

Aerosol Deposition Impacts on Land and Ocean Carbon Cycles

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Abstract

Purpose of Review Atmospheric aerosol deposition is an important source of nutrients and pollution to many continental and marine ecosystems. Humans have heavily perturbed the cycles of several important aerosol species, potentially affecting terrestrial and marine carbon budgets and consequently climate. The most ecologically important aerosol elements impacted by humans are nitrogen, sulfur, iron, phosphorus, and base cations. Here, we review the latest research on the modification of the atmospheric cycles of these aerosols and their resulting effects on continental and marine ecosystems. **Recent Findings** Recent studies have improved our understanding of how humans have perturbed atmospheric aerosol cycles and how they may continue to evolve in the future. Research in both aquatic and terrestrial environments has highlighted the role of atmospheric deposition as a nutrient subsidy, with effects on ecosystem productivity. These studies

further emphasize the importance of local biogeochemical conditions and biota species composition to the regional responses to aerosol deposition.

Summary The size of the impact of anthropogenic aerosol deposition on the carbon cycle and the resulting climate forcing is at present not well understood. It is estimated that increases in nutrient subsidies from atmospheric deposition across all ecosystems are causing an increase in carbon dioxide uptake between 0.2 and 1.5 PgC/year. As aerosol emissions from industrial sources are reduced to improve air quality, these enhancements in carbon uptake may be reduced in the future leading to reduced carbon dioxide emission offsets. However, large uncertainties remain, not only because of limited information on how humans have modified and will modify aerosol emissions, but also because of a lack of quantitative understanding of how aerosol deposition impacts carbon cycling in many ecosystems.

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Introduction

Atmospheric aerosols are liquids or solids suspended in the atmosphere. Aerosols are a significant contributor to air pollution, modify shortwave and longwave radiative fluxes, and impact cloud properties [1]. In addition to the atmospheric impacts, aerosol deposition onto land or ocean surfaces can impact various biogeochemical cycles acting as either a source of nutrients or pollutants. [2–6, 7••]. Anthropogenic activities are currently modifying the concentration and chemical composition of aerosols and thus influencing ecosystem biogeochemistry upon deposition [8]. Presently, 50% of anthropogenic carbon dioxide emissions are taken up by the land and

ocean [9]; the future ability of the biosphere to continue acting as a sink for carbon is uncertain and may be in part dependent on the amount and composition of aerosol deposition [10]. A recent study estimated that aerosol impacts on biogeochemistry may contribute as much climate forcing as either the aerosol direct or indirect-cloud radiative effect [9].

Aerosol deposition can be beneficial or detrimental to ecological processes depending on both the amount and composition of deposition and the underlying ecosystem conditions (e.g., soil properties), and thus the impact from aerosol deposition requires knowledge of local conditions [2, 3]. Similarly, as discussed further below, spatial patterns and temporal trends of atmospheric deposition vary greatly, and have a range of impacts on receiving ecosystems. Aerosol deposition of nitrogen (N) and phosphorus (P) on land can stimulate growth in nutrient-limited regions [11, 12] and the deposition of acid-neutralizing minerals can add alkalinity and base cations such as calcium (Ca^{2+}) to depositional regions [13, 14]. In contrast, the deposition of acidic species such as sulfate or nitrate can reduce soil pH and alter cation exchange capacity thereby reducing the availability of required cations such as Ca^{2+} [2]. Nitrogen limitation is thought to be widespread in terrestrial systems [15], while tropical forests and savannahs may be phosphorus or base cation limited [16, 17], and nutrient co-limitation is common in both terrestrial, marine, and freshwater aquatic ecosystems [18]. Anthropogenic deposition of N or P to these regions may enhance carbon uptake [3, 11, 12, 19]. Freshwater aquatic systems are likely to be sensitive to both N and P deposition as well as to deposition of heavy metals, acidic, or acid-neutralizing compounds [20–22, 23••, 24]. Productivity in some marine ecosystems can be limited by phosphorus, nitrogen, or iron [25–28], and will likely respond to enhanced anthropogenic deposition of these nutrients [5, 6]. However, anthropogenic aerosol deposition of certain metals or acidic compounds may decrease ocean productivity [29, 30].

Here, we review the relatively new research field of investigating biogeochemical impacts from aerosol deposition to continental and marine ecosystems and their feedbacks onto climate. Because of the broad nature of this question, we focus primarily on how anthropogenic aerosols impact the carbon cycle and the corresponding climate feedback, along with how these aerosols influence ecosystems (e.g., productivity, water quality). The degree to which humans have perturbed the nitrogen and sulfur cycles and, in some regions, the iron, phosphorus, and other trace metal cycles is an area of active research. While this paper emphasizes the importance of the aerosol deposition, for sulfur and nitrogen, it is often very difficult to separate the magnitude and impact of aerosol deposition from that of gaseous forms of these chemical species.

Much of the literature reviewed below does not distinguish between the gaseous, wet, and aerosol components of N and S deposition, and thus we include them in this discussion. The first section of this paper reviews recent work dedicated to understanding the magnitude of anthropogenic perturbation to aerosol sources and deposition, as well as the importance of aerosol size and chemical state. The second part of the paper focuses on the impacts of aerosol deposition onto terrestrial, freshwater aquatic, and marine ecosystems. This new area of research seeks to investigate the combined impact of multiple constituents, for example, the impact of local conditions, varying time scales, and the differential response of biotic species to aerosol deposition. Throughout the paper, we emphasize recent work, address open questions and areas of controversy, and highlight important research questions.

Aerosol Sources

Aerosols are highly heterogeneous in space, time, and chemical composition [1]. The atmospheric aerosol mass budget is dominated by natural desert dust and sea-salt aerosols [1]. The direct radiative effects and indirect cloud effects (whereby the radiative balance is modified directly by aerosols or indirectly through modifying cloud albedo) are mainly controlled by aerosol surface area and tend to be dominated by fine mode particles (0.1 to 2 μm diameter). In contrast, some biogeochemically important aerosols (e.g., those containing Fe, P, base cations) have most of their mass in the coarse mode ($>2 \mu\text{m}$) [31]. Common methods for determining aerosol properties, such as aerosol optical depth obtained from satellite and remote sensing approaches, readily capture the fine mode aerosols but satellite methods tend to work less well for coarse mode aerosols, and thus are not sufficient to estimate aerosol-biogeochemistry impacts [31, 32]. The magnitude of the current atmospheric budgets for different elements considered here are quite different; as described below, the best estimates are 110 Tg N/year, 50.5 Tg S/year, and 60 Tg Fe/year, but only 1 Tg P/year (Fig. 1). Note particularly the large N:P deposition ratio of 110:1, whereas the Redfield ratio is 16:1. Here, we are focused on the anthropogenic aerosols and their impact. There are multiple methods for identifying natural versus anthropogenic sources, including bottom-up emission estimates, [e.g. 40], as well as top-down estimates based on observed spatial distributions, correlations between elements [e.g. 45], or isotope-based discrimination [e.g., 46, 47]. The ongoing work to separate the role of natural and anthropogenic processes in contributing to aerosol fluxes and the work on characterizing chemical composition and particle size distribution of aerosols will be the focus of this section.

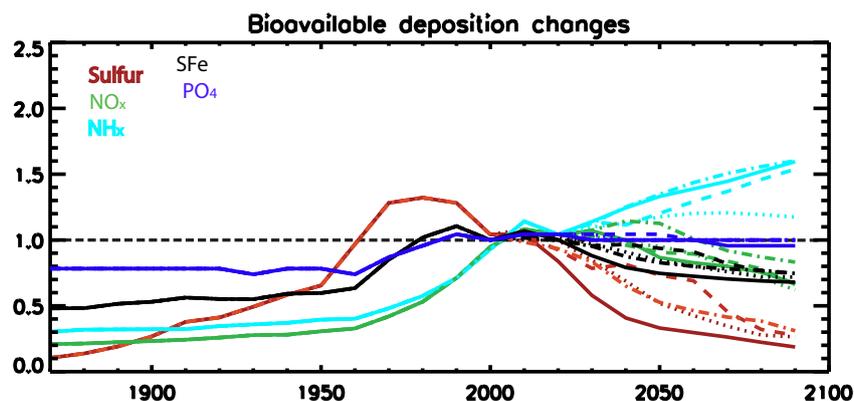


Fig. 1 Relative changes in emission changes for aerosol species relative to emissions in 2000. For the future, estimates are based on the representative concentration pathways estimates for four different scenarios [33–39], represented by the *four lines* into the future. Deposition changes for important species, all normalized to 2000 values (b) for sulfate (*red*), nitrogen oxides (*green*), ammonia (*cyan*), soluble iron (SFe) (*black*), and soluble phosphorus (PO₄) (*blue*) based on historical estimates [40], and for future estimates, the representative concentration pathways [33–39]. Changes for SFe and PO₄ use estimates of desert

dust changes from [41], assuming in the future the model mean for no carbon dioxide fertilization changes in source area from [42], with iron solubility estimates from [43, 44], scaled by the sulfate deposition changes shown and for combustion soluble iron sources from [44] scaled by the black carbon emission changes. For soluble phosphorus (phosphates), desert dust estimate changes are the same as described for iron, and combustion sources of phosphorus come from [45], and are scaled by black carbon emissions. Adapted from [8]

Nitrogen and Sulfur Aerosol Sources

Both nitrogen and sulfur atmospheric cycles have been heavily perturbed by human activities, and their sources include both gas phase primary emissions (emitted directly into the atmosphere) and secondary aerosol formation (formed in the atmosphere from precursor gas species, e.g., NO, NH₃, or SO₂) [48] (Fig. 1). Emission and deposition data for N and S constituents are included in the standard historical and future datasets created for the Climate Model Intercomparison Project (CMIP) [40, 49]. Emission and deposition estimates are critical to understanding the effects of aerosols on ecosystems because, as noted below, uncertainties in these fluxes lead to uncertainties in the assessment of impacts.

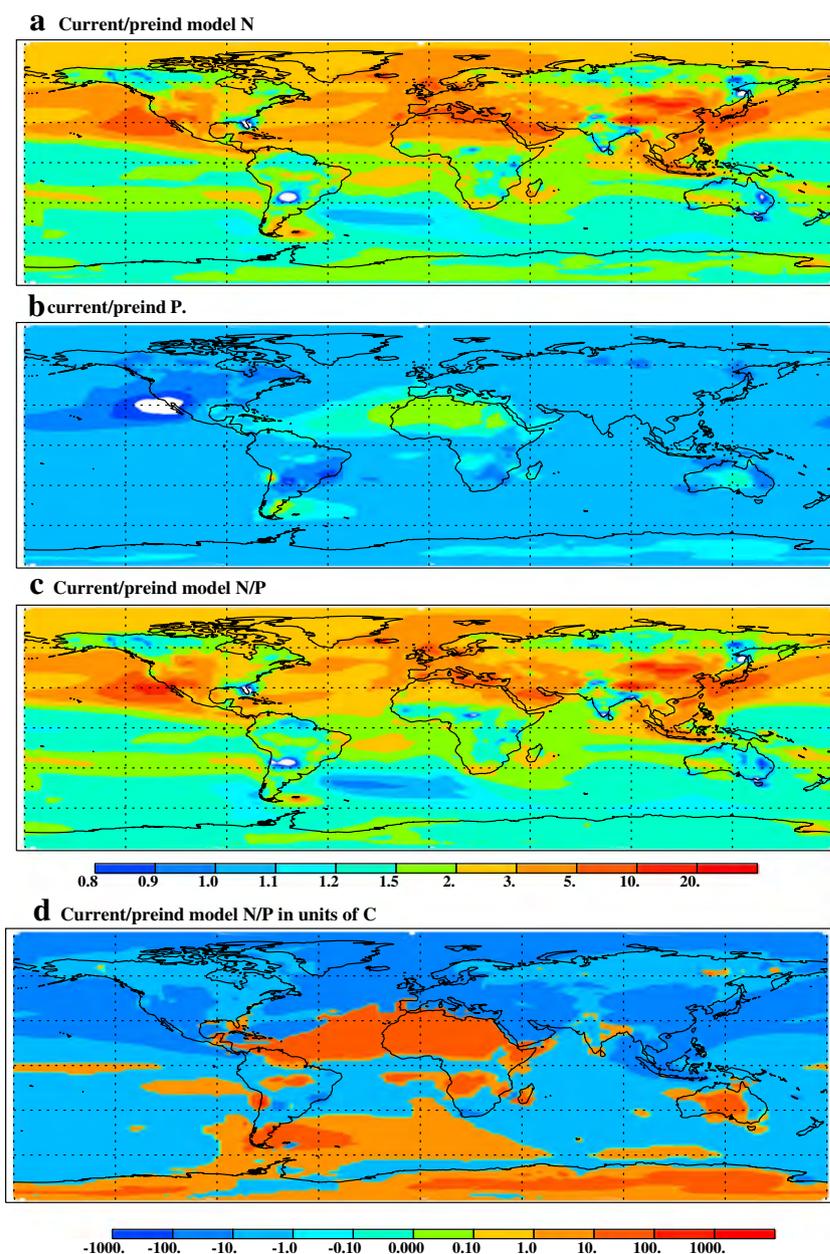
Estimates suggest that nitrogen aerosol and precursor emissions to the atmosphere have increased by approximately 250% over preindustrial conditions, with nearly half of these emissions being processed into aerosols in the atmosphere [50, 51] (Fig. 1) with much of the increase occurring near industrialized regions in the Northern Hemisphere (Fig. 2). The primary emission sources of N aerosols are nitrogen oxides (NO and NO₂) and ammonia (NH₃) released during fossil fuel combustion and as the result of agricultural activities including manure production and the use of fertilizers (both synthetic fertilizers and manure). Human activity is thought to have increased sulfate precursor emissions by a factor of 5 [40]. Naturally occurring sulfate aerosols are directly emitted from volcanoes while a large fraction of sulfur-bearing compounds (e.g., dimethyl sulfide and hydrogen sulfide) from ocean emissions are chemically transformed into sulfate aerosols in the atmosphere. The most significant biogeochemical

impact from increased sulfur emissions is the acidification of precipitation and subsequent impacts on the receiving ecosystems.

A recent observational synthesis has characterized the global spatial distribution and the recent changes in precipitation chemistry and deposition fluxes, and includes the wet and dry, gas and aerosol deposition of sulfur, nitrogen, and phosphorus along with estimates of pH [52••]. This work provides important constraints on ecosystem impacts; however, reliable dry deposition fluxes are difficult to measure, and these fluxes may contribute significantly to deposition budgets in many locations [52••]. Because of the short lifetime of aerosols, paleorecords at multiple locations must be synthesized in order to obtain regional signals [53] and thus we are not able to measure paleo time series for N and S aerosol deposition except at very unique locations (e.g., [54]). Therefore, for most regions, we must rely on estimates of emissions and models to estimate the magnitude of deposition changes in the past.

Global emissions of sulfur dioxide, nitrogen oxides, and ammonia have changed in the last several decades with strong regional variations. These changes are likely to have induced rather dramatic variations in aerosol composition and deposition, especially in regions close to industrial or agricultural activity [52••]. Global anthropogenic sulfur emissions have generally decreased since the mid-1970s (estimated emissions in 2011 are 50 Tg S/year) but with strong regional variations in the emission trends [55] (Fig. 1). Most notably, Asian emissions have been estimated to have peaked in 2006, while European and North American emissions have been decreasing since the mid-1970s [55]. On the other hand, global emissions of nitrogen oxides (NO and NO₂) have changed little

Fig. 2 **a** Modeled ratio of current to preindustrial N deposition from the Community Atmospheric Model (CAM4). **b** Modeled ratio of current to preindustrial phosphorus deposition. **c** Modeled results of the current to preindustrial N:P ratios in deposition. The hemispheric difference in the mass flux changes of N versus P is clearly shown. **d** Stoichiometric representation of the relative impact of P versus N displayed as a ratio of the current to preindustrial deposition rates based on results from the CAM (v4). Values are calculated as current/preindustrial in units of C, such that P is represented as $P \times 106$, and N as $N \times 6.625$. Values are shown as positive (reds) when increases in P deposition dominate, and negative (blues) when increases in N dominate. Adapted from [23]



over the last decade (current emissions are 48.4 Tg N/year), but with large increases regionally in China and India offset largely by decreases in Western Europe and the USA [56, 57] (Fig. 1). Decadal trends in ammonia emissions are difficult to estimate (current estimated emissions are 58 Tg N/year) [58••], but recent simulations suggest ammonia emissions have increased since the preindustrial era by a factor of 7 (from 3 to 21 Tg N/year) due to the increased production of manure (from 0 to 12 Tg N/year) and due to the use of synthetic fertilizer [59–61]. Current estimates of N and S emissions from fossil fuel combustion sources are relatively well characterized [59], but emission changes from soils and agricultural practices are generally harder to estimate. The latter

emissions depend on the interplay among climate, land use and land use conversion, wildfire suppression [60–62], and agricultural practices. Uncertainty in nitrogen budgets may also arise from neglecting organic nitrogen species [51]. Satellite measurements have been interpreted to deduce quantitative NO and NO₂ emission estimates [56] and NO₂ tropospheric trends [63].

Globally, anthropogenic NO, NO₂, and S emissions are projected to decrease in the next century due to more stringent air pollution controls [49]. On the other hand, ammonia emissions are widely projected to increase due to increases in population and food equity, and possibly increased use of biofuels, although these drivers could be partially offset by

increases in agricultural efficiency [64] and diet optimization (Fig. 1). Future changes in aerosol composition are thus also expected to be substantial with ammonia nitrate aerosols expected to be of increased importance [65]. While this has been hypothesized to reduce overall atmospheric acidity, one study suggests aerosol acidity will remain at current levels until atmospheric sulfate reaches near preindustrial levels [66]. Climate also impacts ammonia volatilization with recent studies estimating approximately 3–7.5% increase in agricultural emissions of ammonia per degree Celsius warming in the future [67, 68]. Under the representative concentration pathway (RCP) 8.5 scenario [69] for climate change and middle of the road assumptions for agricultural fertilization accounting for population change, per capita caloric intake and meat consumption a recent study predicts emissions of ammonia will double again by 2100, with 23% of this increase attributable to climate change [70]. Understanding the climate response due to emissions from agriculture and animal management is likely to require additional field observations and modeling studies.

Iron, Phosphorus, and Base Cations

In contrast to N and S species, iron (Fe), phosphorus (P), and base cations occur in the atmosphere almost entirely in the aerosol phase; they are predominantly coarse mode primary aerosols such as mineral dust [43, 45, 71–77]. Because coarse mode aerosols have short lifetimes (hours to days), they are usually deposited close to the source regions, although they can sometimes travel long distances [78, 79]. Recent studies have highlighted the importance of considering the large size fraction of primary aerosols for biogeochemistry [73, 74, 78–80], especially when comparing observed deposition to aerosol model deposition. Models and in situ concentration observations typically only includes long-range transported aerosol modes (<10 μm) [23••, 81, 82] as this is the size range that dominates the aerosol optical depth and are thought to be most important for health effects [83], and thus neglect the large aerosol sizes which will be dominant close to primary aerosol sources.

Iron, phosphorus, and base cations are likely to have some contribution from industrial and biomass burning combustion sources [84–87] and volcanoes. [88]. The sources of base cations such as Ca^{2+} to the atmospheric in interior continental regions is mostly wind-blown dust [14] whereas base cations are associated with marine aerosols in coastal and maritime settings. [89]. Because phosphorus is a required macronutrient, biota tend to accumulate phosphorus, causing primary biogenic aerosols from both ocean and land to contribute ~10% of the aerosol P [45, 90, 91]. Iron, phosphorus, and other metals in aerosols are less studied than N and S aerosols, and of these, iron is the most studied. Thus, estimates for Fe, P,

and base cations such as Ca^{2+} are not available in the standard CMIP emission and concentration datasets [40, 49] but are estimated from individual model and observational synthesis [43, 45], and are likely to have greater uncertainty. For example, a constant climatological iron deposition to the oceans was used in the CMIP5 studies [92] to look at ocean carbon cycling.

Because particles fall through the ocean mixed layer within a few hours to days, only the soluble or bioavailable fraction of the iron, phosphorus, or calcium may be available to the ocean biota [5, 93, 94]. The spatial distribution of different minerals and elements can be deduced from soil maps, limited soil elemental data, and from size distribution assumptions [95–100], and provide some increased ability in the simulation of fractional Fe, Al, and Ca in dust [101]. Previously, most studies assumed 3.5% of dust was iron with a mean dust solubility of 2% [5]; however, recent studies have highlighted the importance of the various chemical states of iron in dust minerals on the solubilization rate of iron [102–105, 106••]. Different mechanisms for solubilization of iron have been proposed, including photochemical-, acidic-, and organic ligand-mediated reactions [107–111]. Studies have also identified the potential importance of more soluble forms of iron from biomass burning, coal, and other combustion sources [84, 85, 112], since in some regions of the globe, long-range transported combustion iron may be quite important (for example, parts of the Southern Ocean) [44, 113]. Based on local measurements [109, 110] metals in aerosols from mining and smelting operations may be important but are not yet included in model estimates and global budgets.

Observations suggest an inverse relationship between the amount of iron and the solubility of the iron [94, 114]. While intriguing, these observations do not uniquely constrain which processes are most important, since several hypothesis are consistent with this relationship. For example, if fine particles, with longer lifetimes, have higher solubilities (e.g., fine combustion particles versus coarse dust particles) [84, 85, 112], or if atmospheric processing takes time to solubilize aerosols [107, 108, 115], higher iron amounts will occur closer to the sources, with higher solubilities occurring farther from the sources. The emissions atmospheric iron is likely to be ~60 Tg Fe/year, with soluble iron being about 1–2 Tg Fe/year [43], with large ranges possible depending on the size of the aerosol considered [116] (Fig. 1). Our understanding of the distribution and processes important for the formation of soluble iron is constrained by the very limited observations [117, 118], largely as daily averages over the oceans from cruises [93, 94].

The limited paleo-observations suggest a 40% increase in dust over the twentieth century, which would suggest a large increase in atmospheric iron deposited to the oceans [41, 119] (Fig. 1). This large increase in dust may in part be explained by an anthropogenic land use source, which detailed model-

data comparisons based on satellite-retrieved aerosol distributions suggest is 25% of current dust sources [120], and/or by increased aridity associated with climate change [42]. In addition, the increase in combustion iron, as well as increased acidity [110, 111] from S and N emissions likely has increased the soluble iron deposition to the oceans 3–4-fold over the twentieth century [41]. Improvements in air quality in the future may reduce soluble iron deposition from combustion iron [113] although future scenarios for whether iron deposition from dust sources will increase or decrease is subject to competing processes such as increased dust due to land use change [121, 122] or increased aridity [42, 123], decreased dust due to wind changes [124], or some combination of these processes (Fig. 1).

Phosphorus sources and concentrations have received less attention than N, S, and even less than Fe from the atmospheric community [52••]. A recent synthesis of limited available observations and modeling focused on long-range transportation was able to simulate the fine and coarse mode aerosols (<10 μm), as well as the observed source apportionment, based on emission factors from the literature [45]. More recent compilations of deposition fluxes in terrestrial and fresh water ecosystems show the enhanced deposition associated with local sources of primary biogenic aerosols [23••, 125]. Models can make simple assumptions for the sources of super-coarse mode (>10 μm diameter) aerosols in terrestrial ecosystems that are consistent with the few studies of primary biogenic particles [90, 126–128], allowing model simulations to match observations for fine, coarse, and super-coarse mode, the latter of which dominates the deposition close to the source regions [23••, 125]. Differences in the details of how size distributions are considered can result in very large differences in the estimates from different sources (contrast the 20-fold higher combustion P estimate from [129] compared with [23••, 125]) highlighting the sensitivity of the P budget to assumptions in the size distributions of P aerosols, especially when comparing to deposition data in primary aerosol source regions.

Similar to Fe, not all P is equally available to aquatic biota, and there is even less solubility data for phosphorus in either marine or lacustrine systems [45, 130]. The limited data suggests a difference in source solubility, with mineral dust sources being more insoluble than other sources [45]. Recent observational data suggest phosphorus undergoes atmospheric processing to make it more soluble as it moves downwind [130], with acidic processing hypothesized to be the dominant mechanism [131, 132]. The total atmospheric phosphorus in the fine and coarse modes (<10 μm), emitted in aerosol form is likely to be about 1 Tg P/year, and only 0.25 Tg P/year is likely to be soluble [45], with large ranges possible if different sizes are considered [116, 125, 129].

Because of the link between P and mineral aerosols, P deposition is likely to have increased with the increased sources of dust over the twentieth century [41]. Acidification

through anthropogenic emissions is also likely to have directly enhanced the solubility of P [132], in addition to the increase in direct emissions of anthropogenic combustion P [8] (Fig. 1), with a distinct spatial signal than increases in N deposition, thus impacting N:P ratios in deposition (Fig. 2). To better understand atmospheric deposition of soluble iron and phosphorus, additional field and laboratory studies in cooperation with modeling studies are required.

Human activity has likely perturbed base cations and other aerosol metals (e.g., Ca, Cu, Mn). While some of these trace metals may serve as nutrients, others may act as pollutants in terrestrial and aquatic ecosystems [30, 133]. The amount of change in deposition of such trace metals from aerosol, as well as their significance on the carbon cycle, has not yet been established.

Aerosol Deposition Impacts on Ecosystems

The study of the impact of aerosol deposition onto ecosystems began in earnest with heightened awareness of acid rain issues in the 1980s and 1990s [2], and has evolved by considering the influence of anthropogenic aerosol deposition, which can modify land, freshwater, and marine productivity [2–6, 7••, 134] and potentially significantly affect climate through the carbon cycle [10]. Recent evidence has highlighted the necessity of considering ecosystem impacts as a combination of the strong spatial gradients in aerosol deposition, local conditions, different time scales, multi-element interactions with carbon, and the varied impacts on different biotic species, which will be the themes of this section.

Terrestrial Ecosystems

Nitrogen limitation is thought to be widespread in many systems, especially temperate forests [15], and thus the large increase in anthropogenic deposition of N may enhance carbon uptake, but the magnitude of this N-induced C sink has been contentious [3, 11, 12, 19, 135]. N impacts are the most well studied of all the deposition-driven terrestrial carbon feedbacks, and some earth system models include some of the relevant N cycling processes [136]. Recent model estimates suggest between 0.24 and 0.7 Pg of carbon (PgC)/year are taken up by terrestrial ecosystems due to anthropogenic nitrogen deposition effects on plant growth [3, 11, 137]. None of these models simulate the effects of N addition in suppressing soil decomposition rates. In temperate and boreal forests, this effect can yield increases in soil C storage as large as the effects of N on tree C gain [138••, 139, 140]. Nitrogen deposition not only affects C storage, but can also contribute to ecosystem emissions of nitrous oxide and nitric oxide and can suppress the uptake of atmospheric methane in surface

soils [141–143]. Nitrogen can act as both a nutrient and a pollutant depending on the amount of deposition and plant and soil conditions [2, 3].

Acid rain, in contrast, which predominately comes from the deposition of sulfate and some forms of nitrogen, can reduce the availability of required base cations such as Ca^{2+} and reduce productivity in forest ecosystems that lack sufficient buffering [2, 144]. For forests growing on calcium-poor sites, high loads of acid deposition can reduce soil calcium availability and cause a slowdown in growth and increase mortality [145]. Chronic acidification can also increase soil C storage by slowing decomposition, a process reversed when ecosystems recover from reductions in acid deposition [146]. As acid deposition has decreased across northern Europe and eastern North America, losses of dissolved organic carbon (DOC) from soils to streamwater have increased, likely due to increased dissolved organic carbon solubility at higher pH [147]. This deacidification-driven change in DOC loss generally has a small effect on terrestrial carbon balance but can substantially alter the DOC inputs to aquatic ecosystems [e.g., [148]. Overall, the net global carbon impact of acid rain onto changes in greenhouse gas emissions has yet to be established, and regional responses will vary with the direction and magnitude of changes in acidic deposition and loading, as discussed in the emission section. Deposition of sulfate onto peatlands can suppress their emissions of methane, although the net global effect on methane budgets is not thought to be large [149].

Certain bedrock and ecosystem types (e.g., tropical forests and savannas) can lead to phosphorus limitation and low base-cation availability [16, 17, 150]. On long time scales, atmospheric aerosols, especially dust, may provide essential nutrients [151]. Studies have shown that North African dust is a source of P, Fe, and base cations to the heavily weathered tropical soils of the Amazon. These inputs provide nutrients and potentially enhance productivity over long time scales [12, 152] and may be essential to the long-term survival of these ecosystems [153]. Similarly, Hawaiian forests have been shown to be dependent on atmospheric inputs of base cations [154], and alpine forests in Europe derive substantial portions of their base-cation subsidies for growth from dust [155]. Recent studies have shown substantial perturbations to atmospheric phosphorus in the Amazon due to deforestation-related biomass burning [19] and land use [156] suggesting that anthropogenic phosphorus deposition in the Amazon may be enhancing productivity (i.e., net primary production) in the remaining forests, by between 0 and 0.5 PgC/year [10]. Modeling studies of the influence of phosphorus deposition onto carbon uptake are in the beginning stages [157, 158]; based on ecosystem budgets, they suggest a smaller role for phosphorus deposition than for nitrogen deposition [157, 158].

Overall, it is clear that nutrient-rich aerosols play an important role in the growth of forests and subsequent uptake of carbon around the world, with substantial regional variations. It is, however, substantially less clear how important these fluxes are to rates of tree growth or how much human activity may have modified these impacts. In a general sense, we know that human perturbation of the N cycle exceeds that of any other biogeochemical cycle and therefore the N deposition effects on ecosystems are likely to be greater than for any other deposition flux. However, over longer timescales (e.g., centuries to millennia) even subtle shifts in deposition of P, Fe, and the base cations could have profound implications for forest development and growth. These issues of both short- and longer term impacts of aerosol deposition are important open questions deserving further research.

Freshwater Aquatic Ecosystems

Anthropogenic nitrogen deposition has long been recognized as an important modifier of freshwater ecosystems, particularly in remote oligotrophic (low-nutrient) systems. N deposition has been linked to acidification and shifts from N- to P limitation [21, 159]. More recent studies have focused on the role of phosphorus, specifically the potential for phosphorus deposition to increase productivity, alter species composition, and shift freshwater systems toward N limitation [7••, 22, 160]. Using a strong observed correlation between the N:P of atmospheric deposition and lake water, one recent study argues that aerosol deposition is a much more important factor in controlling relative nutrient availability than previously thought [23••]. Though humans are modifying N much more than P, considering the Redfield ratio of C:N:P (106:16:1) as a measure of average biological nutrient requirements suggests that P deposition is likely to be more important than N in many ecosystems (Fig. 2).

Though atmospheric deposition of nutrients has been linked to lake eutrophication [161, 162], the potential effects on the carbon budget are less clear. Increases in lake productivity can lead to increases in carbon burial rates [163, 164]. However, lakes are sites of complex carbon processing and whether or not carbon is respired, transported, or buried within the sediments is heavily influenced by other factors including morphology, hydrology, latitude, food webs, and vertical mixing regime [165–167]. Although freshwater systems occupy only a small fraction of the landscape, their impact on the carbon cycle is significant. Although the area they cover and total carbon storage may be smaller, lake systems may be able to take up more carbon annually than marine sediments [164, 168, 169]. Given the magnitude of carbon processed and sequestered in lakes, and the likely sensitivity of aquatic systems to aerosol deposition, more work is needed to understand how

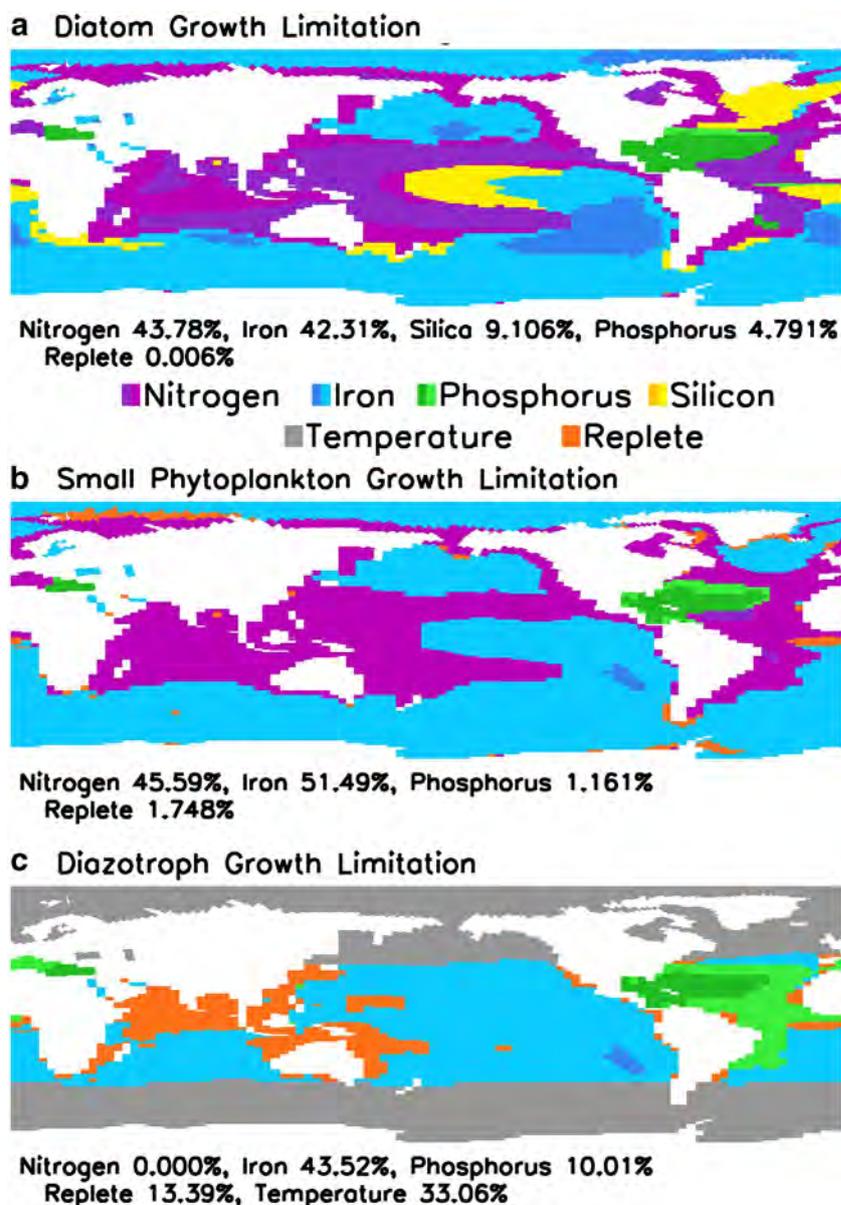
increased anthropogenic aerosol nutrient deposition may interact with climate change to affect the global carbon budget.

Marine Ecosystems

Nitrogen, phosphorus, and iron are known to limit biological productivity and carbon export in the oceans [5, 21, 129]. Thus, atmospheric deposition of these nutrients can directly impact ocean carbon fluxes, particularly where the nutrient deposited is the growth-limiting nutrient for some of the phytoplankton (Fig. 3). When a non-growth-limiting nutrient is deposited, circulation may transport it long distances before

directly impacting productivity [171]. While a few studies have argued that anthropogenic nitrogen deposition is very important for ocean biogeochemistry [6, 172], most consider anthropogenic iron deposition to be the more important factor for carbon uptake [173, 174] because of iron limitation and the potential for large productivity increases in high-nutrient, low-chlorophyll (HNLC) regions of the open ocean [25, 28, 106••, 175•]. Nitrogen fixers extend this sensitivity to atmospheric iron deposition to oligotrophic ocean regions [22, 132, Fig. 3]. Smaller basins may be more sensitive to atmospheric deposition; for example, temporal trends in N and P suggest that an initial increase in nitrogen in the Mediterranean resulted in lower N concentration in response to higher atmospheric deposition, while the phosphorus budgets seem more driven by

Fig. 3 Nutrient limitation map displays the main growth-limiting nutrient over the annual time scale during the 1990s for different phytoplankton functional groups. Darker shade of each color indicates stronger nutrient limitation (growth reduced by >50% from maximum). Nutrient replete means nutrient concentrations are sufficient for the phytoplankton to grow at >90% of their maximal rates, typically indicating light and/or grazing controls on biomass. **a** Diatom growth limitation. **b** Small phytoplankton growth limitation. **c** Diazotroph growth limitation. Adapted from [170] using a recent simulation



the riverine inputs [176, 177]. Atmospheric deposition often results in an oscillatory deposition and response cycle that needs to be considered in understanding short and long time scale responses, especially in low-nutrient low-chlorophyll regions [178]. The larger increases in N deposition than P in many regions of the globe (Figs. 1 and 2) can cause shifts from N to P limitation [175•]. Quantitatively linking carbon fluxes to atmospheric nutrient deposition is complicated by uncertainties in the input rates and internal cycling of each key nutrient, and by interactions and feedbacks across these biogeochemical cycles within the oceans.

Iron cycling within the oceans is less well understood than other nutrients, due to large uncertainties in the inputs of dissolved iron, scavenging losses and interactions with organic ligands, and a limited global observational dataset [137, 139, 179, 180•, 181]. A recent marine Fe cycle model intercomparison [180•] documented orders of magnitude uncertainties in iron sources and residence time across 13 global ocean models. Observational iron-related datasets are rapidly expanding through the GO-SHIP and GEOTRACES programs, providing new insights into iron sources and sinks [182•, 183]. GEOTRACES measures a wide suite of trace elements, some of which may serve as micro-nutrients. Zinc is released from mineral dust deposition, and may influence the ability of phytoplankton to access dissolved organic phosphorus pools [184], illustrating an additional potential link between mineral dust deposition, phosphorus, and carbon.

Nitrogen-fixing plankton species (diazotrophs) have elevated iron requirements, and their growth may be iron-limited over large areas of the oceans, where most of the community is N-limited (Fig. 3). Nitrogen fixation rates can be sensitive to atmospheric iron inputs [22, 105, 132, 135] linking marine N, Fe, and C cycles. Declining biological nitrogen fixation can substantially offset increasing N deposition to the oceans [129, 133, 134]. Recent studies have found considerable regional variations and plasticity in plankton elemental stoichiometry [140•, 175•, 185, 186]. Thus, the stoichiometry of the nutrient deposition and the stoichiometric nutritional demands of different plankton species modify the impacts of nutrient deposition on marine carbon fluxes [175•, 187]. Planktonic organisms will respond differentially to the atmospheric deposition of nutrients (and toxins) impacting plankton community structure and the efficiency of carbon export [141, 170, 188, 189]. Recent estimates suggest an average of 0.07 PgC/year ($\pm 100\%$) draw down due to soluble iron deposition increases over the twentieth century [41]. Combined field, laboratory, and modeling studies of the impacts of aerosol deposition onto the ocean are required to further reduce the uncertainties and better understand the future evolution of ocean biogeochemistry.

Summary and Conclusions

The impacts of aerosols especially N, S, Fe, P, and base cations on climate through terrestrial, freshwater, and marine carbon cycling may be as important as aerosol direct and aerosol-cloud impacts on radiation [10]. However, the biogeochemical processes linking aerosol deposition to carbon cycling have not been extensively investigated. Here, we review current research in this area and identify important research questions.

Though N and S sources are well studied, present and future agricultural emissions of N are still uncertain. For marine and aquatic ecosystems, the soluble forms of Fe and P are likely to be more important. The solubility of atmospheric forms of iron and phosphorus is likely enhanced by anthropogenic activity directly through combustion emissions, as well as through increased emissions of acidic species (e.g., sulfate and nitrate-forming compounds), but the relative role of these different processes is not well understood. For this reason, understanding the relative proportion of natural and anthropogenic emissions, and their solubility is still a priority.

Aerosol impacts on terrestrial, freshwater aquatic, and marine systems are still uncertain in terms of their carbon cycle feedbacks and thus climate. The impacts of increased anthropogenic nitrogen deposition (about half of which is thought to be aerosol form) onto land is thought to increase uptake of carbon by 0.2 to 0.7 PgC/year [3, 11, 137]. Increased phosphorus emissions from biomass burning may be fertilizing the remaining tropical rain forests, causing an increase in carbon uptake between 0 and 0.5 PgC/year [10]. While potentially regionally important, the magnitude of the impact of increased sulfate deposition on the carbon cycle is not clear. The increase in atmospheric soluble iron deposition to the ocean is likely to be causing an uptake of carbon between 0 and 0.6 PgC/year [41]. The overall size of the impact of anthropogenic aerosol deposition onto the carbon cycle and resulting forcing is likely causing an increased uptake of carbon dioxide between 0.2 and 1.5 PgC/year in the current climate. Only the impact of anthropogenic nitrogen deposition on growth is currently considered in IPCC estimates. Future changes in aerosols are likely to be large, as industrial emissions are reduced to improve air quality and how these will influence the uptake of anthropogenic carbon by ecosystems is uncertain. Better constraining the impact on the carbon cycle should be a high research priority.

In addition to the general carbon cycle responses reviewed here, recent studies have emphasized the biotic species-specific response to different elements, the differing time scales of responses, and the impact of combinations of elements onto the ecosystems. Further field, laboratory, and modeling studies, as well as the synthesis between these methods are required to better understand the potential feedbacks of aerosol-biogeochemistry interactions onto climate.

Studies need to be undertaken both within fields (i.e., atmospheric chemistry and aerosols or ecosystem biogeochemical cycling) as well as integrating across fields to better constrain our understanding of this fundamental cross-media process.

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Compliance with Ethical Standards

Conflict of Interest There are no conflicts of interest.

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- Of major importance

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