Aerosol Delivery and Transport Study Intended for Marine Cloud Brightening: Current Status of the Science

Gourihar Kulkarni (Gourihar.Kulkarni@pnnl.gov), Colleen Kaul (Colleen.Kaul@pnnl.gov), Renato Pinto Reveggino (Renato.Pintoreveggino@pnnl.gov), Chandru Dhandapani (Chandru.Dhandapani@pnnl.gov), Kyle Pressel (Kyle.Pressel@pnnl.gov), Hailong Wang (Hailong.Wang@pnnl.gov), and Jingyi Chen (Jingyi.Chen@pnnl.gov) (Pacific Northwest National Lab, Richland, WA, USA)

Background/Objectives. It is important to understand the sensitivity to aerosol source type (size distribution and composition), evolution by physical and chemical processes, and transport/mixing mechanisms that eventually control the aerosol (number concentration, size distribution and other properties) ingested into boundary layer clouds as cloud condensation nuclei (CCN). Particle generation, physics (coalescence, evaporation, deposition, etc.), mixing, and transport need to be examined to expose the important processes tying near-surface aerosol emissions to the state of the cloud-topped marine boundary layer. Such framework using sea-salt water particles (SSPs) provides a novel and useful platform to measure and model aerosols from emission to activation, and also insight into how cloud changes can affect sub-cloud layer turbulence and transport that could be used as a component of parameterizations in lower resolution models. The two main objectives that could be relevant to both geoengineering approaches such as marine cloud brightening (MCB) and traditional field studies are: a) to understand how particles near strong sources evolve as they undergo collision, coalescence, evaporation and turbulence and to provide the "benchmark data" that is used to challenge the highest resolution models of particle evolution, and b) to gain a better understanding of the evolution of SSP plumes from their source to cloud base and to characterize the LES modeling capabilities required to faithfully simulate their evolution across a range of meteorological conditions.

Approach/Activities. Our approach to this multiscale, multi-process problem relies on a combination of unique laboratory observations, computational fluid dynamics (CFD) modeling, machine learning (ML) models, and large eddy simulations (LES). Lab studies are used to generate SSP that are ubiquitous in the marine boundary layer, important to the Earth System, as effective CCN, and it is possible to treat them in simpler ways than many other aerosol components. Advanced computational models are used to characterize aerosol evolution from formation (through natural or engineered processes) to their delivery at cloud base where they serve as CCN. The initial distribution is chosen to be of simple composition and to have a controllable relatively narrow size range that is optimal as a CCN. The idea is that one could reproduce the same type of perturbation in a lab, a model or possibly in an eventual field study to allow a classic "experiment" and "control" paradigm (in different cloud regimes) that has not been easy to do in the past when studying particle evolution, because the source properties, amount, location, time of particle release, etc. vary in uncontrolled ways.

Results/Lessons Learned. Lab studies show promising SSP with lognormal particle size distributions that had a first peak count at 90 to 100 nm median diameter with geometric standard deviations approximately 2. The total aerosol flux 10¹² particles per second could be made. CFD study shows that smaller droplets (< 1000 nm) evaporate within a few centimeters downstream of the nozzle. ML provides averaged properties of fluid flow for LES. LES calculations show that, under expected conditions for MCB applications, evaporation of droplets in the SSP plume can be reasonably approximated as instantaneous and represented by a bulk cooling term at the emission source. Simulations show that choice of numerical scheme to track

the SSP are sensitive to the simulation of cloud properties. The results describing the processes that might influence the SSP distribution and impact on cloud properties will be discussed. We will also discuss the implications for aerosol injection and MCB studies using Earth system models.