PROF. JOSEP PENUELAS (Orcid ID : 0000-0002-7215-0150)

DR. JORDI SARDANS (Orcid ID : 0000-0003-2478-0219)

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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

Josep Penuelas[1,2,3*], Ivan Jannssens[4], Philippe Ciais[5], Michael Obersteiner[6], Jordi Sardans[1,2,3]

[1] CSIC, Global Ecology Unit CREAF-CSIC-UAB, Bellaterra, 08193 Catalonia, Spain.

[2] CREAF, Cerdanyola del Valles, 08193 Catalonia, Spain.

[3] Global Change Research Institute, Czech Academy of Sciences, CZ-60300 Brno, Czech Republic

[4] Research Group Plants and Ecosystems (PLECO), Department of Biology, University of Antwerp, B-2610 Wilrijk, Belgium.

[5] Laboratoire des Sciences du Climat et de l'Environnement, IPSL, 91191 Gif-sur-Yvette, France.

[6] International Institute for Applied Systems Analysis (IIASA), Ecosystems Services and Management, A-2361 Laxenburg, Austria.

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* Corresponding author: Josep Penuelas CSIC, Global Ecology Unit CREAF-CSIC-UAB, Bellaterra, 08193 Catalonia, Spain and CREAF, Cerdanyola del Valles, 08193 Catalonia, Spain.
E-mail address: josep.penuelas@uab.cat

Abstract

The availability of carbon (C) from high levels of atmospheric carbon dioxide (CO_2) and anthropogenic release of nitrogen (N) is increasing, but these increases are not paralleled by increases in levels of phosphorus (P). The current unstoppable changes in the stoichiometries of C and N relative to P have no historical precedent. We describe changes in P and N fluxes over the last five decades that have led to asymmetrical increases in P and N inputs to the biosphere. We identified widespread and rapid changes in N:P ratios in air, soil, water, and organisms and important consequences to the structure, function, and biodiversity of ecosystems. A mass-balance approach found that the combined limited availability of P and N was likely to reduce C storage by natural ecosystems during the remainder of the 21st Century, and projected crop yields of the Millennium Ecosystem Assessment indicated an increase in nutrient deficiency in developing regions if access to P fertilizer is limited. Imbalances of the N:P ratio would likely negatively affect human health, food security, and global economic and geopolitical stability, with feedbacks and synergistic effects on drivers of global environmental change, such as increasing levels of CO_2, climatic warming, and increasing pollution. We summarize potential solutions for avoiding the negative impacts of global imbalances of N:P ratios on the environment, biodiversity, climate change, food security, and human health.

Keywords

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

1. Introduction

The availability of carbon (C) from high levels of atmospheric carbon dioxide (CO2) and anthropogenic inputs of nitrogen (N) on ecosystems are increasing. These increases are, however, not paralleled by those of phosphorus (P), and current inexorable changes in the stoichiometry of C and N relative to P have no historical precedent (Peñuelas et al., 2013). The shifts in organisms' N:P ratio resulting from different environmental conditions are strongly related with shifts in ecosystems structure and function (Sterner & Elser, 2002; Loladze & Elser, 2011 Peñuelas et al., 2013). Imbalances between these two nutrients, N and P in natural, semi-natural, and managed ecosystems (Liu et al., 2010; Sardans & Peñuelas, 2012; Carnicer et al., 2015; Ulm et al., 2016; Peñuelas et al., 2013, Delgado-Baquerizo et al., 2017; Hu et al., 2018). reduce C capture and global food provision and security (Van der Velde et al., 2014; Lu & Tian, 2017; Peñuelas et al., 2017a; Wang et al., 2018a; Kahsay, 2019). These effects may be further exacerbated in cropland in the future by limited access to reserves of mineable P (Cordell et al., 2011; MacDonald et al., 2011; Li et al., 2016; Mew, 2016; Weikard, 2016; Lun et al., 2018).

Changes in the global P cycle, status and resources, together with associated economic impacts, were first debated at least a century ago (Liu et al., 2017). More recent studies have recognized that increases in N:P ratios with rising anthropogenic release have consequences for P and N cycling in soil and water, biodiversity, and ecosystem function (Elser et al., 2010a, b; Peñuelas et al., 2012; Peñuelas et al., 2013). The link between increasing imbalances in biospheric N:P ratios and their impacts on global ecology and socioeconomics is supported by evidence from many studies that have identified clear relationships between drivers of global change and anthropogenic N and P releases and with shifts in ecosystem N:P ratios. These studies have also demonstrated feedbacks and synergies of shifts in the N:P ratios in soil, water, and organisms with increases in atmospheric CO_2 concentrations, climate change, species invasions, ecosystem eutrophication, and changes in soil use (Sardans & Peñuelas, 2012; Sardans et al., 2012a, 2013, 2016a, 2017a; Zhang et al., 2013; Ferretti et al., 2014; Gargallo-Garriga et al., 2014; He & Dijstra, 2014; Deng et al., 2015; Yuan & Chen, 2015; Chen et al., 2016; Delgado-Baquerizo et al., 2016; Jiao et al, 2016; Zhu et al., 2016; Kruk & Podbielska, 2018; Schmitz et al., 2019; Yuan et al., 2018; Peng et al., 2019).

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

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

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

Further evidences accumulated in the last six years after Penuelas et al (2013) robustly confirm the inexorable changes in the stoichiometry of C and N relative to P, which have no historical precedent (Fig. 1). Furthermore, the increasing emissions of NO_x and NH_3 to the atmosphere lead to large imbalances in the ratios of total atmospheric N:P deposition, with higher ratios for total atmospheric N:P than standard averages for soil, water, and organisms (Fig. 2).

Activities involved in food production, such as the application of fertilizer, cultivation of N_2-fixing species of crop plants, livestock husbandry, and the release of N and P to the atmosphere from the combustion of fossil fuels, which are re-deposited on the surface, are key historical and contemporary contributors of bio-active N and P and drivers of these nutrient imbalances (Peñuelas et al., 2012; 2013; Yuan et al., 2018). For example, the N:P ratios of atmospheric total depositions are higher than the average N:P ratios of waters, soils, and organisms (Fig. 3).

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

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

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

The P cycle and N:P ratios are affected by many drivers of global change other than anthropogenic emissions of N and P (Table 1). Higher concentrations of atmospheric CO_2 are correlated with decreases in plant N and P concentrations and increases in the ratios of C:N and C:P (Peñuelas & Matamala 1990;

Peñuelas & Estiarte 1997; Sardans et al., 2012b; Deng et al., 2015), but the effects on plant N:P ratios are less clear. For example, recent meta-analyses have found that rising CO_2 concentrations have led to decreases in N:P ratios in different plant tissues (Deng et al., 2015) and woody plants but not herbaceous plants or mosses (Yue et al., 2017). Yuan and Chen (2015) in a meta-analysis of 315 studies with non-differentiation of plant organs observed an overall decrease in N:P ratios in controlled field conditions under elevated levels of CO_2. However, another review of 215 studies (Sardans et al. 2017b), mostly under controlled field conditions, revealed that increased atmospheric concentrations of CO_2 led to decreased N:P ratios in roots, but not in leaves. Moreover, King et al. (2015) reported increased N:P ratio in one phytoplankton species, decreased N:P ratio in three other species, and no change in N:P ratio in other three species under high levels of CO_2, thus suggesting that the effects of CO_2 enhancement on stoichiometry appear to be species-dependent. It is thus likely that the ongoing increases in atmospheric CO_2 concentrations are reducing N:P ratios in plants, which would be apparently consistent with the GRH for plants under favorable growth conditions (Sterner and Elser, 2002). The hypothesis that atmospheric increases in CO_2 stimulate higher plant uptakes of P than N (Deng et al., 2015) thus remains to be unequivocally demonstrated but begins to have some observational and experimental support (Table 1).

Less information is available regarding the relationships of the rise in atmospheric CO_2 concentration with N and P concentrations and N:P ratio in soil. Huang et al. (2014) observed that a rise in atmospheric CO_2 concentration did not change total soil P concentrations but increased P-available to plants and decreased more recalcitrant soil-P. Increased CO_2 concentrations can Indirectly decrease soil N and P concentrations by several mechanisms including higher plant N and P demands, higher N and P resorption rates and higher exudates production and N and P uptake (Jin et al. 2015; Liu et al. 2018; Van Vuuren et al. 2018). However, the potential impact of CO_2 enhancement of soil N:P ratios also remains inconclusive.

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

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

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

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

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

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

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

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

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

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

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

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

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

3.1 Cascading effects

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

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

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

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

Uncertainty of the effects of N:P ratios on community diversity derives from studies in which higher plant-community diversity has been correlated with higher N:P ratios and lower variation of plant N:P ratios. Higher plant-community diversity may be driven by optimizing nutrient uptake (Abbas et al., 2013), but other studies have found higher variation in N:P ratios among sympatric species (Alexander et al., 2015; Urbina et al., 2015; 2017), indicating that these species tend to maintain different elemental stoichiometries to avoid direct competition. For example, greater partitioning of resources among niches (in this case, N and P) has been demonstrated in sympatric species of diatoms under field conditions, where the expression of genes in the N and P metabolic pathways varied (Alexander et al., 2015).

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

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

Recent studies of the C:N:P ratios in mammalian dung have found strong impacts on plant diversity (Váldes-Correcher et al., 2019), indicating that top-down effects of changes to ecosystem community structure may be driven by N:P ratios and nutrient cycling. More research, however, is needed to support this hypothesis. Several drivers of global change, such as N deposition and increasing aridity, together with imbalances in anthropogenic N:P ratios, are generally shifting ecosystem N:P ratios that in turn affect species community composition and diversity. Soil, water, and organismic N:P ratios have thus been associated with basic traits of ecosystem structure and function, such as growth, photosynthetic activity, investment in reproduction, structure of trophic webs, life-history strategy, and species diversity (Sardans et al., 2012b; Peñuelas et al., 2013; Carnicer et al., 2015, Peñuelas, et al., 2017 and references therein).

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

N:P ratios in ecosystems with the largest capacity to accumulate large amounts of C, such as forests and major estuaries, have tended to increase, including tropical forests that are usually P limited (Sardans et al, 2012a; Peñuelas et al., 2013; Du et al., 2016). These increases in N:P ratios may limit the capacity of terrestrial ecosystems, mainly tropical forests, to store C (Goll et al., 2017; Peñuelas et al., 2017a; Wang et al., 2019). The availability of key nutrients, such as K and P, are predicted to decrease the sensitivity of ecosystems to increasing CO_2 emissions and warming (Fernandez-Martinez et al., 2014; Peñuelas et al., 2017a; Wang et al., 2019). For example, climate-system models have predicted that limited P availability and corresponding imbalances in N:P ratios will decrease the capacity of terrestrial ecosystems to remove CO_2 (Peñuelas et al., 2013, 2017; Goll et al., 2017; Sun et al., 2017; Wang et al., 2019). Similarly, other studies report that recent climatic warming has increasingly decreased the capacity of the biosphere to store C (Fernandez-Martínez et al., 2019), and only forests with nutrient-rich soil had higher net primary production (NPP) in response to increases in gross primary productivity (Fernández-Martinez et al., 2014). Recent improvements to models, such as including N and P cycles in C-cycling models, have predicted that the capacity of the biosphere to store C will decrease when N:P ratios become unbalanced (Wang et al., 2018). Recent studies of the feedbacks and interactive effects of shifts in N:P ratios on climate change mediated by effects on the capacity of ecosystems to store and release CO_2, where N and P cycles have been incorporated into general C and climatic models (Peñuelas et al., 2013; Goll et al., 2017; Wang et al., 2017a), challenge current understanding of the impacts of the interactive effects of global change. Closing this knowledge gap is a priority for future studies. These models have questioned whether changes in P and N availability and N:P ratios may alter the capacity of the biosphere to fix C from anthropogenic CO_2 emissions. Simulated changes in NPP and increases in vegetation and soil-C storage in response to rising CO_2 levels and longer growing seasons in the Northern Hemisphere have likely been overestimated (Hungate et al., 2003; Peñuelas et al., 2017a). Recent progress in implementing mechanistic N and P schemes in models of the terrestrial C cycle, however, underscores the importance of nutrient feedbacks, with reductions in productivity of up to 50% in the 21st Century (Goll et al., 2012). No consensus, though, has yet been reached on future spatial patterns, the degree of nutrient limitation (Zaehle & Dalmonech, 2012), and associated interactions with the coupled system of climate and the C cycle, despite these advances.

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

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

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

4.1. *Food security*

Agriculture may face a potential long-term scarcity of P (McDonald et al., 2011; Obersteiner et al., 2013), likely due to the exhaustion of mineable P reserves (Cordell & White, 2011) and lack of financial access to P fertilizers in poorer countries due to high and fluctuating market prices (Obersteiner et al., 2013). The scarcity of P has long been debated, but ongoing increases in global reserves of mineable P have obscured the potential risk of physical long-term P scarcity (Cordell & White, 2011), although the limited access of many countries still poses a risk to global food security (Fig. 7). The emergence of the global biospheric imbalanced N:P ratio has increased the complexity of the implications of P scarcity (Peñuelas et al., 2013; Lu & Tian, 2017), including risks to food production in agroecosystems (van der Velde et al., 2014; Lu & Tian, 2017). Most P reserves are in only three countries, with Morocco estimated to contain 85% of the global share, followed by China with 6% and the USA with 3% (MacDonald et al., 2011), exacerbating the global problem of supplying P fertilizers.

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

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

Geopolitical tensions associated with P scarcity (Obersteiner et al., 2013) are likely to increase between economically rich and poor P consumers, food-insecure P consumers, and P-producing countries (Obersteiner et al., 2013; Matsubae et al., 2011). These tensions indicate the increasing imbalances in N:P ratios due to socioeconomic and asymmetric (access to N vs P) differences in anthropogenic inputs of biologically active N and P to the biosphere (Peñuelas et al., 2013). Imbalances in total emitted anthropogenic N:P ratios to the biosphere increased exponentially during 1961–2013, with multiple detrimental effects. For example, P limitation has increased in several crops, predominantly in Africa and Asia, which may affect future responses to N fertilization (Lu & Tian, 2017). The accumulated addition of P for 2000–2050 has been estimated at 1232 Tg P across the four Millennium Ecosystem Scenarios (Peñuelas et al., 2013), so the P deficit for cereal crops may increase exponentially, especially in large areas of Africa and Russia (Peñuelas et al., 2013; van der Velde et al., 2014).

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

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

4.2 Human health

Changes in N, P, and N:P ratios cascade up the trophic chain, potentially to humans from food production, when the effects of over-fertilization and imbalances in N:P ratios in crops may become apparent (Peñuelas et al., 2017b; Peñuelas et al., 2019b). N fertilization has historically been excessive in rich countries and has led to the over-production of food, and the low use of fertilizers has staved off malnutrition in poor countries (Smil, 2002). Men born in rich countries in the 1980s were an average of 1.5 cm taller than men born in the 1960s, whereas the height of males born in the same decades in poor countries did not differ (Peñuelas et al., 2017b). Differences in per capita N, P, and N:P intake explained these differences in the height of men born in rich countries better than did socioeconomic and sanitary variables, such as gross domestic product, the human development index and birth weight according with FAO, OCDE and WHO integrated data analyses (Peñuelas et al., 2017b). Some malign neoplasms, particularly of the colon and lung, contain higher concentrations of P and lower N:P ratios than do healthy organs and surrounding tissue (Elser et al., 2007a, b). High N and P intakes from an increased consumption of animal-based foods in some developed countries would therefore likely lead to higher heights, albeit with a higher risk of mortality from cancer.

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

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

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

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

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

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

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

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

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

Reduction of livestock production has been suggested as the most effective approach to reduce global P demand and ensure global food security (MacDonald et al., 2011; Schröeder et al., 2011; Withers et al., 2018b). The three-fold increase in livestock production in the last five decades (Liu et al., 2017) has led to decreased P-use efficiency of inorganically fertilized forage crops and P surpluses from inputs of animal urine and manure (MacDonald et al., 2011; Nesme et al., 2015). A global reduction in livestock production for dietary consumption would decrease the demand for P and its associated environmental problems (Neset & Cordell, 2011; Wu et al., 2016; Bai et al., 2016; Wang et al., 2018e). Decreases in animal production would increase the availability of cropland for producing crops for direct use in human diets, shortening the food chain and increasing resource-use efficiencies, including P, but also N and water (Neset & Cordell, 2011; Rowe et al., 2016). Reducing the consumption of monogastric livestock would increase the sustainable use of P for food production, because such livestock do not efficiently absorb P from forage (Prasad et al., 2015; Wang et al., 2018e).

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

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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 (±SE) anthropogenic inputs of reactive nitrogen (N) and phosphorus (P) to the biosphere (Tg y1) since the industrial revolution. b) Mean (±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; Grübler, 2002; Bouwman et al., 2013a; Fowler et al., 2013; Gu et al., 2013; Lu & Tian, 2017; Yara Fertilizer, 2018), N2 fixation in cropland (Delwiche, 1970; Burns & Hardy, 1975; McElroy et al., 1976; Söderlund & Svennson, 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 & Svennson, 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.

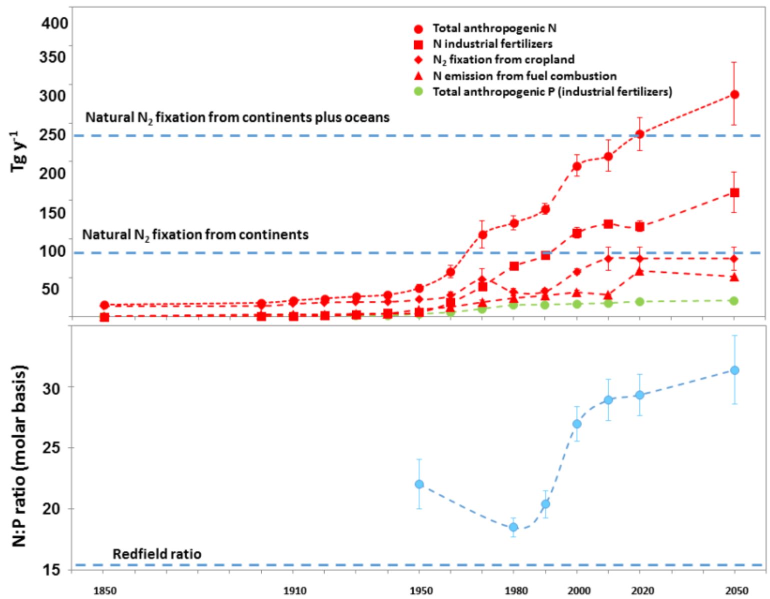
Figure 6. Numbers of studies from the 'Web of Science' search that report effects of increased availability of environmental nitrogen on increased environmental N:P ratios, increased organismic N:P ratios, decreased growth rates, and changes in community structure and ecosystem functioning. The effects 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_2 concentrations	-	*Decrease in soil [N] and [P]*	*Decrease in [N] and [P]* *Decrease or no change in N:P depending on plant type and plant organ*	*Decrease in [N] and [P]* *Decrease or no change in N:P depending on plant type and plant organ*	*Decreases, increases or no change in [N], [P] and N:P depending on phytoplankton species*
Warming	*Heterogeneous effects on soil N and P concentrations and N:P ratios, but most studies reported lower soil [N] and [P] with higher temperatures*	*Heterogeneous effects on soil N and P concentrations and N:P ratios, but most studies reported higher soil [N] with higher temperatures*	*Heterogeneous effects on plant [N] and [P] and in N:P ratios*	*Heterogeneous effects on plant [N] and [P] and in N:P ratios*	*Changes in [N] and [P] depending on multi-functions allocation which also depend on each ecosystem trophic level and biotic and abiotic particular conditions*
Drought/aridity	*Decreases in[N] and maintenance or increase in [P]* *Decreases in N:P ratio*	*Increases in [N] and larger increases in [P]* *Decreases in N:P ratio*	*Increases in [N] and [P]* *Decreases in N:P ratios*	*Decreases in [N] and [P]* *Increases in N:P ratios*	-
N deposition	*Increases in [N]*	*Increases in [N]*	*Increases in [N]*	*Increases in [N]*	*Increases in [N]*

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

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



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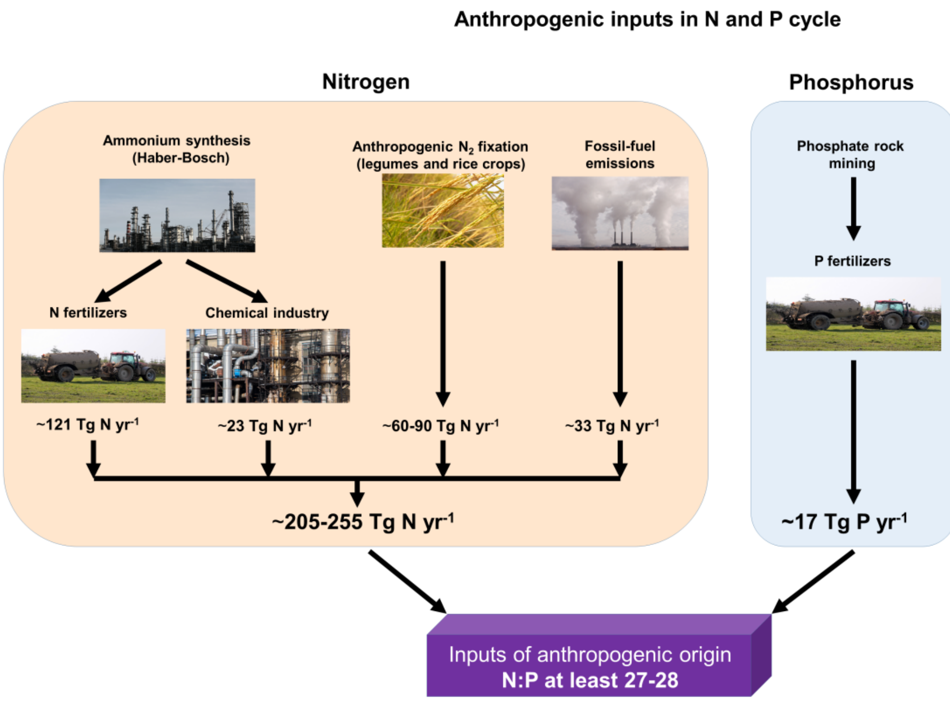

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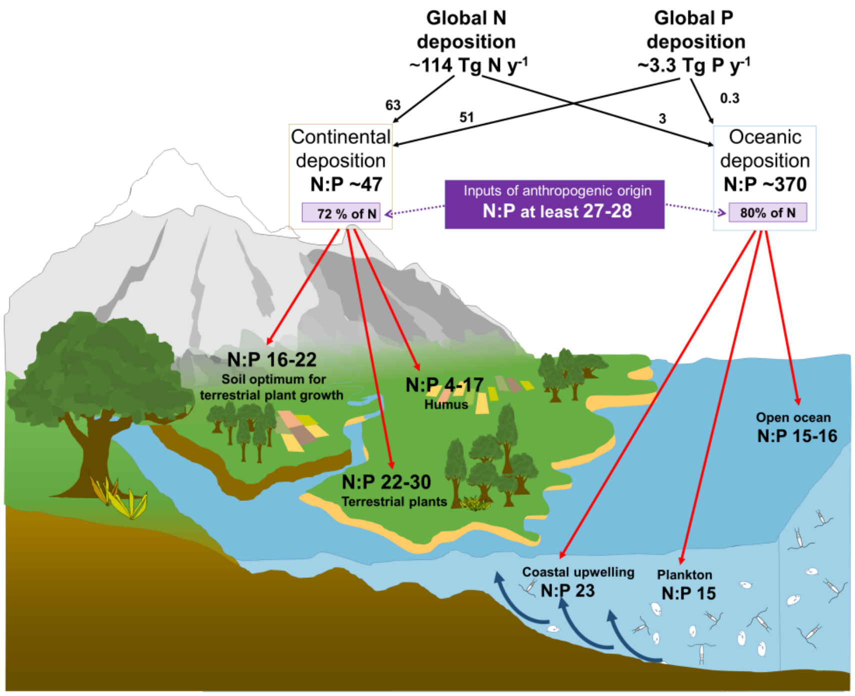

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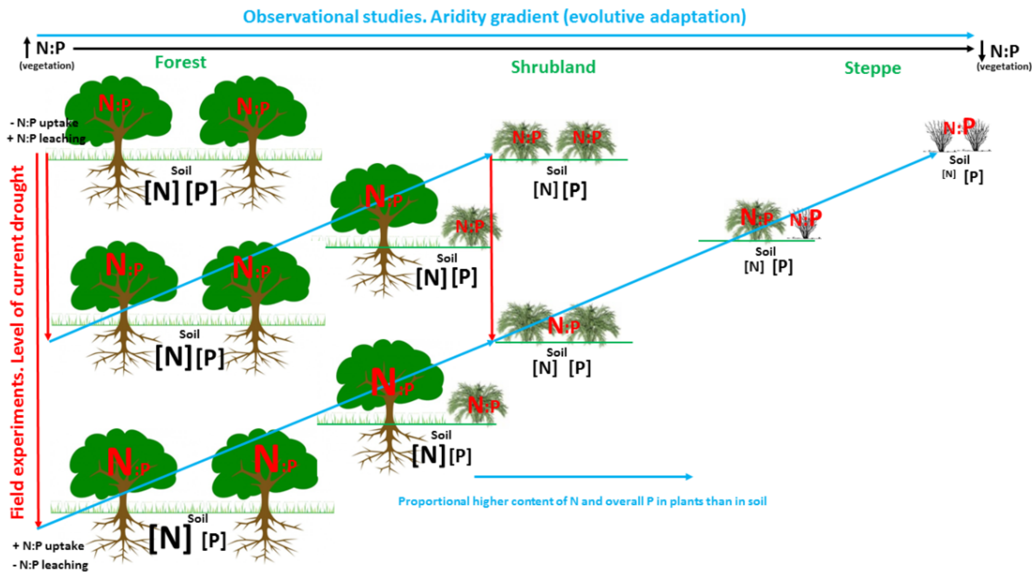

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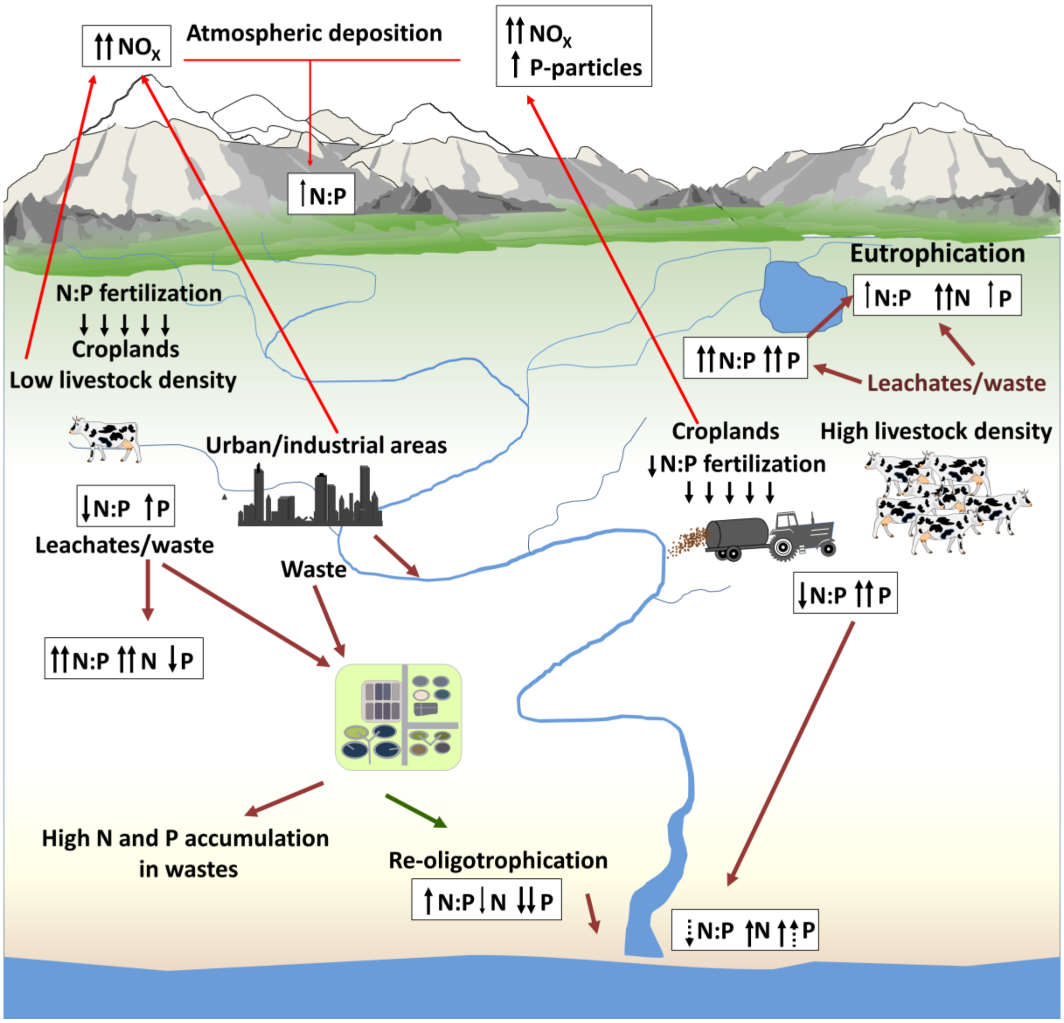

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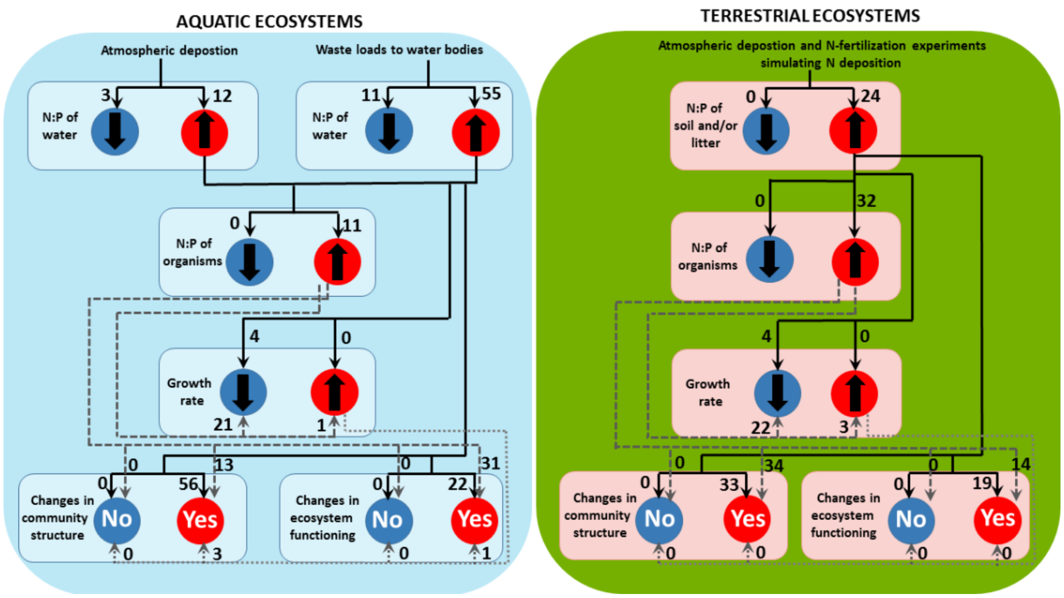

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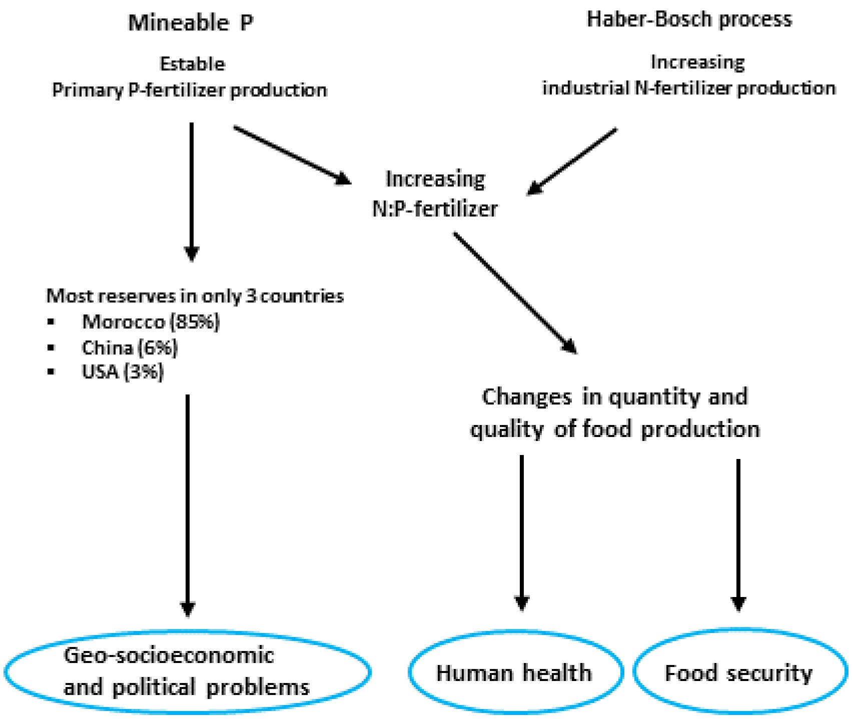

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