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REVIEW

An aerosol odyssey: Navigating nutrient flux changes to marine ecosystems

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This perspective piece on aerosol deposition to marine ecosystems and the related impacts on biogeochemical cycles forms part of a larger Surface Ocean Lower Atmosphere Study status-of-the-science special edition. A large body of recent reviews has comprehensively covered different aspects of this topic. Here, we aim to take a fresh approach by reviewing recent research to identify potential foundations for future study. We have purposefully chosen to discuss aerosol nutrient and pollutant fluxes both in terms of the journey that different aerosol particles take and that of the surrounding scientific field exploring them. To do so, we explore some of the major tools, knowledge, and partnerships we believe are required to aid advancing this highly interdisciplinary field of research. We recognize that significant gaps persist in our understanding of how far aerosol deposition modulates marine biogeochemical cycles and thus climate. This uncertainty increases as socioeconomic pressures, climate change, and technological advancements continue to change how we live and interact with the marine environment. Despite this, recent advances in modeling techniques, satellite remote sensing, and field observations have provided valuable insights into the spatial and temporal variability of aerosol deposition across the world's ocean. With the UN Ocean Decade and sustainable development goals in sight, it becomes essential that the community prioritizes the use of a wide variety of tools, knowledge, and partnerships to advance understanding. It is through a collaborative and sustained effort that we hope the community can address the gaps in our understanding of the complex interactions between aerosol particles, marine ecosystems, and biogeochemical cycles.

Keywords: Aerosol nutrients, Biogeochemical cycles, Iron cycle, Ocean health, Interdisciplinary research

Introduction

Research over the past few decades has focused on identifying and observing the processes involved in aerosol nutrient transport to the ocean, its impacts on marine biogeochemical cycles, and the mechanistic pathways for these impacts. There have been several recent comprehensive reviews on different aspects of this topic (e.g., see Meskhidze et al., 2019; Baker et al., 2021; Hamilton et al., 2022 and references within), which we aim to not overly repeat here. In this article, we review recent research to identify potential foundations for future research. We identify gaps in our knowledge and explore

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some of the necessary research, tools, and partnerships required to advance the field. This is a timely assessment that falls within the time frame outlined by the UN Ocean Decade (2021–2030) and at a time when countries are actively working to achieve wider sustainable development goals also set by the UN. As various socioeconomic pressures, climate changes, and advancements in technology progressively transform humanity's way of living, the amount and geographical distribution of anthropogenic emissions to the atmosphere also change. It is necessary to trace the effects of such changes in the atmosphere and examine their implications for biogeochemical processes in the ocean.

Navigation tools: Looking back

The importance of aerosol deposition on the marine environment was first recognized in the mid-1980s, with the publication of the seminal review by Duce (1986). This study represented a culmination of 2 decades of research, where technological development improved the collection of aerosols from remote regions of the world and the analysis of their chemical composition. Duce provided a first "big picture" take on the role of nutrients in atmospheric aerosols on marine biogeochemical cycles; in particular, those particles carrying essential nutrients, such as nitrogen, phosphorus, silica, and/or iron. This synthesis had far-reaching implications for climate science, as it provided the foundation for a potential explanation to the long-standing question: What caused the sudden decrease in atmospheric carbon dioxide during past glacial periods?

The rapid decline in atmospheric carbon dioxide during past glacial periods coincided with an increase in observed Southern Ocean marine productivity along with abundant dust deposition as recorded in marine sediments and ice cores (Lambert et al., 2008; Wolff et al., 2010). Martin (1990) pulled these threads together and proposed that the release of iron from deposited dust aerosol relieved nutrient stress in the surrounding waters. This release of nutrient stress would then stimulate marine productivity and the subsequent removal of atmospheric carbon dioxide. This was a seminal study in providing not only a plausible explanation for an important climate phenomenon but also in stimulating broad interdisciplinary research around the topic of the role of aerosol in marine biogeochemical cycles.

Motivated by this early work and by advances in instrumentation and analytical techniques, a number of researchers throughout the world initiated a new breadth of aerosol nutrient research at the start of this century. The Surface Ocean Lower Atmosphere Study (SOLAS) international research initiative was born in 2004, encouraging cross-laboratory collaborations on atmosphere–ocean interactions. Following SOLAS, a second successful international initiative was formed called GEOTRACES, whose mission is: "To identify processes and quantify fluxes that control the distributions of key trace elements and isotopes in the ocean, and to establish the sensitivity of these distributions to changing environmental conditions." In 2017, GEOTRACES included aerosol sampling guidance in their well-used "cookbook" resource (Cutter et al., 2017) after publishing initial recommendations on aerosol sample processing a few years earlier (Morton et al., 2013). The combined effort of these 2 international research programs has resulted in a broad coverage of observational data on aerosol nutrients, some of which is incorporated in the GEOTRACES Intermediate Data Product. They have also been successful vehicles for supporting interdisciplinary research. However, some oceanic regions, especially in the Southern Hemisphere, remain less studied—both in space and time—due to accessibility reducing the number of research campaigns sailing southern and polar oceans and the difficulty of collecting suitable atmospheric samples in rough sea conditions.

One topic that has been slower to develop is the study of how changes in aerosol deposition impact marine productivity at the wider (basin) scale. Taking a measure of the integrated marine ecosystem response to changes in aerosol nutrient delivery in modern times can provide insights into past climate events and improve understanding on how marine ecosystems are responding to the pervasive human–pollution–climate modification of natural biogeochemical cycles (Mahowald, 2011; Jickells et al., 2017), including the possibility of future marine-based carbon dioxide removal initiatives undertaken for climate mitigation (e.g., Bach et al., n.d.). Understanding this phenomenon becomes even more crucial when considering the possibility of "nutrient robbing." Modeling suggests that while aerosol nutrient addition does increase local productivity in the receiving water, this productivity boost can, in turn, lead to a downstream depletion of previously consumed macronutrients. As a result of the nutrient robbing, changes in the wider spatial scale of marine productivity may be limited (Tagliabue et al., 2008; Hamilton et al., 2020a; Ito et al., 2020b); influencing the spatial distribution of productivity rather than the integrated amount. Such balanced productivity is especially notable in the central Pacific region because equatorial upwelling is the major internal source of nutrients. A more comprehensive regional-scale monitoring of the aerosol–ocean connection is thus much needed and can be achieved by using a wide range of tools including aerosol observations, satellite technology, marine autonomous platforms, and modeling capabilities.

It seems clear that multidisciplinary teams are needed to diagnose aerosol nutrient processes that extend from land emissions to remote ocean deposition and from the upper troposphere to the ocean depths (**Figure 1**). To better understand the flux and composition of aerosols from a wide variety of natural and anthropogenic sources, marine biogeochemists have worked in tandem with atmospheric chemistry and aerosol transport researchers to map the spatial variation and the magnitude of nutrient deposition over time. Among the first to take a multidiscipline/tool approach, Erickson et al. (2003) used a global aerosol transport model and satellite observations. Modeling provided a dust deposition estimate to the Southern Ocean, with Patagonia identified as the main dust supplier. They then found a positive temporal and spatial correlation between the dust aerosol deposition and satellite-derived chlorophyll concentration. While not necessarily proving causality, as atmospheric circulation

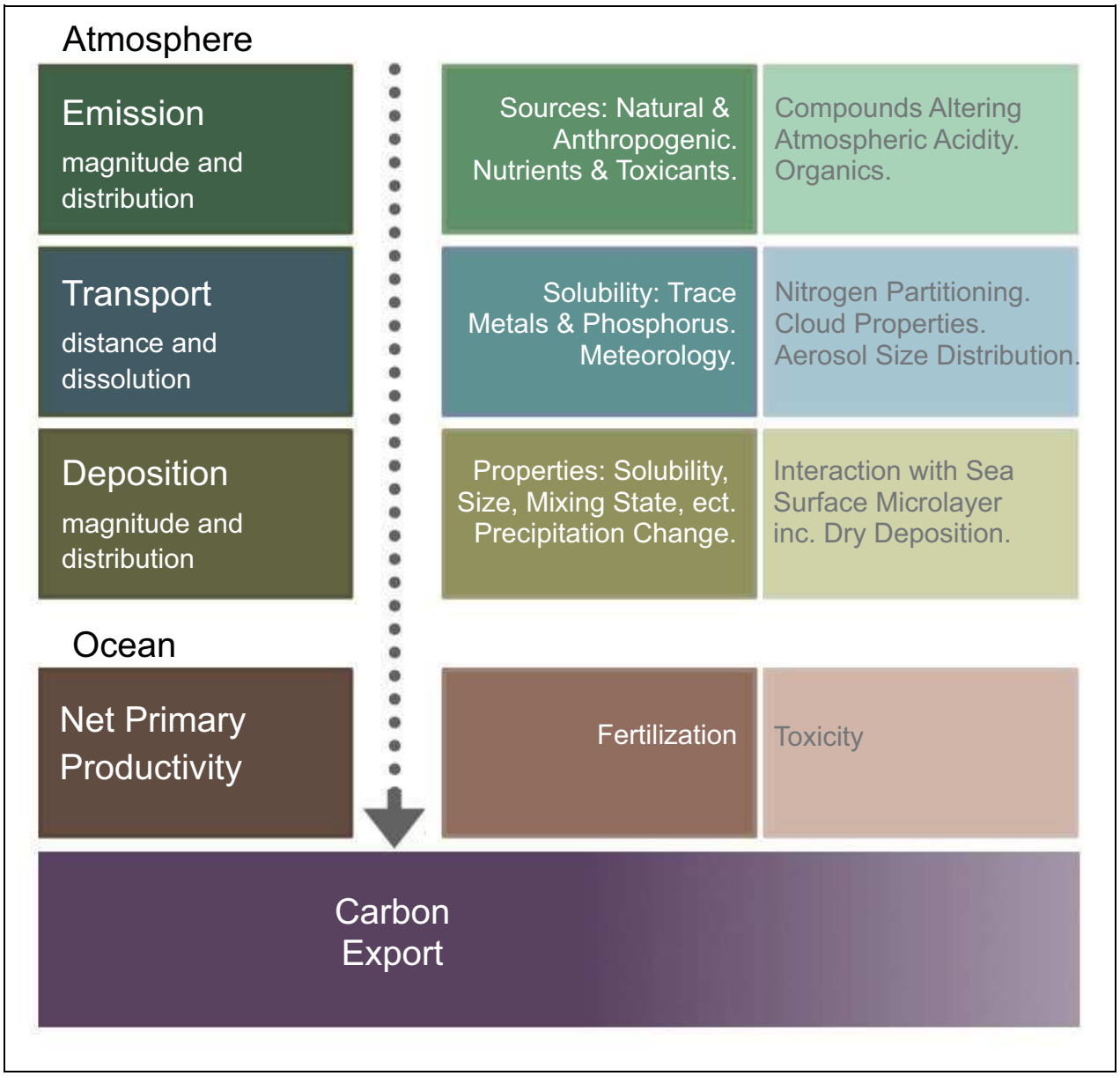

Figure 1. Flowchart of topics relevant to the study of the role of aerosol in marine biogeochemical cycles (left of arrow). This is coupled with a schematic representation of primary and secondary drivers of change in the magnitude, distribution, and availability of atmospheric aerosol nutrient deposition and its impacts on marine biogeochemistry (right of arrow), as discussed in this article. Box shading represents the level of scientific study to date.

may simultaneously enhance both Patagonian dust emissions and the upwelling of nutrient-rich waters (Meskhidze et al., 2007), this study was the first to bring together 2 different tools: models and satellites. Other examples of interdisciplinary teams include paleoclimatologists researching the provenance of dust in ice cores by working more closely with terrestrial and marine geochemists using isotopic techniques (Delmonte et al., 2008; Han et al., 2022) and modeling and remote sensing experts collaborating with atmospheric and marine biogeochemists to understand the transport of aeolian nutrients and impact on primary productivity during large wildfire events (Tang et al., 2021; Ardyna et al., 2022). While these interactions have clearly resulted in scientific transdisciplinary growth, 30 years of research have not yet established the extent to which aerosol deposition regulates atmospheric carbon dioxide levels through the biological carbon pump. One of the reasons is a lack of knowledge on the multiple biological, physical, and chemical processes and couplings at play continue to hinder progress, which is compounded by a somewhat low number of marine observations relative to the time span of research (Hamilton et al., 2022). This is, in part, due to the extremely episodic and localized nature of natural aerosol deposition events in marine areas expected to be sensitive to this input (Mahowald et al., 2009; Hamilton et al., 2019) and the difficulty of collecting observations in remote ocean regions (Anderson et al., 2016).

Setting forth: Emission of biogeochemically significant substances to the atmosphere

Nutrient-bearing aerosols originating from natural sources (e.g., dust storms, volcanic eruptions, wildfires) differ significantly in their fluxes, composition, and properties from those produced by human activities (e.g., industrial emissions, transportation, mining, agriculture) (Baker and Jickells, 2017; Barkley et al., 2019; Ito et al., 2021; Hamilton et al., 2022; Zhang et al., 2022; Shi et al., n.d.). Globally, dust sources dominate the emission of mineral nutrients like iron and phosphorus to the atmosphere while anthropogenic activity is the main source of reactive nitrogen (Jickells et al., 2017; Kanakidou et al., 2018). Regionally and particularly in places upwind of micronutrient (e.g., iron and manganese) limited marine regions other sources can be equally important. As part of identifying future research directions, this emission section focuses on 3 less-studied aerosol nutrient emission sources: fire, volcanoes, and anthropogenic activity.

Fire has been an ancestral tool for wildland management in Indigenous cultures, but record-breaking wildfires worldwide have increased research interest in the impact of fire emissions on climate, ecosystems, society, and planetary health (Shuman et al., 2022). It has recently been acknowledged that fire plumes contain an ideal "cocktail" of essential nutrients for aquatic life (Tang et al., 2021; Ardyna et al., 2022; Perron et al., 2022). While this source of nutrient-bearing aerosol is not new, the intensity and spatial distribution of wildfires are rapidly changing on every inhabited continent (Jones et al., 2022). How much fire can increase aerosol nutrient deposition in oceanic regions, including those that currently receive low aerosol loads, is therefore a topic of growing interest. Yet, several questions must first be addressed before a detailed mechanistic understanding of the impact of fire on marine biogeochemical cycles can be proposed. It is currently unclear how nutrient content is related to fire severity, vegetation biome, and the type and properties of the biomass burning. The relative source contributions of nutrients observed in smoke plumes are also unclear as elements can originate from both the biomass consumed in the fire and from dust that has been entrained from local soils via pyroconvective updrafts (Hamilton et al., 2022; Perron et al., 2022). How coemitted compounds, such as acidic and organic aerosol, influence the in-plume chemistry relevant to nutrients during atmospheric transport is also not readily understood. The story of fire and nutrient supply also does not end when a fire ceases, rather the removal of vegetation by a fire can expose the soil surface and thus enhance dust emissions until the ecosystem recovers (Hamilton et al., 2022; Yu and Ginoux, 2022)—dust that is also potentially enriched with ash from the fire or modified by its heat. Such complex interactions between fire and dust emission pose an issue for current models to simulate, particularly where emission sources are traced separately.

Volcanoes are well recognized for their importance to the Earth System. Volcanic gas, aerosol, and ash emission have widespread impacts, affecting the economy, through impacts on aviation and tourism; human health, through air quality reductions; and climate, through regional weather changes. Sulfur emissions have historically taken the focus in the studies of volcanic emissions, and currently, volcanic ash is not included in standard global climate model configurations. However, volcanic ash has potentially been shown to be relevant in ocean biogeochemistry due to intermittent deposition of large quantities of nutrients, for example, nitrogen, phosphorus, and iron (Achterberg et al., 2021; Perron et al., 2021; Yoon et al., 2023) that are limiting primary production in different regions of the ocean. For example, Olgun et al. (2011) showed that oceanic locations with a high potential for ash deposition overlap with iron limited marine productivity regions, and Achterberg et al.'s (2013) controlled fertilization experiments support the possibility of a positive biogeochemical response to volcanic ash deposition. To better understand the impacts that ash has on regional weather, climate, and ocean biogeochemistry, volcanic ash must be included in global Earth System models. This involves acquiring data to describe the volcanic ash in terms of eruptive quantity, rate, and plume height, as well as its composition. Relevant data can be acquired by satellite and ground-based observations of ash and related eruptive parameters (Gudmundsson et al., 2012; Webley et al., 2012; Bisson et al., 2023). Additional information is needed to verify and optimize such a model, for example, aerosol optical depth (AOD) as detected by satellite instruments, such as the Moderate Resolution Imaging Spectroradiometer (MODIS) on National Aeronautics and Space Administration's (NASA) Aqua and Terra satellites. Model information can then provide insight as to what factors may be affected by changes to the climate system, and the spinoff effects of these changes could be explored by providing the data to collaborators in adjacent fields, such as oceanography or social science. Additionally, such a modeling framework could be used to look at predicted future eruption scenarios—including how a mega volcanic eruption would impact marine ecosystems and planetary health.

Given the potential importance of anthropogenic sources in emitting aerosols that are rich in iron and other metals, the quantification of human activity driven nutrient and pollutant emission magnitudes and properties is a fast-developing research area (Mahowald et al., 2018; Matsui et al., 2018; Ito et al., 2021). Most research to date has focused on iron, including identification of where primary anthropogenic emissions to the atmosphere are dominated by fossil fuel combustion, mining, and metal smelting activities (Rathod et al., 2020; Ito et al., 2021). The magnitude of anthropogenic iron emission estimates has been revised several times in recent years, reflecting both difficulties in constraining this iron source and an increasing research interest in its impacts. An early anthropogenic iron emission estimate by Luo et al. (2008) of 0.66 Tg a^{-1} was revised upward by a factor of 5 to be 3.4 Tg a^{-1} by Matsui et al. (2018) in order to capture observed magnetite concentrations. Most recently, metal smelting was identified as an additional source of iron emission, and current updated best estimates range from 2.2 to 2.6 Tg a^{-1} (Rathod et al., 2020; Ito and Miyakawa, 2023), but given the numerous uncertainties could rise as high as 7.4 Tg a^{-1} (Ito and Miyakawa, 2023). Anthropogenic iron is distinct from

mineral dust iron in several ways. The magnitude of total iron emissions is several orders of magnitude lower than mineral dust (Hamilton et al., 2020b), but iron fractional solubility (soluble/total iron) is typically much higher (Ito et al., 2019). The reason for higher observed solubilities in anthropogenic iron may be due to differences in the mineral composition of iron, with some fossils fuels such as oil being composed of highly soluble iron forms such as ferric sulphate (Schroth et al., 2009; Fu et al., 2012; Ito, 2013). When iron mineral composition is similar, for example, coal is composed of a large fraction of iron oxides (e.g., hematite and magnetite) with low solubility (Li et al., 2022), differences in solubility can occur because of rapid in-plume dissolution chemistry (Baldo et al., 2022). The role of in-plume chemistry in enhancing solubility remains unclear but is likely important due to the presence of acid compounds coemitted in combustion (Li et al., 2017) and the smaller particle size of anthropogenic iron (Ito et al., 2021).

Another example of a metal whose natural cycle has been modified by anthropogenic activity is mercury. Mercury emissions have been declining in some regions, largely due to decreasing coal combustion and the implementation of emissions controls and are expected to decline further due to the implementation of the Minamata Convention. However, natural emissions and resuspended legacy emissions from previously contaminated soils constitute approximately 70% of total emissions, so that mercury deposition will remain high (Pacyna et al., 2016).

While pollution from fossil fuel sources is targeted for reduction, societies' demand for steel and other metal products continues to rise unabated. Metal manufacturing is a complex process that mobilizes metals sequestered for millennia. The extraction, refining, and processing of metals demand significant amounts of energy and natural resource utilization. Mining operations, therefore, result in the release of large amounts of metals into the environment through various pathways, including atmospheric emissions and discharge of wastewater into nearby waterways. To meet growing metal product demands, existing mines will expand and new mines will be developed (Arndt et al., 2017). Until recently, atmospheric aerosol iron modeling did not account for mining and metal smelting because of a lack of understanding of these sources, yet they are likely to be large (Pacyna and Pacyna, 2001; Rauch and Pacyna, 2009). Additionally, metal-containing aerosols originating from mining and smelting activities can be elevated near operation sites due to the suspension of dust and soils that had previously been contaminated—but such a metallic composition enhancement is likely mistaken for "natural" dust in observations and thus needs accounting for in budgets and modeling.

Research Directions:

1. Laboratory and field campaigns aimed at the quantification and classification of biogeochemically significant aerosol emitted from a wide variety of natural and anthropogenic sources.
2. Identifying and understanding climate and human-associated drivers to changes in dust and fire activity and how that changes the emitted distribution of nutrients.
3. Development of a volcanic ash model framework based on the emission and properties of ash for well-observed and characterized eruptions, such as Eyjafjallajökull in 2010.
4. Further observations of various types of volcanic eruptions are needed and should include information on the eruptive mass over time, elemental composition, size distribution, and vertical distribution of the ash.
5. A deeper understanding of how anthropogenic activity alters the atmospheric loading of nutrients and how the properties of anthropogenic aerosol differ from natural aerosol nutrient sources.

The journey: Transport, transformation, and traits of biogeochemically significant substances in the atmosphere

Aerosols undergo a wide range of physicochemical transformations during their transit through the atmosphere. It is while nutrient-bearing aerosols are in the atmosphere (i.e., in transit still) that most samples, usually for subsequent lab analysis, are taken. Thus, much of what we know about the properties of aerosol nutrients and other trace elements over the remote ocean comes from spatially and temporally limited atmospheric snapshots, often taken aboard research vessels. Attaining data to quantify the properties of aerosols from natural sources face a practical limitation, however, in the episodic nature of emission events, such as dust storms, wildfires, and volcanic eruptions. In particular, the unpredictable nature of extreme events makes it challenging to plan measurement campaigns at sea. One approach is the establishment of more long-term observational sites at strategic vantage points able to capture aerosol plumes that are transported over them, including at island locations (e.g., López-García et al., 2021). Another approach is the local collection of ash after a wildfire or volcanic event has occurred. While in situ ash collection may remove seaborne logistical issues, the ground-collected ash chemical composition and particle morphology are likely be different from the aerosol lofted to the atmosphere and care is needed in correlating the two.

It is crucial to understand and quantity an element's solubility (the ratio of the soluble/total concentration) and its evolution in the atmosphere because solubility links atmospheric aerosol deposition to marine ecosystem effects. The soluble fraction of an element in the atmosphere is often used synonymously as the fraction that is bioaccessible to marine biota, and although there may be differences (Meskhidze et al., 2019), we adopt this terminology here. Atmospheric solubility is element specific; for example, on average, iron tends to exhibit low fractional solubilities, while other micronutrients such as manganese tend to have higher fractions (Mahowald et al., 2018). However, quantifying the aerosol trace element (including iron) solubility within different ocean regions is challenging as it can only be estimated from laboratory

leaching experiments. A compounding challenge is that a wide variety of leaching experiments have been used since the late 1990s to quantify what percentage (i.e., solubility) of the total (digestion) element is soluble (leach) in a sample (Perron et al., 2020). The wide variety of leaching method protocols used to operationally estimate solubility results in a large range of reported aerosol iron solubility estimates, which cannot be accurately compared between studies (Shelley et al., 2018). Indeed, each leaching protocol accesses slightly different soluble fractions and represents different processes or events occurring on different timescales (Perron et al., 2020). To date, there has been little cross-laboratory intercomparison of these methods, although efforts are underway under the Reducing Uncertainty in Soluble aerosol Trace Element Deposition (RUSTED) Scientific Committee on Oceanic Research (2022) working group 167. The lack of standardization of aerosol leaching protocols is thought to have hindered the accurate model representation of aeolian trace element fluxes to surface waters (Anderson et al., 2016; Meskhidze et al., 2019) as data produced using different leaching protocols are most often considered as equal in the validation of global aerosol iron models.

An important factor affecting multiple aerosol elements is atmospheric acidity, which includes the acidity from aerosols, cloud droplets, and precipitation (Pye et al., 2020). It is now understood that acidity controls nutrient transport and its bioavailability upon deposition (Baker et al., 2021). For nitrogen, acidity affects the partitioning of ammonia (NH_3)/ammonium (NH_4^+) and nitric acid/nitrate between their gas (former) and particulate (latter) forms (Guo et al., 2016; Uno et al., 2020), impacting atmospheric lifetimes, transport, and mode of deposition (i.e., wet or dry) to the ocean. For example, the atmospheric lifetime of NH_4^+ is much longer than that of NH_3, so that transport distances of NH_4^+/NH_3 are longer under more acidic conditions, although the total amount of NH_4^+/NH_3 deposition is unchanged. For phosphorus, iron, and other trace metals, acidity affects solubility through insoluble minerals readily dissolving under acidic conditions relevant to atmospheric aerosol. The impact of particulate solubility on aerosol in-cloud scavenging, and thus wet deposition, is uncertain due to the many factors influencing the precipitation scavenging of aerosol. In general, the more soluble the element is the more effectively scavenged in clouds it becomes. Yet, solubility likely has a greater influence on rain scavenging compared to snow scavenging and is likely to be more significant for externally mixed aerosol than the internally mixed ones (Cheng et al., 2021). The activation of aerosol into cloud droplets also likely halts the acid dissolution process (Shi et al., 2015). Overall, increased acidity increases the amount of available phosphorus and trace metal deposition to the ocean (Baker et al., 2021).

There is growing interest in understanding the water-soluble fraction of organic nitrogen in the atmosphere. Water-soluble organic nitrogen is estimated to constitute approximately 30% ($\sim$20% from land and $\sim$10% recycled from the ocean) of the total atmospheric reactive nitrogen (soluble organic nitrogen + NH_4^+ + NH_3) deposited to the ocean (Altieri et al., 2021), although contributions vary in space and time. This soluble organic nitrogen appears to be composed of a wide range of different organic nitrogen compounds (Jickells et al., 2013). There is also evidence for water-insoluble organic nitrogen in the atmosphere (Kanakidou et al., 2016), which may include soil organic matter and biological debris, and may ultimately also become bioavailable after aerosol aging (Myriokefalitakis et al., 2020).

The role of aerosol iron in marine biogeochemical cycles has received much attention due to the soluble (bioaccessible) fraction playing a crucial role in phytoplankton growth and productivity in the open ocean. Indeed, ocean biogeochemistry modeling suggests that in the absence of atmospheric soluble iron deposition, nearly all open ocean productivity south of the Arctic Ocean would become limited by low iron availability (Mahowald et al., 2018). The variability in aerosol iron properties between different oceanic regions can be understood by considering differences in source component contributions. In general, iron sources can be divided into 3 components: (1) a primary dust iron source that is characterized by coarse sized particle number concetrations (mostly >2 μm; Adebiyi and Kok, 2020) with a large mass emission flux but low solubility (Jickells et al., 2005); (2) a primary pyrogenic iron source, produced from anthropogenic and natural combustion processes such as industrial and vehicular combustion or wildfires, that is characterized by fine sized particle number concentrations (mostly <2 μm) with a smaller mass emission flux but higher solubility than dust sources (Ito et al., 2019; Ito et al., 2021); and (3) a secondary soluble iron source, which is produced during atmospheric transport due to acid or organic processes liberating soluble iron from insoluble iron-containing particles from any source (Johnson and Meskhidze, 2013; Shi et al., 2015; Li et al., 2017). Recently developed observational tools have assisted in differentiating iron aerosol sources based on the observed properties of iron aerosol itself, as opposed to more traditional methods that use the presence of other tracers measured in the airmass alongside iron to help in distinguishing sources. These new tools include the use of iron isotopes and iron oxide measurements. Accuracy in iron isotope ratio measurements significantly improved with the adoption of multicollector inductively-coupled-plasma mass-spectrometry (MC-ICP-MS) in the early 2000s (Johnson et al., 2020). The isotopic end-member signature of iron in mineral dust aerosol is equivalent to that of the upper continental crust at +0.1‰ ± 0.1‰, while the iron in combustion-sourced aerosol is often isotopically much lighter at values between −4.1‰ and +0.3‰ (Fitzsimmons and Conway, 2023). Using this information, the differentiation of iron aerosol sources has been attempted through the examination of its isotopic composition (Kurisu et al., 2016; Conway et al., 2019). Iron isotopes present promise but more measurement and process understanding are needed, including that atmospheric organic processing of iron in mineral dust is suggested to produce heavier isotope signatures and that the end-member signature of iron in fire smoke is largely

unknown at present (Fitzsimmons and Conway, 2023). Another recent measurement advancement is the modification of the single-particle soot photometer (SP2) instrument (Lamb, 2019), normally used in measuring and characterizing black carbon aerosol in real time (Stephens et al., 2003). The modified SP2 has been used to measure iron oxide particles at low concentrations with subsequent analysis separating mineral from anthropogenic iron oxide sources (Lamb et al., 2021). In both examples, a characteristic of iron, that is sufficiently different between sources, was identified and exploited. To date, only a couple of studies have used these techniques to constrain aerosol modeling of iron, one focused on the North Atlantic (Conway et al., 2019) and another focused on the Southern Atlantic (Liu et al., 2022). In addition to the development of observational techniques and their adoption by modelers in model validation and development, research in different ocean regions is therefore needed.

Research Directions:

1. Development of aerosol trace element measuring techniques that can be used in situ at low atmospheric concentrations.
2. Intercomparison and standardization of different aerosol leaching protocols.
3. Further development and evaluation of aerosol trace element modeling.
4. Development of techniques that support fingerprinting of observed aerosol trace element sources.
5. More long-term, time series orientated, observational sites to aid identifying patterns or trends in aerosol, including episodic and extreme events.
6. A deeper understanding of aerosol nutrient aging and its control on observed solubility under different atmospheric conditions.
7. Linking measured solubility in the atmosphere to bioavailability in the ocean.

The return: Deposition of biogeochemically significant substances from the atmosphere

After their journey, aerosols finally deposit on land or at the surface of the ocean. At the turn of the 21st century, ocean iron fertilization experiments, in particular, the SOIREE experiment in the Southern Ocean (Boyd et al., 2000), confirmed that it is possible to detect increased marine productivity after artificial deposition of nutrients with satellite-derived chlorophyll observations. Shortly after, Gao et al. (2001) made a global estimation of iron deposition based on in situ measurements and proposed using satellites to carry out such a task. The use of satellite platforms was then optimistically viewed as a method to observe aerosol iron deposition and the marine response over large spatial scales. However, the initial enthusiasm generated from these experiments contrasted with the practicality of exploring whether a marine fertilization effect can be observed downwind from major nutrient aerosol source regions. The location of in situ iron fertilization experiments is known with very high precision, yet this is not the case for atmospheric aerosol deposition estimates derived from satellite platforms with once or twice-a-day overpasses. Current satellite data measure where aerosols are in suspension in the atmosphere, yet cannot estimate the flux of aerosol reaching the ocean surface at a given location. Another major impediment in high-latitude ocean regions sensitive to aerosol inputs (e.g., the Northeast Pacific and the Southern Ocean) is the pervasive cloudiness; there can be no direct satellite observation of the ocean surface for several days, or weeks, at such latitudes. Even when clear sky patches are present, the detectability of a signal of interest is not assured. As discussed in the later section "Navigation Tools: Looking Forwards," new satellite platforms are being designed to overcome some of these limitations and the rest of this section focuses on atmospheric aerosol measurements and modeling.

To date, most research has focused on investigating the impact of iron deposition on marine ecosystems, followed by nitrogen and then phosphorus. Current best estimates of deposition to the ocean range between 241 and 377 Gg/a for soluble iron, 45 and 108 Gg/a for phosphate (Hamilton et al., 2022), and between 105 and 198 Tg/a for total nitrogen ($NO_x + NH_y$ + organic nitrogen; Kanakidou et al., 2018). Windblown atmospheric particles reach the ocean surface through wet and dry deposition processes. Modeling suggests that dry deposition is likely to be the primary route of nutrient transfer from the atmosphere to the ocean (**Figure 2**). Dust is modeled with a lower hygroscopicity than other modeled coarse-sized aerosol types such as sea salts (Fanourgakis et al., 2019), and the spatial pattern of higher dry deposition reflecting major dust transport pathways is therefore most likely due to both the larger size of dust particles and their lower ability to uptake water in these simulations.

Results in **Figure 2** represent, to our knowledge, the only global estimate of wet and dry aerosol deposition for all 3 nutrients undertaken to date in a single model framework that also contains a representation of the atmospheric dissolution of iron and phosphorus. However, **Figure 2** iron and phosphorus results can be compared with the more general dust aerosol deposition modeling in other climate models, assuming that the major source of each in many regions is dust (Hamilton et al., 2022). Additionally, iron results can be compared to another Earth System model containing an iron aerosol scheme with atmospheric processing (Hamilton et al., 2019). **Figure 2** agrees with most climate models; of the 13 climate models examined by Zhao et al. (2022), 12 were found to have dry deposition as the major (62%–88% of total deposition) loss pathway for dust (and thus iron and phosphorus). The one outlier is the Community Earth System Model, which is also the host model for the alternate iron aerosol scheme in an Earth System model framework, where simulations of both dust and iron show wet deposition as the major (62%–64% of total dust/iron deposition) loss pathway (Hamilton et al., 2020b).

Beyond iron, the aerosol deposition of other potentially limiting trace metal nutrients, such as manganese or cobalt (Fishwick et al., 2018), are likely important for ocean biogeochemical cycles (Mahowald et al., 2018), but have received less study to date. For example, while light

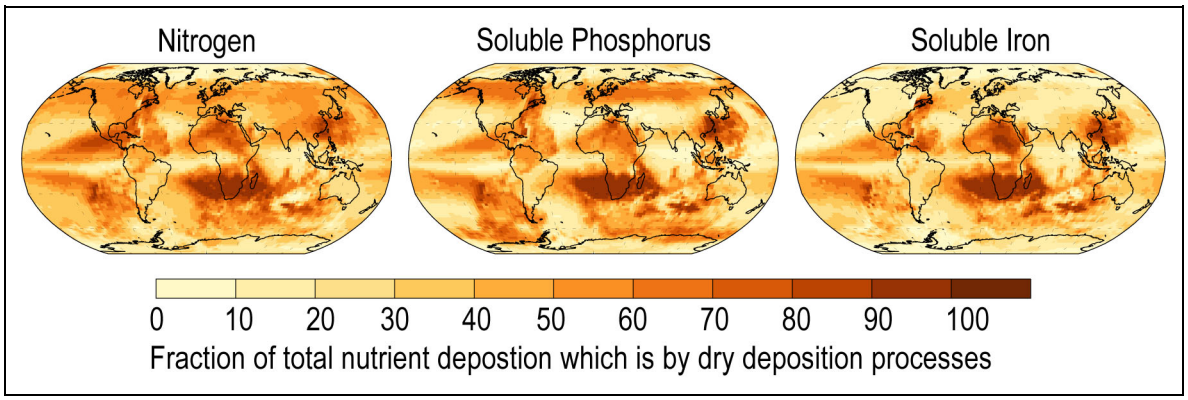

Figure 2. The spatial distribution of the fraction of dry deposition to total deposition for 3 primary aerosol nutrients. Results from TM4-ECPL global atmospheric chemistry transport model simulations (Myriokefalitakis et al., 2020). Emission, transport, and wet and dry deposition fluxes of major aerosol components, including nitrogen (Kanakidou et al., 2016), soluble phosphorus (Myriokefalitakis et al., 2016), and soluble iron (Myriokefalitakis et al., 2015), were simulated globally. The model considered gas-phase chemistry, various natural and human emissions sources, aerosol microphysics, and the chemical conversion, dissolution, and aging of nutrients, providing a comprehensive insight into the distribution of aerosol deposition to the ocean.

and iron have been recognized as the main growth limiting factors for primary producers around the Drake Passage region of the Southern Ocean, new studies have shown that manganese is also limiting for a wide range of Southern Ocean phytoplankton, particularly during the springtime (Latour et al., 2021; Hawco et al., 2022). This phenomenon may also become more pronounced under climate change, as polar phytoplankton adapts to changing conditions, and is discussed further in the next section. A multielement understanding of the complex interactions and interdependencies between potentially limiting elements now becomes necessary for accurately assessing the impact of deposition on marine ecosystems (BOX).

The sea surface microlayer (SML) is the boundary layer between the ocean and the atmosphere, where aerosols finish their airborne journey and begin their oceanic one. The SML is a thin skin (1–1,000 μm) at the surface of the ocean, which harbors a specific ecology and chemistry in comparison to the underlying water just below (Engel et al., 2017 and reference therein). The biogeochemical and physical processes there are critical to air-sea exchanges of energy, gases, and primary aerosols, which affect climate (Engel et al., 2017; Barthelmeß et al., 2021; Hendrickson et al., 2021). Atmospheric aerosol deposition can accumulate at the SML (Astrahan et al., 2016; Tovar-Sánchez et al., 2020), where the aerosol can continue its transformation through photoreaction and interaction with the SML organic matter content, before being exported to deeper waters. However, little is known about the residence time of elements leaching from atmospheric particle at the SML and how they may alter physicochemical properties of the SML and thus air-sea exchange and surface biogeochemical processes. The SML is a gel matrix (Cunliffe and Murrell, 2009), where aerosol can incorporate into transparent exopolymer particle and acts as ballast, enhancing particle sinking rates and carbon export (Bressac et al., 2014; van der Jagt et al., 2018). Despite the extent of the SML covering most of the ocean surface, it is still seldom studied due to the cumbersome sampling methods involved. The promising technology advancement of floats, uncrewed aerial vehicles, and satellites observations (Ribas-Ribas et al., 2017; Ribas-Ribas et al., 2021; Nichol et al., 2023) will likely enable better assessments of the role the SML play in surface aerosol accumulation, transformation, and sinking processes.

The atmospheric transport of aerosol nutrients from the land to the ocean depends highly on prevailing weather patterns (Smith et al., 2017; Hamilton et al., 2020b). Once deposited, ocean nutrient concentrations can further fluctuate depending on abiotic factors, such as ocean mixing transport patterns, and biotic factors, such as the local biological activity (Baker and Croot, 2010; Bressac and Guieu, 2013; Boyd et al., 2017). As a result, the prediction of the precise impact of major aerosol emission events (e.g., large dust storms, a volcanic eruption, or wildfire outbreaks) following deposition to the marine environment requires multiple pieces of information obtained from a wide range of disciplines. Bridging this knowledge gap is key to enhancing global understanding of the interactions between aeolian nutrient deposition and the biological carbon pump within the Earth System. Research teams embracing this venture use ship-based measurements and laboratory studies (Meskhidze et al., 2019) to determine the impact of aerosol deposition on marine productivity and to assess which nutrients are limiting that productivity (BOX). Mesocosm studies are one example of a complex but critical approach to acquire mechanistic understanding of the chemical and biological reactions to aerosol deposition. They have successfully been used for studying dust (Gazeau et al., 2021) and anthropogenic aerosols deposition in seawater (Herut et al., 2016). Recently, an European team of researchers has carried out the first known mesocosm experiments using wildfires ash in the project PYROPLANKTON, funded by the European Space Agency (PYROPLANKTON, 2023). Although the results of this

Nutrient Limitation: Toward a Holistic Multielement Understanding

To understand how future changes to aerosol composition and deposition fluxes impact marine ecosystem productivity and services, it is important to consider that aerosol is often a mixture of elements—with the potential for both nutrients and toxicants to be present. In the Figure circle color indicates the nutrient(s) limiting growth, as inferred from chlorophyll and/or primary productivity increases following artificial nutrient addition. The benefits of taking a multielement aerosol approach become even more evident when considering that marine nutrient colimitation is a common feature across the world's ocean. While single-colored circles indicate that no secondary limiting nutrient was identified, this may also reflect a lack of testing. Additionally, to better understand the impact of nutrient colimitation on production and respiration, the overall microbial food web needs considering – not only the phytoplankton response to aerosol deposition.

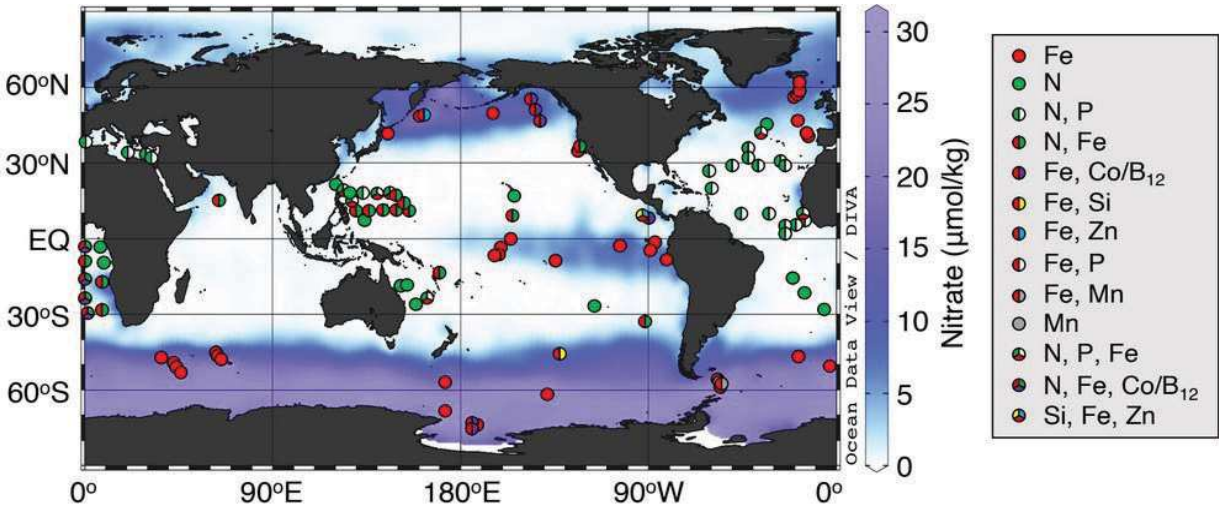

Figure: The spatial pattern of marine nutrient limitation, adapted from Hashihama et al. (2021). Data from: Moore et al. (2013) and references therein; Browning et al. (2017, 2018, 2021, 2022); Hattori-Saito et al. (2010); Dreux Chappell et al. (2016); Kondo et al. (2013); Li et al. (2015); Mackey et al. (2014); Saito et al. (2014); Takeda et al. (1995). Surface nitrate concentration taken from the World Ocean Atlas (2018).

experiment are work-in-progress (Guieu et al., 2023), the work done during the experimental design showed a high uncertainty on the chemical composition of ash with regard to its size, origin, and collecting methods (Llort, personal communication, 19/07/2023). Next steps in this direction will be to use ash from different origins and field campaigns to sample the aerosols at both emission and deposition.

Research Directions:

1. How nutrients are transferred from the atmosphere to the ocean contains many unresolved questions and future work to improve deposition estimates includes:
   a. Improving observational capabilities and model parameterizations of wet and dry deposition.
   b. Model inclusion of data-driven methods with existing physics-driven parameterizations to improve performance, such as in the representation of subgrid scale or high temporal frequency processes.
   c. A more complete understanding of aerosol properties, including size, shape, hygroscopicity, and composition that influence the deposition process and how to best represent them in models.
   d. Reducing uncertainties in surface characteristics, such as roughness, and how they influence the deposition velocities of aerosol.
   e. Increased observational data for model evaluation, particularly over the remote ocean.

f. Increased understanding of phoretic effects. Gradients in temperature, water vapor, and electricity near the surface represent an additional influence on the dry deposition of particles on water compared to terrestrial vegetative surfaces.

g. Further study of the isotopes such as beryllium (^{7}Be) and thorium ($^{232/230}$Th) that can be used to deduce aerosol deposition fluxes and have potential to constrain and improve models.

h. Solutions to overcome issues with the current opportunistic nature of rainwater sampling at sea.

2. Taking a more holistic approach to aerosol science that includes a multielemental (i.e., beyond nitrogen, phosphorus, and iron) understanding of the complex interactions and interdependencies between potentially limiting elements.
3. Explore the control of dust mineralogy on the aerosol chemical aging process, and thus influence (a) the deposition solubility of the mineral nutrients contained within dust particles and (b) the uptake rate of dust in clouds and subsequent loss via wet deposition processes.
4. More focus on the integrated spatiotemporal response of marine ecosystems and nutrient availability to changes in aerosol nutrient deposition.
5. Field campaigns, laboratory studies, and mesocosm experiments designed to characterize aerosol properties and the response of the SML and marine ecosystems to their deposition.

The battle: Climate change, pollution, and future ocean health

The modern relationship between aerosols, marine biogeochemical cycles, and ocean health cannot be understood without considering the context of climate change and human pollution. Covering more than 70% of the Earth's surface, the ocean has absorbed around 25% of the human-induced carbon dioxide emissions (Gruber et al., 2019). Both Earth System model projections (Fu et al., 2016) and global observations (Li et al., 2020; Sallée et al., 2021) show that the ocean is becoming increasingly stratified under climate warming, leading to a reduction in vertical nutrient flux to surface phytoplankton (Fu et al., 2016) and a potential shift toward smaller sized phytoplankton groups. While smaller particles, in general, sink more slowly into the deep ocean (Steinacher et al., 2010), small-sized phytoplankton, that is, picophytoplankton, are now understood to contribute substantially to carbon export and may be able to adapt to climate change driven decreases in ocean mixed layer depths (Lomas et al., 2022). The quantification of carbon export efficiency therefore depends not only on the knowledge of the biomass accumulation due to aerosol deposition but also the rate of carbon export across different phytoplankton populations.

The future pattern of nutrient aerosol deposition contains many uncertainties: in the impacts of climate mitigation strategies; how population, consumerism, and economics will drive manufacturing and agriculture practices; how aridification will impact dust production; and how wildfire activity will change in a warmer world with changing precipitation patterns. Here, we use wildfire as an example of the complex interconnection between climate change and nutrient aerosol supply. Despite a global 25% decrease in burned area over the past 2 decades (Andela et al., 2017), the occurrence and severity of fire is rapidly changing on every vegetated continent with large forest fire emissions increasing at mid-to-high latitudes (McCarty et al., 2021; Zheng et al., 2021). The speed of fire activity and emission change is linked to the level of climate change, and even under low warming projections fire can be expected to increase in the extra-tropics, impacting Earth's ecosystems and societies. In the Northern Hemisphere, the Boreal Forest is among the most vulnerable to global warming in the coming decades (McCarty et al., 2021). Rapid warming is increasing Boreal Forest fires and thawing the permafrost, exposing large quantities of peat, a type of soil composed mainly of decomposed plant material. This peat contains vast nitrogen stocks stored away for a long time—the same nutrient limiting Arctic Ocean phytoplankton growth (Willis et al., n.d.). Ardyna et al. (2022) suggested a link between Arctic warming, permafrost thawing, increased atmospheric nitrogen release from increased forest and peat fires, and Arctic marine ecosystems (via relieving phytoplankton nutrient stress due to increased nitrogen deposition from fire). The effects of peat burning on Arctic ecosystems will likely be even more pronounced if model predictions about reductions in nitrogen deposition in the Arctic relative to phosphorus deposition come to fruition (**Figure 3a**). In the Southern Hemisphere, a region with a high potential for increased future fire activity is Australia. A recent catastrophic megafire burned around 7 Mha in Southeastern Australia between September 2019 and March 2020 (Filkov et al., 2020). By consuming at least 21% of the country's temperate and broadleaf forest, this megafire produced massive aerosol laden smoke plumes that were transported toward the southern Pacific Ocean. The aerosol deposited to the ocean surface here induced a widespread phytoplankton bloom in these iron-limited waters (Tang et al., 2021). Understanding such complex multidimensional interactions between fire events and the Earth System, including both polar oceans experiencing rapid environmental change (Willis et al., n.d.), requires expertise from across the geosciences and social sciences to fully appreciate and identify the main challenges that need to be addressed (Shuman et al., 2022). However, the future impacts of fires on the Earth System remain poorly constrained due to a lack of communication between relevant fields of expertise.

In considering how patterns of ocean nutrient limitation may change in response to future changes in aerosol nutrient fluxes, it is insightful to investigate the changes in the magnitude of aerosol fluxes in concert with the changes in the ratio of elements, both nutrient (**Figure 3**) and pollutant. A multielement approach achieves a more comprehensive understanding of the complex interactions

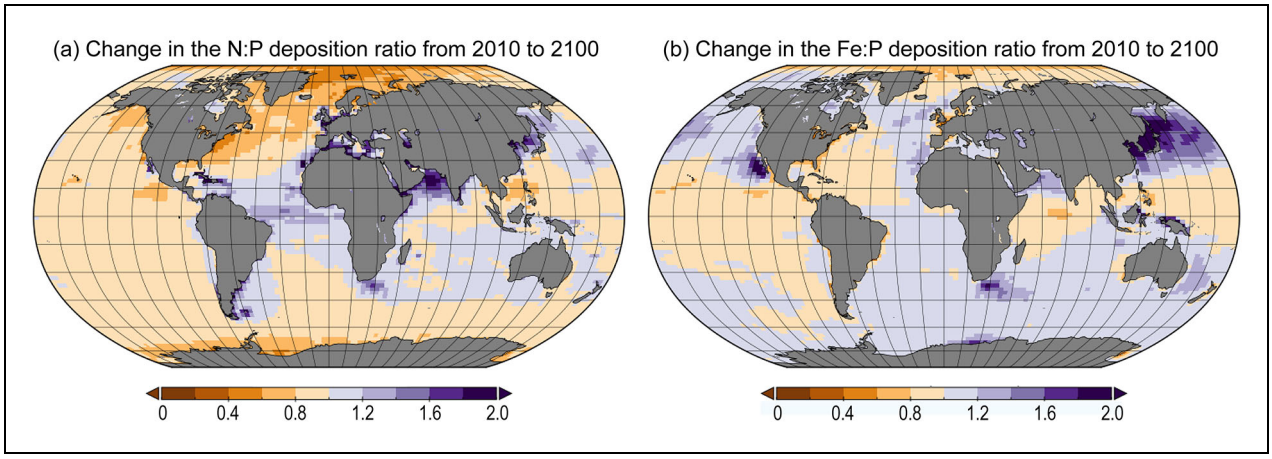

Figure 3. Modeled fractional change in the total (wet + dry) deposition ratio over this century (2010–2100) for (a) nitrogen (N) to phosphorus (P) and (b) iron (Fe) to phosphorus (P). A ratio value >1 suggests that, with all other considerations remaining equal, nutrient limitation pressure moves toward phosphorus. A ratio value <1 suggests that, with all other considerations remaining equal, nutrient limitation pressure moves toward either nitrogen (a) or iron (b). Model results from Myriokefalitakis et al. (2020), see also **Figure 2** caption. Only 21st century emission changes are considered in these simulations, that is, the additional uncertainty due to climate change on transport pathways or the dissolution rate (for phosphors and iron) is omitted.

between marine ecosystems and atmospheric inputs (BOX). This is especially important given how human activities alter atmospheric composition, and thus element ratios, in different ways in different regions. Focusing on the North Pacific region in **Figure 3** helps highlight some of the complexity and challenges to forecasting potential impacts of project changes in nutrient aerosol stoichiometry. The North Pacific can be divided into 3 subregions: the eastern, central, and western Pacific. Each region is likely to be sensitive to changes in aerosol nutrient deposition in differing ways. The western North Pacific is located directly downwind of the industrially active East Asian region with significant and increasing iron emissions. In this region, the changes in nitrogen deposition alter the level of surplus nitrogen available in the surface water, providing a link between anthropogenic activity that emits most of the nitrogen to the atmosphere (Jickells et al., 2017) and the level of primary productivity in downwind ocean regions (Taketani et al., 2018; Zhang et al., 2019a). However, many Asian countries are rapidly working to reduce anthropogenic emissions due to strong air quality control policies (Uno et al., 2020). The level of mitigation achieved therefore introduces uncertainty in projections, for example, successful policy could result in reduction in nitrogen deposition similar to the North Atlantic. The central Pacific region is dominated by the North Pacific Gyre that oscillates between iron and phosphorus limitation, due mainly to changes in dust deposition (Letelier et al., 2019). The combination of lower nitrogen and higher iron deposition across the central North Pacific suggests that this region may move toward a more sustained phosphorus limitation pattern in the future (Kim et al., 2011; Kim et al., 2014). The changes in pollution emissions once again are important to consider because of their secondary impacts on nutrient deposition. Reductions in SO_2 and NO_x (e.g., from China) are not matched by NH_3 emission reductions, which are much more difficult to control. This may increase the pH of aerosol/cloud/fog water and lead to changes in nutrient transport, deposition, and solubility (Baker et al., 2021). The eastern North Pacific region is influenced by westerly winds transporting Asian dust (often mixed with anthropogenic aerosol and gases) as well as easterly dust-anthropogenic aerosols from the U.S. coast. This region is also seasonally influenced by a strong deposition contribution from wildfires that are predicted to increase in the future.

The Southern Ocean is another important ocean region in which to consider future changes to aerosol deposition and related impacts on biogeochemical cycles. For example, the manganese/iron elemental ratio in dust is generally lower than the manganese/iron nutrient ratio requirement in polar phytoplankton. This suggests that, with all other factors constant, manganese limitation in the Southern Ocean increases with increasing dust deposition, a relationship that has been shown for the last glacial maximum where dust fluxes to the ocean were several-fold greater than today (Hawco et al., 2022). The same relationship can be hypothesized to also be true for other aerosol sources, where the ratio of manganese/iron in aerosol is lower than the required ratio or similar to that in dust, such as in fire and anthropogenic aerosol (Mahowald et al., 2018). However, the changes in the Southern Ocean deposition flux of soluble iron from dust, wildfire, and anthropogenic sources by the end of this century are uncertain. One Earth System model study projected increasing deposition fluxes across all sources (Hamilton et al., 2020a), while another study projected similar or less dust deposition but similar or more fires and anthropogenic deposition (Bergas-Massó et al., 2023). These differences depend not only differences on how aerosol processes are represented across different climate models but also on the level of climate mitigation

achieved, its impact on aerosol emissions, and the socioeconomic pathway humanity follows (Bergas-Massó et al., 2023).

Substances previously designated as nutrients for marine biota are, conversely, often designated as pollutants in terms of human health. As such, they can become a subject of concern for air quality legislation, either directly or indirectly by policy aimed at lowering fossil fuel combustion from which nutrient aerosol emissions can be considered a by-product. Over recent decades, air quality legislation has reduced the total mass of emissions from fossil fuel burning, but abatement methods for larger particles are more effective and cheaper than for smaller particles, for example, cyclone filters versus bag filters (Klimont et al., 2002); resulting in their preferential use and shifting the mean aerosol iron particle size distribution to smaller diameters. Fine-sized aerosol particles have longer atmospheric lifetimes than coarse-sized aerosol particles, partly due to them being lofted higher into the atmosphere more readily. For the case of anthropogenic iron aerosol, modeling suggests that this trend toward relatively more fine-sized aerosol particles has increased the total anthropogenic aerosol lifetime by approximately 40% over the past 4 decades (Hamilton et al., 2020b). Increased long-range aerosol transport can increase both the magnitude of total deposited iron to remote marine regions and its fractional solubility (Longo et al., 2016). The impact of air pollution legislation, and resulting mitigation methods, on the magnitude of aerosol nutrients delivered to the ocean highlights the need to consider the interconnectedness between policies aimed at improving human health and their potential indirect effects on ocean health, emphasizing the importance of a holistic transdisciplinary approach to this subject.

While pollution can increase nutrient supply to the oceans, not all aerosol deposited in the ocean are beneficial to phytoplankton. The deposition of certain metals, such as copper (Jordi et al., 2012), cadmium (Hindarti and Larasati, 2019), and mercury (Kershaw and Hall, 2019; Zheng et al., 2019), can be toxic in large amounts. These toxic metals negatively impact the growth and productivity of marine species because they interfere with the normal functioning of essential cellular processes, including photosynthesis and respiration (Mahowald et al., 2018). Different phytoplankton may however exhibit varying sensitivities to aerosol-borne trace metals (Hamilton et al., 2022) and understanding the bioavailability of toxic substances is likely as important as understanding bioavailability for nutrients. While some of the same approaches can be used, this will likely require advancements in analytical techniques, experimental approaches, molecular tools, and interdisciplinary collaborations. Understanding species-specific responses to metal exposure, including variations in toxic metal tolerance and the potential for physiological adaptation, is also essential. The toxicity of certain metals such as copper is also affected by its local environment, including the availability of other elements and ocean pH. For example, the presence of iron can reduce copper toxicity (Yang et al., 2019), while future ocean acidification may enhance copper toxicity for certain marine biota (Yang et al., 2019; Cao et al., 2022). Investigating how future changes in toxic aerosol deposition relates to physiological responses, growth inhibition, cellular damage, and oxidative stress induced by metal exposure is needed and provides insight into the potential for wider impacts on phytoplankton community composition, species succession, and ecosystem dynamics. By addressing these research topics, we can enhance our knowledge of the ecological consequences of future human activity and help inform effective management and conservation strategies for marine ecosystems in the coming decades.

An additional anthropogenic aerosol type is microplastics. Microplastics have been identified in riverine systems discharging to the ocean (Liu et al., 2021) and the atmosphere has also recently been identified to be a significant vector for the transport of primary and resuspended microplastics to the ocean (Brahney et al., 2021). The magnitude and mechanisms of microplastic transport are very poorly understood at present (Allen et al., 2022). Atmospheric transport is likely an important pathway for the accumulation of microplastics in remote areas (Zhang et al., 2019b), including the world's ocean. The study of microplastics is complicated by the fact that "microplastics" refers to a vast array of diverse particle types, defined by different chemical compositions and physical structures (Rochman et al., 2019). The physicochemical properties of biogeochemically relevant plastics need classifying and assessing in terms of the risk that different classes pose to ocean health. Traditionally, airborne transport is considered as the physical transport of gases and particles through the atmosphere following prevailing wind flows. But species with both terrestrial and marine habits, notably seabirds, likely play an interesting role in microplastic transport through the atmosphere. Many seabird species cover vast stretches of ocean habitat as central-place foragers regularly return to land to roost and reproduce or wander the ocean for months in between breeding cycles. Seabird ingestion of plastics invariably results in aerial transport of up to 10,000 miles (Clay et al., 2023) from initial point of contact. Macro- and microplastics are increasingly found in seabird digestive systems, and plastic ingestion is predicted to occur in 99% of seabird species by 2050 (Wilcox et al., 2015). The addition of a biologically mediated microplastic transport method presents an opportunity for interdisciplinary research between the physical aerosol and ecological communities. Regardless of aerial transportation route, microplastics are now a ubiquitous sedimentary signature of the Anthropocene (Zalasiewicz et al., 2016) and their impacts on biogeochemical cycles, primary productivity, and the marine food web are still debated and in need of further study.

Once deposited into the ocean, plastic debris—including microplastics—can be buoyant, neutrally buoyant, or sinkable, which allows it to freely move throughout the water column, encountering numerous marine species and environments (Cole et al., 2011). Microplastics could modify biogeochemical cycles through metals adsorbing onto them (Zhang et al., 2020), potentially altering

residence times and the chemistry they can undergo. There is growing concern about the environmental implications of microplastics in marine environments, including that nonplastic-based organic contaminants that have adhered to plastic particles may have long-term environmental health impacts within ocean food webs, potentially extending upward to humans through the processes of bioaccumulation (Andrady, 2011); this is an area of active research and debate (Smith et al., 2018; Susanti et al., 2020). The small size of microplastics makes them available in different trophic levels leaving marine biota vulnerable to exposure (Wright et al., 2013). It is estimated that 49% of fish exhibit microplastic ingestion (Wootton et al., 2021), and microplastic ingestion by larval- (Gove et al., 2019) and adult-form fish (Jovanović, 2017) has also been observed.

While discussion has focused on aerosol deposition to the ocean and the response of phytoplankton, increased marine productivity can also lead to increased aerosol emission back to the atmosphere. This marine aerosol source and its interactions with climate are reviewed in depth by Sellegri et al. (n.d.), as part of this SOLAS special issue. Beyond their climate impacts, some marine emissions may also pose a threat to ecosystems or human populations (Sharoni et al., 2015; Abdullah et al., 2022; Sha et al., 2022; Tong et al., 2023) because these aerosols can include a range of pollutants and irritants, such as bioaerosols, allergens, pathogens, or toxins. The risk of toxic aerosol emissions is increasing, underscoring the importance of further research in this area in collaboration with biologists, public health experts, and social scientists. By studying harmful aerosol dispersal and deposition, we can better predict the impact on ecosystems, society (such as food security and public health), and the economy.

One final research connection to encourage is between this community and those working on the topic of carbon dioxide removal for climate mitigation, also called negative emissions. Due to the ocean's high carbon storage capacity, it has the potential to help meet global climate goals by promoting methods to enhance carbon uptake and its longer term sequestration (Scott-Buechler and Greene, 2019; National Academies of Sciences and Medicine, 2022). Artificial ocean fertilization (often via the addition of iron to stimulate phytoplankton growth) is one of the currently considered marine carbon dioxide removal (mCDR) methods. For more details on this mCDR method and others, see the reviews by Bach et al. (n.d.) and Johnson et al. (n.d.) as part of this special issue. Part of the interest of using iron to stimulate the biological carbon pump is due to the large effort already spent in undertaking artificial ocean iron fertilization experiments. The 13 past deliberate iron fertilization experiments showed that phytoplankton biomass accumulation is possible following artificial iron addition (Boyd et al., 2007). However, there is debate around the strength of response and any resultant increase in carbon export to deep ocean layers, although the European Iron Fertilization Experiment (EIFEX) conducted in the Southern Ocean (Smetacek et al., 2012) suggested its potential. At least 3 conditions have been identified as being necessary to detect the carbon export response to aerosol addition (Yoon et al., 2018): (1) changes in the size of phytoplankton community from pico- and nanophytoplankton to microphytoplankton, shifting communities toward a diatom-dominated bloom; (2) low rates of bacterial remineralization and grazing pressure; and (3) a sufficient experimental duration, enabling monitoring for both immediate and delayed response to iron addition. EIFEX was accompanied by aggregation of diatoms and their subsequent rapid sinking, suggesting future fertilization methods to derive a significant carbon export aim to repeat such a response (Smetacek et al., 2012). An important concern in mCDR is that side effects absolutely must be considered; offsets must be compared against the benefits (Williamson et al., 2012). Therefore, an approach to transparent and robust monitoring, reporting, and verification (MRV) on the benefits and risk of ocean fertilization is needed to assess its feasibility as a carbon removal method. Exploring ocean biogeochemical and ecological response to aerosol deposition under natural or anthropogenic aerosol fertilization events, for example, the science herein, can provide comprehensive data sets (e.g., modeling, satellite observations, and field experiments) and the scientific knowledge to help address challenges associated with the implementation of MRV, informing the future of artificial ocean fertilization as a viable (or not) method of mCDR.

Research Directions:

1. Assessing bioavailability and toxicity of biogeochemical significant aerosols under different atmospheric and oceanic conditions.
2. The chemical speciation and complexation of nutrient and toxic metals with other substances, such as organic ligands and plastics, that affect residence times in seawater as well as interactions with phytoplankton cells.
3. How the uptake of metals in biota and subsequent accumulation through the food chain affects the toxicity of other organisms needs more understanding.
4. Assessing plastic particle properties and classifying their impacts on ocean health.
5. Assessing the impact of potentially harmful aerosols on ecosystems and society, either due to their inherent toxicity or as vectors of pathogens and other pollutants.
6. Increased data and knowledge sharing between modelers and observationalists, for example, those working on artificial iron addition to improve MRV and mCDR or on human health via changes in air quality.

Navigation tools: Looking ahead

Recent and upcoming developments in satellite remote sensing, marine autonomous platforms, and nutrient

aerosol modeling provide optimism for advancing understanding of the aerosol-ocean biogeochemistry connection in the coming decades. Here, we review some of the most promising of these new tools for the community to use either now or in the short-term future. Due to the large effort needed to keep track of the ever-expanding number of satellites, we have also summarized many of the current and upcoming satellite platform capabilities in **Table 1**. We note however that missing in this description are comments on the current private satellite deployment initiatives and a myriad of satellite sensors already deployed by a number of countries (China, Russia, India, and others). While as a group they represent a very exciting and alternative source of information, we purposely omitted their description because at the time of this writeup, there are too many unknowns regarding data production, availability, and accessibility to make the usage of such datasets practical to the scientific community.

Newer data from recently deployed satellite platforms remain to be fully explored from the viewpoint of aerosol and ocean biology detection. The current generation of geostationary sensors has now, compared to the previous generation of sensors, more spectral bands and higher spatial resolution making them suitable for monitoring aerosol transport at subhourly resolution. Currently available products follow the heritage of those existing from polar sensors (Levy et al., 2013; Kondragunta et al., 2019; Yoshida et al., 2021). Among the ocean products measured by geostationary satellites, the satellite Geostationary Ocean Color Imager (GOCI), centered over the Korean Peninsula, is the first operational sensor with spectral channels (8 bands; 412–865 nm range) optimized for ocean biology observations. A more advanced geostationary sensor is planned for monitoring coastal areas north of the Equator in the Americas (the Geostationary Littoral Imaging and Monitoring Radiometer, 2023). While these sensors are adequate for aerosol and ocean observations, they are limited in the number of products they offer, and the coverage over high latitudes is poor (poleward of $\sim 55^{\circ}$). Another instrument to highlight is the currently deployed Tropospheric Ozone Monitoring Instrument (TROPOMI; Veefkind et al., 2012). This sensor is novel in that it provides hyperspectral observations from the ultraviolet (UV) to the near-infrared (NIR) range at an unprecedented spatial resolution (for UV sensors), making it unique in its class. While the spectral coverage offers exciting possibilities to explore new aerosol and ocean biology products, such potential is not realized yet as the only products currently available from this sensor are atmospheric aerosol and cloud properties from modified heritage algorithms (Loyola et al., 2018; Torres et al., 2020). Ocean biology signals of interest are present in the UV range (Werdell et al., 2018), but there are no official ocean products from this sensor. Finally, spaceborne lidar satellites normally used for atmospheric applications are also suitable for subsurface remote sensing of intensive optical properties (Behrenfeld et al., 2013; Hostetler et al., 2018) and can provide a plethora of new information (Behrenfeld et al., 2017), including simultaneous atmospheric and ocean observations during day and nighttime. However, as of today, no study has used these observations in aerosol deposition-ocean biology analyses nor standard products are available from data providers.

Of the upcoming satellite technologies soon to be deployed, there are number of encouraging developments. For the sake of space, we highlight one mission because it will be specifically designed to study the ocean and the atmosphere with new and improved sensors, and it will produce relevant products. The Plankton, Aerosol, Cloud, ocean Ecosystem (PACE, 2023) Mission is a multinational effort led by NASA to provide a complete suite of atmospheric and ocean observations using a variety of active and passive technologies. It is scheduled to be launched by early 2024. The main sensor in the satellite is the Ocean Color Instrument (OCI), and the 2 secondary payloads are the Hyper-Angular Rainbow Polarimeter 2 (HARP2) and the Spectro-Polarimeter for Exploration (SPEXone) polarimeters (Gorman et al., 2019). OCI is a hyperspectral sensor from the UV to NIR ranges and a nadir pixel resolution of 1 km. Atmospheric and ocean products from OCI will include heritage products developed from NASA's Earth Observing System (EOS) era sensors such as MODIS and OMI and a new cadre of products (PACE, 2023). Among the latter, new phytoplankton community composition and pigment concentration products will enable the identification of specific types of phytoplankton populations. As different plankton communities are expected to react differently to aerosol deposition property differences, this information may prove helpful. For example, to better understand why some ecosystems do not seem to show a noticeable change (as observed by current satellite products) upon a known aerosol deposition event to the ocean. New atmospheric products include proxies for aerosol types (aerosol shape, complex refractive index, and spectral single scattering albedo), improving current techniques by providing more information on the aerosol composition reaching marine ecosystems. One caveat is that many of these new products are provisional at this point, and their quality will not be ascertained until the mission is in orbit and validation campaigns are carried out.

One important challenge for the user is to decide whether to work with satellite data online or directly downloading data locally. Embracing cloud services (a trend already initiated by NASA and ESA) will enable more convenient data access and will reach a broader range of researchers. The data access interface should provide preview functionality to assist users in evaluating the data set for their purposes prior to download.

Another set of observational tools that can provide new perspectives on the aerosol–ocean–biology relationship are marine autonomous platforms, which include profiling floats, unmanned surface vehicles (USVs), and gliders (Chai et al., 2020). Each platform type is characterized by different time and spatial coverage and can carry different sensors. USVs, such as Saildrones or WaveGliders, are naturally the platforms that can more easily be adapted to studying aerosol above the ocean. USV sensors can measure the properties of the air layer above the ocean, air-sea

Table 1. Satellite sensors used in the study of aerosol and marine biogeochemical cycles

Platform/Sensor and Operation Period		Spatial Coverage and Observation Frequency	Vertical Resolution	Sensor Technology	Key Products	Key Property Proxy (Atmosphere and/or Ocean)	Research Product Retrieved	Refs (#) and Website
Terra MODIS-MISR Aqua-MODIS	1999–2022+	Daily global coverage	Column integrated	Visible to Near Infrared Multiple-broad bands Single angle (MODIS) Multiple angle (MISR)	Standard atmospheric composition and ocean biology products MISR reports atmospheric particle shape	Atmosphere: Aerosol mass concentration and dominant particle size Ocean: Surface chlorophyll, particle organic, and inorganic carbon	Aerosol refractive index and shape Phytoplankton functional types	1, 2, 3, 4, 5, a
ESR1-2/ATSR Envisat/AATSR	1995–2012	Daily global coverage	Column integrated	Selected in visible, near infrared, and infrared Dual camera broad band	AOD and fine mode fraction	Atmosphere: Aerosol mass concentration and dominant particle size		6, b
IASI	2007–2015	Daily global coverage	Vertically resolved	Selected in infrared Passive broad band	Dust optical depth	Atmosphere: Aerosol mass concentration for detected dust		7, c
PACE OCI	2024+	Daily global coverage	Column integrated	Passive hyperspectral	Standard ocean biology products	Ocean: Standard ocean biology products. Surface chlorophyll, particle organic, and inorganic carbon	Standard atmospheric composition products over land and ocean	8, 11, 12, 16, d
PACE HARP2	2024+	Bidaily global coverage	Column integrated	Passive multiangle polarization	Standard atmospheric composition	Atmosphere: Standard atmospheric composition	Aerosol layer height, aerosol shape, single scattering albedo, size distribution, and dominant aerosol composition Ocean roughness and ocean refractive index	9, 10, 11, 13, d
PACE SPEXone	2024+	Approx. monthly (every 30 days) global coverage	Column integrated	Hyperspectral	Standard atmospheric composition	Atmosphere: Standard atmospheric composition	Aerosol layer height, aerosol shape, single scattering albedo, size distribution, spectral aerosol single scattering albedo, and dominant aerosol composition	9, 10, 11, 13, d
Sentinel3 A/B/C-OLCI	2016–	Daily global coverage	Column integrated	Visible to near infrared Multiple-broad bands	Standard ocean biology products	Ocean: Standard ocean biology products		21, 22, 25, e, f

(continued)

Table 1. *(continued)*

Platform/Sensor and Operation Period		Spatial Coverage and Observation Frequency	Vertical Resolution	Sensor Technology	Key Products	Key Property Proxy (Atmosphere and/or Ocean)	Research Product Retrieved	Refs (#) and Website
Envisat MERIS	2002–2012	Bidaily global coverage	Column integrated	Visible to near infrared 15 Broad bands	Standard atmospheric composition and ocean biology products	Atmosphere: Standard atmospheric composition Ocean: Standard ocean biology products		24, 26, g
Seastar SeaWifs	1997–2010	Daily global coverage	Column integrated	Visible to near infrared 8 Broad bands	Standard ocean biology products	Ocean: Standard ocean biology products	Atmosphere aerosol retrieval of 550-nm AOD and fine mode fraction	27, 28, h
CALIPSO CALIOP	2004–2022	1/1–2 weeks depending on latitude, global coverage	Vertically resolved	Atmospheric lidar polarization 1 channel	Aerosol backscattering, depolarization, and spectral dependance	Atmosphere: Aerosol concentration, sphericity, and particle size	Subocean surface biological and nonbiological particle properties and concentration	14, 15, 17, 18, 19, i, j
EarthCare ALID	2023–	Monthly global coverage 1 overpass/25 days	Vertically resolved	Atmospheric lidar polarization 1 channel	Vertical profiles of aerosol extinction, boundary layer height, and aerosol type	Atmosphere: Aerosol concentration, sphericity, and particle size. Aerosol type	N/A	29, k
EarthCare MSI	2023–	Monthly global coverage 1 overpass/25 days	Column integrated	2× visible, 2× near infrared, and 3× infrared 8 Band medium resolution spectrometer	Standard atmospheric composition	Atmosphere: Standard atmospheric composition	N/A	29, 30, k
COMS GOCI	2010–2021	South Korea regional coverage 8/day	Column integrated	Multiple-broad band	Standard ocean biology products	Ocean: Chlorophyll concentration, the optical diffuse attenuation coefficients, the concentration of dissolved organic material or yellow substance, and the concentration of suspended particles	Aerosol retrievals using ML (research product)	20, 23, l, m

ABI GOES16-17-18	2016–	North and South America regional coverage 6/hour (operational) 60/hours (on demand)	Column integrated	16 Multiple-broad band	AOD and aerosol detection product	Atmosphere: Atmospheric aerosol mass concentration and distinction between smoke and dust	N/A	31, n
GLIMR	2026+	Only North American coastline coverage 1/hour min	Column integrated	Hyperspectral band	No atmospheric products. No list published but legacy products expected	No list published but legacy products expected	TBD	o
MetOP-SG/3MI	2025+	Daily global coverage	Column integrated	Passive multiangle polarization, broad band, heritage on POLDER	Standard atmospheric composition	Atmosphere: Atmospheric aerosol mass concentration and dominant size, plus improved distinctions between aerosol types through sensitivity to particle shape and refractive index	Standard ocean color products are listed as secondary products. Likely to be based on algorithms previously developed for POLDER	32, 33, p, q
PARASOL POLDER	2004–2013	Daily global coverage	Column integrated	Passive, multiangle polarization	AOD and fine mode fraction	Aerosol size, complex refractive index, fraction of non-sphericity, and scale height		34,35, r

Vertical resolution is either column integrated or vertically resolved. Column integrated products use passive sensors to measure the total radiance originating from the atmosphere and surface. Vertically resolved products use both passive sensors and active sensors with lidar pulses to sample discreet vertical aspects of the atmosphere and ocean. We only report IASI for passive as it has the longest time series. Visible (VIS) to near infrared (NIR) band range refers to a sensor with channels typically ranging from 400 to 2,100 nm. Research products are where different research groups have algorithms that create these products but are not currently available by satellite data providers. Standard Atmospheric Composition Products typically refers aerosol optical depth at one or more wavelength and a measure of dominant aerosol size in the pixel. IASI = Infrared atmospheric sounding interferometer; GOCI = geostationary ocean color imager; GLIMR = Geostationary Littoral Imaging and Monitoring Radiometer; HARP2 = Hyper-Angular Rainbow Polarimeter 2; SPEXone = Spectro-Polarimeter for Exploration.

Web (Data) Portals:

a. https://www.earthdata.nasa.gov/

b. https://climate.esa.int/en/projects/aerosol/data/

c. https://climate.esa.int/en/projects/aerosol/data/ (data currently only available on request)

d. https://pace.oceansciences.org

e. https://sentinels.copernicus.eu/web/sentinel/user-guides/sentinel-3-olci

f. https://sentinels.copernicus.eu/web/sentinel/sentinel-data-access

g. https://earth.esa.int/eogateway/instruments/meris

h. https://oceancolor.gsfc.nasa.gov/about/missions/seawifs/

i. https://www.icare.univ-lille.fr/calipso/

(continued)

Table 1. *(continued)*

j. https://asdc.larc.nasa.gov/project/CALIPSO

k. https://earth.esa.int/eogateway/missions/earthcare

l. http://kosc.kiost.ac.kr/index.nm?menuCd=43

m. https://oceancolor.gsfc.nasa.gov/about/missions/goci/

n. https://registry.opendata.aws/noaa-goes/

o. https://eos.unh.edu/glimr

p. https://www.eumetsat.int/eps-sg-3mi

q. https://www.eumetsat.int/metop-sg

r. https://www.icare.univ-lille.fr/parasol/

All URLs listed under "a–q" were last accessed and found active on August 15, 2023.

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fluxes, and a few meters below the water surface. Their main advantage is spending all their time at the surface, where optical sensors can measure changes in atmospheric properties, such as AOD. On the other hand, the wave-induced movement of USV hinders the use of photometers of the type deployed on land. Even when using a mobile sensor, pointing toward the sun can be highly challenging for a sensor that needs to work within the constrained energy and space of a USV. This hindrance could be overcome with recent advances in instrumentation and optical modeling, which show that zenith sky radiometers can be used to measure AOD, achieving accuracies similar to the ones from AERONET sensors (Almansa et al., 2017; Almansa et al., 2020). Compared to sun photometers, zenith radiometers are more robust and without moving parts, hence more adapted to marine conditions (Leymarie et al., 2018). Although the wave-induced movement is an obvious problem, the addition of existing compass and accelerometer sensors on the USV can be used to determine the radiometer's measurement angle with respect to the zenith. The combination of the different data sources allows using only the zenith-looking measurement or, for better performance, using all the data to retrieve AOD thanks to advanced atmospheric optical models (e.g., GRASP; Dubovik et al., 2014). Zenith-looking radiometers could also be deployed in profiling floats, such as Argo. The latest models of biogeochemical-Argo floats (BGC-Argo) are equipped with an oxygen sensor positioned at the top of the float that samples both below and above the water (Bushinsky et al., 2016). A similar setup for a radiometer could be used to measure the optical properties of the atmosphere. Although the setup and validation of this approach still need to be tested, incorporating atmospheric measurement in the Argo program has the potential to provide aerosol measurements in step with ocean biogeochemistry measurements.

Profiling floats, and BGC-Argo in particular, hold an important advantage with respect to USV as they can measure the full impact of aerosol deposition on the transformation and vertical export of organic carbon. Floats can measure the changes in phytoplankton accumulation, primary production, and particulate/dissolved carbon from the surface to 1,000–1,500 m deep. Besides, floats are quasi-Lagrangian platforms that naturally follow and monitor the changes in the water mass enriched with aerosols. Gliders provide a mixed option between USV and profiling float, with high-resolution measurements in a particular area, but they cannot be used to create a global network of sentinels, as is the case for floats. While satellite technology is making progress in capturing the variability of phytoplankton below the ocean surface, the use of marine autonomous platforms for aerosol-related measurements above the ocean surface remains unexplored.

Despite these quickly developing technologies showing much promise for the study of the ocean–aerosol link, they cannot measure aerosol deposition from the atmospere to the ocean surface or diagnose many of the complex aerosol properties and processes, which ultimately determine the bioaccessibility of those nutrients being deposited to marine biota. This is one area where aerosol and Earth System modeling can help to bridge the gap.

Recent developments in the state-of-the-art chemistry-transport models (CTMs) have enabled the simulation of the lifecycle of soluble aerosol nutrients, such as phosphorus and iron. These models have been successful in reproducing the main features of present-day soluble nutrient concentrations compared to in situ observations (Myriokefalitakis et al., 2016; Herbert et al., 2018; Myriokefalitakis et al., 2018; Ito et al., 2020a), improving our understanding of the relative contribution of acidic and organic dissolution nutrient release from atmospheric processing compared to source inherited solubility for a wide range of nutrient aerosol sources. CTMs have also highlighted a strong relationship between air quality and dissolved nutrient deposition fluxes to the oceans (Ito, 2013; Ito et al., 2019). CTMs do not, however, couple changes in the atmosphere's energy balance with meteorology and climate, such as changes to winds, precipitation, and temperature. Therefore, major nutrient emissions sources such as desert dust and wildfires are insensitive to climate change and any related land cover change. As a result, the estimates of past and future nutrient deposition into the ocean are often highly unclear.

Many biogeochemistry model studies rely mainly on dust deposition mass fluxes to estimate the atmospheric input of iron and phosphorus, assuming constant fractions in the nutrient content, 3.5% for iron and 750–800 ppm for phosphate, and its fractional solubility at around 1% (e.g., Aumont et al., 2015). However, neglecting the importance of nutrient-containing combustion aerosol inputs to the global ocean, which can be 50% or more of the total input for important in remote areas such as the Southern Ocean (Barkley et al., 2019; Tang et al., 2021; Liu et al., 2022; Ito and Miyakawa, 2023), biases the spatial deposition pattern toward the Northern Hemisphere dominated dust sources. For the case of iron, increasing biogeochemical cycling modeling complexity to reflect multiple iron sources leads to improved comparison to observations than simpler representations (Tagliabue et al., 2016). On the other hand, simplifications in marine biogeochemistry may lead to less reliable estimates of productivity in the deposition regions, such as when applying a constant Redfield ratio (C:N:P:Fe = 122:16:1:0.1-to-0.001), estimating carbon export efficiency (g CO_2 exported per gram of nutrient aerosol deposited) which is potentially sensitive to aerosol source (Hamilton et al., 2020a; Ito et al., 2020b), or the redistribution of nutrients to lower latitudes (Tagliabue et al., 2009). The most integrated atmosphere-ocean model experiments to date have mainly relied on driving ocean biogeochemical models with complex atmospheric input files that were derived offline (Krishnamurthy et al., 2009; Hamilton et al., 2020a; Myriokefalitakis et al., 2020; Hamilton et al., 2022); these studies indicate that productivity is sensitive to changes in the atmospheric supply of nutrients, with different rates of change depending on region; from a few percentages at the global scale (<1%–3%) to slightly higher within high nutrient low chlorophyll regions (<1%–5%), reaching a potential

maximum of up to tens of percentage within selected subtropical gyre regions (10%–20%). Such simplifications call for more intricate interactions between atmosphere and ocean model components.

As stated above, acidity is a key factor in determining the bioavailability of aerosol nutrients since it is assumed that they must be primarily in a soluble (bioaccessible) form in order to be utilized by marine biota. In global models, acidity is still a significant source of uncertainty, particularly in oceanic regions. Global models reveal significant differences for both ammonium and nitrate over oceanic basins, as Nault et al. (2021) recently pointed out. The process of aerosol dissolution during their long-range transport in the marine boundary layer may be adversely affected by uncertainties in oceanic ammonia sources (i.e., too high reduced nitrogen emissions; Paulot et al., 2015). Yet, the bioavailability of aerosols and, consequently, their significance for ocean biogeochemistry in Earth System models may also depend on the assumptions made about microphysics and chemical speciation. The state-of-the-art global models typically estimate the acidity of fine aerosols as internal mixed with submicron sea salt aerosols, resulting in unavoidably higher aerosol pH values due to the presence of high sodium concentrations in the marine environment. These discrepancies in aerosol pH, along with other uncertainties typically observed in Earth System models (such as temperature, cloud liquid water content, and relative humidity), may have an impact on the rate of nutrient deposition over the ocean, particularly nitrogen, as they have a significant impact on the gas/particle equilibrium of semi-volatile compounds, such as nitrate and ammonium.

Earth System models have recently, nevertheless, been able to couple sophisticated descriptions of the atmospheric nutrient inputs to the global ocean based on explicit parameterizations of their primary sources (i.e., anthropogenic or natural emissions) and the secondary source from dissolution of insoluble minerals during transport (Hamilton et al., 2019; Hajima et al., 2020; Myriokefalitakis et al., 2021). Detailed transient simulations within the Coupled Model Intercomparison Project (CMIP) framework have not yet been undertaken but are ready for the next round of experiments that are proposed to include a more interactive Earth System modeling approach. One advantage of explicitly accounting for dynamic nutrient cycling is that biogeochemistry disturbances affecting marine productivity over the coming centuries can be predicted with greater detail than before. Concurrently, the changes in marine primary productivity can feedback on climate by biota producing those aerosol and precursor gases that modify the atmospheric energy budget through scattering or absorbing radiation or modifying cloud processes (Charlson et al., 1987). Therefore, a combined aerosol-biogeochemistry-climate modeling framework can now be used to help address long-standing questions regarding the influence of atmospheric composition on marine productivity and climate.

One practical hurdle to improving modeling of the long-range transport of aerosols over remote ocean environments is how to best use the current suite of mainly shipborne in situ observations to constrain model simulations in remote ocean regions. As stated above, each marine aerosol nutrient observation during the research ship's deployment usually represents a single day worth of continuous air measurement, resulting in few measurements within many key remote ocean areas (Hamilton et al., 2019). Creating suitable empirical distributions of aerosol nutrient properties, particularly under diverse meteorological conditions and/or anthropogenic forcings, is challenging with such sparse data sets, hindering model validation. Additionally, a model's spatial resolution has a larger footprint than a single observation site or ship track; a representation error not unique to this field, but pervasive across the geosciences (Schutgens et al., 2017). To help address this shortfall in data and model constraints, alternate methods include temporal averaging over seasonal or annual time periods and spatially aggregating observations, sometimes called "super-obbing" to produce climatologies with a statistically useful number of observations in a given region. These 2 methods can be combined for a more complete spatiotemporal solution and Hamilton et al. (2019) have shown how capturing the small-scale regional properties of observed soluble iron concentrations can help overcome some of the sampling challenges in this field. However, differences in observations and model simulations can arise when investigating individual case studies, particularly those focused on larger or more extreme natural episodic events that are more difficult to capture with ship-based surface observations. To help mitigate the bias present in small data sets, comparing median aerosol concentration values between observation and simulation climatologies is likely to be more appropriate than means. A combination of satellite and in situ data, along with improved model capacity in simulating extreme events, represents a way forward until more data become available. Such a combined methodology was recently used to identify that increasing Boreal fire activity likely stimulated Arctic phytoplankton blooms via fire aerosol nutrient (nitrogen) transport and deposition (Ardyna et al., 2022).

One example of how recent satellite data advance will improve Earth System modeling is in the representation of dust aerosol mineralogy, a critical component for understanding the iron cycle. Dust is a mixture of minerals with different physicochemical properties that show significant regional variations. These minerals have varying iron content, chemical structure, and typical grain sizes, thus affecting the total iron emitted and its susceptibility to atmospheric dissolution processes (Journet et al., 2008; Shi et al., 2012). Minerals in dust also contain alkaline elements (e.g., calcium, potassium) that alter aerosol pH, and hence, the acidic dissolution potential of iron in the atmosphere. Early models aimed at representing the contribution of dust sources to the atmospheric iron cycle neglected this complexity, using a constant iron content from dust instead (e.g., Luo et al., 2008). This simplification is still assumed in many biogeochemical ocean models by applying dust climatologies with a fixed iron ratio (e.g., 3.5%; Aumont et al., 2015). Advances have been made in recent years to characterize the mineralogy-

dependent iron emissions from dust sources (Scanza et al., 2018). However, the mineralogical composition of dust sources at the global scale is highly uncertain and derived from the extrapolation of a limited set of measurements (Claquin et al., 1999; Journet et al., 2014). As a result, the current modeling representation of dust mineralogy, size distribution, and iron content can be improved. The ongoing NASA Earth Surface Mineral Dust Source Investigation (EMIT) mission (Green and Thompson, 2020) aims to characterize the mineralogy of dust sources through high-quality hyperspectral spectroscopy techniques. The EMIT sensor, installed in the International Space Station in mid-2022, allows the quantification of 10 different minerals in the soil with an unprecedented level of detail (spatial resolution and geographical coverage). Following current evidence (Shi et al., 2011), most atmospheric iron models classify the lithogenic iron in 3 mineralogy-dependent dissolution pools (fast, intermediate, and slow dissolution). This partition aims to mimic observations that suggest that a small fraction of the iron, in the form of ferrihydrite and/or nanoiron oxides, is highly reactive and bioavailable. In contrast, structural iron in the matrix of phyllosilicates and the larger crystalline particles of iron oxides are increasingly difficult to dissolve (Shi et al., 2012; Ito and Xu, 2014). EMIT will bring information on the abundance of nanoiron oxides in soils, allowing Earth System models to transition toward a more realistic characterization of iron dissolution pools.

Positive model developments in the last few decades include the discovery and the importance of Earth System phenomena previously overlooked (such as wildfires, volcanoes, and high latitude dust) as significant natural sources of aerosol nutrients. Indeed, 2 independent modeling studies both concluded that wildfire iron was likely to be 6–8× more efficient at sequestering atmospheric CO_2 to ocean depths than dust iron deposits (Hamilton et al., 2020a; Ito et al., 2020b). Numerical models incorporating anthropogenic iron sources have also been recently developed, allowing for simulations of the distribution and impact of human activity on iron deposition and ocean biogeochemistry (Myriokefalitakis et al., 2018). However, it is important to note that model validation of iron emissions is limited by the sparsity of observational data, especially in the Southern Hemisphere. Concurrent, the community studying aerosol nutrients is embracing the idea that a more realistic depiction of the land–atmosphere–ocean continuum will be best achieved by improved integration of observational and modeling efforts across a wide variety of disciplines. For example, high latitude dust activity (emission from glaciers and cold deserts at high latitudes) has been recently reported as a major source of aeolian material to nutrient depleted marine regions. However, mainstream climate and Earth System models do not include such sources in their simulations (Bullard, 2017; Meinander et al., 2022), although when explicitly simulated in custom-made model versions their impacts on the local environment can be significant (Tobo et al., 2019).

Understanding how future changes in nutrient limitation patterns, due to changes in deposition as discussed above, could impact ocean biogeochemical cycles requires advanced modeling and observational capabilities. Earth System models able to make projections are a product of decades of cumulative knowledge from across the physical sciences, often including information gained from adjoining fields, such as social science, economics, or policy. Vehicles for gathering and directing such large intellectual efforts include the UN, and while the discussion here falls within the Ocean Decade (2021–2030), the topic of aerosol and ocean biogeochemistry is underrepresented in the program. With growing interest in topics such as the Blue Economy, artificial iron fertilization, and the ocean's health, this is an ideal time for this community to foster more partnerships across disciplines and share expertise.

Future Directions:

1. We identified 4 difficulties to be addressed in using the existing satellite platforms:
   a. Uncertainty in whether an observed bloom was at the receiving end of deposited nutrient aerosols in the days preceding to the observation and thus can be classed as an aerosol fertilized patch.
   b. A low airborne aerosol loading does not have enough spectral contrast to enable remote aerosol type detection. That is, smoke, dust, or volcanic ash have different characteristics, but existing aerosol algorithms can only make aerosol source distinction at moderate-to-very high loadings.
   c. If aerosol deposition does not result in an observable chlorophyll change, as appears to be the case in many instances, the use of additional ocean biology satellite products to monitor changes, such as community shifts, biomass accumulation, or improvements to phytoplankton physiological status and health, is needed—yet not straightforward or routinely undertaken (Behrenfeld and Michigan, 2013; Westberry et al., 2023).
   d. The ability to observe the atmosphere and ocean in cloudy regions using the (combined) features of hyperspectral, polarization, and angular measurements onboard new satellite missions (**Table 1**).
2. For an average *nonsatellite-savvy* data user, access to data sets remains a practical hurdle. Some suggested developments in aiding satellite data use include:
   a. The development of standardized data products that incorporate spectral or vertical profiling features would enable easier access and utilization of the data by the wider scientific community.
   b. Improving the accessibility of data sets with user-friendly interfaces.
   c. Operational agencies, particularly those utilizing sensors like the Geostationary Operational Environmental Satellites (GOES) and Himawari satellites for forecasting, could consider establishing science teams responsible for updating algorithms and calibration constants. This would ensure that the data produced remain relevant

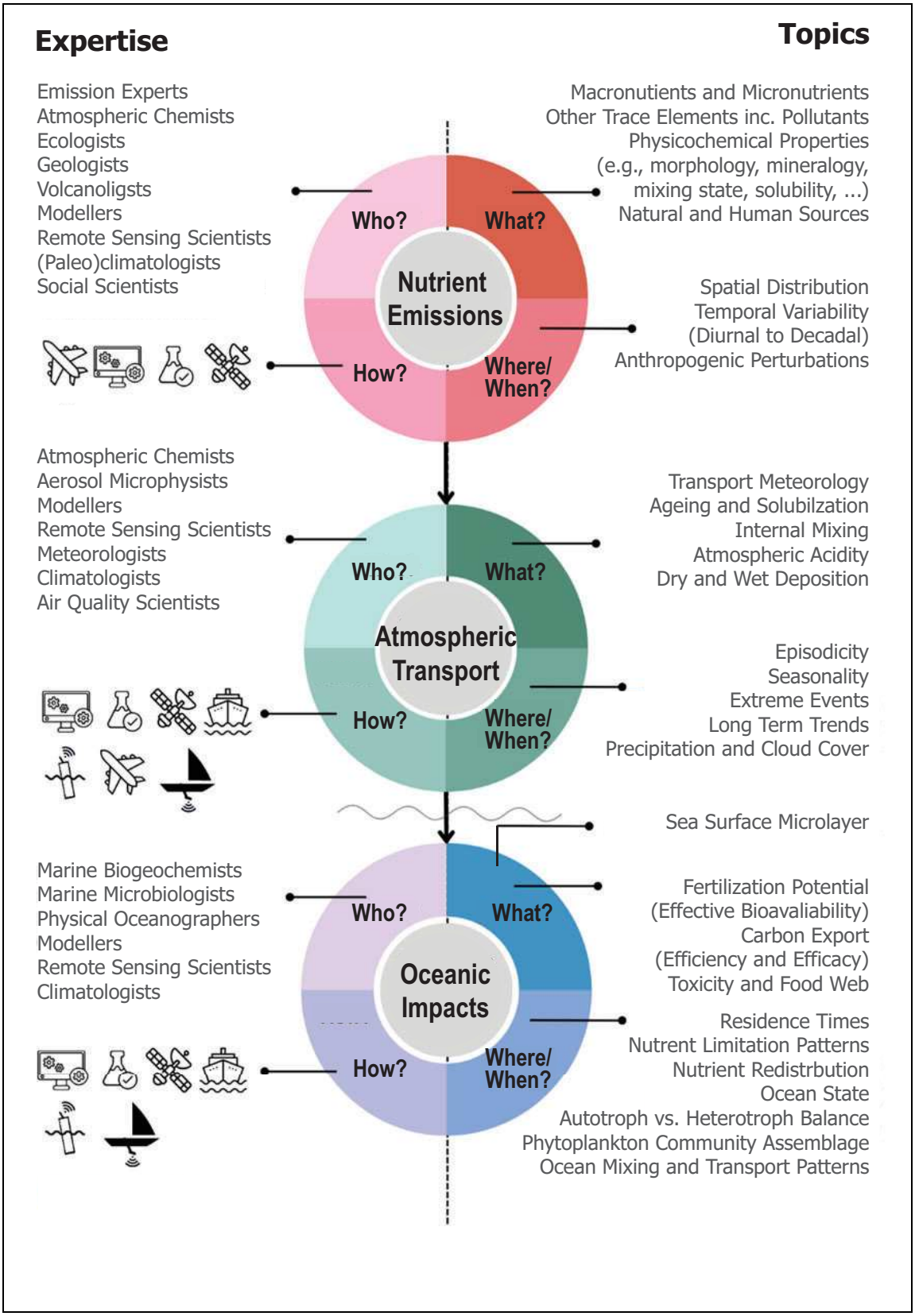

Figure 4. Summary schematic of the expertise and topics needing investigating to further understand the aerosol nutrient–ocean biogeochemistry story. All topics are likely to be influenced by climate change. "How?" icons pictorially represent the tools needed, as identified in the text, such as increased computing resources, laboratory measurements, and observational tools.

and accurate for scientific applications over time, not only for deposition studies but for aerosol studies in general.

d. Collaborative initiatives between technology-focused deployments and scientific data production plans are to be encouraged. For example, the unique capabilities of CubeSats can be further explored to detect features currently not observed in traditional satellite-based (aerosol) studies.

3. Investigate methodologies to improve nutrient aerosol modeling and the coupling of processes within Earth System models, including:

   a. Given the critical role of pH in nutrient transport and deposition fluxes, explore strategies to incorporate more detailed pH-related processes, while considering the complexity and computational expense associated with acidity-dependent variables.

   b. Improved mechanistic understanding of natural and anthropogenic nutrient (and toxicant) aerosol, from emissions to interactions with both atmospheric constituents and ocean biogeochemistry.

   c. Mesocosm approaches, using aerosol samples from different origins and field campaigns, are essential to advance our understanding of aerosol impacts on marine ecosystems and the global carbon cycle.

   d. Develop strategies that balance computational efficiency with the development of new or more detailed processes, ensuring that model simulations achieve acceptable performance levels while capturing the essential physicochemical characteristics of nutrient aerosol and their spatiotemporal deposition pattens.

4. All the efforts listed across all sections need interdisciplinary teams with expertise across the geosciences. Creation of forums and workshops are needed to establish such a community.

"The journey, not the destination matters"

T. S. Eliot's words are suitably applicable to this discourse on the future of the study of aerosol nutrient changes; the journey of discovery and understanding is just as important as the destination. This odyssey requires collaboration and cross-disciplinary partnerships, as well as the ability to adapt and evolve as new information is uncovered (**Figure 4**). With each new discovery, perspectives and approaches must be refined and updated. The journey requires a spirit of open-mindedness and a willingness to evolve conventional approaches to understanding how the world works. The result is not just a clearer understanding of the subject but a deeper appreciation of the complex interplay between science, society, and the world around us. Exploring aerosol nutrient changes in the years ahead promises to be both exciting and enlightening, and "Those who arrive at the end of the journey are not those who began" speaks to the very nature of this transdisciplinary research area.

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Competing interests

The authors declare that they have no conflict of interest.

Author contributions

Contributions to conception and design: DSH, ARB, YI.

Acquisition and analysis of figure and table data: DSH, SG, SM, EB-M, YK.

Reviewing the literature, drafting, and revising the article: All authors.

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