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PRIMARY MARINE AEROSOL FLUXES

Progress and Priorities

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Primary marine aerosols generated from sea spray constitute the largest fraction of atmospheric aerosol loading over the remote open oceans. They have a direct climate effect through their influence on both solar and terrestrial radiation and an indirect climate effect because of their role in providing cloud condensation nuclei (CCN); they act as a sink for some atmospheric trace gases and may significantly modify heat, moisture, and momentum exchange across the air–sea interface in oceanic storms. Further, marine aerosol, sometimes known simply as sea spray, contributes to regional air quality by adding to the total particulate mass and through sea-to-air transfer of bacteria and viruses.

Eighteen atmospheric, oceanic, and aerosol scientists met for a day and a half at the National University of Ireland, Galway, to review current research of the production of sea spray at the air–sea interface. The meeting was divided into three sessions—micrometeorological methods for measuring the rate of particle production, the enrichment of aerosol with organic matter, and global estimates of sea spray production—and the findings of each section are summarized here. A list of workshop presentations is available online (www.cost-735.org/meetings/minutes/wg2may10galway.pdf.)

WORKSHOP ON PRIMARY MARINE AEROSOL FLUXES

WHAT: Representatives from nine countries met to review the state of the art for predicting the rate at which sea spray aerosol is created at the air–sea interface.
WHEN: 10–11 May 2010
WHERE: Galway, Ireland

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MICROMETEOROLOGICAL TECHNIQUES. In situ estimates of the sea spray source function (SSSF) have commonly been derived from an equilibrium approach in which a balance between production and deposition at the surface is assumed. The method relates production to near-surface particle concentration through a velocity scale. Multiple processes contribute to deposition: gravitational sedimentation, impaction on the surface via turbulent and Brownian motions, and scavenging by other spray droplets. All vary in magnitude with particle size. The appropriate velocity scales are not well defined: most rely on theoretical or modeling approaches and are largely unverified. This approach is falling out of favor as new technology makes more direct micrometeorological techniques viable.

Direct eddy covariance measurements and flux-profile techniques, while well established for other scalar fluxes, have been applied to marine aerosol fluxes only within the last 10 years and in only a handful of studies. Although the measurements are promising, they still require validation. Both techniques rely on the tenets of the the Monin–Obukhov similarity theory—namely, that the eddy covariance flux is constant with height near the sea surface when conditions are stationary and horizontally homogeneous and that the vertical gradient in concentration can be related to the surface flux. Neither assumption has been validated, however. Moreover, because aerosol particles have inertia, they may not follow turbulent motions perfectly; eddy covariance measurements may thus require corrections for inertial effects. As for profile measurements, current theory has not established a direct relation between the concentration profile and the surface flux of sea spray—that is, a flux-gradient relation. Some empirical evidence does suggest, however, that the vertical aerosol concentration profile near the surface is semilogarithmic, as are profiles that do obey Monin–Obukhov similarity.

Eddy covariance measurements are limited in their current applicability to particles with radii of about 5 μm and less. While larger particles typically dominate the sea spray mass, their number concentrations are so low that the rapid sampling required for eddy covariance measurements yields sampling statistics that are too noisy to provide reliable turbulent fluxes. Conversely, measurements based on average concentration can yield reliable profiles because of the longer averaging times that are permissible.

ORGANIC ENRICHMENT. It is increasingly recognized that organic enrichment of sea spray aerosol is important, influencing its effectiveness as CCN and its role in heterogeneous chemical interactions with trace gases. For submicron particles, there is general agreement that the organic enrichment increases with decreasing size, with some studies reporting enrichment factors of 70%–80% for the smallest sizes. With the advent of sophisticated online aerosol mass spectrometry measurements, it is becoming evident that organic spray aerosol mass can substantially exceed that of other submicron marine aerosol types produced via secondary pathways (e.g., non-sea-salt sulfate and methane sulfonic acid) under certain conditions. Substantial recent progress has been made in characterizing the chemical and physicochemical characteristics of organic sea spray. For example, sea spray has been shown to have characteristics quite different from terrestrial (natural and anthropogenic) primary and secondary organic aerosols. However, the disparate information from individual studies requires clearer interpretation, generalization, and synthesis.

The contribution of organic components of bubble-derived primary particles from both seawater and from algal exudate material has been characterized using state-of-the-art instrumentation down to dry diameters of tens of nanometers; the contribution has a clear dependence on bubble distribution characteristics. There is evidence that changes in the physics of bubble bursting by the presence of organic material (OM) determines the particle size distribution, but the important relationships (e.g., wind speed dependencies, aging of the organic material, algal bloom evolution) remain unquantified.

An investigation of the relationship between organic mass or mole fraction and key particle properties (e.g., dry to wet size) is desirable, as is the resolution of discrepancies between field and laboratory measurements (perhaps in terms of the ageing processes in the former). Deriving generic molar properties of marine OM would be most useful for predicting particle properties. There are also unresolved discrepancies between laboratory measurements arising in large part from the use of very different bubble-tank systems for generating the aerosol.

ASSESSING GLOBAL PRODUCTION. Sea spray's role in numerous physical and chemical processes brings a need to assess global production, including the organic fraction, and to incorporate spray production in general circulation, chemical transport, and climate models. To estimate global sea spray production, a suitable parameterization of the SSSF is combined with global datasets from satellites or models. For example, wind speed data from the National Aeronautics and Space Administration's (NASA's) Quick Scatterometer (QuikSCAT) and Special Sensor Microwave/Imager (SSM/I) or from weather prediction models can be used with existing SSSFs to derive sea spray flux maps. Satellite and in situ data are also increasingly being used to complement each other in the development of new SSSFs. A new development has been built on a combination of in situ measurements of organic matter enrichment and satellite retrievals of chlorophyll *a* concentrations from ocean color measurements [via the Moderate Resolution Imaging Spectroradiometer (MODIS) or Sea-viewing Wide Field-of-view Sensor (SeaWiFS) instruments] to produce a joint organic–inorganic SSSF that allows global estimates of the organic mass flux from sea spray to be made.

The accuracy of the estimates depends on both the quality of the global datasets and the choice of

SSSF. Sensitivity studies of sea spray flux show that although part of the uncertainty comes from error propagation from the satellite retrievals or model fields used, poorly constrained parameterizations of the SSSF cause the largest deviations from base estimates. Global and regional predictions of water insoluble organic material (WIOM)—or OM, dependent on the WIOM extraction used—show substantial sensitivities to both the chlorophyll fit and the SSSF. Comparison of organic aerosol loading in air masses with contributions from ocean waters with low and high biological productivity with low- and high-concentration bubble-tank measurements are required to evaluate and better constrain the parameterizations. It was noted that using chlorophyll as a measure of productivity is not unambiguous, but segregation of different algal dissolved organic material contributions from NASA level 2 satellite products (or level 4 where available) may help, as may high-spectral-resolution products from instruments such as the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) on the European Space Agency's Environmental Satellite (*Envisat*).

Another new development is the attempt to include forcing parameters other than wind speed in SSSFs. The performance of SSSFs based on whitecap fraction improves when sea state information, such as a wave-breaking parameter or dissipation energy, is included. Modeling results of sea spray production on a regional scale also emphasize the importance of wind fetch and local wave conditions on the predicted flux. Retrievals of whitecap fraction from satellite microwave emissivity measurements provide a more extensive dataset and much greater understanding of the geophysical variability of whitecaps than those obtained from in situ photographs. Whether for in situ validation of satellite retrievals or development of new and improved parameterizations, a major problem in these studies is the scarcity of high-quality in situ and satellite data pairs matched in time and space.

CONCLUSIONS. A final discussion session focused on assessing gaps in current knowledge and approaches to addressing them. The workshop identified the following needs:

- Validation studies and fundamental work on the physics of micrometeorological techniques applied to sea spray aerosol
- Development of new techniques to estimate the production rate of larger spray droplets where concentrations are too low for eddy covariance and gravitational sedimentation is significant
- An evaluation of the impact of different techniques on the organic enrichment of aerosols in laboratory studies, and development of recommendations for standardizing practice
- Determination of which environmental factors are most important in controlling aerosol production and organic enrichment and how they can be related to laboratory conditions
- An evaluation of key satellite and surface variables for global estimates and predictions of sea spray emissions and aerosol loading, and of satellite proxies for organic enrichment of sea spray
- In situ validation of satellite data products, along with development of corrections for biasing factors (e.g., bubbles on ocean color)
- An evaluation of techniques for interpolating over gaps in satellite data fields

As a closing bit of irony, almost every one of the scientists involved in this marine aerosol workshop had his or her travel disrupted, and in some cases cancelled, because of the aerosol cloud from the erupting Icelandic volcano Eyjafjallajökull.

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