Workshop on Iron at the Air-Sea Interface

26–30 July 2021
Online and onsite in Asheville, North Carolina, USA

From 26–30 July 2021, members of the international community with expertise in atmospheric chemistry, organic geochemistry, photochemistry, chemical and biological oceanography, dust/combustion aerosol geochemistry, remote sensing, and various aspects of atmospheric, ocean, and Earth system modeling were brought together to discuss the complicated problems of iron (Fe) speciation in the atmosphere, Fe biogeochemistry at the atmosphere-ocean interface and in the upper ocean, and the effects of atmospheric Fe deposition on ocean primary productivity and carbon dioxide (CO₂) uptake. Due to travel restrictions related to the ongoing pandemic, the meeting was conducted in a hybrid mode. The in-person portion of the meeting was held in Asheville, North Carolina and the online participants were able to join using two streaming platforms. Overall, seventy-four scientists from twenty-three countries registered to participate and to present their experimental, modeling, and remote sensing studies related to atmospheric supply and speciation of aerosol Fe, its contribution to the dissolved Fe inventory of the ocean, and its potential impacts on primary production and CO₂ uptake. The workshop brought together established scientists and young researchers (M.Sc.

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and Ph.D. students and early career researchers who received their Ph.D. within 7 years) to provide opportunities for professional interactions in a focused and productive forum. More than one-third of the participating scientists were female.

Aerosol Fe is a crucial source of micronutrients to the remote marine environment. Improved understanding of how aerosol Fe impacts the various biogeochemical-physical interactions and feedbacks between the atmosphere and the ocean is a key component of the United States SOLAS Science Plan and SOLAS-Japan.

The workshop was dedicated to an improved understanding of how atmospheric bioaccessible Fe moves across the atmosphere-ocean interface and becomes bioavailable for ocean ecosystems and at scales important for the carbon cycle.

The workshop agenda can be viewed here. The individual presentations and a keynote speech were followed by two discussion sessions and a summary session. All presentations were broadcast and recorded through Zoom. The two discussion sessions were focused on Fe biogeochemistry in the atmosphere and in the ocean, while the summary session focused on progress achieved since the 2018 workshop (Meskhidze et al., 2019) and future directions in the field. Charges to the working groups were laid out and distributed to all participants ahead of time. Three breakout groups (two online and one in person) were created for each discussion session. Each breakout group had a moderator/discussion leader and a rapporteur. The discussions in the breakout groups were organised around the workshop themes. The first discussion session was designed to identify the critical unresolved questions concerning the sources, chemical forms and transformations, lifetime, ocean deposition, and organic complexation of atmospherically delivered Fe. The second discussion session was designed to address ocean biological uptake mechanisms of Fe by the microbial community (phytoplankton and bacteria), chemical forms and distribution of Fe-binding ligands in seawater, and toxicity of aerosols. These discussions were summarised in presentations and short write-ups delivered by the breakout group leaders. The discus-
sions in the summary session were related to identifying and documenting the progress achieved since 2018 by modifying the level of understanding (as ‘low’, ‘medium’, or ‘high’) assigned to each topic in the Science Prioritization Matrix developed during the previous workshop (Meskhidze et al., 2019). In the end, a plenary discussion focused on identifying research priorities for advancing knowledge in the field.

Electronic poster presentations were carried out virtually using the SpatialChat virtual platform. Active discussions took place regarding understanding the atmospheric chemistry of Fe with organics relevant to cloud droplets and wet aerosols, diel variability of Fe in an estuarine surface microlayer, the effect of ocean acidification on the chemical speciation of Fe, the contribution of combustion Fe in marine aerosols over the north-western Pacific Ocean, soluble Fe deposition under the Coupled Model Intercomparison Project Phase 6 (CMIP6) scenarios, and machine learning approaches at the air-sea interface: neural networks for biogeochemistry. Participants were able to move freely between the posters and participate in online discussions by dragging their avatars towards each poster.

A few important take-home messages from the meeting were:

• The community should assemble a glossary that better defines and reconciles the different terminologies used by the atmospheric and oceanographic research communities.

• There is a need for standardisation of different aerosol materials (i.e., processed mineral dust, combustion aerosols, and various Fe-bearing organic and inorganic species) and experimental methods (i.e., the range of ocean microbes in laboratory measurements, mesocosms, and in-situ communities) that are used to assess the bioaccessibility and bioavailability of aerosol Fe. High-precision Fe isotope measurements can be used to aid such studies.

• Experimental approaches also need to consider the possible toxicity and biological competition /co-limitation effects of other trace elements (e.g., Cu, Cd, Mn, Pb), ligands, and organic compounds in aerosols.

• More research is needed on the longer-term fate of aerosol Fe during its weeks to months residence time in the surface mixed layer. This includes the effects of marine particles and the sea-surface microlayer on aerosol Fe biogeochemistry. In particular, in the sunlit water column the roles played by microorganisms in altering aerosol Fe bioavailability and residence time, particle micro-environments, aggregation/disaggregation, size (including colloids), the chemical composition and thickness of the surface microlayer, and the effect of microplastics should be investigated.

• It was recognised that reproducible methods now exist to characterize some of the organic compounds and classes complexing Fe in seawater and aerosols. However, as most of the operationally- (electrochemically-) defined Fe binding ligands in seawater remain uncharacterized, laboratory and field incubations and process studies are recommended to target certain ligands and oceanic regimes. There is also a need to better quantify organic ligands through intercomparison of the various measurement techniques in the ocean, aerosols and rain.

• In light of the recent advances in identifying specific Fe-organic ligand complexes in the atmosphere and seawater, there is a distinct need for a review or synthesis paper on the impacts of photochemistry on Fe redox speciation and complexation in both aerosols and seawater.

• Despite the enormous progress achieved over the past decade, it was recognised that atmospheric models don’t always include the complexities associated with aerosol Fe solubilities (especially the high values inferred from some field studies), size fractionation occurring during atmospheric transport, mineral aerosol composition at the source regions, and different sources and forms of aerosol Fe. Oceanic models need to better treat post-depositional
processes (e.g. in the sea-surface microlayer, dissolution, scavenging, complexation, aggregation) and Fe ligand types, their concentrations, and spatiotemporal distributions. It was suggested that the community should continue to recognize and utilize existing aircraft and satellite data.

To better disseminate the workshop results the attendees agreed to write a review paper. The paper will summarise the achievements since the last workshop and chart a path for improved understanding and characterisation of the mechanisms affecting Fe at the air-sea interface. To keep the momentum going, the attendees also agreed to hold virtual meetings once every two-to-three months with an invited speaker presentation.

Reference

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Cassandra Gaston is an Assistant Professor of Atmospheric Sciences at the Rosenstiel School of Marine and Atmospheric Science at the University of Miami, USA. Her research interests include physical and chemical measurements of aerosols, aerosol impacts on marine biogeochemical cycles, and heterogeneous reactions. She received her Ph.D. in 2012 in Oceanography from the Scripps Institution of Oceanography. She was then a postdoctoral researcher at the University of Washington in the Department of Atmospheric Sciences from 2012-2015.

Atmospheric transport of North African dust-bearing supermicron freshwater diatoms to South America: implications for iron transport to the equatorial North Atlantic Ocean

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Atmospheric aerosols from Africa are transported by the trade winds to the western equatorial North Atlantic Ocean every winter and spring and can contain nutrients, such as iron (Fe). In this study, we measured the size and composition of supermicron (particle diameter >1μm) aerosols collected at a site on the northeast coast of South America. Using electron microscopy, we found three distinct Fe-containing particle types: mineral dust, freshwater diatoms from African paleolakes, and pollen grains; all three particle types extended into the super-coarse mode with particle diameters >10μm. Particle asphericity, measured as the aspect ratio (AR_{perp}), increased with increasing particle size (see Figure 2, Barkley et al., 2021) and likely explains the long-range transport of super-coarse particles. Electron mapping of freshwater diatoms also revealed surficial Fe-rich inclusions. Once deposited in the ocean, the asphericity (high AR_{perp}) and light density of freshwater diatom particles likely increases their residence time and therefore, the time for Fe dissolution in the surface ocean compared to dust.

Reference
Catarina Guerreiro did her Ph.D. in Geology, in Portugal, after which she moved to Germany to start exploring the effects of Saharan dust on calcifying phytoplankton (coccolithophores) across the tropical Atlantic in 2015. In 2018, she was awarded a Marie Sklodowska-Curie Actions (MSCA) to continue her research at the University of Lisbon. She is currently leading the project ‘CHASing the environmental Effects of dust deposition across the Atlantic and Southern Ocean: a coccolithophore perspective (CHASE)’ in Lisbon, expanding her research towards the entire Atlantic Ocean to explore the role of dust in modulating the composition and distribution of coccolithophore communities and subsequent impact on the biological carbon pump.

Influence of dust on the export production of calcifying phytoplankton (coccolithophores): implications for the biological carbon pump

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Ongoing climate warming is likely to hamper marine phytoplankton productivity and change their community structure, by reducing the supply of nutrients from deep waters to the euphotic zone due to enhanced ocean stratification. This has cascading effects not only for the remaining marine food web, but also for the regulation of atmospheric carbon dioxide (CO₂) via the “biological carbon pump” (BCP). Amongst marine phytoplankton, coccolithophores (Haptophyta) are the main primary producers covering their cells with tiny calcite plates (the coccoliths), through a biogeochemical process that incorporates carbon in calcite and releases CO₂ into the environment. This provides them a unique ability for interacting with the marine carbon cycle in three ways: photosynthesis (CO₂ sink), calcification (CO₂ source) and carbon-burial in oceanic sediments (coccolith-ballasting). By crucially contributing to modulate the rain ratio, i.e., ratio of particulate inorganic carbon (PIC) to particulate organic carbon (POC) fluxes, a key parameter for biogeochemical models exploring the long-term efficiency of CO₂ drawdown (Hutchins, 2011), any changes in the coccolithophore communities will almost certainly contribute to change the earth’s climate. To explore the role of dust deposition in modulating their influence on the rain ratio and functioning of the BCP, we have quantified 1 year of species-specific coccolith- and coccolith-calcium carbonate (CaCO₃) export fluxes collected at four sediment traps moorings (i.e., CB-20°N/21°W; M1-12°N/23°W; M2-14°N/37°W; M4-12°N/49°W) along a transatlantic transect underneath the largest dust plume originating from Africa.

The studied transect was characterised by very striking basin-scale ecological gradients, with
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fluctuations of deep-dwelling species Florisphaera profunda and Gladiolithus flabellatus clearly increasing towards west and fluxes by of fast-blooming surface-dwelling species Emiliania huxleyi and Gephyrocapsa oceanica increasing in the opposite direction (Figure 3). Such distribution is reflecting the geostrophic deepening of the nutrient further west, forced by the trade winds, and the increasing proximity to yearlong upwelling conditions towards offshore NW Africa (Guerreiro et al., 2019). Highest fluxes were unexpectedly found at the westernmost site M4, where the sea surface temperature (SST) is higher and the nutrient is deepest along the tropical North Atlantic (Figure 3). Ca 3-5 times higher coccolith fluxes, and 2-3 higher coccolith-\(\text{CaCO}_3\) fluxes at M4 compared to the other sites were mostly produced by deep-dwelling species, contributing to nearly half of the total carbonate flux at M4 (45%), much higher compared to 23% at M2 and 15% at M1 and CB. Highest Coccolith-\(\text{CaCO}_3/\text{POC}\) ratios and weak correlations between the carbonate of

Figure 3: Spatiotemporal variation of (a) daily precipitation rates (blue) and wind speed (black line), (b) seasonal mixed layer (MLD) from the NASA Ocean Biogeochemical Model (NOBM) (light blue, 2012–2013), MLD monthly climatology obtained from the in situ data measured by Argo floats (dashed curve, 2000–2018), and sea surface temperature (SST-red line), (c) UPZ/LPZ ratios (light green), sea surface Chlorophyll-a (Chl-a) concentrations (dark green line), (d) relative abundance of the most abundant taxa (>3%), (e) total coccolith export fluxes (black) at sites M4, M2, M1 and CB. Taxa with a similar spatiotemporal distribution were grouped (Gm=G. muellerae; Ge=G. ericsonii; Helico=H. spp.; Ress=R. sessilis; Rhabdo=R. spp.; Umbello=U. spp.; Eh=E. huxleyi; Go=G. oceanica; Clep=C. leptoporus; Umbili=U. spp.; Calcio=C. spp.; Syraco=S. spp.; Fp=F. profunda; Gi=G. flabellatus). UPZ/LPZ calculated from the ratio between the UPZ-species E. huxleyi and G. oceanica, and LPZ-species F. profunda and G. flabellatus (where UPZ and LPZ = Upper- and Lower Photic Zone). All data from Guerreiro et al. (2017, 2019).
deep-dwelling species and POC at M4 suggest that increasing productivity in the lower photic zone in response to ocean warming might enhance the rain ratio and reduce the coccolith-ballasting efficiency, thereby contributing to weaken the BCP (Guerreiro et al., 2019; 2021).

Superimposed to these large-scale ecological gradients, and despite the markedly stratified tropical ocean conditions and the greater distance to the dust sources in Africa, site M4 showed evidence suggestive of short-term dust-related enhanced productivity in the upper photic zone. The first event occurred in spring, with fluxes of POC, E. huxleyi and planktonic foraminifera strikingly increasing in response to dry dust deposition combined with some degree of wind-stirred water mixing. The second event occurred later in the fall, when the studied region was under the seasonal influence of the Intertropical Convergence Zone, with E. huxleyi and G. oceanica, as well as diatoms, strikingly increasing in response to wet dust deposition (Figure 4). The presence of a sea surface salinity minimum during this period suggests that the Amazon waters were probably contributing both nutrients and buoyancy for retaining the dust-born nutrients in the sunlit layer of the ocean at M4, thereby providing optimum nutrient and light conditions at the surface (Guerreiro et al., 2017). Interestingly, both dust-related productivity events resulted in lower Coccolith-CaCO$_3$/POC ratios (Figure 4) and higher biogenic silica (bSiO$_2$)/CaCO$_3$ ratios. In addition, and despite their smaller-sized coccoliths, carbonate fluxes produced by these surface-dwellers were found to have a stronger correlation to POC along the transect, even compared to larger-sized coccolith taxa and to the dominant deep-dwelling species, indicating that the efficiency of coccolith-ballasting is ecologically dependent (Guerreiro et al., 2021). Overall, our study suggests that increasing Saharan dust outbreaks (e.g., Mirzabaev et al., 2019) might contribute to compensate the projected weakening of the BCP, either by providing nutrients to fuel fast-blooming phytoplankton and/or by stimulating the export of POC via both dust- and coccolith-ballasting.

Figure 4: Spatiotemporal distribution of (a) fluxes of particulate organic carbon (POC) (in green) and calcium carbonate (CaCO$_3$) produced by UPZ species (black line); (b) fluxes of mineral dust (in orange) and Coccolith-CaCO$_3$/POC ratio (black line), at trap sites M4 and M2. Data from Korte et al. (2017), van der Does et al. (2020) and Guerreiro et al. (2021).

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How fast does the iron cycle spin in the open ocean?

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Iron (Fe) is an essential component of the photosynthetic apparatus, nitrogenase, and many other enzymes that drive biogeochemically relevant reactions. Thanks to the International GEOTRACES program, we know much more about the spatial distribution of Fe across the global ocean than we did 10 years ago, but the timescales that characterise the Fe cycle remain poorly resolved. This has led to concern about how well biogeochemical models can simulate past or future changes in the marine Fe cycle and how such changes would affect phytoplankton growth and carbon storage in the oceans.

In June 2019, the EAGER Chief Scientist Training cruise provided an opportunity to build an Fe budget at Station ALOHA (A Long-Term Oligotrophic Habitat Assessment; 22.75°N, 158°W), the site of the Hawaii Ocean Time-series (HOT) within the North Pacific Subtropical Gyre. During this cruise, dissolved Fe inventories were similar to past investigations (Boyle et al., 2005; Fitzsimmons et al., 2015), showing a maximum in the surface, a relative minimum at 100 m near the deep chlorophyll maximum layer, followed by increasing concentrations through the mesopelagic zone. The sinking flux of Fe was measured at multiple depths in the upper water column, allowing total Fe turnover times to be calculated. Finally, the uptake of dissolved Fe onto particles was estimated using a novel low-level stable isotope approach enabled by the precision of multi-collector inductively coupled plasma mass spectrometry (MC-ICPMS).
The resulting Fe budget provides several key constraints on the Fe cycle at North Pacific Subtropical Gyre. First, it takes ~3.5 days for dust to sink out the surface mixed layer, a duration that should be considered when conducting solubility experiments (which are typically performed for seconds to hours). Second, the dissolved Fe pool in the surface ocean appears to be recycled by the microbial ecosystem many times before being exported. Third, roughly 10% of aerosol Fe must dissolve into seawater to balance our budget. This is higher than observed in leaching experiments with desert dust aerosols and could mean that ecosystem processes that are not replicated by leaching experiments (e.g. zooplankton grazing and acidic digestion in their gut) contribute to Fe solubilisation. Alternatively, there could be a significant and highly soluble contribution of anthropogenic Fe to Station ALOHA, in addition to desert dust (Ito et al., 2019). This may alter the ecology observed there, especially the success of nitrogen fixing cyanobacteria, which have high Fe requirements (Letelier et al., 2019).

References


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Joan Llort is a biogeochemical oceanographer interested on the influence of dust and wildfires aerosols on phytoplankton growth and carbon export. He obtained his Ph.D. at the Laboratoire d’Océanographie et du Climat: Expérimentations et Approches Numériques (LOCEAN) laboratory (Paris, France) and spent three years as a postdoc at the Institute for Marine and Antarctic Studies (IMAS, Hobart, Australia). In 2019, he joined the Barcelona Supercomputing Centre in Spain.

Large-scale phytoplankton response to pyrogenic iron


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Droughts and climate-change-driven warming are leading to more frequent and intense wildfires (Abatzoglou et al., 2019, Bowman, 2020), arguably contributing to the severe 2019–2020 Australian wildfires (Figure 7; Abram et al., 2021). Aerosol emissions from wildfires can lead to the atmospheric transport of macronutrients and trace metals such as nitrogen and iron (Fe), respectively (Guieu et al., 2005, Schlosser et al., 2017). It has been suggested that the oceanic deposition of wildfire aerosols can relieve nutrient limitations and, consequently, enhance marine productivity (Ito et al., 2011, Hamilton et al., 2022), but direct observations are lacking. Here we use satellite and atmospheric reanalysis data to evaluate the effect of 2019–2020 Australian wildfire aerosol deposition on phytoplankton productivity. We find anomalously widespread phytoplankton blooms from December 2019 to March 2020 in the Southern Ocean downwind of Australia. Aerosol samples originating from the Australian wildfires contained a high Fe content and atmospheric trajectories show that these aerosols were likely to be transported to the bloom regions, suggesting that the blooms resulted from the fertilization of the iron-limited waters of the Southern Ocean. Our

Figure 7: Satellite image of fires burning in the Eastern Coast of Australia in December 2019 (Source: ESA-Sentinel).
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Wildfires, a significant contributor to atmospheric iron and other essential nutrients to seawater South-East of Australia

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In the Southern Ocean (south of 30°S), sub-nanomolar concentrations of bioavailable iron (Fe) in surface waters drastically limits phytoplankton growth and its associated feedback on the carbon cycle and climate (Boyd, 2015). Aeolian deposition is a major pathway for Fe supply to this remote oceanic region.

While dust dominates the total atmospheric Fe loading, it is highly insoluble (therefore poorly bioavailable) in seawater (Jickells et al., 2005). Although aeolian transport is known to result in enhanced mineral Fe solubility in aerosols down the atmospheric pathway, this phenomena alone cannot explain high aerosol Fe solubility (>10%) reported over the Southern Ocean (Ito et al., 2020). Combustion emissions, be it from anthropogenic fuel and biomass burning or from wild forest fires, also contain a small fraction of Fe, the latter which is thought to be significantly more soluble in seawater compared to mineral Fe (Oakes et al., 2012). In addition, combustion sources release acids (i.e., sulfur and nitrogen oxides) and organic (i.e., oxalate) compounds to the atmosphere which are known to facilitate Fe dissolution at the surface of mineral aerosols, resulting in enhanced Fe bioavailability upon deposition to the ocean (Baker et al., 2021).

Atmospheric deposition to the Southern Ocean is either dominated by dust or fire emissions (Hamilton et al., 2020), depending on the season. However, the highly episodic and hardly predictable nature of these sources make it difficult to constrain their variability in space and time. In 2019, Meskhidze and co-workers (2019) recommended the development of atmospheric time-series monitoring stations in the Southern Hemisphere in order to capture such episodic atmospheric features (Meskhidze et al., 2019).

In our recent study (Perron et al., manuscript in preparation), we report analysis of weekly aerosol sampling from 2016 to 2020 at the time-series atmospheric sampling station of kunanyi/Mt Wellington, in Tasmania (Australia, Figure 9, 10). Observations revealed a significant increase in atmospheric concentrations of bio-essential nutrients (including Fe, manganese (Mn), nitrate (NO₃⁻), and ammonium (NH₄⁺)), as well as lithogenic tracers (aluminium (Al) and titanium (Ti)), and anthropogenic aerosols (lead (Pb), vanadium, (V)) on

Morgane Perron studied Marine Chemistry in France and moved to Australia in January 2015 to undertake a Master’s degree internship. She continued her research in Hobart, Tasmania, Australia, to undertake a Ph.D. then a 2-year postdoc investigating trace metals contained in aerosols and their transport to the Southern Ocean.
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Fire-dominated days when compared to dust-dominated days. Further distinction between the atmospheric signal from local and proximal fire events (Tasmanian, summer 2018-2019) compared to large-scale distant (mainland Australia, summer 2019-2020) wildfires highlighted an important role of atmospheric processing in enhancing atmospheric solubility of aeolian Fe and creating secondary organic (NO$_3^-$) and inorganic (NH$_4^+$) aerosols during the aeolian transport of the fire plume. Estimates of atmospheric dust loading based on measurements of the lithogenic tracer, Ti, in aerosols highlighted significant dust entrainment occurs along with the pyrogenic cloud. This hypothesis was further confirmed by the constant enrichment factor in Fe, close to that of the averaged Earth crust, in all kunanyi aerosols regardless of their prevailing source. As the magnitude and occurrence of large forest fires will likely increase with changing climate, it is essential that these findings are implemented into global modelling studies (both atmospheric and oceanic) in order to better constrain the response of marine ecosystems to atmospheric deposition of both vital nutrients (Fe, Nitrogen (N)) and pollutants (Pb) from pyrogenic sources.

**Figure 9:** Whisker representation of the estimated atmospheric dry deposition of (a) total Iron (Fe), F(T$_{Fe}$), (b) labile Fe, F(L$_{Fe}$), and (c) soluble Fe, F(S$_{Fe}$), in micromole of Fe per square meter per day ($\mu$mol m$^{-2}$ d$^{-1}$) as well as (d) mineral dust flux estimates in milligram per square meter per day (mg m$^{-2}$ d$^{-1}$) at kunanyi over autumn, spring, summer and fire seasons.

**Figure 10:** Summary map of published and new estimates of average atmospheric dry deposition of total (T$_{Fe}$) and soluble Fe (S$_{Fe}$), in micromole per square meter per day, based on field measurements over and south of Australia. Contours colors indicate this study (red), land-based studies (blue) and ship-based studies (green). A star indicates that the flux unit was converted from the original literature to match units in this study.

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