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*Editorial*

# Ocean Contributions to the Marine Boundary Layer Aerosol Budget

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Projections of future climate remain an important scientific goal for much of the Earth science community. However, uncertainty in the origin, number size distribution, and chemical composition of background aerosol complicates the assessments of direct and indirect anthropogenic aerosol forcing and makes it difficult for Earth system science models to accurately predict 21st century climate change. Because effects on climate are estimated from the differences between model simulations with present-day and preindustrial aerosol and precursor emissions, concerns over the global aerosol budget in the absence of anthropogenic influence have sparked renewed interest in the natural emissions of trace gases (acting as particle precursors) and aerosols. A reduction of the uncertainties in the representation of natural aerosols in climate models requires an improved quantification of their number, size distribution, chemical composition, and hygroscopic properties in the marine boundary layer (MBL). However, measurements of particle properties over the ocean are challenging because of the vast spatiotemporal variability and low concentration. Due in part to a lack of the sort of measurements needed to constrain models and also, in part, to the complex feedbacks between ocean and atmosphere responding to a wide range of stress factors, marine aerosol–cloud interaction remains the largest uncertainty in current climate models.

This Special Issue is motivated by our perceived need to provide a platform for current discussions regarding the oceanic contribution to the MBL aerosol, cloud condensation nuclei (CCN), and ice nucleating particle (INP) number concentration over different parts of the oceans. Significant research was performed in the 1950s–1990s following the discovery and initial characterization of the bubble bursting process and suggested link between oceanic phytoplankton and dimethyl sulfide (DMS) emissions to the atmosphere. Size-dependent production flux of primary sea spray aerosol (SSA) particles, along with its dependence on environmental variables such as wind speed, have by now been introduced into models. Models also include oceanic DMS production and its effect on the atmospheric sulfur cycle.

Renewed interest in the subject was sparked in the 2000s, when a number of studies identified large fractions of organic material in the ambient marine aerosol. Globally diverse settings were involved and this motivated an upsurge in the laboratory, field, satellite, and modeling research. These studies aimed to better characterize marine primary organic aerosol production mechanisms, aerosol mass and number fluxes, size-dependent sea spray enrichment factors, the split between water-soluble and -insoluble organic fractions, the chemical composition of organic aerosol, as well as the effect of marine organics on cloud microphysical properties. Parameterizations for oceanic emission rates of

biogenic trace gases (e.g., DMS, isoprene, monoterpenes, and iodocarbons) which lead to secondary aerosol production in the MBL have also been developed.

Recent studies have highlighted yet another dimension of the problem. It was shown that achieving high confidence in Earth system model simulations critically depends upon the implementation of more realistic ocean ecosystem–aerosol interactions. Feedbacks that operate on multiple spatiotemporal scales must be incorporated. Studies have shown that the production rates and physiological properties of SSA and the resulting efficacy to act as CCN or INP can be strongly influenced by the ocean's biogeochemical state (nutrient availability, pH, bacterial and phytoplankton abundance, eco-physiological factors, and evolutionary drivers). Our Special Issue presents a snapshot of current research topics in this study area. It comprises seven peer-reviewed, open access articles spanning the breadth of the field.

The importance of marine biogeochemical process representations in Earth system models is highlighted in the study by Ogunro et al. [1] The authors developed an International Ocean Model Benchmarking package and used it to evaluate surface ocean concentrations and the sea–air fluxes of DMS. This study shows that with a dynamic representation of DMS, models tend to over-predict sea surface concentrations in the productive region of the eastern tropical Pacific by almost a factor of two, and the sea–air fluxes by a factor of three. It is proposed that systematic model-data benchmarking will help to identify and lead to improved subgrid-scale parameterizations and Earth system model development.

A number of contributions address nascent and aged sea spray aerosol and offer indications or suggestions for their improved representation in Earth System Models. Carter-Fenk and Allen [2] use myristic, palmitic, and stearic acids to study the effect of fatty acids on nascent SSA. They also varied the pH of laboratory aqueous media to mimic the aging of nascent SSA in the MBL. The results of their study provide improved characterization of interfacial morphological changes throughout various stages of SSA aging in the marine boundary layer. Improved characterization of particle aging may lead to better assessments of SSA radiative effects and the reduction of the degree of uncertainty in their representation in climate models. Schill et al. [3] investigated the impact of divalent cations on the enrichment of soluble saccharides in primary SSA. By adding saccharides (glucose and glucuronic acid) and surfactants (palmitic acid) to artificial seawater, these authors show that divalent cations (i.e., $Mg^{2+}$) mediate the co-adsorption of soluble organics to insoluble surfactants at the ocean surface. This interaction may well contribute to the enrichment of soluble saccharides in SSA. While the proposed mechanism of ion-mediated co-adsorption of soluble organics to insoluble surfactants may not be solely responsible for the differences between models of SSA production and observations, it will undoubtedly lead to the improved representation of SSA chemical composition in current approaches. Elliott et al. [4] applied the principles of Gibbs phase plane chemistry across the entire ocean–atmosphere interface to investigate aerosol generation and geophysical transfer issues. Their study shows that the incorporation of marine surface tension at the sea–air boundary leads to (1) the reduction in turbulence and (by extension) laminar gas–energy diffusion; and (2) the alteration of the bubble film mass emission into the boundary layer. Their concepts for water–air adsorption theory will be included in the OCEANFILMS aerosol emissions package and likely lead to more accurate SSA emissions in climate models.

The study by Merkulova et al. [5] is devoted to the investigation of the effect of ocean emissions on aerosol optical depth (AOD) using remotely sensed data. By combining Moderate Resolution Imaging Spectroradiometer (MODIS) and Aerosol Robotic Network (AERONET) AOD retrievals with 2 m wind speed data, the study develops a new power–law relationship between AOD and surface–wind over the oceans.

The contribution by Menzo et al. [6] utilizes a reduced-complexity climate model known as Hector, but applies it in a completely novel manner. The authors conduct a regional scale analysis quantifying the possible effects climate change may have on DMS emissions within the oceans. Their findings suggest that natural sulfur emissions may exert a forcing up to 4 times that of the $CO_2$ marine feedback,

and that it may be possible to reverse the radiative forcing sign at low latitudes. This study is an attempt to illustrate the importance of a dynamic ocean–atmosphere feedback within the sulfur cycle on the global radiative budget.

Due to the extraordinary complexity of contemporary scientific research, it is often challenging to communicate the results, such as the above, to the general public. This is particularly true for policymakers, who may be dedicated professionals in their own right but have only limited understanding of the feedback-intensive global climate system. To address this problem, Menzo [7] has developed a web-based strategy specifically designed to convey marine biogeochemical feedback concepts to lawmakers and their colleagues as may be necessary. The information is provided as an interactive, public domain website. It is firmly grounded in the technical literature yet structured specifically for accessibility to non-scientists. Every page on the website includes images or videos to foster engagement with its technically challenging (and often heavily aerosol-oriented) content. It is hoped and expected that such a website with its tailored pedagogical value may contribute to better-informed policy formulation and legislation.

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