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FIRST REACTIONS

Ocean Sea Spray, Clouds, and Climate

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Sea spray aerosol (SSA) particles provide the connection between the ocean and the atmosphere with a profound and not yet fully understood impact on climate; in this issue, in related papers, teams lead by Kimberly Prather and Timothy Bertram have completed studies that together shed light on important factors in SSA composition and properties.[1] What has been known is that aerosols scatter solar radiation and nucleate clouds and ice particles, which further scatter, reflect, and absorb radiation efficiently, depending upon wavelength. For the last 20 years, measurements of the composition of atmospheric aerosols at different locations found that they consist of rich mixtures with a large organic content. The oceanic marine phytoplankton is an important yet little understood ecosystem controlling the organic composition at the sea surface where SSA are generated.[2] The transfer of organic material from the ocean to these aerosols is influenced by the ocean surface, which acts as a global separator of soluble and surface active organics. At the same time, the sea surface acts as an integrator of fluxes of material from the atmosphere, the ocean floor, and the bulk sea itself. Organic surfactant films exist on the surface of oceans as well as the large population of SSA affecting their atmospheric processing.[3] However, field observations have shown that the size, concentration, and composition of SSA particles varies dramatically between field studies as a consequence of an as yet unknown balance of chemical, sunlight driven, biological, and physical processes.[4]

In this issue, Wang et al.[1a] and Schill et al.[1b] present experimental results examining some of the assumptions used in "state-of-the-art" models and convincingly demonstrate that the simplifications used are inadequate to explain observations in the real atmosphere. Key to these studies is the oceanic water enclosure developed at Scripps Institute of Oceanography at UC San Diego[5] (Figure 1), which provides close to natural conditions while allowing control over environmental factors. This mesocosm (middle scale) experimental tool attempts to link the micro (molecular) and macro (ocean and atmosphere system) scales and consequently provides a connection between laboratory experiments, field, and model studies of SSA aimed at exploring their natural and anthropogenic effects on climate.

How do phytoplankton influence the makeup of sea spray aerosol? How does that affect cloud formation? Two papers this month begin to answer these questions.

Figure 1. The 3,400 gallon wave machine in which the studies were completed. Image credit: the American Chemical Society. Reprinted from ref 1a.

Model studies using simplifying assumptions in the treatment of the organic content of SSA fail to explain discrepancies between different field studies.

Model studies using simplifying assumptions in the treatment of the organic content of SSA fail to explain discrepancies between different field studies. The types of mesocosm studies presented here offer a rich opportunity to improve our understanding of the mechanisms connecting the chemistry of the ocean and the atmosphere. For example, historically, the influence of biological processes to aerosol

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organic content was estimated in models to be a single biological parameter, using chlorophyll a as a proxy for phytoplankton concentrations.[6] However, field observations give conflicting results as to the correlation between the organic content in SSA and chlorophyll a concentrations. This recent work[1a] provides evidence that chlorophyll a is not sufficient and points to other factors contributing to the composition of aerosols.

Using 3,400 gallons of natural seawater in a tank capable of mimicking natural breaking waves, two successive phytoplankton blooms were induced in a controlled environment over twenty-nine days. Each bloom gave SSA with different composition and properties.[1a] In the first bloom, aliphatic rich organics were enriched in submicrometer particles in correlation with chlorophyll a concentrations; by contrast, no such organic enrichment was found in the second bloom. These results are significant in informing the debate generated by field studies and challenge the simple relationship used in models between organic content of SSA and chlorophyll a concentration;[6] the overall chlorophyll a content was similar between the two blooms, but the organic SSA aerosol components were different—clearly something more is at play. Essentially what Prather and co-workers showed was that variations in the dynamics between the various types of microbes and their associated metabolic enzymes affected the concentrations of less water-soluble species such as lipids in seawater, resulting in smaller SSA particles. In turn, bursting bubbles from the waves launched these molecules into the atmosphere (Figure 2), explaining the variability in the organic content of aerosol produced in the two blooms.

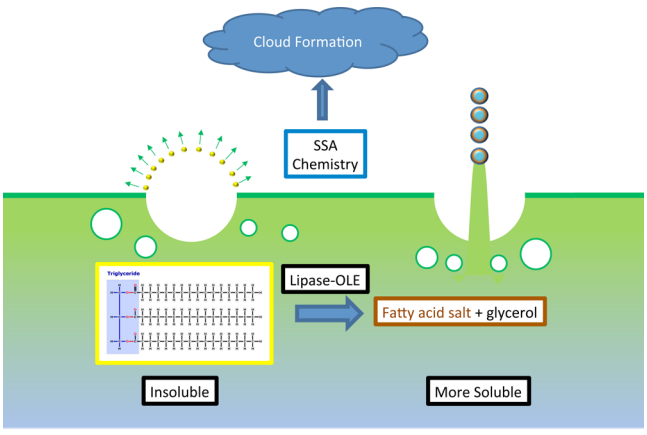

Figure 2. Differential enzymatic breakdown of phytoplankton metabolites results in populations of varying solubility. Insoluble species lead to small sea spray aerosol (SSA) particles while the more soluble products are associated with larger particles. The chemical makeup and particle size distribution of SSA influences cloud formation and climate. Image credit: Kimberly Prather.

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Additionally, one important factor in cloud nucleation and climate is the hygroscopicity of particles.[7] In treating this property, climate models are challenged by the heterogeneity in the particle's chemical composition and resulting SSA hygroscopicity since they typically assume that particles of the same size have identical composition. Schill et al.[1b] report results of high resolution measurements of cloud condensation nuclei (CCN) and show that the standard literature approach is greatly oversimplified. For this reason, models fail to explain the observed temporal changes in SSA particle hygroscopicity. The results presented make a compelling case for treating the variation in particle chemical composition to allow for a large number of particles of different composition in the real environment.

The interdisciplinary studies of Wang et al.[1a] and Schill et al.[1b] combine oceanography, biogeochemistry, and atmospheric chemistry to study some of the factors that determine the size, composition, and organic content of sea spray aerosol. Overall, the results expose the weakness of oversimplified model assumptions in understanding the variability in SSA organic content recorded in field observations. They demonstrate that all the tools available to scientists from laboratory single component and multicomponent systems to SSA mimics and nascent SSA from synthetic and mesocosm phytoplankton bloom experiments and field studies are required to understand the chemical complexity of sea spray aerosol particles needed to inform the development of predictive atmospheric chemistry and climate models.

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