inputs of  
592 animal urine and manure (MacDonald et al., 2011; Nesme et al., 2015). A global reduction in livestock  
593 production for dietary consumption would decrease the demand for P and its associated environmental



594 problems (Neset & Cordell, 2011; Wu et al., 2016; Bai et al., 2016; Wang et al., 2018e). Decreases in  
595 animal production would increase the availability of cropland for producing crops for direct use in human  
596 diets, shortening the food chain and increasing resource-use efficiencies, including P, but also N and water  
597 (Neset & Cordell, 2011; Rowe et al., 2016). Reducing the consumption of monogastric livestock would  
598 increase the sustainable use of P for food production, because such livestock do not efficiently absorb P  
599 from forage (Prasad et al., 2015; Wang et al., 2018e).

600 National and international environmental agencies and policy makers have failed to confront the  
601 recognized global risks of unbalanced N:P ratios to the biosphere and humankind. N and P cycles and  
602 associated ratio imbalances are starting to be incorporated into climatic and C-cycling models, but they  
603 must be addressed by a coordinated international policy and forum of global change.

604

605

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### Captions to Tables and figures

Table 1. Summary of the relationships of global change drivers with N and P concentrations and N:P ratio of soil, plants and freshwater plankton.

Figure 1. a) Mean ( $\pm$ SE) anthropogenic inputs of reactive nitrogen (N) and phosphorus (P) to the biosphere ( $\text{Tg y}^{-1}$ ) since the industrial revolution. b) Mean ( $\pm$ SE) N:P ratios of inputs of reactive N and P to the biosphere since the industrial revolution. Data are for N industrial fertilizers (Galloway et al., 1995, 1998; 2004, 2008; Smil, 2000; Tilman et al., 2001; Mackenzie et al., 2002; Fields, 2004; FAO, 2008, 2015, 2017; Gruber & Galloway, 2008; Mogollón et al., 2018a; Canfield et al., 2010; Grubler, 2002; Bouwman et al., 2013a; Fowler et al., 2013; Gu et al., 2013; Lu & Tian, 2017; Yara Fertilizer, 2018), N<sub>2</sub> fixation in cropland (Delwiche, 1970; Burns & Hardy, 1975; McElroy et al., 1976; Söderlund & Svernnson, 1976; Fields, 2004; Galloway et al., 2004, 2008; Herridge et al., 2008; Canfield et al., 2010; Bouwman et al., 2013a; Fowler et al., 2013; Gu et al., 2013), N emissions from fuel combustion (Eriksson, 1959; Robinson & Robbins, 1970; Söderlund & Svernnson, 1976; MacKenzie et

al., 2002; Fields, 2004; Galloway et al., 2004; Gruber & Galloway 2008; Reay et al., 2008; Canfield et al., 2010; Grübler, 2011; van Vuuren et al., 2011; Gu et al., 2013), and P industrial fertilizers (Smil, 1999; Mackenzie et al., 2002; FAO, 2008, 2015, 2017; Bondre, 2011; McDonald et al., 2011; Bouwman et al., 2013a; Lu & Tian, 2017; Lun et al., 2018; Yara Fertilizer, 2018).

Figure 2. Annual anthropogenic inputs to the global nitrogen and phosphorus cycles and contribution to the N:P ratios (molar basis) of biospheric compartments. Data are from references reported in Figure 1.

Figure 3. N:P ratios (molar basis) of total atmospheric deposition in continents and oceans compared with ratios in plants, plankton, soil, and water. Data derived from Graham & Duce (1979), Smil (2000), Galloway et al. (2004, 2008), Duce et al. (2008), Mahowald et al. (2008), and Schlesinger et al. (2009).

Figure 4. Impacts of short-term (field experiments) and long-term (natural gradients) of drought and aridity on plant and soil N and P concentrations and N:P ratios. Letter size is proportional to concentration.

Figure 5. Current N and P imbalances linked to human activity in river basins.

Figure 6. Numbers of studies from the 'Web of Science' search that report effects of increased availability of environmental nitrogen on increased environmental N:P ratios, increased organismic N:P ratios, decreased growth rates, and changes in community structure and ecosystem functioning. The effects of nitrogen deposition and eutrophication and of increased environmental N:P ratios are indicated by solid lines, the effects of increased organismic N:P ratios are indicated by dashed lines, and the effects of increased growth rates are indicated by dotted lines. See Table S1 for detailed information on these studies.

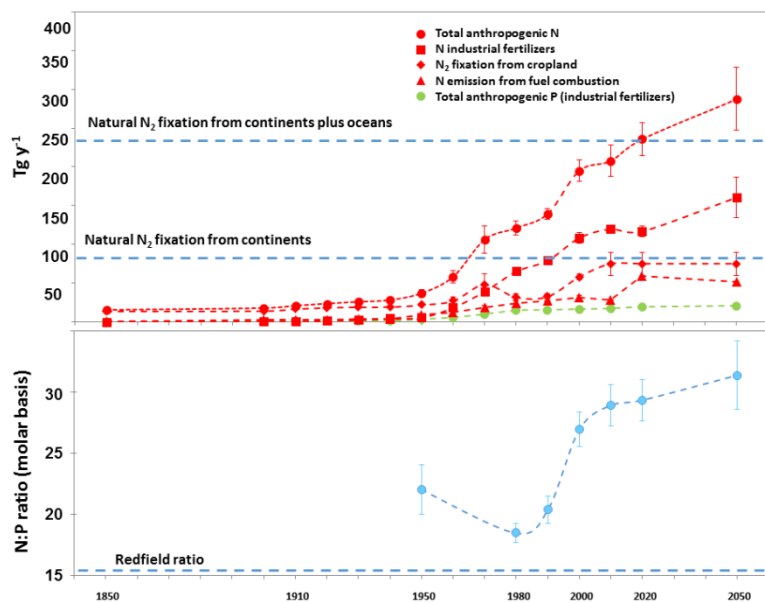
Figure 7. Schematic of the increased imbalance in N and P fertilizers and the negative impacts of N:P-ratio imbalances and P scarcity on food security, human health, and sociopolitical stability.



| Global change drivers                                 | Effects on N and P concentrations and N:P ratios  |  |   |   |  |
|---|---|--|---|---|--|
|   | Soil  |  | Plants  |   | Plankton   |
|   | Natural gradients   | Field experiments  | Natural gradients   | Field experiments   |  |
| Increasing atmospheric CO <sub>2</sub> concentrations | -   | <i>Decrease in soil [N] and [P]</i>  | <i>Decrease in [N] and [P]<br/>Decrease or no change in N:P depending on plant type and plant organ</i> | <i>Decrease in [N] and [P]<br/>Decrease or no change in N:P depending on plant type and plant organ</i> | <i>Decreases, increases or no change in [N], [P] and N:P depending on phytoplankton species</i>  |
| Warming   | <i>Heterogeneous effects on soil N and P concentrations and N:P ratios, but most studies reported lower soil [N] and [P] with higher temperatures</i> | <i>Heterogeneous effects on soil N and P concentrations and N:P ratios, but most studies reported higher soil [N] with higher temperatures</i> | <i>Heterogeneous effects on plant [N] and [P] and in N:P ratios</i>                                     | <i>Heterogeneous effects on plant [N] and [P] and in N:P ratios</i>                                     | <i>Changes in [N] and [P] depending on multi-functions allocation which also depend on each ecosystem trophic level and biotic and abiotic particular conditions</i> |
| Drought/aridity                                       | <i>Decreases in [N] and maintenance or increase in [P]<br/>Decreases in N:P ratio</i>   | <i>Increases in [N] and larger increases in [P]<br/>Decreases in N:P ratio</i>   | <i>Increases in [N] and [P]<br/>Decreases in N:P ratios</i>   | <i>Decreases in [N] and [P]<br/>Increases in N:P ratios</i>   | -  |
| N deposition  | <i>Increases in [N]</i>   | <i>Increases in [N]</i>  | <i>Increases in [N]</i>   | <i>Increases in [N]</i>   | <i>Increases in [N]</i>  |

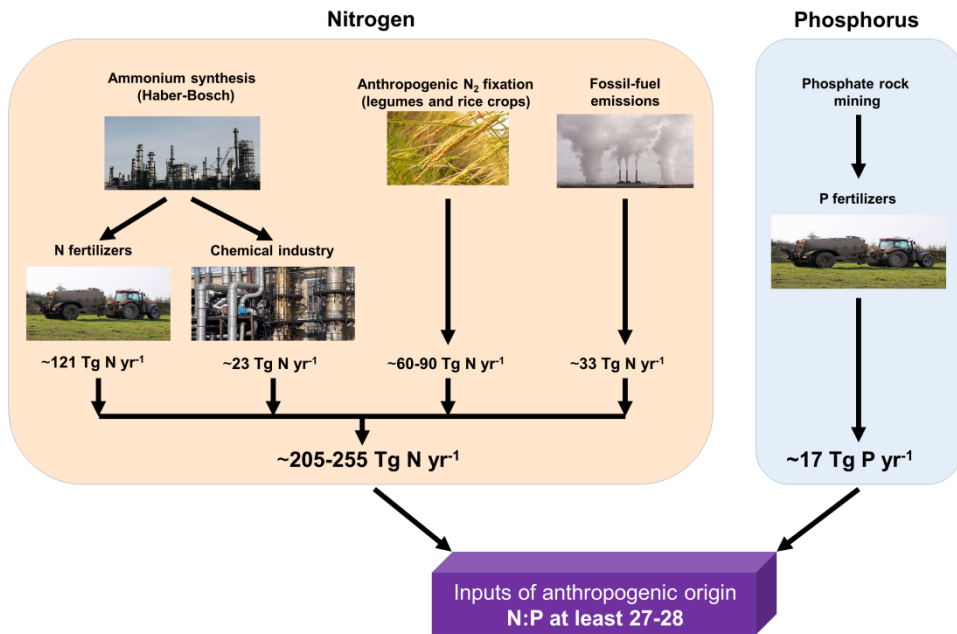
|                        | <i>No changes or decrease in [P]<br/>Increases in N:P</i>  | <i>No changes or decrease in [P]<br/>Increases in N:P</i> | <i>No changes or decrease in [P]<br/>Increases in N:P</i>  | <i>No changes or decrease in [P]<br/>Increases in N:P</i> | <i>No changes or decrease in [P]<br/>Increases in N:P</i> |
|------------------------|--|---|--|---|---|
| P deposition           | -  | -   | -  | -   | No change in [N]<br>Increases in [P]<br>Decreases in N:P  |
| Plant species invasion | <i>Increases in [N]<br/>Increases in [P], but also dependent on natural soil N and P status.<br/>Not enough data to infer changes in N:P ratio</i> | -   | <i>Increases in [N]<br/>Increases in [P], but also dependent on natural soil N and P status.<br/>Not enough data to infer changes in N:P</i> | -   | -   |

- not sufficient data reported or no sense in inferring some effect.

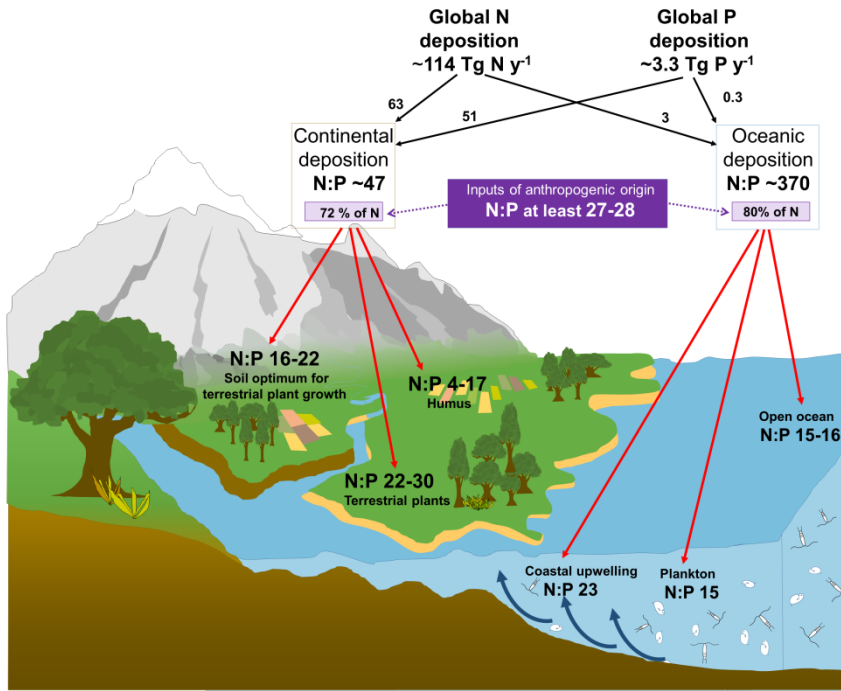


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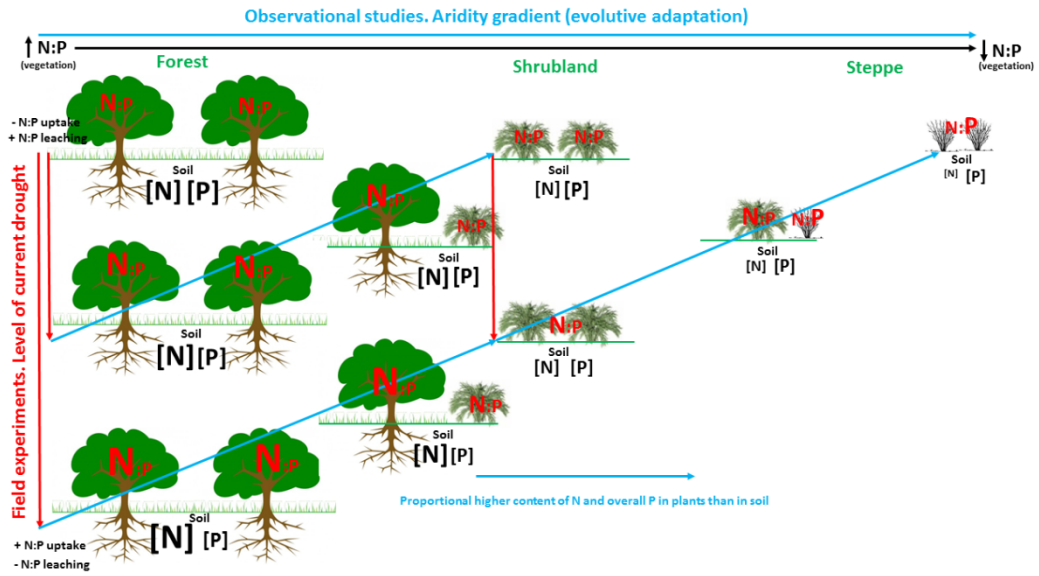
Anthropogenic inputs in N and P cycle



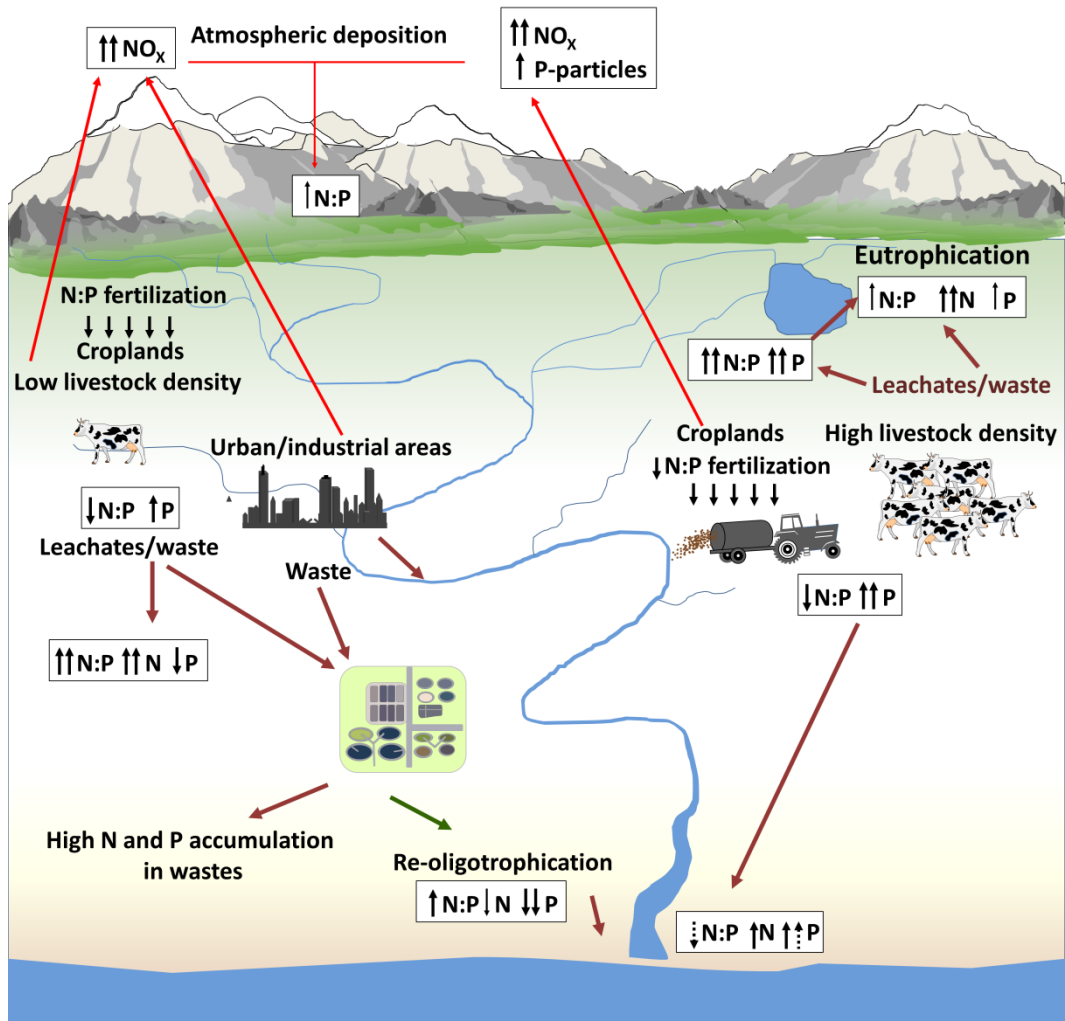
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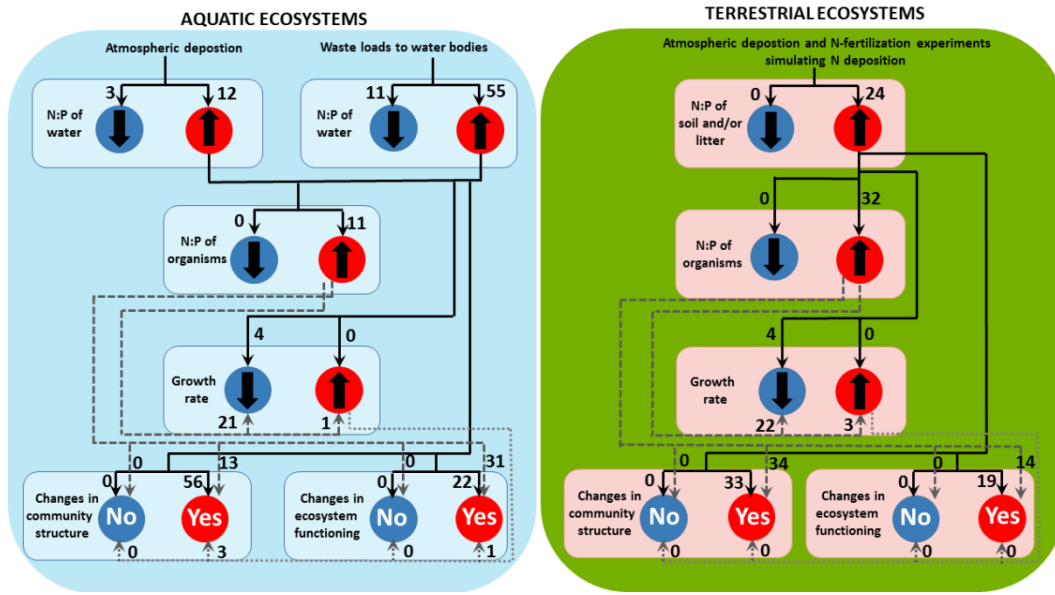
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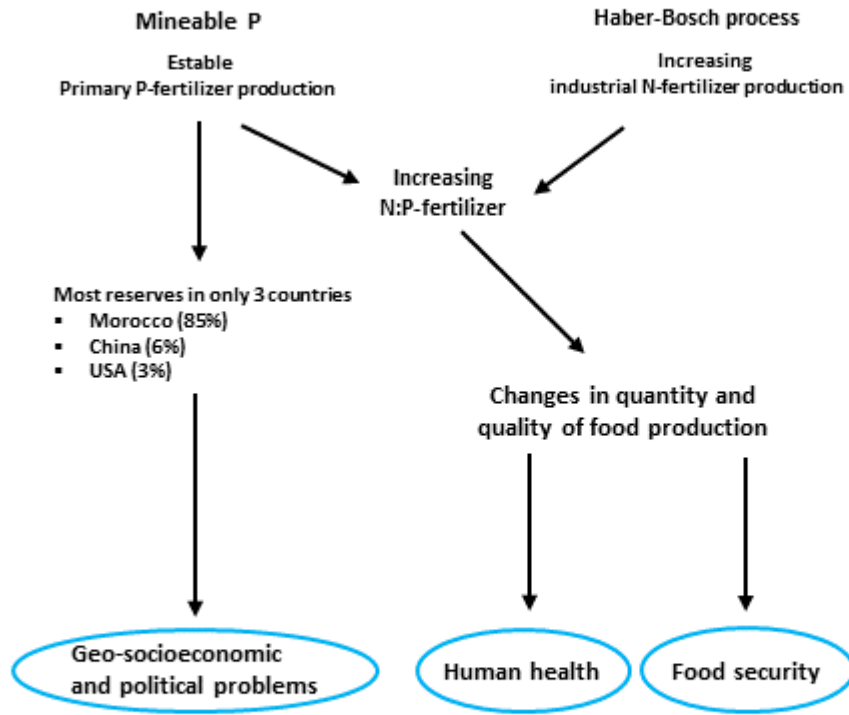


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