

Biogeochemical Coupling between Ocean and Atmosphere—A Tribute to the Lifetime Contribution of Robert A. Duce

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ABSTRACT

To mark the publication of the special collection in honor of Robert (Bob) A. Duce in the *Journal of the Atmospheric Sciences*, we have summarized his most important contributions to the subject of biogeochemical coupling between the atmosphere and ocean. Here we have divided these contributions into four themes—deposition from the atmosphere and its effects on the oceans, volatile elements emitted from the oceans, sea surface biology and aerosol formation, and marine aerosols and clouds. It is our intent that this summary along with the papers in this special collection provide an overview of the enormous contributions that Bob Duce has made to the subject during his distinguished scientific career.

1. Deposition from the atmosphere and its effects on the oceans

Bob Duce was one of the first people to understand the importance of the interaction of the oceans and the atmosphere, in particular, the transfer of chemical elements between the two reservoirs. Atmospheric deposition has been known to reach and impact the oceans for a long time. Darwin observed dust storms on the *Beagle* expedition and pondered their significance (Darwin 1846) and the importance of atmospheric dust inputs to marine sediments has been evident for many years (e.g.,

Griffin et al. 1968). There has been evidence since the 1960s that contaminants from land can reach the oceans via the atmosphere, for instance radionuclides from atmospheric weapons testing (e.g., Druffel 1981) and persistent organic pollutants (e.g., Aguilar et al. 2002). However, the Sea/Air Exchange Program (SEAREX) marked the first major coordinated study of the long-range atmospheric transport of biogeochemically important material from land to the ocean (including desert dust, persistent organic pollutants, nitrate, sulfate, and trace metals such as lead and particulate mercury) and set a benchmark for future studies (Duce 1989).

The significance of these atmospheric inputs for global biogeochemical cycles has become increasingly evident. Problems arising from the atmospheric transport of

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persistent, or slowly degrading, and bioaccumulating organic pollutants to the oceans persist, despite many years of regulation of some of these contaminants, compounded by the arrival of some new organic chemical contaminants of concern (Farrington and Takada 2014). Similarly, there are still concerns over the impact on the oceans of terrestrial mercury emissions because its bioaccumulation in fish may pose a human health risk. Mercury concentrations in the oceans have been increased by atmospheric transport and deposition of anthropogenic emissions, leading to a legacy of mercury that cycles within the oceans (Lamborg et al. 2014) and volatilizes back into the atmosphere.

There are, however, some more encouraging stories in which the recognition of the importance of atmospheric contamination has led to effective control measures. Lead, for example, was an important part of the SEAREX program, leading to Clair Patterson's seminal demonstration of the global impact of lead contamination (Patterson and Settle 1987). The restrictions on lead usage from the mid-1970s, coupled to its relatively short lifetime in the oceans due to particle scavenging, means that lead concentrations in the oceans (as recorded in coral skeletons) have now declined to similar concentrations to those seen in 1900 (Boyle et al. 2014).

Over the last couple decades, it has been realized that the atmosphere is not only a source of contaminants to the oceans, but also a source of nutrients, particularly nitrogen and iron (and to a lesser extent phosphorus), which can influence ocean productivity and hence Earth's biogeochemical and climate system (Jickells et al. 2017). The input of nitrogen (in oxidized, reduced, and organic forms) to the oceans from land via the atmosphere (39 Tg N yr^{-1}) is now comparable to riverine inputs (34 Tg N yr^{-1}) and greatly elevated over preindustrial levels (Jickells et al. 2017). The effect of this atmospheric nitrogen input is to create a low-level fertilization of the surface ocean (over about a half of which primary production is thought to be nitrogen limited) and to increase primary production and ocean CO_2 uptake and N_2O emissions from anoxic and low-oxygen zones (Duce et al. 2008; Jickells et al. 2017).

The input of dust to the ocean was an important component of the SEAREX program (Prospero et al. 1989). Moore et al. (1984) suggested that the dust transport quantified in SEAREX is a key source of iron to the oceans, and the work by Duce (1986) was the first to show clearly that atmospheric transport is the source of most of the iron and possibly nitrogen in the surface waters of the North Pacific and North Atlantic gyres. From the pioneering work of Martin (1990) and Martin and Fitzwater (1988), it is now well established that the deficiency of iron limits productivity over almost half of the world oceans and drives patterns of nutrient limitation and

nitrogen fixation (Jickells et al. 2005; Boyd and Ellwood 2010; Okin et al. 2011; Ward et al. 2013; Jickells and Moore 2015). The atmospheric deposition of iron into the global ocean is estimated to be $10\text{--}30 \text{ Tg Fe yr}^{-1}$ for total iron, of which $0.2\text{--}0.4 \text{ Tg Fe yr}^{-1}$ is labile (Myriokefalitakis et al. 2018). These new insights contributed to atmospheric deposition becoming a central plank of the GEOTRACES international research program (<http://www.geotraces.org>). Despite this progress, the relative importance and solubility of dust and anthropogenic sources of iron (and many other trace metals), the controls on their solubility in atmospheric deposition and cycling within the ocean are still not well understood (Baker and Croot 2010).

Work by Bob Duce and Rich Arimoto in SEAREX involved a range of trace metals and aerosols. Those metals also play important but poorly understood roles in regulating ocean productivity (Moore et al. 2013). There is evidence, for instance, that manganese inputs may play a role in regulating productivity in iron deficient areas (Browning et al. 2014). The productivity of the oceans responds to multiple drivers including the atmospheric deposition of various nutrients, along with fluvial inputs and internal nutrient cycling processes (e.g., Krishnamurthy et al. 2010; Okin et al. 2011; Moore et al. 2013; Ito et al. 2016; Yang and Gruber 2016).

2. Volatile elements emitted from the oceans

Although much of Bob Duce's work was and continues to be on the input of chemicals, both natural and man-made, from the atmosphere into the oceans, he also showed that active processes in the oceans transfer material in the opposite direction, that is, from the ocean to the atmosphere. Clear examples are the easily volatilized elements including halogens (particularly iodine), mercury, selenium, and sulfur. It should be noted that matter is also transferred from sea to air in the form of sea salt. This represents another large yet poorly quantified flux of major seawater constituents to the atmosphere and then to land. Important early work on estimating the fluxes was pursued in the Duce Group (e.g., Erickson and Duce 1988; Duce et al. 1983); this topic is still an active field of research.

The early evidence for cycling of volatiles came from examination of the enhanced concentration of some elements in atmospheric aerosols and rain collected over or near the oceans (from research vessels, islands, and other coastal sampling sites)—such as the measurement campaign of SEAREX initiated and led by Bob Duce. In particular, measurements of the chemical composition of marine aerosols can be used to derive an enrichment factor for individual elements, by calculating the ratio of the measured elemental concentration in the aerosol to

the concentration in the source materials, which include sea salt and transported continental dust. An enrichment factor greater than one means that the element is enriched relative to its composition in the major sources at the area of study. Some of the highest enrichment factors measured over the sea are for the easily volatilized elements, including sulfur, selenium, mercury, and iodine, where enrichments range up to 10 or 100 and not infrequently even several thousand [i.e., for selenium (Duce et al. 1975) and for iodine (Duce et al. 1965)]. Such enrichments are often attributed to a flux of a volatile form of the element formed in seawater to the atmosphere where it is incorporated into aerosols leading to the observed enrichments. Although much has still to be learned about how the volatiles are formed, the importance for some elements has been well established, for example, dimethyl sulfide (DMS-CH₃SCH₃) for sulfur (Liss et al. 2014), the elemental form for mercury (Costa and Liss 1999) and mono and dimethylated forms for selenium (Amouroux et al. 2001).

The establishment of whether a steady-state balance of the geochemical budget of an element exists requires quantitative assessment of the fluxes of the element to the atmospheric system. The results for several elements showed that balance was not possible without a significant flux of the elements out of the oceans, which was generally assumed to be in the form of volatile gases, as illustrated by examples of such budgeting exercises for sulfur (Brimblecombe 2004) and selenium (Mosher and Duce 1987). In some cases, direct confirmatory evidence has been obtained from gas fluxes estimated from measurements made at sea from research vessels. This involves measuring the concentrations of the gas, identified as being important, in both surface seawater and the overlying air and using a transfer velocity parameterization (e.g., Garbe et al. 2014) to convert the air-sea concentration difference into a flux (Liss and Slater 1974). For the elements considered here, flux calculations have been made, for example, for volatile selenium species in the North Atlantic (Amouroux et al. 2001) and using the large database of DM measurements assembled by Lana et al. (2011). The results show significant fluxes of the volatiles out of the oceans and add credibility to the ideas that the oceans are an important source.

The existence of fluxes of volatile forms of particular elements can have significance in a wider environmental context and even globally. Some examples include the importance of marine-derived volatile forms of iodine, sulfur, and selenium not only in helping to close their global elemental balances but also in recycling the elements onto the terrestrial environment, which can have importance for mammalian (including human) health, for example, selenium (Rayman 2000) and iodine

(Fuge and Johnson 2015). Further, the emission of marine sulfur and iodine volatiles can lead to new particle formation in the atmosphere with potential implications for climate (Charlson et al. 1987; Liss and Lovelock 2007), the oxidation capacity of the marine troposphere (Saiz-Lopez et al. 2012), and iodine injection to the stratosphere (Saiz-Lopez et al. 2015). Also, the work of Carpenter et al. (2013) showed that atmospheric iodine levels are significantly influenced by sea surface emissions of inorganic iodine, accounting for 75% of observed iodine oxide levels over the tropical Atlantic Ocean. In addition, the Duce group made pioneering observations of the organic forms of nutrients and in particular of the organic nitrogen and organic phosphorus (e.g., Duce et al. 1972; Graham and Duce 1979; Mace et al. 2003a,b,c), which led to the first global modeling of organic nitrogen and organic phosphorus atmospheric input to the ocean (Kanakidou et al. 2012).

3. Sea surface biology and aerosol formation

Life in the ocean is predominantly microbial (Karl 2007) and concentrated near the surface where there is sufficient light to support photosynthesis. Marine phytoplankton fix an estimated 45–55 Pg Cyr⁻¹ (1 Pg = 10¹⁵g) (Longhurst et al. 1995; Field et al. 1998; Carr et al. 2006), which is approximately 40% of the total photosynthetic production on Earth. The distribution of phytoplankton across the ocean is uneven and largely determined by nutrient supply. One of Bob Duce's key contributions was recognizing the significance of atmospheric deposition in supplying essential nutrients (such as iron and biologically available nitrogen) and the role that atmospheric deposition plays in regulating marine biogeochemical processes, including the carbon cycle (Duce and Tindale 1991; Duce et al. 2008; Jickells et al. 2005; Owens et al. 1992).

Marine aerosols are either emitted directly into the atmosphere (primary) or produced from new particle formation via gas-to-particle conversion (secondary) in the atmosphere. As illustrated in Fig. 1, primary aerosols and freshly nucleated particles undergo chemical and physical transformations and are subjected to transport, cloud processing, and removal. A wide variety of aerosol types, which originate from new particle formation, primary production from sea spray, ship emissions, and long-range transport of natural and anthropogenic particles from the continents, exist in marine environments. In the clear marine atmosphere, the oxidation products of DMS, iodine compounds, and amines are believed to be the important biogenic precursors for new particle formation (e.g., Charlson et al. 1987; O'Dowd and de Leeuw 2007; Quinn and Bates 2011; Zhang et al. 2012; Qiu and Zhang 2013; Sellegri et al. 2016). Furthermore, it is plausible that photochemical oxidation products of

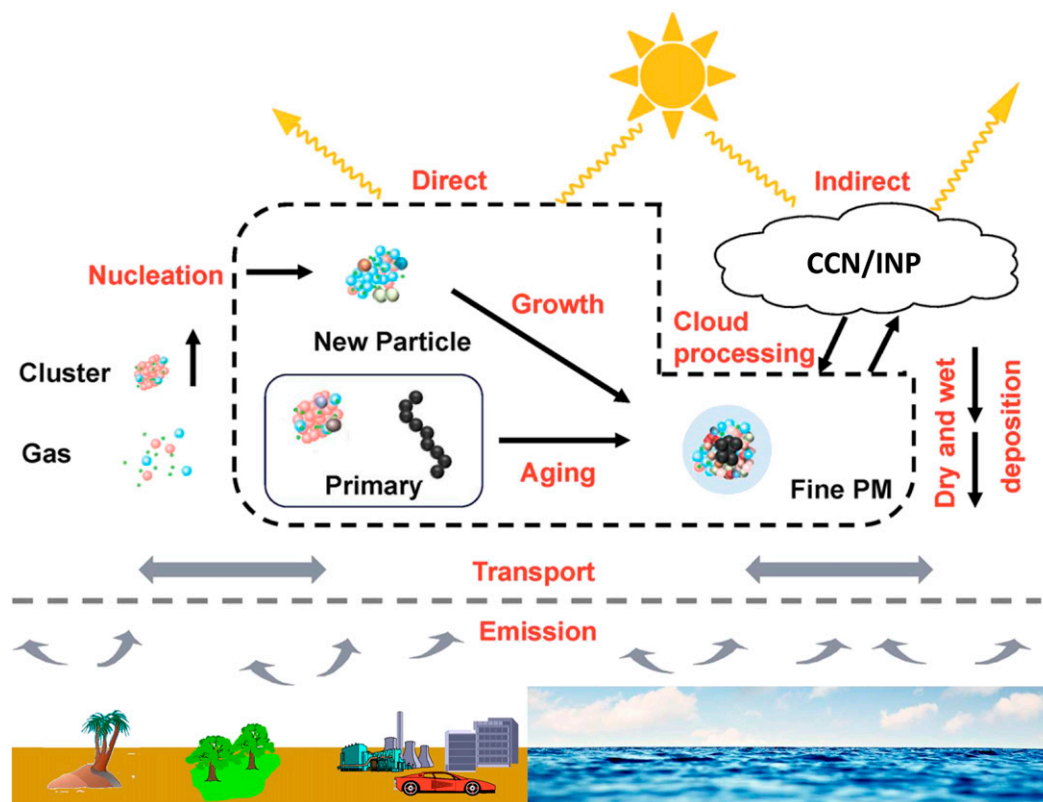


FIG. 1. Life cycles and impacts of atmospheric aerosols [modified from Zhang et al. (2015)].

volatile organic compounds, which enter the atmosphere via air–sea exchange, play a role in nucleation and growth of nanoparticles (Zhang 2010; Zhao et al. 2009; Xu and Zhang 2012).

In addition to sea salt, organic matter may also be transferred to the atmosphere in sea spray aerosols (SSA). In the last 15 years, there has been considerable interest in the coupling between biological processes and the organic matter content of SSA (O'Dowd et al. 2004, 2015; Quinn et al. 2014; Brooks and Thornton 2018), with the hope that this will provide insights into both the formation and complexity of marine aerosols.

Processes at the very surface of the ocean may be key to understanding the connectivity between biological processes, atmospheric aerosol properties, and clouds. The sea surface microlayer (SML) is defined as a 1–1000- μm -thick interface (Liss and Duce 1997) and has measurably different properties from the underlying waters (Hunter 1997). Bob Duce recognized the significance of the SML showing that heavy metals and organic compounds were enriched by factors of 1.5–50 in it (Duce et al. 1972). Subsequent work has shown that many constituents (particularly those in particulate form) occur at higher concentrations in the SML than in the underlying waters (Cunliffe et al. 2011; Engel et al. 2017); including bacteria

and viruses (Aller et al. 2005), dissolved organic carbon (DOC) (Sieburth et al. 1976; Hunter 1997), and exopolymer gels (Wurl and Holmes 2008; Thornton et al. 2016). When bubbles burst at the sea surface, the SML is skimmed off to produce film and jet drops, accounting for the enrichment of marine aerosol in organic matter compared to subsurface water (Liss and Duce 1997; Russell et al. 2010; Quinn et al. 2015).

Recent research has shown a direct link between the composition of aerosols and biological activity, which is usually indicated by chlorophyll concentrations in seawater (O'Dowd et al. 2004, 2015; Rinaldi et al. 2013; Russell et al. 2010). For example, O'Dowd et al. (2004) found that the organic fraction of submicron aerosol mass increases from a winter value of 15%–63% during the spring bloom in the North Atlantic Ocean. In contrast, other work has shown little or no link between the chlorophyll concentrations and the organic matter content or properties of SSA (Quinn et al. 2014); these authors proposed that the carbon content and properties of fresh SSA are relatively constant and reflect the large background pool of organic matter in the ocean. Indeed, the majority of the large inventory of DOC in the ocean (662 Pg C; Hansell et al. 2009) is uncoupled from ecosystem activity in the surrounding water on the scale of

days to years as it is recalcitrant, with a mean radiocarbon age of 4000–6000 years (Hansell 2013). These conflicting observations illustrate the challenges and multidisciplinary approaches required to develop a mechanistic understanding of how biological processes affect the composition of organic matter in the ocean and subsequent properties of SSA and marine clouds (Brooks and Thornton 2018).

4. Marine aerosols and clouds

Bob Duce's pioneering work on the atmospheric pathway for long-range transport of natural and pollutant aerosols from the continents to the ocean has inspired research on the interaction of marine aerosols with clouds and climate (e.g., Zoller et al. 1974; Duce et al. 1975; Duce et al. 1980; Duce et al. 1991). Marine aerosols influence climate directly by absorbing or scattering solar radiation (Satheesh and Moorthy 2005) and indirectly by acting as cloud condensation nuclei (CCN; Quinn and Bates 2011; Quinn et al. 2014) or ice nucleating particles (INPs; Wang et al. 2015; Wilson et al. 2015). For example, the contribution of new particle formation to the total absorption of solar shortwave radiation is estimated to be -2.2 W m^{-2} over the oceans, which is much larger than that over the continents (Kazil et al. 2010). The global annual mean top-of-the-atmosphere radiative forcing by DMS aerosol is estimated to be -2.03 W m^{-2} , whereas over the Southern Ocean during SH summer the mean DMS aerosol radiative forcing reaches -9.32 W m^{-2} (Thomas et al. 2010). However, poor understanding of aerosols and clouds remain the major sources of uncertainty in climate models (IPCC 2013; National Academies of Sciences, Engineering, and Medicine 2016). The activation of marine aerosols as CCN has been identified as the most important factor in determining the formation and microphysical properties of shallow marine clouds (Hudson and Noble 2014).

As discussed above, aerosols produced locally over regions of biological activity enter the atmosphere through gas-phase emissions mainly of DMS and volatile organic compounds or as SSA directly injected into the atmosphere through bubble bursting (Ayers and Caine 2007; Quinn and Bates 2011). Despite more than 35 years of debate, the relative importance of these sources as CCN or INP remains uncertain. Both in situ (Ayers and Gras 1991; Hegg et al. 1991) and remote sensing methods (Sorooshian et al. 2009; Meskhidze et al. 2011) provided evidence of increased CCN concentrations over regions of high biological activity, although the amplitude of increases in CCN varies between locations.

Assessment of marine biota–CCN interactions is complicated by the observation that SSA can simultaneously

possess high CCN ability and low hygroscopicity (Ovadnevaite et al. 2011). This puzzling combination of properties is consistent with the presence of surfactants such as proteins, macromolecules, or exopolymer gels that partition to the surface of droplets, reducing the surface tension required for CCN activation (Li et al. 2013; Burrows et al. 2014). Significant reductions in the surface tension of marine aerosols containing biosurfactants have been observed in situ (Ekstrom et al. 2010). In addition, modeling of exopolymer gels suggests them to be hydrophobic yet CCN active (Li et al. 2013; Sun et al. 2014).

In the case of mixed-phase and cirrus clouds, the impacts of marine aerosols on microphysics are highly uncertain. This is due to the combination of limited knowledge of marine aerosols to act as INPs and the few in situ measurements of ice nucleation in the marine environment (Hoose and Mohler 2012; Wilson et al. 2015; DeMott et al. 2016). As shown by Bob Duce and coworkers, continental mineral dust and soils are significant sources of transported primary aerosols over Earth's oceans (Duce et al. 1980; Uematsu et al. 1985; Duce and Tindale 1991). Dust is injected into the atmosphere with an annual amount of 1000–4000 Tg, of which about 500 Tg is deposited in the ocean (Huneeus et al. 2011). About 25% of dust particles lifted annually from the Saharan Desert (about 1150 Tg) are transported to the Atlantic Ocean (Shao et al. 2011). In addition to increasing the primary productivity through ocean fertilization (Schulz et al. 2012), mineral dust and aerosolized soil impact climate by facilitating the formation of ice-containing clouds over remote regions of the ocean. An experiment determined that while marine biogenic sources most likely dominate INP populations over the Southern Ocean, over the majority of the global ocean, both marine and continental contributions to INP are important (Burrows et al. 2013). While this latter study demonstrated the potential importance of marine aerosols as INPs, due to a dearth of data the calculations were performed with the assumption that marine biological particles capable of inducing freezing are equally distributed around the globe. Recently, several laboratory, field, and modeling studies have developed parameterizations of INPs from the marine biogenic source and K-feldspar dust, showing that organic material from sea spray in the SML and K-feldspar dust transported over the ocean from desert regions nucleate ice in mixed-phase and high-altitude clouds (Wilson et al. 2015; Vergara-Temprado et al. 2017).

The impacts of long-range transport of natural and anthropogenic aerosols on oceanic weather and climate have received increasing attention (Zhang et al. 2007). Dust particles absorb and scatter sunlight, and the relative importance of absorption and scattering by dust

particles plays an important role in determining regional radiative budget (Wang et al. 2013). For example, Saharan dust exhibits significant impacts on regional longwave and shortwave radiation, cloud formation, convective systems, and hurricanes over the tropical Atlantic (Pan et al. 2018). Also, increasing long-range transport of anthropogenic aerosols from Asia in recent decades has been attributed to climatically significant changes in winter storm patterns over the Pacific, as reflected by observed increasing deep convective clouds and precipitation, as well as a decadal variation of midlatitude cyclones (Zhang et al. 2007; Li et al. 2008; Wang et al. 2014a,b). In addition, anthropogenic aerosols transported from the continents have been shown to impact tropical cyclones, by modifying radiative budget and microphysical processes (Wang et al. 2014c). The coupled microphysical and radiative effects of anthropogenic aerosols result in delayed development, weakened intensity, and early dissipation of tropical cyclones, but an enlarged rainband and increased precipitation under polluted conditions (Wang et al. 2014c). Another recent observational analysis has shown enlarged rainfall areas of tropical cyclones over the western North Pacific by anthropogenic aerosols (Zhao et al. 2018).

5. Conclusions

Bob Duce has made fundamental contributions to understanding of atmospheric transport of chemicals from the continents, their deposition to the ocean, and their impact on marine biogeochemistry and climate, with field and numerical studies in Antarctica, the Arctic, and all the major oceans. His pioneer research has fundamentally altered the research direction in the chemical interactions between the atmosphere and the oceans. He has provided crucial leadership to the atmospheric–oceanic sciences community nationally and internationally (Duce et al. 1984; Duce and Liss 2002; National Academies of Sciences, Engineering, and Medicine 2016).

Elucidation of the biogeochemical coupling between ocean and atmosphere remains challenging under human-induced changes in the global environment and climate. There is still much to learn about the atmospheric inputs to the ocean. For example, in the case of nitrogen inputs these gaps in our understanding include (i) the nature and source of the organic nitrogen, (ii) the significance of air–sea ammonia and organic nitrogen exchange in the overall cycle, (iii) the impact of changing emissions with ammonia emissions projected to increase while NO_x declines over the next century, (iv) the impact of atmospheric nitrogen deposition on surface water biological nitrogen fixation, and (v) the impact on the overall ocean

nitrogen cycle and the emission of the powerful greenhouse gas N₂O from the oceans.

The atmospheric inputs to the ocean and the cycles in both the atmosphere and ocean are likely to evolve with future global change, but our capacity to predict the overall effect on the oceans and the climate system is limited. There is a need for better understanding of the key atmospheric emission, transport, transformation and deposition processes, and of the subsequent cycling of these biogeochemically significant compounds in the ocean. Coupled atmospheric and oceanic models are needed to understand these interacting biogeochemical cycles, scale-up fluxes, and predict future changes.

In addition, marine aerosols are highly complex, and that complexity must be carefully considered in assessing the formation of warm and cold clouds and the properties of marine clouds, which impact oceanic weather, climate, and deposition of materials to the oceans. New particle formation in the marine atmosphere remains a subject of considerable uncertainty, particularly concerning the roles of the oxidation products of DMS, volatile organic compounds, iodine compounds, and amines, which enter the atmosphere via the air–sea exchange, as well as their synergistic effects.

More marine CCN and INP measurements are needed, especially in undersampled regions such as the Southern Ocean and the Arctic. To accurately quantify the impacts of marine aerosols on clouds and climate requires future in situ measurements of the abundance, spatial and temporal distributions, and chemical/physical properties of marine aerosols, CCN, and INP as well as the microphysical parameters of hydrometeors to constrain and validate atmospheric models. Clearly, future research will require comprehensive atmospheric–oceanic field campaigns that match the ambition of SEAREX, as pioneered by Robert Duce.

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REFERENCES

- Aguilar, A., A. Borrell, and P. J. H. Reijnders, 2002: Geographical and temporal variation in levels of organochlorine contaminants in marine mammals. *Mar. Environ. Res.*, **53**, 425–452, [https://doi.org/10.1016/S0141-1136\(01\)00128-3](https://doi.org/10.1016/S0141-1136(01)00128-3).
- Aller, J. Y., M. R. Kuznetsova, C. J. Jahns, and P. F. Kemp, 2005: The sea surface microlayer as a source of viral and bacterial

- enrichment in marine aerosols. *J. Aerosol Sci.*, **36**, 801–812, <https://doi.org/10.1016/j.jaerosci.2004.10.012>.
- Amouroux, D., P. S. Liss, E. Tessier, M. Hamren-Larsson, and O. F. X. Donard, 2001: Role of oceans as biogenic source of selenium. *Earth Planet. Sci. Lett.*, **189**, 277–283, [https://doi.org/10.1016/S0012-821X\(01\)00370-3](https://doi.org/10.1016/S0012-821X(01)00370-3).
- Ayers, G. P., and J. L. Gras, 1991: Seasonal relationship between cloud condensation nuclei and aerosol methanesulfonate in marine air. *Nature*, **353**, 834–835, <https://doi.org/10.1038/353834a0>.
- , and J. M. Caine, 2007: The CLAW hypothesis: A review of the major developments. *Environ. Chem.*, **4**, 366–374, <https://doi.org/10.1071/EN07080>.
- Baker, A. R., and P. L. Croot, 2010: Atmospheric and marine controls on aerosol iron solubility in seawater. *Mar. Chem.*, **120**, 4–13, <https://doi.org/10.1016/j.marchem.2008.09.003>.
- Boyd, P. W., and M. J. Ellwood, 2010: The biogeochemical cycle of iron in the ocean. *Nat. Geosci.*, **3**, 675–682, <https://doi.org/10.1038/ngeo964>.
- Boyle, E. A., and Coauthors, 2014: Anthropogenic lead emissions in the ocean: The evolving global experiment. *Oceanography*, **27**, 69–75, <https://doi.org/10.5670/oceanog.2014.10>.
- Brimblecombe, P., 2004: The global sulphur cycle. *Biogeochemistry*, W. H. Schlesinger, Ed., Vol. 8, *Treatise on Geochemistry*, Elsevier, 645–682.
- Brooks, S. D., and D. C. O. Thornton, 2018: Marine aerosols and clouds. *Annu. Rev. Mar. Sci.*, **10**, 289–313, <https://doi.org/10.1146/annurev-marine-121916-063148>.
- Browning, T. J., H. A. Bouman, G. M. Henderson, T. A. Mather, D. M. Pyle, C. Schlosser, E. M. S. Woodward, and C. M. Moore, 2014: Strong responses of Southern Ocean phytoplankton communities to volcanic ash. *Geophys. Res. Lett.*, **41**, 2851–2857, <https://doi.org/10.1002/2014GL059364>.
- Burrows, S. M., C. Hoese, U. Poschl, and M. G. Lawrence, 2013: Ice nuclei in marine air: Biogenic particles or dust? *Atmos. Chem. Phys.*, **13**, 245–267, <https://doi.org/10.5194/acp-13-245-2013>.
- , O. Ogunro, A. A. Frossard, L. M. Russell, P. J. Rasch, and S. M. Elliott, 2014: A physically based framework for modeling the organic fractionation of sea spray aerosol from bubble film Langmuir equilibria. *Atmos. Chem. Phys.*, **14**, 13 601–13 629, <https://doi.org/10.5194/acp-14-13601-2014>.
- Carpenter, L. J., and Coauthors, 2013: Atmospheric iodine levels influenced by sea surface emissions of inorganic iodine. *Nat. Geosci.*, **6**, 108–111, <https://doi.org/10.1038/ngeo1687>.
- Carr, M. E., and Coauthors, 2006: A comparison of global estimates of marine primary production from ocean color. *Deep-Sea Res. II*, **53**, 741–770, <https://doi.org/10.1016/j.dsr2.2006.01.028>.
- Charlson, R. J., J. E. Lovelock, M. O. Andreae, and S. G. Warren, 1987: Oceanic phytoplankton, atmospheric sulphur, cloud albedo and climate. *Nature*, **326**, 655–661, <https://doi.org/10.1038/326655a0>.
- Costa, M., and P. S. Liss, 1999: Photoreduction of mercury in sea water and its possible implications for Hg⁰ air–sea fluxes. *Mar. Chem.*, **68**, 87–95, [https://doi.org/10.1016/S0304-4203\(99\)00067-5](https://doi.org/10.1016/S0304-4203(99)00067-5).
- Cunliffe, M., R. C. Upstill-Goddard, and J. C. Murrell, 2011: Microbiology of aquatic surface microlayers. *FEMS Microbiol. Rev.*, **35**, 233–246, <https://doi.org/10.1111/j.1574-6976.2010.00246.x>.
- Darwin, C., 1846: An account of the fine dust which often falls on vessels in the Atlantic Ocean. *Quart. J. Geol. Soc.*, **2**, 26–30, <https://doi.org/10.1144/GSL.JGS.1846.002.01-02.09>.
- DeMott, P. J., and Coauthors, 2016: Sea spray aerosol as a unique source of ice nucleating particles. *Proc. Natl. Acad. Sci. USA*, **113**, 5797–5803, <https://doi.org/10.1073/pnas.1514034112>.
- Druffel, E. M., 1981: Radiocarbon in annual coral rings from the eastern tropical Pacific Ocean. *Geophys. Res. Lett.*, **8**, 59–62, <https://doi.org/10.1029/GL008i001p00059>.
- Duce, R. A., 1986: The impact of atmospheric nitrogen, phosphorus, and iron species on marine biological productivity. *The Role of Air-Sea Exchange in Geochemical Cycling*, P. Buat-Menard, Ed., Reidel, 497–529.
- , 1989: *SEAREX: The Sea/Air Exchange Program*. Academic Press, 404 pp.
- , and N. W. Tindale, 1991: Atmospheric transport of iron and its deposition in the ocean. *Limnol. Oceanogr.*, **36**, 1715–1726, <https://doi.org/10.4319/lo.1991.36.8.1715>.
- , and P. S. Liss, 2002: The surface ocean—lower atmosphere study (SOLAS). *Atmos. Environ.*, **36**, 5119–5120, [https://doi.org/10.1016/S1352-2310\(02\)00327-8](https://doi.org/10.1016/S1352-2310(02)00327-8).
- , J. W. Winchester, and T. W. Van Nahl, 1965: Iodine, bromine, and chlorine in the Hawaiian marine atmosphere. *J. Geophys. Res.*, **70**, 1775–1799, <https://doi.org/10.1029/JZ070i008p01775>.
- , B. J. Ray, J. G. Quinn, T. L. Wade, S. R. Piotrowicz, and C. E. Olney, 1972: Enrichment of heavy-metals and organic compounds in surface microlayer of Narragansett Bay, Rhode Island. *Science*, **176**, 161–163, <https://doi.org/10.1126/science.176.4031.161>.
- , G. L. Hoffman, and W. H. Zoller, 1975: Atmospheric trace metals at remote northern and southern hemisphere sites: Pollution or natural? *Science*, **187**, 59–61, <https://doi.org/10.1126/science.187.4171.59>.
- , C. K. Unni, B. J. Ray, J. M. Prospero, and J. T. Merrill, 1980: Long-range atmospheric transport of soil dust from Asia to the tropical North Pacific—Temporal variability. *Science*, **209**, 1522–1524, <https://doi.org/10.1126/science.209.4464.1522>.
- , V. A. Mohnen, and D. G. Zimmerman, 1983: Organic material in the global troposphere. *Rev. Geophys.*, **21**, 921–952, <https://doi.org/10.1029/RG021i004p00921>.
- , and Coauthors, 1984: *Global Tropospheric Chemistry: A Plan for Action*. National Academy Press, 194 pp.
- , and Coauthors, 1991: The atmospheric input of trace species to the world ocean. *Global Biogeochem. Cycles*, **5**, 193–259, <https://doi.org/10.1029/91GB01778>.
- , and Coauthors, 2008: Impacts of atmospheric anthropogenic nitrogen on the open ocean. *Science*, **320**, 893–897, <https://doi.org/10.1126/science.1150369>.
- Ekstrom, S., B. Noziere, M. Hultberg, T. Alsberg, J. Magner, E. D. Nilsson, and P. Artaxo, 2010: A possible role of ground-based microorganisms on cloud formation in the atmosphere. *Biogeosciences*, **7**, 387–394, <https://doi.org/10.5194/bg-7-387-2010>.
- Engel, A., and Coauthors, 2017: The ocean’s vital skin: Toward an integrated understanding of the sea surface microlayer. *Front. Mar. Sci.*, **4**, 165, <https://doi.org/10.3389/fmars.2017.00165>.
- Erickson, D. J., and R. A. Duce, 1988: On the global flux of atmospheric sea salt. *J. Geophys. Res.*, **93**, 14 079–14 088, <https://doi.org/10.1029/JC093iC11p14079>.
- Farrington, J. W., and H. Takada, 2014: Persistent organic pollutants (POPs), polycyclic aromatic hydrocarbons (PAHs), and plastics examples of the status, trend, and cycling of organic chemicals of environmental concern in the ocean. *Oceanography*, **27**, 196–213, <https://doi.org/10.5670/oceanog.2014.23>.
- Field, C. B., M. J. Behrenfeld, J. T. Randerson, and P. Falkowski, 1998: Primary production of the biosphere: Integrating terrestrial and oceanic components. *Science*, **281**, 237–240, <https://doi.org/10.1126/science.281.5374.237>.

- Fuge, R., and C. C. Johnson, 2015: Iodine and human health, the role of environmental geochemistry and diet. *Appl. Geochem.*, **63**, 282–302, <https://doi.org/10.1016/j.apgeochem.2015.09.013>.
- Garbe, C. S., and Coauthors, 2014: Transfer across the air–sea interface. *Ocean–Atmosphere Interactions of Gases and Particles*, P. S. Liss and M. T. Johnson, Eds., Springer, 55–112.
- Graham, W. F., and R. A. Duce, 1979: Atmospheric pathways of the phosphorus cycle. *Geochim. Cosmochim. Acta*, **43**, 1195–1208, [https://doi.org/10.1016/0016-7037\(79\)90112-1](https://doi.org/10.1016/0016-7037(79)90112-1).
- Griffin, J. J., H. Windom, and E. D. Goldberg, 1968: The distribution of clay minerals in the World Ocean. *Deep-Sea Res. Oceanogr. Abstr.*, **15**, 433–459, [https://doi.org/10.1016/0011-7471\(68\)90051-X](https://doi.org/10.1016/0011-7471(68)90051-X).
- Hansell, D. A., 2013: Recalcitrant dissolved organic carbon fractions. *Annu. Rev. Mar. Sci.*, **5**, 421–445, <https://doi.org/10.1146/annurev-marine-120710-100757>.
- , C. A. Carlson, D. J. Repeta, and R. Schlitzer, 2009: Dissolved organic matter in the ocean a controversy stimulates new insights. *Oceanography*, **22**, 202–211, <https://doi.org/10.5670/oceanog.2009.109>.
- Hegg, D. A., R. J. Ferek, P. V. Hobbs, and L. F. Radke, 1991: Dimethyl sulfide and cloud condensation nucleus correlations in the northeast Pacific Ocean. *J. Geophys. Res.*, **96**, 13 189–13 191, <https://doi.org/10.1029/91JD01309>.
- Hoese, C., and O. Mohler, 2012: Heterogeneous ice nucleation on atmospheric aerosols: A review of the results from laboratory experiments. *Atmos. Chem. Phys.*, **12**, 9817–9854, <https://doi.org/10.5194/acp-12-9817-2012>.
- Hudson, J. G., and S. Noble, 2014: Low-altitude summer/winter microphysics, dynamics, and CCN spectra of northeastern Caribbean small cumuli, and comparisons with stratus. *J. Geophys. Res.*, **119**, 5445–5463, <https://doi.org/10.1002/2013JD021442>.
- Huneus, N., and Coauthors, 2011: Global dust model intercomparison in AeroCom phase I. *Atmos. Chem. Phys.*, **11**, 7781–7816, <https://doi.org/10.5194/acp-11-7781-2011>.
- Hunter, K. A., 1997: Chemistry of the sea-surface microlayer. *The Sea Surface and Global Change*, P. S. Liss and R. A. Duce, Eds., Cambridge University Press, 287–320.
- IPCC, 2013: *Climate Change 2013: The Physical Science Basis*. Cambridge University Press, 1535 pp., <https://doi.org/10.1017/CBO9781107415324>.
- Ito, T., A. Nenes, M. S. Johnson, N. Meskhidze, and C. Deutsch, 2016: Acceleration of oxygen decline in the tropical Pacific over the past decades by aerosol pollutants. *Nat. Geosci.*, **9**, 443–447, <https://doi.org/10.1038/ngeo2717>.
- Jickells, T., and C. M. Moore, 2015: The importance of atmospheric deposition for ocean productivity. *Annu. Rev. Ecol. Evol. Syst.*, **46**, 481–501, <https://doi.org/10.1146/annurev-ecolsys-112414-054118>.
- , and Coauthors, 2005: Global iron connections between desert dust, ocean biogeochemistry, and climate. *Science*, **308**, 67–71, <https://doi.org/10.1126/science.1105959>.
- , and Coauthors, 2017: A reevaluation of the magnitude and impacts of anthropogenic atmospheric nitrogen inputs on the ocean. *Global Biogeochem. Cycles*, **31**, 289–305, <https://doi.org/10.1002/2016GB005586>.
- Kanakidou, M., and Coauthors, 2012: Atmospheric fluxes of organic N and P to the global ocean. *Global Biogeochem. Cycles*, **26**, GB3026, <https://doi.org/10.1029/2011GB004277>.
- Karl, D. M., 2007: Microbial oceanography: Paradigms, processes and promise. *Nat. Rev. Microbiol.*, **5**, 759–769, <https://doi.org/10.1038/nrmicro1749>.
- Kazil, J., and Coauthors, 2010: Aerosol nucleation and its role for clouds and Earth’s radiative forcing in the aerosol-climate model ECHAM5-HAM. *Atmos. Chem. Phys.*, **10**, 10 733–10 752, <https://doi.org/10.5194/acp-10-10733-2010>.
- Krishnamurthy, A., J. K. Moore, N. Mahowald, C. Luo, and C. S. Zender, 2010: Impacts of atmospheric nutrient inputs on marine biogeochemistry. *J. Geophys. Res.*, **115**, G01006, <https://doi.org/10.1029/2009JG001115>.
- Lamborg, C., and Coauthors, 2014: Mercury in the anthropocene ocean. *Oceanography*, **27**, 76–87, <https://doi.org/10.5670/oceanog.2014.11>.
- Lana, A., and Coauthors, 2011: An updated climatology of surface dimethylsulfide concentrations and emission fluxes in the global ocean. *Global Biogeochem. Cycles*, **35**, GB1004, <https://doi.org/10.1029/2010GB003850>.
- Li, G., Y. Wang, K.-H. Lee, Y. Diao, and R. Zhang, 2008: Increased winter precipitation over the North Pacific from 1984–1994 to 1995–2005 inferred from the Global Precipitation Climatology Project. *Geophys. Res. Lett.*, **35**, L13821, <https://doi.org/10.1029/2008GL034668>.
- Li, X., T. Hede, Y. Q. Tu, C. Leck, and H. Agren, 2013: Cloud droplet activation mechanisms of amino acid aerosol particles: Insight from molecular dynamics simulations. *Tellus*, **65B**, 1–13, <https://doi.org/10.3402/tellusb.v65i0.20476>.
- Liss, P. S., and P. G. Slater, 1974: Flux of gases across the air-sea interface. *Nature*, **247**, 181–184, <https://doi.org/10.1038/247181a0>.
- , and R. A. Duce, 1997: Preface. *The Sea Surface and Global Change*, P. S. Liss and R. A. Duce, Eds., Cambridge University Press, xiii–xvi.
- , and J. E. Lovelock, 2007: Climate change: The effect of DMS emissions. *Environ. Chem.*, **4**, 377–378, <https://doi.org/10.1071/EN07072>.
- , and Coauthors, 2014: Short-lived trace gases in the surface ocean and the atmosphere. *Ocean–Atmosphere Interactions of Gases and Particles*, P. S. Liss and M. T. Johnson, Eds., Springer, 1–54.
- Longhurst, A., S. Sathyendranath, T. Platt, and C. Caverhill, 1995: An estimate of global primary production in the ocean from satellite radiometer data. *J. Plankton Res.*, **17**, 1245–1271, <https://doi.org/10.1093/plankt/17.6.1245>.
- Mace, K. A., N. Kubilay, and R. A. Duce, 2003a: Organic nitrogen in rain and aerosol in the eastern Mediterranean atmosphere: An association with atmospheric dust. *J. Geophys. Res.*, **108**, 4320, <https://doi.org/10.1029/2002JD002997>.
- , R. A. Duce, and N. W. Tindale, 2003b: Organic nitrogen in rain and aerosol at Cape Grim, Tasmania, Australia. *J. Geophys. Res.*, **108**, 4338, <https://doi.org/10.1029/2002JD003051>.
- , P. Artaxo, and R. A. Duce, 2003c: Water-soluble organic nitrogen in Amazon Basin aerosols during the dry (biomass burning) and wet seasons. *J. Geophys. Res.*, **108**, 4512, <https://doi.org/10.1029/2003JD003557>.
- Martin, J. H., 1990: Glacial–interglacial CO₂ change: The iron hypothesis. *Paleoceanography*, **5**, 1–13, <https://doi.org/10.1029/PA005i001p00001>.
- , and S. E. Fitzwater, 1988: Iron deficiency limits phytoplankton growth in the northeast Pacific Subarctic. *Nature*, **331**, 341–343, <https://doi.org/10.1038/331341a0>.
- Meskhidze, N., and Coauthors, 2011: Global distribution and climate forcing of marine organic aerosol: 1. Model improvements and evaluation. *Atmos. Chem. Phys.*, **11**, 11 689–11 705, <https://doi.org/10.5194/acp-11-11689-2011>.
- Moore, C. M., and Coauthors, 2013: Processes and patterns of oceanic nutrient limitation. *Nat. Geosci.*, **6**, 701–710, <https://doi.org/10.1038/ngeo1765>.

- Moore, R. M., J. E. Milley, and A. Chatt, 1984: The potential for biological mobilization of trace elements from aeolian dust in the ocean and its importance in the case of iron. *Oceanol. Acta*, **7**, 221–228.
- Mosher, B. W., and R. A. Duce, 1987: A global atmospheric selenium budget. *J. Geophys. Res.*, **92**, 13 289–13 298, <https://doi.org/10.1029/JD092iD11p13289>.
- Myriokefalitakis, S., and Coauthors, 2018: Reviews and syntheses: The GESAMP atmospheric iron deposition model intercomparison study. *Biogeosciences*, **15**, 6659–6684, <https://doi.org/10.5194/bg-15-6659-2018>.
- National Academies of Sciences, Engineering, and Medicine, 2016: *The Future of Atmospheric Chemistry Research: Remembering Yesterday, Understanding Today, Anticipating Tomorrow*. National Academies Press, 226 pp., <https://doi.org/10.17226/235730>.
- O'Dowd, C. D., and G. de Leeuw, 2007: Marine aerosol production: A review of the current knowledge. *Philos. Trans. Roy. Soc.*, **365A**, 20072043, <https://doi.org/10.1098/rsta.2007.2043>.
- , and Coauthors, 2004: Biogenically driven organic contribution to marine aerosol. *Nature*, **431**, 676–680, <https://doi.org/10.1038/nature02959>.
- , and Coauthors, 2015: Connecting marine productivity to sea-spray via nanoscale biological processes: Phytoplankton dance or death disco? *Sci. Rep.*, **5**, 11, <https://doi.org/10.1038/srep14883>.
- Okin, G. S., and Coauthors, 2011: Impacts of atmospheric nutrient deposition on marine productivity: Roles of nitrogen, phosphorus, and iron. *Global Biogeochem. Cycles*, **25**, <https://doi.org/10.1029/2010GB003858>.
- Ovadnevaite, J., and Coauthors, 2011: Primary marine organic aerosol: A dichotomy of low hygroscopicity and high CCN activity. *Geophys. Res. Lett.*, **38**, L21806, <https://doi.org/10.1029/2011GL048869>.
- Owens, N. J. P., J. N. Galloway, and R. A. Duce, 1992: Episodic atmospheric nitrogen deposition to oligotrophic oceans. *Nature*, **357**, 397–399, <https://doi.org/10.1038/357397a0>.
- Pan, B., and Coauthors, 2018: Impacts of Saharan dust on Atlantic regional climate and implications for tropical cyclones. *J. Climate*, **31**, 7621–7644, <https://doi.org/10.1175/JCLI-D-16-0776.1>.
- Patterson, C. C., and D. M. Settle, 1987: Review of data on eolian fluxes of industrial and natural lead to the lands and seas in remote regions on a global scale. *Mar. Chem.*, **22**, 137–162, [https://doi.org/10.1016/0304-4203\(87\)90005-3](https://doi.org/10.1016/0304-4203(87)90005-3).
- Prospero, J. M., M. Uematsu, and D. L. Savoie, 1989: Mineral transport to the Pacific Ocean. *SEAREX: The Sea/Air Exchange Program*, J. P. Riley, R. Chester, and R. A. Duce, Eds., Vol. 10, *Chemical Oceanography*, Academic Press, 187–218.
- Qiu, C., and R. Zhang, 2013: Multiphase chemistry of atmospheric amines. *Phys. Chem. Chem. Phys.*, **15**, 5738–5752, <https://doi.org/10.1039/C3CP43446J>.
- Quinn, P. K., and T. S. Bates, 2011: The case against climate regulation via oceanic phytoplankton sulphur emissions. *Nature*, **480**, 51–56, <https://doi.org/10.1038/nature10580>.
- , and Coauthors, 2014: Contribution of sea surface carbon pool to organic matter enrichment in sea spray aerosol. *Nat. Geosci.*, **7**, 228–232, <https://doi.org/10.1038/ngeo2092>.
- , D. B. Collins, V. H. Grassian, K. A. Prather, and T. S. Bates, 2015: Chemistry and related properties of freshly emitted sea spray aerosol. *Chem. Rev.*, **115**, 4383–4399, <https://doi.org/10.1021/cr500713g>.
- Rayman, M. P., 2000: The importance of selenium to human health. *Lancet*, **356**, 233–241, [https://doi.org/10.1016/S0140-6736\(00\)02490-9](https://doi.org/10.1016/S0140-6736(00)02490-9).
- Rinaldi, M., and Coauthors, 2013: Is chlorophyll *a* the best surrogate for organic matter enrichment in submicron primary marine aerosol? *J. Geophys. Res.*, **118**, 4964–4973, <https://doi.org/10.1002/jgrd.50417>.
- Russell, L. M., L. N. Hawkins, A. A. Frossard, P. K. Quinn, and T. S. Bates, 2010: Carbohydrate-like composition of submicron atmospheric particles and their production from ocean bubble bursting. *Proc. Natl. Acad. Sci. USA*, **107**, 6652–6657, <https://doi.org/10.1073/pnas.0908905107>.
- Saiz-Lopez, A., J. M. C. Plane, A. R. Baker, L. J. Carpenter, R. V. Glasow, J. C. G. Martin, G. McFiggans, and R. W. Saunders, 2012: Atmospheric chemistry of iodine. *Chem. Rev.*, **112**, 1773–1804, <https://doi.org/10.1021/cr200029u>.
- , and Coauthors, 2015: Injection of iodine to the stratosphere. *Geophys. Res. Lett.*, **42**, 6852–6859, <https://doi.org/10.1002/2015GL064796>.
- Satheesh, S. K., and K. K. Moorthy, 2005: Radiative effects of natural aerosols: A review. *Atmos. Environ.*, **39**, 2089–2110, <https://doi.org/10.1016/j.atmosenv.2004.12.029>.
- Schulz, M., and Coauthors, 2012: Atmospheric transport and deposition of mineral dust to the ocean: Implications for research needs. *Environ. Sci. Technol.*, **46**, 10 390–10 404, <https://doi.org/10.1021/es300073u>.
- Sellegrì, K., and Coauthors, 2016: Evidence of atmospheric nanoparticle formation from emissions of marine microorganisms. *Geophys. Res. Lett.*, **43**, 6596–6603, <https://doi.org/10.1002/2016GL069389>.
- Shao, Y., and Coauthors, 2011: Dust cycle: An emerging core theme in Earth system science. *Aeolian Res.*, **2**, 181–204, <https://doi.org/10.1016/j.aeolia.2011.02.001>.
- Sieburth, J. M. N., and Coauthors, 1976: Dissolved organic matter and heterotrophic microneuston in surface microlayers of North Atlantic. *Science*, **194**, 1415–1418, <https://doi.org/10.1126/science.194.4272.1415>.
- Sorooshian A., and Coauthors, 2009: On the link between ocean biota emissions, aerosol, and maritime clouds: Airborne, ground, and satellite measurements off the coast of California. *Global Biogeochem. Cycles*, **23**, GB4007, <https://doi.org/10.1029/2009GB003464>.
- Sun, L., X. Li, T. Hede, Y. Q. Tu, C. Leck, and H. Agren, 2014: Molecular dynamics simulations reveal the assembly mechanism of polysaccharides in marine aerosols. *Phys. Chem. Chem. Phys.*, **16**, 25 935–25 941, <https://doi.org/10.1039/C4CP03423F>.
- Thomas, M. A., and Coauthors, 2010: Quantification of DMS aerosol-cloud-climate interactions using the ECHAM5-HAMMOZ model in a current climate scenario. *Atmos. Chem. Phys.*, **10**, 7425–7438, <https://doi.org/10.5194/acp-10-7425-2010>.
- Thornton, D. C. O., S. D. Brooks, and J. Chen, 2016: Protein and carbohydrate copolymer particles in the sea surface microlayer (SML). *Front. Mar. Sci.*, **3**, 135–143, <https://doi.org/10.3389/fmars.2016.00135>.
- Uematsu, M., R. A. Duce, S. Nakaya, and S. Tsunogai, 1985: Short-term temporal variability of eolian particles in surface waters of the northwestern North Pacific. *J. Geophys. Res.*, **90**, 1167–1172, <https://doi.org/10.1029/JC090iC01p01167>.
- Vergara-Temprado, J., and Coauthors, 2017: Contribution of feldspar and marine organic aerosols to global ice nucleating particle concentrations. *Atmos. Chem. Phys.*, **17**, 3637–3658, <https://doi.org/10.5194/acp-17-3637-2017>.

- Wang, X. F., and Coauthors, 2015: Microbial control of sea spray aerosol composition: A tale of two blooms. *ACS Cent. Sci.*, **1**, 124–131, <https://doi.org/10.1021/acscentsci.5b00148>.
- Wang, Y., A. Khalizov, M. Levy, and R. Zhang, 2013: Light absorbing aerosols and their atmospheric impacts. *Atmos. Environ.*, **81**, 713–715, <https://doi.org/10.1016/j.atmosenv.2013.09.034>.
- , and Coauthors, 2014a: Assessing the effects of anthropogenic aerosols on Pacific storm track using a multiscale global climate model. *Proc. Natl. Acad. Sci. USA*, **111**, 6894–6899, <https://doi.org/10.1073/pnas.1403364111>.
- , R. Zhang, and R. Saravanan, 2014b: Asian pollution climatically modulates mid-latitude cyclones following hierarchical modeling and observational analysis. *Nat. Commun.*, **5**, 3098, <https://doi.org/10.1038/ncomms4098>.
- , K.-H. Lee, Y. Lin, M. Levy, and R. Zhang, 2014c: Distinct effects of anthropogenic aerosols on tropical cyclones. *Nat. Climate Change*, **4**, 368–373, <https://doi.org/10.1038/nclimate2144>.
- Ward, B. A., S. Dutkiewicz, C. M. Moore, and M. J. Follows, 2013: Iron, phosphorus, and nitrogen supply ratios define the biogeography of nitrogen fixation. *Limnol. Oceanogr.*, **58**, 2059–2075, <https://doi.org/10.4319/lo.2013.58.6.2059>.
- Wilson, T. W., and Coauthors, 2015: A marine biogenic source of atmospheric ice-nucleating particles. *Nature*, **525**, 234–238, <https://doi.org/10.1038/nature14986>.
- Wurl, O., and M. Holmes, 2008: The gelatinous nature of the sea-surface microlayer. *Mar. Chem.*, **110**, 89–97, <https://doi.org/10.1016/j.marchem.2008.02.009>.
- Xu, W., and R. Zhang, 2012: Theoretical investigation of interaction of dicarboxylic acids with common aerosol nucleation precursors. *J. Phys. Chem.*, **116**, 4539–4550, <https://doi.org/10.1021/jp301964u>.
- Yang, S., and N. Gruber, 2016: The anthropogenic perturbation of the marine nitrogen cycle by atmospheric deposition: Nitrogen cycle feedbacks and the ¹⁵N Haber-Bosch effect. *Global Biogeochem. Cycles*, **30**, 1418–1440, <https://doi.org/10.1002/2016GB005421>.
- Zhang, R., 2010: Getting to the critical nucleus of aerosol formation. *Science*, **328**, 1366–1367, <https://doi.org/10.1126/science.1189732>.
- , G. Li, J. W. Fan, D. L. Wu, and M. J. Molina, 2007: Intensification of Pacific storm track linked to Asian pollution. *Proc. Natl. Acad. Sci. USA*, **104**, 5295–5299, <https://doi.org/10.1073/pnas.0700618104>.
- , A. F. Khalizov, L. Wang, M. Hu, and W. Xu, 2012: Nucleation and growth of nanoparticles in the atmosphere. *Chem. Rev.*, **112**, 1957–2011, <https://doi.org/10.1021/cr2001756>.
- , and Coauthors, 2015: Formation of urban fine particulate matter. *Chem. Rev.*, **115**, 3803–3855, <https://doi.org/10.1021/acs.chemrev.5b00067>.
- Zhao, C., and Coauthors, 2018: Enlarging rainfall area of tropical cyclones by atmospheric aerosols. *Geophys. Res. Lett.*, **45**, 8604–8611, <https://doi.org/10.1029/2018GL079427>.
- Zhao, J., A. F. Khalizov, R. Zhang, and R. McGraw, 2009: Hydrogen bonding interaction of molecular complexes and clusters of aerosol nucleation precursors. *J. Phys. Chem.*, **113**, 680–689, <https://doi.org/10.1021/jp806693r>.
- Zoller, W. H., E. S. Gladney, and R. A. Duce, 1974: Atmosphere concentrations and sources of trace metals at the South Pole. *Science*, **183**, 198–200, <https://doi.org/10.1126/science.183.4121.198>.