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Meeting Report

Production mechanisms, number concentration, size distribution, chemical composition, and optical properties of sea spray aerosols

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Background

The impact of sea spray aerosols on global climate remains one of the most uncertain components of the aerosol–radiation–climate problem, but has received less attention than the impacts of terrestrial and anthropogenic aerosols. The last decade has produced a large body of information regarding the sources and composition of marine aerosols, resulting in a reassessment of the complex role that sea spray particles play in climate and various geophysical phenomena. As sea spray aerosol contributes substantially to the preindustrial, natural background which provides the baseline on top of which anthropogenic forcing should be quantified, and because the ocean covers over 70% of the Earth's surface, the representation of sea spray aerosol in climate models strongly influences the predicted impact on climate of anthropogenic aerosols via direct and indirect effects. In addition, climate change affects atmospheric parameters, such as wind speed which has controlling effect on the production of sea spray aerosol. Recent reviews on sea spray aerosol production and composition (de Leeuw *et al.*, 2011) summarized the state of the art and remaining uncertainties.

Over forty scientists from six countries convened in Raleigh, NC on June 4–6 2012 to review the status and prospects of sea spray aerosol research. Participants were researchers from the oceanography and atmospheric science communities, including academia, private industry, and government agencies. The workshop was held with the primary objectives of (1) identifying the most critical open questions regarding sea spray aerosol and developing a list of priorities for conducting and facilitating novel research and (2) ranking the most pressing science questions based on their feasibility impact on reducing the current uncertainty ranges for different processes. The four main focus groups followed by the three breakout sessions determined the most urgent questions that would improve quantification of sea spray aerosol-radiation-climate interactions, with special emphasis on the production flux, number concentrations, chemical composition, nucleation properties, and optical properties.

Workshop highlights

Instead of relying on individual presentations, the meeting format was structured to emphasize consensus-building among participants who collectively discussed successes, weaknesses, and research goals and methods of achieving them in their specific expertise of sea spray aerosol research. Following a keynote presentation by Gerrit de Leeuw highlighting open questions from his perspective, the workshop participants were divided into four focus groups (Figure 1): *in situ* field measurements, laboratory experiments, remote sensing, and regional and global modeling. Following short presentations by the group leaders in which successes and difficulties of past measurements/modeling/remote sensing efforts were reviewed and important missing parameters were identified, participants deliberated in breakout sessions to discuss three major topics: sea spray source function, atmospheric aging and budget, and optical properties. In each session, participants elaborated on the current sea spray aerosol-related research status and identified future priorities. In the following we reiterate the main discussion points summarized in each of the four main sessions.

In situ field measurements

The discussion in this session focused on methods to measure fundamental quantities of interest for determining the impact of sea spray aerosols on climate, specifically, size-resolved number concentrations and production fluxes with information on size-resolved chemical composition. Various participants described the need for measurements of additional parameters to better understand the sources and sinks of sea spray aerosol. Key among these were background meteorology, biogeochemistry and physics of the ocean mixed layer including surface roughness/wind stress, wave breaking and bubble plume dynamics, and whitecap fraction (defined as the fraction of the ocean surface covered by whitecaps generated by breaking waves). Much of the discussion was centered on the difficulties quantifying the sources of sea spray aerosol, especially the relationship between the ocean surface whitecap fraction and aerosol production. The difficulties of obtaining accurate measurements of sea spray aerosol production flux were also discussed. The group consensus was that the whitecap fraction measurements need to be improved through standardization of measurement protocols, better linked with bubble plume spectra, and performed alongside of direct field measurements of sea spray aerosol production (such as by eddy covariance) over a range of wind speeds.

Organic matter in sea spray aerosols was also noted as an important topic, mainly in the context of the need for a better understanding of the processes that control its amount in sea spray aerosol, whether it is internally or externally mixed with inorganic components, its atmospheric evolution and interaction with secondary species, and the current lack of a good proxy for organic enrichment of marine aerosols. Correlations of chlorophyll-*a* (Chl-*a*) and dissolved organic carbon (DOC) concentrations at the sea surface, the most commonly used proxies, with organic fraction of sea spray aerosol are not conclusive and need to be further examined. In terms of sea spray aerosol removal processes, participants described the difficulty of measuring net aerosol fluxes near the ocean surface. Overall, session participants agreed that interdisciplinary field campaigns evaluating marine aerosol production and processing would benefit from coordinated measurements of ocean conditions (e.g. whitecap fraction, bubble spectra, surface ocean wave properties,

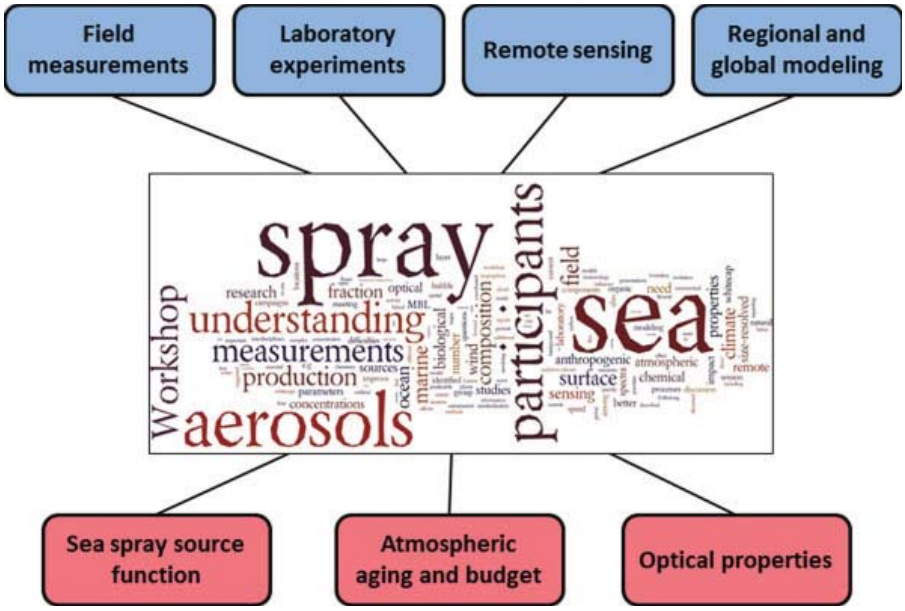

Figure 1. The four main focus groups followed by the three breakout sessions used for the discussion related to current sea spray aerosol-related research status and future priorities. The word cloud was generated through the Wordle software (http://www.wordle.net/) using the workshop discussions. The more frequently a word is used in the text, the larger that word appears in the word cloud.

upper ocean ecology and chemistry, photic-zone optical properties) and routine atmospheric measurements [e.g. aerosol number and mass size distribution, size-resolved aerosol chemistry, aerosol hygroscopicity, cloud condensation nuclei (CCN) spectra, ice nuclei (IN) spectra, aerosol optical properties and optical depth, relative humidity, surface wind speed, cloud fraction, and precipitation rates]. Because entrainment of air and aerosol from the free troposphere to the marine boundary layer (MBL) modifies marine aerosol concentrations within the MBL, assessment of entrainment should also be incorporated. Additionally, measurements of trace components such as black carbon concentrations can indicate the extent to which the pristine MBL aerosol composition may have been perturbed by anthropogenic sources.

Discussion of future major integrated field studies considering the highly nonlinear and nonuniform pattern of sea spray emissions that arises through the combined effect of physical, chemical, and biological parameters emphasized the importance of site selection. Ideal criteria would include (1) minimal continental influence on the marine aerosol (lack of pollution, dust, etc.), (2) stable meteorology and well-defined boundary layer, (3) predictable gradients and seasonality in wind speed, whitecaps, and biological productivity over significant spatial scales that are statistically resolvable by satellite, and (4) favorable logistics for water, airborne, remote sensing, and possibly land measurements taken on a long-term basis. Such locations were identified as the channel between Maui and the Big Island of Hawaii in the equatorial trade wind region, where a natural wind tunnel in the MBL is formed from mountains reaching into the free troposphere which focuses and accelerates clean marine-air-mass winds and the Mace Head in the N E Atlantic, perhaps one of the most utilized and best characterized stations possessing very strong sea-salt signals in periods of low biological activity and the strongest organic-enriched sea spray signals during periods of high biological activity. These and other locations (e.g. Amsterdam Island in the Southern Indian Ocean) representing different climate zones, biological productivities, and degrees of anthropogenic influence should be targeted for coordinated studies of marine aerosol production and evolution. Characterizing seasonal and spatial variation will be essential to improve the input that field campaigns can provide toward validating large-scale models.

Laboratory experiments

This session concentrated on the role of canonical experiments in understanding sea spray emissions. Discussion focused on the representativeness of results of experiments both in the laboratory and in the field, terminology, standardization of seawater, and the need for intercomparison studies. The representativeness of laboratory experimentation was discussed in the context of the scale of physical models (e.g. depth of model ocean and bubble path length) and methods of bubble generation (e.g. frits, jets, falling water, single bubble vs multiple bubbles and associated surface rafts-transient features of all breaking waves that are sustained by bubble plume detrainment). It was suggested that laboratory measurements could help identify a 'universal' parameter (e.g. bubble volume flux) that can be used for scaling laboratory experiments

to model global production of sea spray. Despite the various challenges that hamper such a development, there was a clear consensus that canonical studies leading to improved mechanistic understanding of the emission process are necessary. The terminology discussion concerned ambiguities and confusion that arise from imprecise definitions and lack of clear distinctions; for instance, DOC versus particulate organic carbon (POC), a raft of bubbles versus foam, and dry foam versus wet foam. Terminology uncertainties are largely due to the interdisciplinary nature of sea spray aerosol research, with oceanographers and atmospheric scientists frequently using different nomenclature. While avoiding strict definitions of these terms here, the consensus was that investigators should clearly define all quantities and that the field should conscientiously strive toward convergence on terminology. Some participants expressed the need to develop some standardization of seawater used in bubble bursting experiments so that the effects of different bubble generation systems could be better understood. In addition, it was suggested that there is a need for an intercomparison workshop, possibly similar in structure and design to the recent ice nucleation workshops held at the Aerosols Interaction and Dynamics in the Atmosphere (AIDA) cloud chamber facility in Germany (DeMott *et al.*, 2011). The intercomparison may lead to the development of a 'pod' of the most essential instruments to be deployable in field campaigns or in the laboratory and may provide insight into how different bubble generation mechanisms (e.g. weirs vs frits) can affect sea spray production fluxes.

Remote sensing

This session began with an overview of the contribution of remote sensing to understanding sea spray aerosol, ranging from ground-based systems including the Maritime Aerosol Network (MAN) (Smirnov *et al.*, 2009) a ship-borne data acquisition initiative complementing island-based AEROsol RObotic NETwork (AERONET) measurements to satellites missions such as the MODerate resolution Imaging Spectroradiometer (MODIS), Multi-angle Imaging Spectral Radiometer (MISR), Advanced Along Track Scanning Radiometer (AATSR), POLarization and Directionality of the Earth's Reflectances (PARASOL), MEdium-spectral Resolution Imaging Spectrometer (MERIS), Sea-viewing Wide Field-of-view Sensor (SeaWiFS), and Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO). Ground, satellite, and aircraft-based remote sensing can provide quantities that are or may be useful in estimating sea spray aerosol emissions and budget, including aerosol optical depth (AOD), surface wind speed, wave parameters, Chl-*a* concentration, colored dissolved organic matter (CDOM) concentration, whitecap fraction, and sea surface temperature (SST). Session participants suggested that there is a need for more aircraft data to improve spatial and spectral resolution (e.g. accurate high vertical-resolution aerosol profiles near the ocean surface). It was also suggested that ground- and ship-based systems for measurement of marine aerosol properties should be expanded. Quantities such as aerosol chemical speciation, sea state (roughness and whitecap fraction), and AOD (near clouds and under high-wind conditions) could be determined through improvements in the horizontal, vertical, temporal, and spectral resolution of *in situ* measurements. An open question is the extent to which sea spray aerosols (or even marine organics aerosols) can be distinguished from continental aerosols via remote sensing techniques. If such a distinction can be achieved, it will greatly enhance the ability of remote sensing to evaluate marine aerosol production, processing, and radiative effects.

Regional and global modeling

Discussion by session participants identified and evaluated successes and limitations of sea spray aerosol modeling. Successes included the implementation of interactive sea-salt aerosol emissions into most models (including treatment of the sea spray aerosol organic fraction in some) and the preliminary quantification of changes in CCN spectra associated with the various sea spray aerosol source functions. One of the main difficulties in modeling sea spray aerosol production is the lack of flux and concentration observations, particularly long-term size-resolved aerosol number concentration and chemical composition. Currently, available data compilations are coarsely resolved and include only mass concentrations. The need for a compilation of size-resolved marine aerosol number concentration database, similar to the University of Miami ocean aerosol network dataset was stressed. (This dataset is established by D. L. Savoie and J. M. Prospero and hosted by the Goddard Institute of Space Physics as part of the Global Aerosol Climatoloy Project; *Mishchenko et al.*, 2002). It was pointed out by the participants that many size-resolved observations of number concentration exist, but there has not been a dedicated effort to compile them into an integrated database. Having long-term data with seasonal and spatial variability is essential for input to/validation of large-scale chemical transport and climate models.

Many of the participants discussed the uncertainties in the organic enrichment of sea spray aerosol and issues with the use of satellite-derived surface Chl-*a* concentrations and other oceanic proxies like DOC, POC, or CDOM. More fundamentally, there was a debate about whether the current method of parameterizing sea spray aerosol emissions by using wind speed at 10-m reference height needs to be extended or replaced by incorporating other more directly related quantities, including sea state, wind stress, etc. Development of improved parameterizations is hampered by the lack of information on the production mechanism of sea spray organics, and highlights the need for

Table I. Prioritization matrix.[a]

Parameter	Current understanding	Impact if achieved	Difficulty/resources needed
Source function			
Bulk mass and number emissions	Med	Med-Low	Med-Low
Bulk chemical composition/hygroscopicity	Med-Low	Med-Low	Med-Low
Size-resolved mass and number emissions	Med-Low	High	Med
Size-resolved chemical composition/hygroscopicity	Low	High	High
Mixing state	Low	High	High
CCN number flux	Med-Low	High	Med
Giant CCN	Low	Med	High
IN number emissions	Low	Med	Med
IN sources	Low	Med	Med
Whitecap fraction	Med	High	Med-Low
Bubble spectra	Med	High	Med
Seawater/microlayer chemical composition	Low	High	Med
Size-resolved organic speciation	Low	High	High
Optical properties			
AOD	Med	High	Med-Low
Refractive index	Med	Med	Med-Low
Depolarization	Med	Med-Low	High
Humidified scattering	Med	High	Low
Ångström exponent	Med	Med-Low	Med
Absorption Ångström exponent	Med-Low	Med-Low	Med
Fluorescence	Med-Low	Med	Med-Low
Lidar ratio	Med-Low	Med	Med
Marine boundary layer budget			
Wet removal	Low	High	Med
Dry removal	Med-Low	Med	Med
Photochemical aging	Low	High	High
Volatility	Low	Med	Med
Entrainment	Med-Low	High	Med
Cloud processing	Low	High	High
Transport	Med-Low	Med	Med

[a]Ranking levels were assigned using the following numerical values: 1: Low, 2: Med-Low, 3: Med, 4: Med-High, and 5: High.

a more integrated research approach that strengthens the linkages between modeling, laboratory, and field experiments.

The group also discussed the feasibility of using the same source functions for the inorganic and organic components of sea spray aerosols, as well as issues that are related with the mixing state of these two components and their impact on the model-simulated climate, primarily via aerosol removal and CCN activity. The key question that remains unanswered is how the aerosol production schemes should be modified in the models to consider organic component of sea spray. Some suggestions for future work included (1) a model intercomparison study similar to the AeroCom studies (e.g. *Koch et al.*, 2011), (2) development of size- and composition-resolved source functions for marine aerosols, (3) development of an adjoint model that estimates the sensitivity of model output (e.g. surface concentration) with respect to model inputs (e.g. emissions), and (4) compilation of a global marine aerosol size distribution and chemical composition dataset.

Prioritization matrix

In addition to summarizing the current state of the science and outlining future needs and methods for reducing the uncertainties in the field, the workshop participants also created a prioritization matrix of a number of sea spray aerosol research areas from three broad categories: the source function, optical properties, and the MBL budget. For each topic, the current level of understanding, the impact full understanding would have in the field if achieved, and the potential resources needed, including the difficulty of performing the task, were ranked on a scale ranging from one to five. Individual responses were collected, and 'low', 'medium', and 'high' level of understanding was assigned based on the consensus achieved by majority of workshop participants. These rankings are summarized in Table I. The order of listing of topics does not convey any indication of priority settings by the attendees.

The group identified seven topics that have the lowest understanding and the highest impact if achieved: (1) size-resolved chemical composition/ hygroscopicity, (2) mixing state, (3) wet removal, (4) photochemical aging, (5) cloud processing, (6) seawater/ microlayer chemical composition, and (7) size-resolved organic speciation. Nearly all of these topics are related to the organic fraction/distinct organic species of sea spray aerosol and characterized by the difficulty in measurement techniques and/or the problem of attribution of the measured properties to sea spray emission, either in the laboratory or the field. Although size-resolved number flux (i.e.

production) was not included in this list, we stress that as long as it remains unknown, chemical composition and aging remain inconsequential. As a result, the impact achieved by understanding the size-resolved number flux is high.

Within the source function category, topics pertaining to the size-resolved properties were mostly classified as having a lower level of understanding and higher impact if achieved than the bulk aerosol properties. Topics such as mixing state and the ability of sea spray to serve as ice nuclei were ranked at low level of understanding. It is encouraging to note that no topics in the optical properties section were ranked to have low understanding. The highest impact if achieved was assigned to the topic of humidified scattering. Several topics in the MBL budget category were evaluated at a medium or high impact if achieved because of their importance in determining surface concentrations and lifetime, yet generally had low levels of understanding because relatively little research has been undertaken on them compared with that on the sea spray aerosol source function. Seven topics were identified as high difficulty and/or high cost, and five of these were also ranked as low understanding and high impact if achieved: (1) size-resolved chemical composition/hygroscopicity, (2) size-resolved organic speciation, (3) mixing state, (4) photochemical aging, and (5) cloud processing. Broadly, the first three and the last two are two areas where concerted efforts such as large-scale field campaigns and joint laboratory studies could lead to more cost-effective ways to collectively address these topics. Two other topics, seawater/microlayer chemical composition and wet removal, were ranked as low scientific understanding, high impact if achieved, and medium difficulty, and thus are worth targeting in the near future.

Conclusions and suggestions for future work

Improving the understanding of sea spray aerosols requires additional laboratory studies, field measurements, remote sensing, and modeling research. Several specific ideas were suggested during the course of the meeting, including:

1. Tabulation of a set of terminology involving sea spray aerosols that is consistent among oceanographers, atmospheric scientists, and computer modelers.
2. Standardization of artificial sea water and laboratory intercalibration of sea spray production sources using the same artificial and natural seawater. Several participants felt that a center focused on sea spray aerosols would help ensure standardization.
3. Standard oceanic, aerosol, and meteorological measurements that should be included, if possible, in all field campaigns focusing on sea spray aerosol. As a starting point the group suggested a list presented above in the '*in situ* field measurements' section.
4. A model intercomparison of the bulk and size-resolved number concentration and chemical composition of marine aerosols, preferably in the framework of AeroCom.
5. Creation of a size-resolved sea spray aerosol observational database that includes chemical composition, biological composition, and number concentration.
6. Identifying locations for studies where instrumentation, model results, and remote sensing products could be effectively brought together in a suitable and predictable environment.

Ultimately, the participants of this meeting stressed the need for better communication between interdisciplinary fields of sea spray aerosol research. Collaboration between those doing laboratory studies, field measurements, remote sensing, and modeling efforts was stated as being essential in order to quickly improve the understanding of the complex issues involving sea spray aerosols. The workshop presentations are available online (http://www4.ncsu.edu/nmeskhi/Marine_Aerosol_Workshop/WEBSITE.html).

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