NOAA FORM 17-4 (4-81)

U.S. DEPARTMENT OF COMMERCE
NATIONAL OCEANIC AND ATMOSPHERIC ADMINISTRATION

Form Approved OMB Control No. 0648-0025
Expires 05/31/2021

INITIAL REPORT ON WEATHER MODIFICATION ACTIVITIES

This report is required by Public Law 92-205; 85 Stat. 735; 145 U.S.C. 330b. Knowing and willful violation of any rule adopted under the authority of Section 2 of Public Law 92-205 shall subject the person violating such rule to a fine of not more than $10,000, upon conviction thereof.

Complete in accordance with instructions on reverse and forward one copy to:
National Oceanic and Atmospheric Administration
Office of Oceanic and Atmospheric Research
1315 East-West Highway, WWMC-3, Rm 11216
Silver Spring, MD 20910

1. PROJECT OR ACTIVITY DESIGNATION, IF ANY

CAARE

2. DATES OF PROJECT

a. DATE FIRST ACTUAL WEATHER MODIFICATION ACTIVITY IS TO BE UNDERTAKEN: April 2, 2024

b. EXPECTED TERMINATION DATE OF WEATHER MODIFICATION ACTIVITIES: May 24, 2024

3. PURPOSE OF PROJECT OR ACTIVITY

Study sea salt plume evolution & transport in mari

	4. (a) SPONSOR	4. (b) OPERATOR
NAME	Sarah Doherty, PhD	Same as Sponsor
AFFILIATION	UW College of the Environm	
PHONE NUMBER	206-543-6674	
STREET ADDRESS	Ocean Science Bldg, 1492 NE Boat St	
CITY	Seattle	
STATE	WA	
ZIP CODE	98105	

5. TARGET AND CONTROL AREAS (See Instructions)

	TARGET AREA	CONTROL AREA
LOCATION	Alameda, CA	Alameda, CA
SIZE OF AREA (SQ. MI)	.0024	.0024

6. DESCRIPTION OF WEATHER MODIFICATION APPARATUS, MODIFICATION AGENTS AND THEIR DISPERSAL RATES, THE TECHNIQUES EMPLOYED, ETC. (See Instructions)

A ground-based instrument will generates droplets of 3.5% sea salt water, made with a sea salt standard and purified water. Generated aerosols have dry aerosol size of peak=50nm, sigma=2.5. Emission rate: 200g/sec of saline droplets, or 7g/sec sea salt. System will typically run less than 30min at a time, for a total of 360 kg of saline droplets or 12.6 kg of salt. The generated aerosol size and concentration will be measured in-situ at distances approx. 15m, 100m & 200m downwind of the source, at heights of 2-30m above the surface, using miniSEMS & miniOPCs, and at one of the stations also with full-sized SEMS, OPS and APS instruments.

7. LOG BOOKS *Enter name, affiliation, address, and telephone number of responsible individual from whom log books or other records may be obtained.*

NAME	Sarah Doherty, PhD
AFFILIATION	UW College of the Environment
PHONE NUMBER	206-543-6674
STREET ADDRESS	Ocean Science Bldg, 1492 NE Boat St
CITY	Seattle
STATE	WA
ZIP CODE	98105

8. SAFETY AND ENVIRONMENT

- [ ] YES [x] NO — Has an Environmental Impact Statement, Federal or State, been filed? If yes, please furnish a copy as applicable.
- [x] YES [ ] NO — Have provisions been made to acquire the latest forecasts, advisories, warnings, etc., of the National Weather Service, Forest Service, or others when issued prior to and during operations? If yes, please specify on a separate sheet.
- [ ] YES [x] NO — Have any safety procedures (*operational constraints, provisions for suspension of operations, monitoring methods, etc.*) and any environmental guidelines (*related to the possible effects of the operations*) been included in the operational plans? If yes, please furnish copies or a description of the specific procedures and guidelines.

9. OPTIONAL REMARKS (See instructions. Use Separate Sheet).

CERTIFICATION: I certify that all statements in this report on this weather modification project are complete and correct to the best of my knowledge and are made in good faith.

NAME OF REPORTING PERSON	Sarah Doherty, PhD
AFFILIATION	UW College of the Environment
SIGNATURE	Sarah G. Doherty
STREET ADDRESS	Same as Sponsor
OFFICIAL TITLE	Program Director
CITY	
STATE	
ZIP CODE	
DATE	3/22/2024
PHONE NUMBER	206-543-6674

INSTRUCTIONS FOR INITIAL REPORT ON WEATHER MODIFICATION ACTIVITIES

One completed copy of this form is to be received 10 days* or more prior to actual modification activities. A NOAA file number will be assigned by the Administrator after receipt of the initial report for each project or activity.

A supplemental report in letter form referring to the appropriate NOAA file number must be made to the Administrator if the "Initial Report" is found to contain any material inaccuracies, misstatements, omissions, or if there are changes in plans for the project or activity.

*For exceptions, see Sections 908.4(b) and (c), Part 908 of Title 15, Code of Federal Regulations.

Item 1 Enter designation, if any, used by operator for the project or activity.

Item 2 Enter: (a) Date first actual weather modification activity is to be undertaken;
(b) Date on which final weather modification activity is expected to occur.

Item 3 Enter the purposes of the project or activity: e.g., rainfall increase, hail suppression, cold fog dispersal, etc.

Item 4 Enter: (a) Name, phone number, affiliation, and address of the primary person for whom the project is to be performed (sponsor).
(b) Name, phone number, affiliation, and address of the person primarily responsible for carrying out the project (operator).

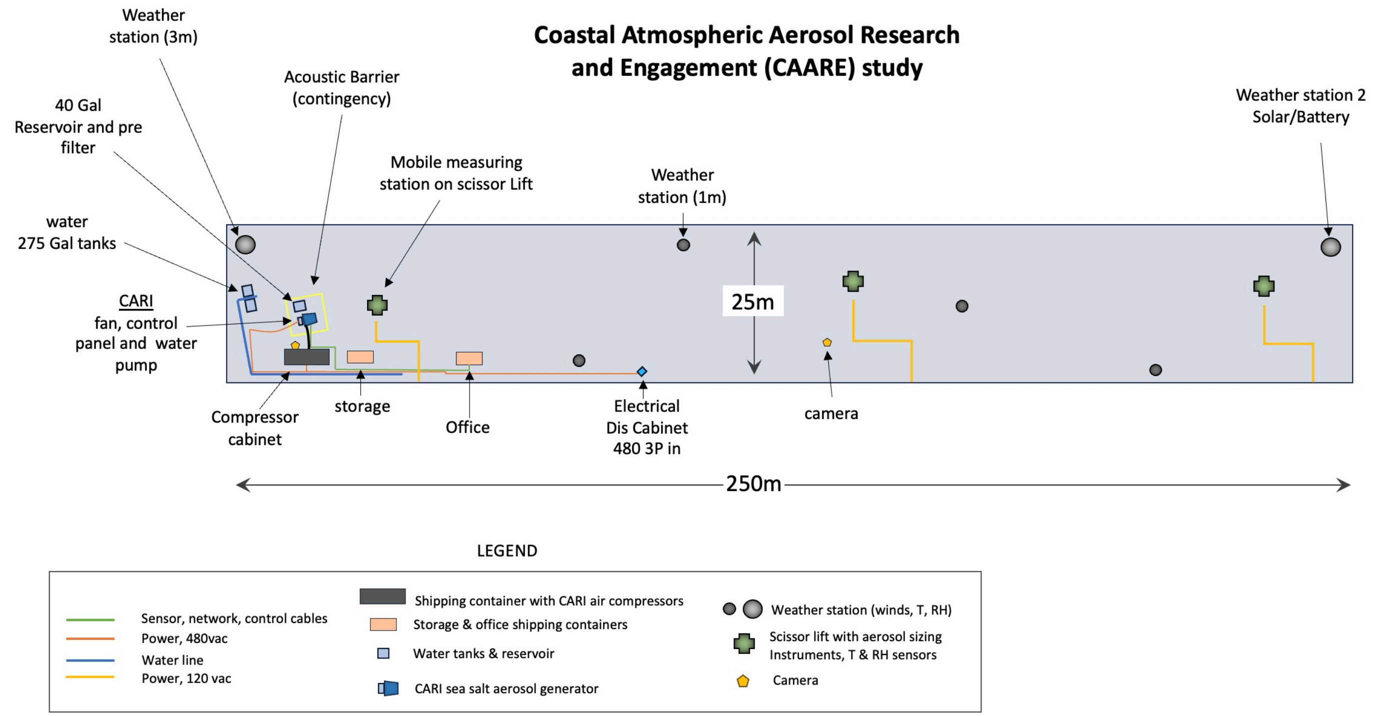
Item 5 A map should be attached showing size and location of target area, control area, coded number and location of each item of ground-based weather modification apparatus and coded number and location of key rain gauges, radars, or other precipitation measuring devices. Also show location of airport for airborne operations.

Item 6 Describe the weather modification apparatus, modification agents, and the techniques to be used. This would include type of ground or airborne apparatus to be used, type of modification material to be dispensed, rate of dispensing material in grams per hour or other appropriate units, type of precipitation gauges to be used in target and control areas, and any other pertinent information such as type of radars, type of aircraft to be used, techniques to be employed (e.g., cloud-based seeding at 10,000 feet msl).

Item 7 List name, phone number, affiliation, and address of the responsible individual from whom log books or other records may be obtained.

Item 8 Provide applicable answers to questions as indicated.

Item 9 This item is to permit the reporting person to include any information not covered by items 1 through 8 but which he feels is significant or of interest. It is also to be used to include any information not covered elsewhere that the Administrator may request.



Coastal Atmospheric Aerosol Research and Engagement (CAARE) study
Weather station (3m)
40 Gal Reservoir and pre filter
Acoustic Barrier (contingency)
Mobile measuring station on scissor Lift
Weather station (1m)
Weather station 2 Solar/Battery
water 275 Gal tanks
CARI
fan, control panel and water pump
25m
Compressor cabinet
storage
Office
Electrical Dis Cabinet 480 3P in
camera
250m
LEGEND
Sensor, network, control cables
Power, 480vac
Water line
Power, 120 vac
Shipping container with CARI air compressors
Storage & office shipping containers
Water tanks & reservoir
CARI sea salt aerosol generator
Weather station (winds, T, RH)
Scissor lift with aerosol sizing Instruments, T & RH sensors
Camera

Reviewer Report for the Proposal "Field Study of a Controlled-Release Sea Salt Aerosol Plume"

Prepared on 31 December 2023

Dilip Ganguly (Indian Institute of Technology Delhi)

Nicolás Huneeus (Universidad de Chile, Chile)

Jim Hurrell (Colorado State University)

Armin Sorooshian (University of Arizona)

Paul Wennberg (California Institute of Technology)

Summary

A committee of six chosen by the project team was contacted to serve as volunteer reviewers for a proposal to conduct a field effort focused on marine cloud brightening. This work was conducted confidentially among the team of reviewers and the project team was not involved with influencing any aspect of this report; they strictly just introduced the reviewers to one another and asked them to work together to reach the point of preparing and sharing a report with the project team. Four of the volunteer members submitted independent reviewer comments to a shared google drive, then Armin Sorooshian organized the comments into this single report. The report was then shared with the entire reviewer committee for editing and commenting. This version reflects the final edited report with unanimous approval from the committee.

The structure of the report below follows the same order of questions that the committee was asked to address (shown in black font), followed at the end by a recommendation as to whether a doppler lidar would be a useful addition (as the committee was requested to do).

Detailed Reviewer Responses

i. the value of the stated scientific objectives, within the larger goals of the MCB Program, including how it could eventually contribute to a reduction in the currently large uncertainty in present-day climate forcing through aerosol-cloud interactions and improving projections of the impacts of proposed marine cloud brightening;

The committee felt the proposal was well-written, justified, and consistent in terms of the scientific goals and activities (observations and modelling) planned for this first experiment. Given that such interactions have been extensively studied in the field previously and, yet, major uncertainties remain, is a strong motivation for undertaking emissions experiments with well characterized aerosol inputs together with proper analysis of the meteorology. The proposed observational and modeling activities could improve not only our understanding of the potential of MCB, but also reduce key uncertainties in present and future climate forcing related to aerosol-cloud interactions. The data collected would be of relevance to global-scale modelers as well as others. In general, the measurements resulting from this experiment will represent an important database that can help reduce the uncertainty on aerosol-cloud interactions.

Having said all this, at least one committee member believed it would have been better if the proposal sea salt aerosol plume experiment could have been examined with more knowledge of the CACIE experiment. Apparently, there is already a document on CACIE. Also, another member commented that given the general applicability of this study to knowledge of current climate (e.g., how much has the reduction in shipping sulfur emissions forced the post 2020 climate), they were surprised that the proposal is so focused on the MCB motivation – perhaps this is a deliberate effort at building a permissions structure for such experiments? Having this as the central motivation does add risks (see e.g., SCOPEX).

ii. whether the planned observations, modeling and analyses are suitable to meet these objectives;

The committee felt that the proposal plan seems adequate. The different activities planned for this first experiment are suitable for the stated objectives. The planned observations and modeling activities are rationale and grounded in firm science. They should yield new insights. One reviewer noted that they appreciate that uncertainties are stated and understood throughout the proposal., commenting "*It will therefore be important to understand whether aerosols produced by a spray system using filtered sea water have similar hygroscopicity to natural sea spray aerosol or behave more like pure sodium chloride. Ascertaining this information will be important in determining the CCN activity of the injected aerosol, which is critical for determining the concentration of cloud droplets formed during activation at cloud base.*"

This aerosol plume experiment is a first step towards achieving the scientific objectives set for the CACIE experiment. An important aspect of this experiment is to have atmospheric conditions similar to those in marine stratocumulus regions, since it will make it easier to apply these results to the CACIE experiment. Several actions and precautions are taken to ensure these conditions, but it is not clear from the main document how likely or often these conditions will occur over the 4-6 weeks planned duration of the experiment.

iii. whether any intentional emissions included in the study could produce a measurable impact on weather or climate;

The reviewers had no concerns with the injection of sea salt aerosol. However, there was some surprise amongst reviewers that the proposed site is directly upwind of a large urban area. Is there concern that the much larger input of urban aerosol will substantially reduce the signal to noise of the experiments? Was it not possible to find a less impacted site?

iv. the feasibility of conducting the planned observations, modeling and analyses as described in the field study plan;

All reviewers felt the project plan is ambitious and were unclear on the personnel and time involved to do everything. In particular, the amount of data to be collected is substantial and not trivial to analyze. Do the investigators have the resources to examine all the data in a comprehensive way? It is difficult to evaluate the feasibility of completing all the planned activities. No information was provided on the members of the team that will be involved in the experiment and therefore it is difficult to determine if the expertise is there to deal with the all the unforeseen events that occur in all field campaigns. Is a team member familiar with each one of the instruments that will be used in the experiment? What about the modelling?

One reviewer noted that it would help to have more information on the site location. Is there local resistance or concerns (whether founded or unfounded) around issues like local air quality, etc.? How many options exist, and how do different options affect the field study plan?

Lastly, one reviewer questioned whether the data will be made public.

v. any proposed improvements to the study plan;

The reviewers noted a couple areas where more information would be helpful. This includes desiring more information on how easily the results from the proposed land-based site can be translated to marine areas. Aside from simpler logistics and potentially reduced costs using the proposed site, what are the ambient conditions over the proposed site (e.g., are there elevated levels of pollutants from humans)? What are the possible limitations?

One reviewer was unsure if the representation of the tethered balloon system in Figure 7 is how it will be at the end, but just in case the proposers aim to use a spherical balloon for the experiment, it is suggested to use something with a more aerodynamical shape, like a zeppelin. Spherical balloons deform easily with even moderate wind velocities risking the buoyancy of the system.

Maybe it has been considered and is just not indicated in the document, but is there any sensor proposed to be included to measure the height of the payload of the tethered balloon, or will it be estimated somehow?

vi. any additional areas of scientific or safety concern raised by SRB members;

The reviewers did not identify additional areas of concern.

vii. any comments on the characterization of the study.

Overall, the reviewers felt the proposal was well done and well thought out. Science objectives seem to be within reach.

All reviewers commented that a doppler lidar would certainly be a valuable addition. One reviewer stated that the lack of a secured deployment of a doppler lidar is a big risk.

USS HORNET
Sea, Air & Space Museum Alameda, California

20 March 2024

To Whom it May Concern,

My name is Laura Fies and I am Executive Director of the USS Hornet Sea, Air & Space Museum, the host site for the Marine Cloud Brightening project being undertaken by the University of Washington, SilverLining, and other partners.

As the study has been described to us, creating a mist of salt water that will be monitored for the length of our Flight Deck (with additional monitoring stations potentially located in our visitor parking lot), the Hornet Museum is comfortable with the project operating during the hours that the Museum is closed to the public and while the Museum is open to the public so long as signage is posted alerting our visitors to the spray.

We do not have any concerns about any effects to our staff, volunteers, or visitors (especially as visitors will be given the active choice to engage with the project should it be active during the Museum's public hours) or to our artifacts and historic vessel. This is due to the in-depth conversations that we have had with project representatives as well as the fact that the active material used is salt water—something the Museum is used to managing and mitigating on our historic Flight Deck. Nothing within this project exceeds a scale or use of machinery, power, or materials beyond the Museum's usual restoration and operational use or goes beyond the scope of our Use Permit as defined within our pier rental agreement with the City of Alameda.

We are looking forward to hosting the Marine Cloud Brightening project, particularly in terms of their interest in outreach and education. The Museum sees an average of 14,000 students through our educational programming every year, and we are excited to integrate information about this groundbreaking research in accompaniment to our traditional STEM workshops.

Best,

Laura Fies
Executive Director
USS Hornet Sea, Air & Space Museum
Laura.Fies@uss-hornet.org

National Weather Service forecasts and warnings for Alameda, California will be checked 3, 2, and 1 day in advance of any operational days, as well as in the mornings of operational days, before commencing operations.

INFORMATION PROVIDED UNDER THE PROVISIONS
OF THE
PAPERWORK REDUCTION ACT OF 1995

The Paperwork Reduction Act o f1995 requires that individuals or organizations be provided with the following information if they provide information on paper forms which are collected by the Federal Government.

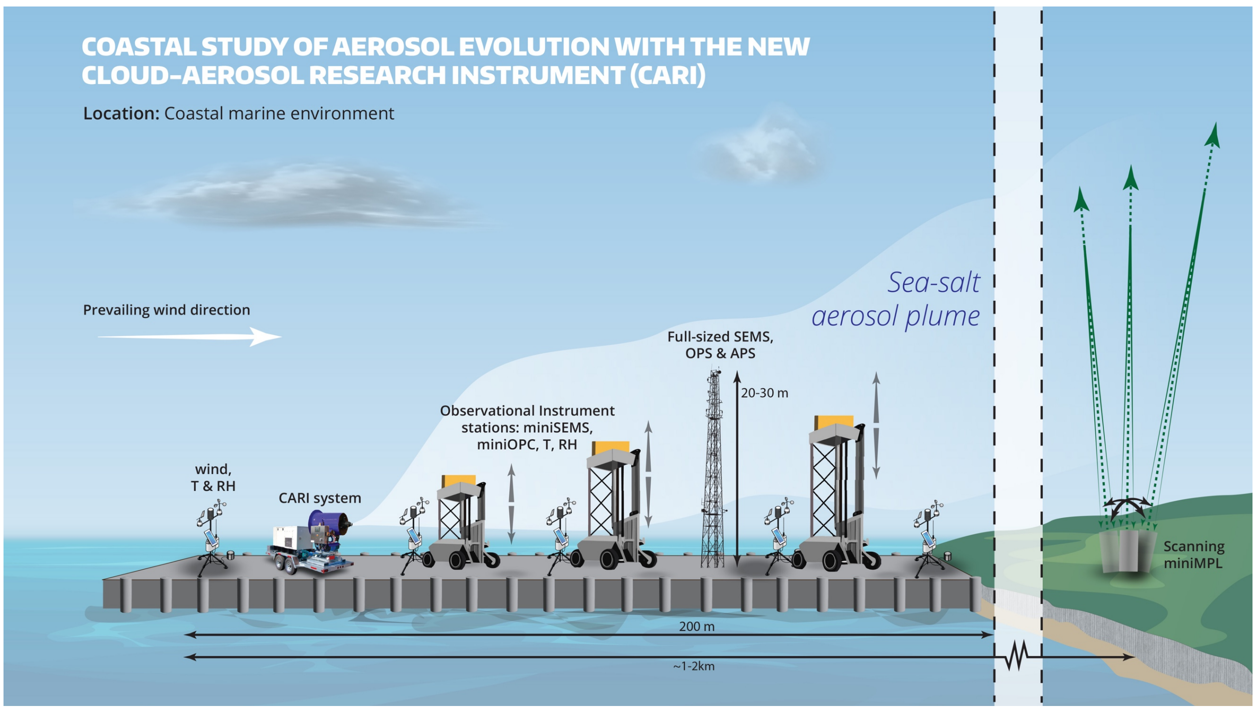
1. Public Law 92-205, enacted December 18, 1971 (amended by Public Law 94-490, Section 6(b), October 15, 1976) requires that all non-federal weather modification activities in the United States and its territories be reported to the Secretary of Commerce. The National Oceanic and Atmospheric Administration has implemented the Act and the current reporting requirements are published in the Code of Federal Regulations (15 CFR 908).

2. The intent of the program is to increase expertise in the field of weather modification, to allow scientists and other concerned persons to have access to information on current and past efforts at weather modification, to help avoid unneeded and wasteful duplications, to aid in preventing territorial overlapping of weather modification operations, to provide data to assess possible harmful or dangerous activities, and to furnish information to check both desirable and undesirable atmospheric changes against records of weather modification efforts. To meet this objective, information is collected on the location and size of the target area, names and addresses of sponsors and operators, beginning and ending dates of the project, specific purpose, description of apparatus and seeding agents to be used, number of days of operations, number of hours of operations of each type of weather modification apparatus, and total amount of seeing agent used.

3. A Federal agency may not conduct or sponsor, and a person is not required to respond to, nor shall a person be subject to a penalty for failure to comply with an information collection subject to the requirements of the Paperwork Reduction Act of 1995 unless the information collection has a currently valid OMB Control Number. The approved OMB Control Number for this information collection is 0648-0025. Without this approval, we could not conduct this information collection. Public reporting for this information collection is estimated to be approximately 30 minutes per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the information collection. All responses to this information collection are mandatory pursuant to Public Law 92-205, enacted December 18, 1971 (amended by Public Law 94-490, Section 6(b), October 15, 1976). Send comments regarding this burden estimate or any other aspect of this information collection, including suggestions for reducing this burden to the OAR Weather Program Office at Weather.Modification@noaa.gov.

Field Study of Controlled-Release Sea Salt Aerosol Plume

Robert Wood & Sarah Doherty
Dept. of Atmospheric Sciences
University of Washington
Seattle, WA
Jessica Medrado, Dongyun Shin, Matt Gallelli, David Johnson & Sean Garner
SRI, International
Palo Alto, CA



ver. March 18, 2024

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1 Motivation

Climate forcing from anthropogenic aerosol emissions via aerosol-cloud interactions is estimated to be between about 10% and 40% that of anthropogenic forcing from greenhouse gases (GHGs) and of opposite sign (Forster et al., 2021), providing an important but highly uncertain offset to GHG climate warming (Figure 1). While the magnitude of present-day climate forcing through aerosol-cloud interactions is highly uncertain (Seinfeld et al., 2016; Bellouin et al., 2019), theoretical and observational studies indicate that aerosols can significantly increase albedo via aerosol-cloud interactions (Twomey 1977; Chen et al., 2014; Diamond et al., 2020; Chen et al., 2022).

The observed impacts of aerosols on clouds led to the hypothesis that marine cloud brightening (MCB), i.e., the targeted addition of aerosols with optimized size and concentration to specific low marine clouds, could be used to reduce climate warming and associated impacts (Latham 1990; Latham et al., 2012). The clouds expected to be most susceptible to the addition of aerosols are clean stratocumulus clouds (Oreopoulos et al., 2008; Alterskjaer et al., 2012), which cover about 20% of the Earth's oceans (Wood, 2012). These clouds are therefore the focus of intentional marine cloud brightening (MCB) studies. It is also estimated that approximately 60% of present-day effective radiative forcing from pollution via aerosol-cloud interactions (ACI) occurs in stratocumulus clouds (Diamond et al., 2020). However, major questions remain regarding the role of cloud macrophysical adjustments to aerosol perturbations, which, depending upon the meteorological and cloud state, can either augment or offset cloud brightening from increases in cloud droplet number concentration identified by Twomey, depending upon the meteorological and cloud state (H. Wang et al., 2011; Bellouin et al., 2019; Chen et al., 2022), and which were the basis for the original MCB concept. **Better quantifying stratocumulus cloud responses to the addition of aerosols under different meteorological and background aerosol conditions will help reduce uncertainties in present-day ACI climate forcing, and therefore the rate of planetary warming due to increasing greenhouse gases alone, as well as being critical to determining whether MCB could feasibly be used as a way to predictably and reliably slow climate warming**.

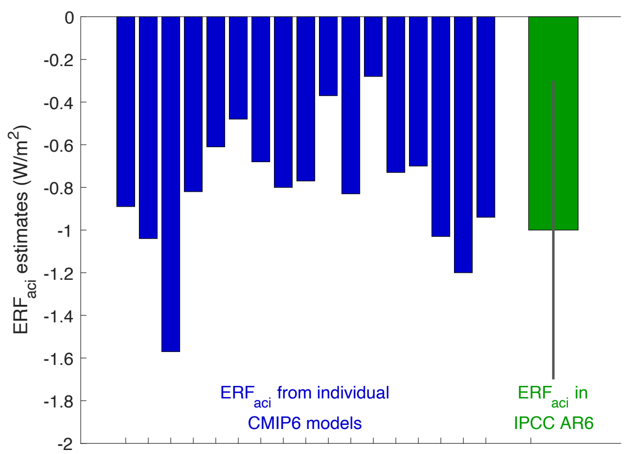

Figure 1. *Global model estimates of climate forcing by aerosol-cloud interactions (ERF$_{aci}$) vary widely. Shown here are estimates of ERFaci from 17 CMIP6 global models (blue bars; from Table 6 of Smith et al., 2020) as well as the best estimate (green bar) and range (gray line) in ERF$_{aci}$ as assessed in IPCC AR6 (Forster et al., 2021). For reference, ERF by greenhouse gases is assessed in AR6 as +3.84 W/m^2 (range: +3.46-4.22 W/m^2).*

Prior observations and utility of controlled perturbation experiments

The importance of ACI as a climate forcing mechanism and the persistent uncertainty in this forcing (IPCC, 2001; IPCC, 2007; IPCC, 2013; IPCC, 2021), has motivated numerous field studies of how pollution and biomass burning smoke are altering cloud properties (e.g. MASE I and II[1], VOCALS-REX 2008[2], E-

[1] Marine Stratus/Stratocumulus Experiment (2005 & 2007)

[2] VAMOS Ocean-Cloud-Atmosphere-Land Study (2008)

PEACE[3], CSET[4], ORACLES[5], LASIC[6], ACE-ENA[7], TRACER[8], ESCAPE[9]). These campaigns, coupled with modeling across a range of scales (M. Wang et al., 2011; Wyant et al., 2015; Glassmeier et al., 2019) and analysis of satellite data (Chen et al., 2014; Wang et al., 2012; McCoy et al., 2017) have significantly advanced our understanding of aerosol impacts on clouds. **However, establishing quantitative causal connections from these observations is hindered by co-variability between changes in aerosol and cloud properties (Stevens and Feingold 2009), because both are sensitive to variations in atmospheric transport and meteorology, as well as to local sources of aerosols, heat, and moisture. This makes it challenging to untangle aerosol-driven effects on clouds from those due to meteorology.**

A notable exception to this co-variability occurs with ship tracks, where cloud albedo changes are confined to narrow corridors defined by the addition of aerosols from transiting cargo ships (Figure 2). These ship tracks provide a type of natural experiment that allows for more effective distinction of aerosol-driven changes in clouds (Christensen et al., 2022), by contrasting the cloud properties and cloud evolution in and adjacent to the track. As such, ship tracks have been the focus of a number of field studies (e.g. MAST in 1994, Durkee et al., 2000a; MASE in 2005 & 2007, Lu et al, 2007, 2009; ACRUISE, ongoing). Such studies provided some of the first evidence of precipitation suppression in ship tracks (Ferek et al., 2000) and that liquid water path often decreases in ship tracks (Coakley and Walsh 2002). But the mechanisms for cloud adjustments involve both cloud responses to precipitation suppression and more efficient entrainment mixing (Ackerman et al. 2004; Wood 2007). Precipitation formation and cloud top entrainment are poorly understood processes and are especially difficult to accurately represent in low resolution global models (Stephens et al., 2010; Guo et al., 2011).

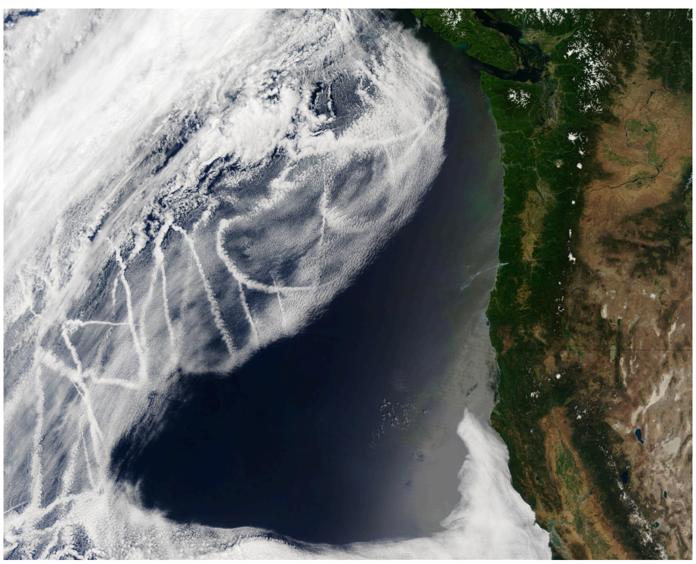

Figure 2. *Ship tracks are a highly visible manifestation of how aerosol emissions can alter clouds, here leading to visibly brightened tracks of clouds in the stratocumulus deck off the west coast of North America. [Credit: NASA Earth Observatory]*

While ship tracks provide a convenient "natural experiment" demonstrating how low marine clouds respond to aerosol injections, relying on such observations of opportunity makes planning the sampling of specific cloud and aerosol regimes difficult. The aerosol particles driving the cloud changes are also from poorly characterized and variable sources. This limits the ability to build statistics on cloud responses under given cloud, meteorological and/or background aerosol conditions. Given the complexity of cloud

[3] Eastern Pacific Emitted Aerosol Cloud Experiment (2011)
[4] Cloud System Evolution over the Trades (2015)
[5] ObseRvations of Aerosols above CLouds and their intEractionS (2016-2018)
[6] Layered Atlantic Smoke Interactions with Clouds (2016 & 2017)
[7] Aerosol and Cloud Experiments in the Eastern North Atlantic (2017-2018)
[8] Tracking Aerosol Convection interactions Experiment (2021)
[9] Experiment of Sea Breeze Convection, Aerosols, Precipitation, and Environment (2022)

responses to aerosols, and the fact that perturbations are occurring in the context of a naturally evolving and variable cloud field, sampling sufficient to build statistics will be essential to reducing uncertainties in aerosol-cloud interactions.

Planned emissive field studies

In the interest of having better observational constraints on aerosol-cloud interactions, we have proposed a field study design that builds on those used for previous field campaigns, with the important difference of being able to control the aerosol perturbation to the cloud. Using a consistent and well-characterized aerosol source will allow us to more definitively separate cloud changes driven by aerosol-cloud *covariability* associated with meteorology from those driven by local sources driving aerosol-cloud *interactions*. Systematically conducting such controlled-perturbation studies over a range of background aerosol and meteorological conditions will further allow for building statistical relationships between aerosol perturbations and cloud responses over a range of timescales.

Important questions also remain about how different meteorological conditions typical in regions of low marine clouds affect transport of aerosol from the surface to cloud base, how well these transport processes can be constrained with observations, and how well even higher-resolution (e.g. large-eddy simulating) models represent these processes.

Controlled-aerosol Aerosol-Cloud Interaction Experiment (CACIE)

The aerosol-cloud interaction study we have proposed, the Controlled-aerosol Aerosol-Cloud Interaction Experiment (CACIE), is described in a separate document.

The CACIE experiment characteristics include:

i) using a single, well-characterized source of aerosols emitted at a rate sufficient to produce a measurable cloud perturbation
ii) a study design targeting process-level insights, with a focus on processes indicated to be important in cloud-resolving modeling simulations, and
iii) building statistics on cloud responses to the generated aerosol across a range of conditions, and co-analyzing the observed responses with simulations of the observed cases.

This requires the use of a spray system capable of generating a plume of aerosol that:

- is well-characterized (aerosol composition, size and number concentration),
- will be efficiently transported to cloud base, and
- will produce a significant cloud perturbation.

The proposed CACIE study involves use of a spray system that would produce sea salt aerosol optimized for marine cloud brightening, targeted for use in marine stratocumulus regions. As described below in more detail, this spray system must be capable of producing a plume of at least 10^{15} sea salt particles per second of sea salt that, just downstream[10] (i.e. ~200 m) of the spray system, is in the ~10-200 nm dry diameter size range (Connolly et al., 2014; Wood, 2021), and that fewer than 0.1-0.2% of the particles exceed 2 μm dry diameter. Further, it must be shown that evaporative cooling of the spray droplets in the plume will not prevent the plume from mixing up to cloud base (i.e., up to 1-2km altitude) without significant loss of the generated aerosols under the atmospheric boundary layer

[10] As shown by Wood (2021) (see their Figure 11), for the size and concentration of aerosols produced by this spray system, the effect of coagulation on particle number concentrations is insignificant within a few 10's of meters downstream of the spray system, due to the rapid dilution of the plume. Thus, the aerosol size distribution within ~100-200m downstream of the spray system will likely be representative of the aerosol size at cloud base.

conditions typical in regions of marine stratocumulus. A spray system designed to meet these requirements, CARI, the Cloud Aerosol Research Instrument, has been developed as part of the University of Washington Marine Cloud Brightening (MCB) Program.

Herein we further describe:

- the basis for the performance requirements we've set for the CARI system, and
- the field studies we propose, which care designed to
  - assure that CARI meets the above requirements and
  - addresses science questions about the evolution and transport of the aerosols within the marine boundary layer.

Land-based controlled-aerosol-release studies

There are several overarching science questions that are driving the development of land-based controlled aerosol release studies:

- Can aerosol production rates be determined using observations of the aerosol size distribution at sites 100-1000 m downstream of the spray system?
- Can coagulation rates immediately downwind of the spray system be constrained through combined analysis of observations and model simulations?
- How does the aerosol size distribution in the plume evolve under different meteorological conditions, such as different wind speeds, background turbulence levels, and relative humidities?
- How do we optimize our measurement suite to be able to constrain the energy costs associated with the spray system?

Joint analysis of observations of the generated aerosol plume and multi-scale modeling will be used to test our understanding of the aerosol evolution and transport in the boundary layer (see Section 5). Results of the study will also be useful for developing improved representation of the transport of a surface-generated plume in models across a range of scales, including the large-eddy simulations (LES) being used to study the potential efficacy of marine cloud brightening (e.g. Glassmeier et al., 2019; Wood, 2021; Hoffmann and Feingold, 2021) and, ultimately, in global models being used to study the climate impacts of MCB.

2 Requirements for generated aerosol

Aerosol-cloud interactions driving spray system requirements

Aerosol-cloud interactions affect cloud albedo initially through changes in cloud microphysical properties (droplet size and droplet number concentration), which in turn can trigger cloud macrophysical responses (total amount of cloud water and how long the cloud lasts, and therefore cloud fraction). It is well-established that adding sub-micron sized aerosols to clean, low clouds increases the number of cloud droplets in the cloud (Martin et al., 1994), increasing the cloud water surface area; for a cloud with a given amount of liquid, this increases the cloud albedo (the Twomey effect; Twomey, 1974). The increase in albedo with aerosol concentration is largest for clouds with otherwise low concentrations of cloud-condensation nuclei (CCN) because the effect asymptotes at higher concentrations of CCN. Aerosol increases in clouds are therefore expected to be most effective at producing a negative radiative forcing in regions with low concentrations of background aerosols.

The original idea of intentional marine cloud brightening was based on the idea of leveraging the Twomey effect (Latham 1990). We now know that the decrease in cloud droplet size with the Twomey effect in turn leads to other cloud responses. In contrast to the Twomey effect, which always produces a negative radiative forcing, these cloud responses can either act to increase or decrease cloud albedo. Smaller droplets at the cloud top can lead to increases in droplet evaporation. The associated evaporative cooling adds to cloud-top turbulence and entrainment of dry, lower free-troposphere air, decreasing humidity in the cloud (Wang et al., 2003; Xue and Feingold, 2006; Jiang et al., 2006; Small et al. 2009; Dagan et al, 2017). In addition, smaller droplets sediment more slowly so more of the cloud water is present in the entrainment zone at cloud top, leading to more cloud-top evaporative and radiative cooling, again increasing the cloud-top entrainment rate (Ackerman et al., 2004; Bretherton et al., 2007; Chen et al., 2014; Michibata et al., 2016; Sato et al., 2018). Both processes can lead to a reduction in cloud liquid water path (LWP), offsetting or possibly even overwhelming Twomey brightening.

On the other hand, reduced cloud droplet size also suppresses precipitation. This reduces moisture loss from the cloud and increases cloud LWP and cloud lifetime and therefore, with time, cloud fraction (Albrecht, 1989; Erfani et al., 2022). This effect appears to be particularly strong in clouds with very low initial aerosol concentrations (H. Wang et al., 2011; Erfani et al., 2022), which are precisely the clouds that are also most susceptible to Twomey brightening and are therefore ideal candidates for marine cloud brightening. The net effect of these cloud responses on cloud reflectivity and lifetime depends strongly on the background aerosol and meteorological conditions, as well as on the size and concentration of the aerosol added to the cloud.

Aerosol size

Requirements on the size and concentration of aerosols targeting marine cloud brightening have been determined based on two types of simulations. Parcel model simulations can account in detail for cloud droplet activation as a function of aerosol size distribution and background meteorological conditions. Parcel models are suitable for running simulations over a large range of meteorological, aerosol size and aerosol concentration combinations because of their lower computational demands. However, they cannot account for the dynamical responses to changes in cloud droplet number concentrations that drive cloud macrophysical responses. Doing so requires the use of large-eddy simulating (LES) models, which are more computationally expensive but can resolve most of the dynamical processes that drive cloud evolution, as well as representing cloud microphysical responses to change in aerosol concentrations. Depending on the LES model and the simulation set-up, they can account to varying degrees for how aerosol size affects cloud responses.

Parcel model studies by Connolly et al. (2014) and Wood (2021) show that aerosols in the 30-60 nm dry diameter range are most efficient at producing cloud brightening through the Twomey effect, where efficiency is measured in terms of forcing per mass of sea salt injected. Using a heuristic model that allows exploration of how cloud droplet number concentration (CDNC) changes as a function of the sub-cloud aerosol size distribution, aerosol concentration and updraft speed Wood (2021) simulated the aerosol perturbation from a collection of point sprayers, accounting for plume overlap. Based on typical clean marine stratocumulus conditions, that study found that the most efficient forcing is produced when a single sprayer is able to produce 10^{15}-10^{16} aerosol per second of 30-60 nm dry diameter, assuming there is negligible scavenging of the aerosol between the spray system and cloud base.

Aerosols smaller than 30 nm dry diameter are less effective at activating as cloud condensation nuclei (CCN), and at higher mass injection rates (see discussion below) the larger number of smaller particles suppresses cloud supersaturation (Alterskjær and Kristjansson, 2013; Wood, 2021). At sizes larger than 60 nm, the aerosols are even more effective as CCN, but the forcing produced for a given mass injection drops off, quite rapidly above 100 nm dry diameter. Injecting aerosols of much large sizes (>2000 nm; i.e. "giant CCN") can induce clouds to precipitate, losing water mass; injecting a sufficient number of these larger aerosols can offset brightening through the Twomey effect by the smaller aerosols, or even lead to sufficient cloud loss that there is scene dimming (Feingold et al., 1999; Hoffmann and Feingold, 2021). The addition of giant CCN to clouds can induce precipitation even at very low (e.g., 10^{-3} cm^{-3}) concentrations in stratocumulus with low to moderate accumulation mode aerosol concentrations (Feingold et al., 1999), exactly the types of clouds likely to be targeted with MCB. As these concentrations of giant CCN can already be present in the ambient environment (Woodcock 1953; Jensen and Lee, 2008), injections for MCB should aim to minimize adding aerosol of this size to cloud base.

Aerosol concentration

For the first field studies of aerosol-cloud interactions proposed as part of the CACIE study, the goal is to produce a measurable change in cloud droplet number concentration (N_c) and cloud albedo (α) with a single plume of sea salt aerosol. Assuming a fixed cloud LWP, the Twomey effect produces a cloud albedo that increases with cloud droplet number concentration as:

$$\frac{d\alpha}{dN_c} = \frac{1}{3}\frac{\alpha(1-\alpha)}{N_c} \qquad [1]$$

(Platnick and Twomey, 1994; Quaas et al., 2008). Clean to moderately polluted marine boundary layers typically have cloud droplet concentrations, N_c, ranging from about 30 cm^{-3} to 150 cm^{-3} (Wood 2012, Latham et al., 2012). For a typical marine stratocumulus cloud albedo, α=0.45 (Wood 2021), the increase in albedo ($\Delta\alpha$) through the Twomey effect as a function of a perturbation (ΔN_c) in cloud droplet number can be calculated by integrating Eqn. [1]. This calculation shows that producing $\Delta\alpha$ in the range 0.05 to 0.15 (i.e., about a 10-30% increase) through the Twomey effect alone would require that the spray system be capable of producing ΔN_c values of about 10 cm^{-3} (for a background N_c of 30 cm^{-3}) to 125 cm^{-3} (for a background N_c of 150 cm^{-3}).

Based on the analyses of Connolly et al. (2014) and Wood (2021), sea salt aerosols in the 10-200 nm dry diameter size range will act as effective CCN. As a first-order approximation we therefore set ΔN_c equal to the change in the number concentration of aerosols in this size range, ΔN_a, at cloud base – i.e. we

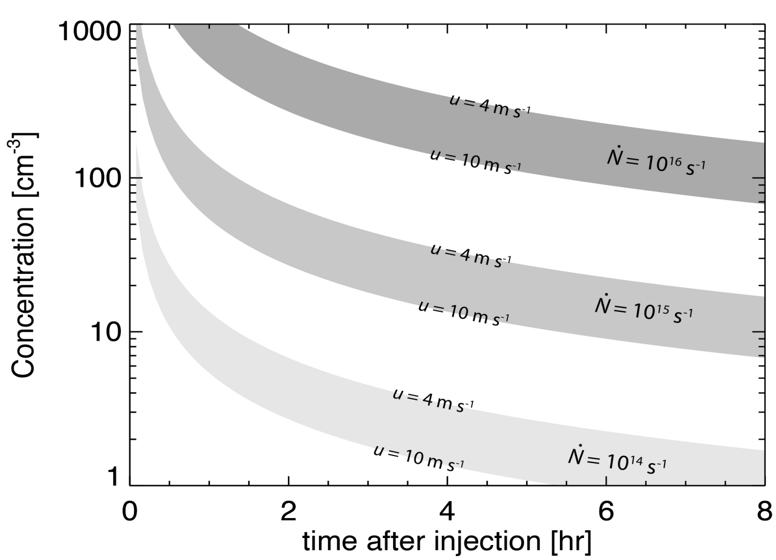

Figure 3. An injection rate of at least 10^{15} particles s^{-1} is needed in order to increase aerosol concentrations at cloud-base by >10 cm^{-3}, as shown here for calculations using a parcel model. Here, it is assumed that the plume rapidly fills the assumed 1 km deep PBL, laterally spreads at a rate of 1.85 km hr^{-1} (Wood 2021), and that there are no aerosol sinks. For reference, at these wind speeds an aerosol plume of neutral buoyancy injected at the ocean surface would take about 20 min to reach cloud base. [Calculations done following the analysis of Wood, 2021]

assume that all injected aerosol in this size range act as CCN. Wood (2021) calculated the required emissions rate from a point-source spray system in order to achieve ranges in ΔN_a at cloud base as a function of time after injection under typical conditions in marine stratocumulus regions. That study shows that achieving ΔN_a at cloud base of >30 cm^{-3} for 2-3 hours following injection requires an injection rate of 10^{15} particles s^{-1} or more (Figure 3), when injecting aerosol into a boundary layer with 4-10 m s^{-1} winds (typical of marine stratocumulus regions), and under the assumptions that the aerosol rapidly (within 10–20 min; Chosson et al., 2008) mixes through the shallow (<2 km deep) boundary layers typical in these regions, and that the plume has a horizontal spreading rate of 1.85 km hr^{-1} (based on LES studies and previous estimates of plume spreading in the literature; see Wood 2021). With time the plume mixes laterally, so that sustaining the aerosol perturbation for longer would require injecting between ~3×10^{15} and 10^{16} particles s^{-1}.

Producing a plume of salt particles that has a sufficient number of smaller (<200 nm dry diameter) particles to achieve a significant cloud brightening while simultaneously producing very few larger particles presents both engineering and scientific challenges. Particle concentrations will necessarily be extremely high in the region immediately downwind of the sprayer, leading to the potential for excessive particle-particle coagulation (Turco and Yu, 1997). To minimize coagulation, turbulence present in the ambient flow will be required to facilitate mixing of air from the sprayer with ambient air to produce the dilution necessary.

Aerosol composition

The efficacy of an aerosol as a cloud condensation nucleus is dictated primarily by its size, and only secondarily by its composition (Dusek et al., 2006; Petters and Kreidenweis, 2007). Nonetheless, more hygroscopic aerosol will more efficiently nucleate cloud droplets. Sea salt is highly hygroscopic, available in abundance and naturally occurring in the marine environment, and it is non-toxic. As such, it is effectively the only aerosol being considered for use in marine cloud brightening, and thus is the only aerosol that we propose using in the spray system for our studies. Nevertheless, it is becoming increasingly understood that sea spray aerosol often contains quantities of marine organic materials, and that these can reduce the hygroscopicity by coating the inorganic salts (e.g., Saliba et al., 2021). It will therefore be important to understand whether aerosols produced by a spray system using filtered sea water have similar hygroscopicity to natural sea spray aerosol, or behave more like pure sodium chloride. Ascertaining this information will be important in determining the CCN activity of the injected aerosol, which is critical for determining the concentration of cloud droplets formed during activation at cloud base.

Summary of spray system requirements

In sum, based on studies to date a spray system optimized to brighten marine stratocumulus clouds will need to produce:

- **a) 10^{15}-10^{16} particles s^{-1} of sea salt aerosol in the nominal 10-200 nm dry diameter size range, ideally with a peak in the size distribution near 30-60 nm dry diameter**
- **b) an aerosol plume that is sufficiently buoyant to effectively mix vertically through the marine boundary layer downstream of the spray system, so it reaches cloud base, and**
- **c) an aerosol plume where fewer than 0.1-0.2% of the generated aerosol particles have dry diameters larger than 2000 nm.**

For initial testing of whether the system meets these aerosol and plume requirements, we further specify that it be able to produce:

- **d) an aerosol plume meeting these aerosol size and injection rate requirements for at least 30 min of continual operation.**

We note that the aerosol specified above is quite a bit smaller than that in pollution or biomass burning plumes, where aerosols typically have dry mean diameters of ~100-300 nm. In this regard it is not a perfect proxy for current anthropogenic aerosol; however, as the aerosol size is optimized specifically for cloud brightening, observing the microphysical aerosol-cloud interactions and cloud responses to this aerosol can help place an upper limit on the negative radiative forcing driven by the less-optimally sized pollution and biomass burning aerosol.

3 Cloud Aerosol Research Instrument (CARI) description

The spray system to be used in this study, CARI, has an array of individual nozzles to generate the needed size and concentration of sea salt aerosols. The array of nozzles is installed in the front end to generate a plume propelled forward by a fan installed on the back end (Figure 4).

Figure 4. *The CARI system is similar in design to a snow making device, with an array of nozzles and generated aerosol accelerated by an integrated fan. (Shown here with a sub-set of nozzles 60 in an outer ring only; the system can accommodate a total of 250 nozzles.)*

Aerosols produced by the nozzles incorporated into CARI have been measured in an enclosed space (volume: approx. 355 m^3), using a 4-nozzle matrix. During these indoor studies, the particle size and concentration of the generated aerosol were continuously measured for approximately one hour, having achieved a high degree of mixing inside the test chamber three to five minutes after emission. The aerosol measured in these studies has an initial geometric mean diameter of 50 nm with a geometrical standard deviation of ~2.5 (Figure 5). The generated aerosol size and concentration shows very little dependence on ambient relative humidity (RH) in the indoor space, for RH ranging from 30% to 80%.

Each nozzle can produce up to 5 x 10^{12} sea salt crystals per second in the desired dry diameter size range of 10-200 nm, with fewer than 0.2% of the generated aerosols in the undesired >2000 nm diameter size. Further improvement of single-nozzle performance is currently being investigated by the team.

These indoor studies also indicate that having multiple nozzles run in close proximity, even when they are angled towards a common point in space, does not measurably affect the properties of the generated aerosol (i.e., the size distribution standard deviation) other than increasing the total aerosol throughput. These results suggest that the size of aerosol produced by the full (many nozzle) spray system will not differ appreciably from that seen in the indoor tests with a single or just a few nozzles. However, these indoor tests can only be operated with a limited number of nozzles running simultaneously (up to four); more than this and aerosol concentrations in the enclosed space become sufficiently high that aerosol coagulation dynamics affect the aerosol size distribution in ways that are not representative of concentrations that will be reached in the outdoor environment. In the outdoor environment, the aerosols will rapidly be dispersed through transport and mixing driven by ambient winds and atmospheric turbulence, such that it is expected there will not be significant coagulation more than a few meters downstream of CARI (Wood 2021).

Based on the ultimate desired aerosol production rate (3×10^{15}-1×10^{16} particles/sec in the 10-200 nm dry diameter size range), CARI is designed to accommodate hundreds of nozzles, with an initial nozzle matrix of 250 nozzle units. The CARI system allows for varying the number of nozzles as well as the operating condition to accommodate different aerosol generation rates. The study will therefore include characterizing how varying the number of nozzles and the operating conditions affect the concentration of aerosols in the generated plume.

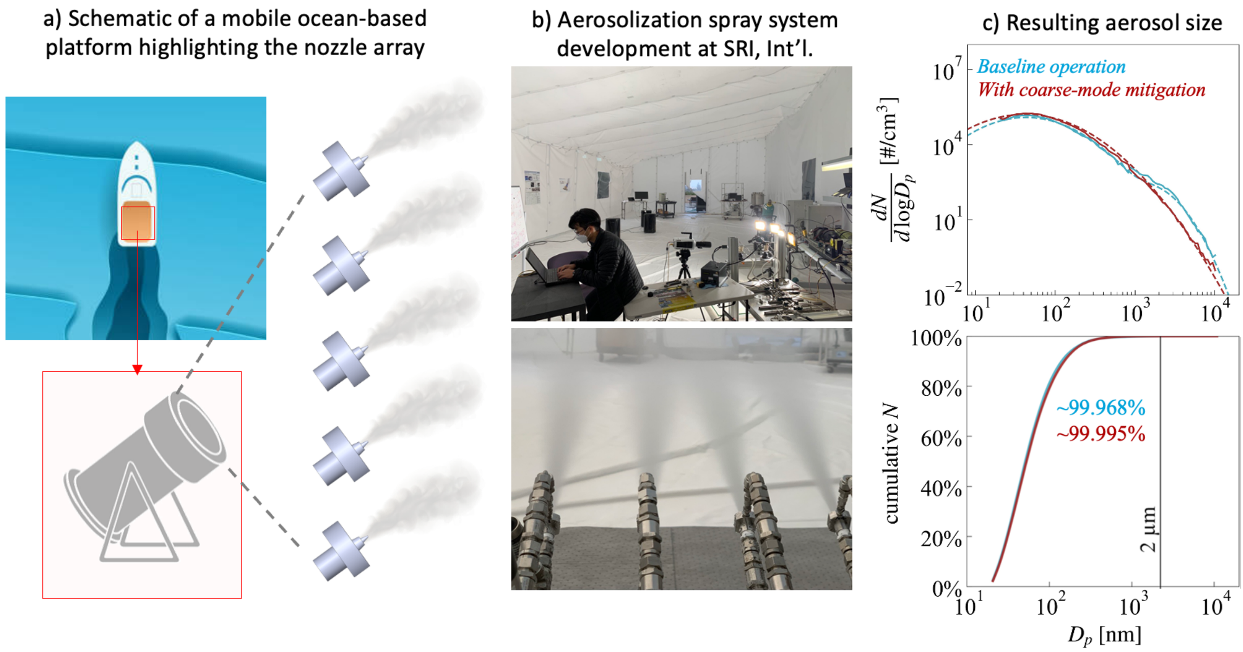

Figure 5. *The CARI system [bottom left] that would be used on a ship [top left] during the CACIE field studies has hundreds of nozzles in order to generate sufficient CCN to measurably perturb cloud droplet number concentrations. The individual nozzles incorporated into CARI have been tested individually and in multiples in different configurations [center photos] to study the impact on the generated aerosol size and concentration. Individual nozzles and arrays of up to four nozzles have been shown in the lab to generate a sea salt aerosol size distribution dominated by the desired 30-200 nm dry diameter range [top right], while producing very few of the undesirable larger (>2 µm dry diameter) size aerosols [bottom right].*

4 Aerosol plume study location

As noted above, the study described here will take place in a coastal location. We have opted to have our initial studies in a coastal location, rather than at sea, due to the greater cost and logistical difficulties of an ocean-based study. Conducting the early studies at a land-based site will allow for extensive testing and characterization of the CARI system generated sea salt plume and how this plume evolves and is transported in an atmospheric boundary layer similar to that in the ocean environment before moving to ocean-based studies.

Site meteorology

Following emission from CARI, the generated sea salt aerosol and how effectively it is mixed into the marine boundary layer and transported to cloud base will be affected by the ambient atmospheric conditions. The aerosol size distribution will be most strongly affected by how quickly the plume disperses and by the ambient relative humidity, as these two factors affect aerosol coagulation rates.

Lofting, dispersal and mixing of the plume vertically in the boundary layer will be affected by atmospheric stability and boundary layer winds.

Ideally, the coastal study described here would take place under conditions where the atmospheric boundary-layer winds, thermodynamic structure and relative humidity are similar to that in marine stratocumulus regions – the target for MCB and the CACIE study. However, conducting field studies at sea generally requires the use of ships, which are costly to operate, and either aircraft or additional ships to make the needed measurements at varying locations in the plume downwind of the spray system. An alternative suitable in particular for first studies using CARI is a land-based coastal site that is consistently influenced by marine air and that is configured to allow for observational platforms located at varying distances downwind of the aerosol generation point.

Desirable characteristics for a coastal land-based site that allows for study of how the generated aerosol would behave in marine stratocumulus regions include an atmospheric boundary layer with meteorological conditions, and in particular lower tropospheric static stability, comparable to that found in the open ocean in marine stratocumulus environments. This would include:

- boundary layer heights of appox. 500-1000 m (Albrecht et al., 1995; Wood and Hartmann, 2006; Wood, 2012)
- relative humidities of ~70-85% R.H. near the surface, and that ideally remain above 60% R.H. throughout the boundary layer (to remain above the sea salt aerosol deliquescence point; see discussion below in the context of lidar retrievals)
- winds ranging from ~2m s^{-1} to ~10-15 m s^{-1}

From a practical standpoint, the selected study site would also have winds that are of a consistent and predictable direction, so that observing instruments can be set up at relatively fixed locations downwind of the spray system.

A difference between a land site and the marine environment is that solar heating on land can appreciably add to lift and turbulence near the surface. Surface heating can be minimized by running the studies earlier in the day and preferably on cloudy days. When there is daytime heating, measurements earlier in the day would also favor boundary layer humidities more typical of that found in marine stratocumulus regions.

Facilities

Logistical considerations for the study site include:

- sufficient space for the CARI system and associated equipment, and for intensive in-situ aerosol observations (see description below) for a distance of at least 200 m downwind of the spray system
- the capacity to store and move the equipment needed for the described study (e.g. scales on the order of a shipping container)
- access to 330 KVA, 264 kW, 460 V power and multiple 30A, 120V standard power outlets for running the CARI system
- access to sea water, or a location for storing a tank of sea water

Study goals & design

The essential goals of this study are to:

1. **quantify how the size distribution of the generated sea salt aerosol from the CARI spray system evolves,**
2. **quantify the plume dispersion, mixing and aerosol concentrations under a range of atmospheric boundary layer conditions, particularly those similar to that in marine stratocumulus regions, at distances from immediately (<10 m) to ~200 m (or ~ 1 minute downwind, depending on wind speed) downstream of the system, and**
3. **constrain and improve simulations of aerosol transport and evolution over this same range.**

These goals are focused on aerosol characterization within 200 m of the spray system because it is expected that aerosol coagulation will rapidly become minimal after emission, so the aerosol size distribution within 200 m of the spray system should be representative of the aerosol that will be transported to cloud base. Similarly, parcel model studies indicate evaporative cooling would affect plume buoyancy only within a few meters of the spray system; beyond this, the plume will have mixed with ambient air such that the generated aerosol will be in equilibrium with the ambient humidity. As such, observing the plume mixing with ambient air and its transport for the first 200 m downstream of the spray system should be sufficient for determining whether negative buoyancy will prevent efficient transport of the generated aerosol to cloud base. Making comprehensive measurements of the aerosol size at greater distances downstream would also require a logistically complex and more costly field study, such as in-situ aerosol measurements from an aircraft.

As described below, a more limited set of measurement at sites downwind of the main study site may be possible. **An additional goal to be achieved as conditions and logistics allow is to:**

4. **characterize plume dispersion and vertical transport within the boundary layer at distances of up to 10 km downstream of the spray system, in order to better quantify efficiency of transport to the top of the boundary layer under different conditions.**

Combined, these goals will allow us to determine the sensitivity of the generated aerosol plume characteristics (size, concentration, plume dispersion and vertical transport) to *ambient meteorological conditions* and *system operational conditions*, and under what conditions the spray system meets the requirements defined in Section 2 above.

Measurements & observed parameters

The study will employ a mix of near-field (within 200-250 m of the spray system) in-situ observations and lidar observations further (~1-10 km) downwind to study the aerosol and plume characteristics and evolution (Figure 6 and Table 1).

Meteorological measurements

Surface temperature, pressure, humidity, and wind data will be collected using five weather stations located from just upwind to a few hundred meters downwind of the spray system. Temperature and humidity at various heights above the platform surface will also be measured from in-situ observational platforms on a series of scissor lifts (see below) to be located at three different distances downwind of the spray system. These will be supplemented by additional atmospheric profile data from the closest balloon sonde, released 00Z and 12Z daily by the U.S. National Weather Service.

As conditions allow, a miniMPL lidar (see below) will be used to retrieve the atmospheric mixed layer height before the sea salt plume is introduced. The atmospheric mixed layer height corresponds to the depth over which aerosols emitted at the surface will ultimately be efficiently mixed by the turbulent layer of atmosphere adjacent to the earth's surface (Stull, 1988; Seibert et al., 2000). It can be retrieved from surface-based lidar using various methods to identify boundaries between layers with high and low aerosol density (e.g., Ware et al., 2016; Solanki et al., 2019). As noted above, a study site that is representative of the marine boundary layer will have a quite shallow (500-1000 m deep) and humid boundary layer, so surface emissions of aerosols and aerosol precursors will be trapped in the boundary layer, and aerosol light scattering will be increased by aerosol humidification. In many cases this may produce a sufficient contrast between the air below and above the inversion to allow for mixing height retrieval from the miniMPL lidar signal. When such retrievals are possible, the mixed layer height will be used as one metric for how representative the study site is of the marine boundary layer in stratocumulus regions.

Aerosol and atmospheric in-situ measurements on tethered balloons

Instrument packages mounted on scissor lifts (Figure 6) will be located at varying distances downwind of CARI to measure near-surface profiles of the atmosphere (T and RH) and of aerosol size and concentration. The lift height will be dynamically adjustable to allow for measurement from the surface up to a height of 5.5 m for lift located immediately downwind of CARI and to just under 10 m for the other two lifts.

Assuming vertical and horizontal spreading rates are equivalent (i.e. assuming homogeneous isotropic turbulence), the aerosol should loft at a rate of about 1 km per hour. If the spreading rate is linear and the wind speed is 5 m sec^{-1}, at 100m downstream of the spray system the plume height will be about 6 m and at 200m will be about 12 m; if spreading follows a log-linear behavior, or the winds lower, it

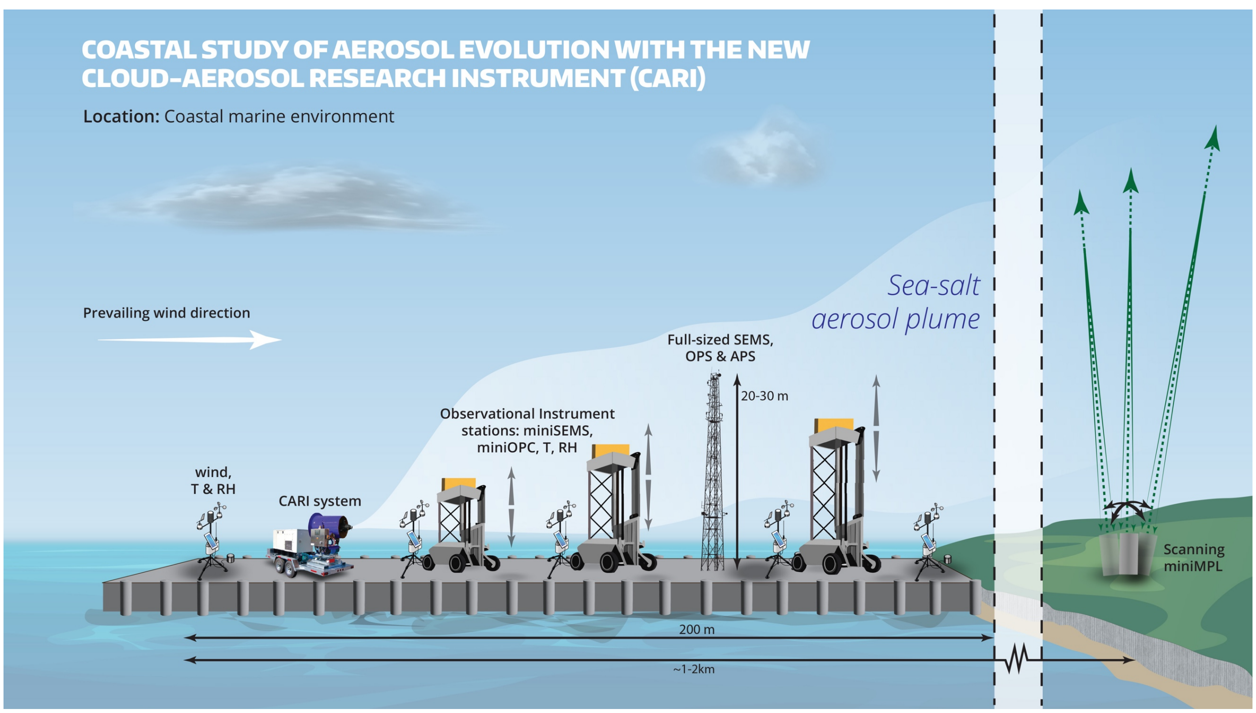

Figure 6. *Observational configuration of the sea-salt spray plume release studies.*

Table 1. *Summary of instruments to be deployed for this study, parameters they will measure, and associated notes (top), as well as observables to be provided by external data sources. More detailed descriptions are given in the main text.*

Scientific Parameter / Property	Accuracy/Specs	Instrument type (#) [location]	Notes
Measurements provided as part of the study			
Temperature	±0.5C	HOBOnet Wireless Temp/RH Sensor *(2) [surface, 10m upwind & 200m downwind from CARI]* *(4) [1-15m altitude on tethered balloons]*	Met data for: • initializing LES simulations • calculating ambient-RH size distributions from measured dry aerosol sizes
Humidity	±2.5% RH		
Winds	Speed: ±2mph (±1.1m/s) Direction:±7°	HOBOnet Anemometer *(2) [surface, 10m upwind & 200m downwind from CARI]*	
Aerosol size distribution [5-375nm]	Size res variable (10:1 typical)	Brechtel miniSEMS *(4) [1-15m altitude on tethered balloons]*	Measurements of dry aerosol size distribution (5nm to 3μm) from tethered balloons at variable altitudes in the plume, at 4 locations within 200m of CARI), and a filter-based measurement to estimate the concentration of giant CCN (aerosols >2μm dry diameter) in both the background and generated aerosol. All miniaturized instruments to allow deployment on tethered balloons. Mini GNI instrument(s) will be on loan from the University of Hawai'i.
Aerosol size distribution [130nm-3μm]	48 bins 0.15-0.5 μm; 24 bins 0.5-3 μm	Brechtel miniOPC Model 9405 *(4) [1-15m altitude on tethered balloons]*	
Giant CCN size distribution [~1-20μm]	dry salt particle size by microscope analysis	Mini Giant Nucleus Impactor (GNI)+ microscope analysis *(1) [1-15m altitude on tethered balloons]*	
Aerosol size distribution 5nm to 1.0μm		SEMS *(1) [5-10m fixed altitude, tower-mounted inlet]*	Measurements of dry aerosol size distribution (5nm to 20μm) from full-size instruments, allowing for a single, more robust measurement of the full aerosol size distribution and cross-calibration with the instruments on the tethered balloons.
Aerosol size distribution 0.5μm to 20μm		APS *(1) [5-10m fixed altitude, tower-mounted inlet]*	
Boundary layer depth	target: ±120m	miniMPL *(1) [1-10km distance from CARI, mobile platform]*	miniMPL vertical range resolution is 30m
Plume spreading & lofting	(see notes)		Plume boundaries will be set where attention-corrected aerosol backscatter is >2x the average background.
Spatial variation in aerosol concentrations across plume	(see notes)		Capability to retrieve concentrations will depend strongly on signal strength and confidence in our ability to constrain *S*, which will in turn depend on variability ability to estimate ambient-RH aerosol size distribution.
Measurements provided by external sources			
Surface temperature (SST) outside immediate study area	±2C	SST from NOAA High-resolution Blended Analysis of Daily SST	Data for initializing LES simulations
		Land surface temperatures from NASA MODIS Land Surface Temperature (LST) product	
Vertical temperature profile surface to 4 km	±1C @ ~10 m vertical resolution	Temperature & wind profiles from Oakland airport soundings (available 2x per day at 0 and 12 UTC, or 4pm and 4am local time)	
Vertical wind profile surface to 4 km	±2 m s^{-1} @ ~10 m vertical resolution		

would be somewhat higher. Hence, the scissor lift furthest from the CARI system may not be able to reach the top of the plume, but the observational packages on the first two lifts should, and all three will be able to reach into the plume.

Measurements will be made both before, during and after operation of the CARI system, to characterize the background conditions, the sea salt plume, and how the two interact.Dry aerosol size and concentration will be measured in packages on all three lifts using the miniaturized Scanning Electrical Mobility Sizer (mSEMS) (Brechtel Mfg.) and the miniaturized Optical Particle Counter (mOPC) (Brechtel Mfg.) with front-end dehumidifiers. The miniaturized version of the SEMS is capable of rapid scans (<~5 sec for the expected aerosol concentrations) of the aerosol size distribution for aerosol dry diameters of 5 nm to 375 nm. The mOPC has good overlap in size with the mSEMS, covering 150 nm - 3μm sized aerosol.

A single Mini Giant Nucleus Impactor (Tiang et al., 2021) will also be include in one of the observational packages to determine the number of super-micron aerosols being generated by CARI. Combined with the measurements from the full-sized SEMS, OPS and APS instruments (see next section) this will allow us to quantify whether the CARI system is meeting the requirement that fewer than 0.1-0.2% of generated aerosol particles are larger than 2 μm dry diameter.

Temperature and humidity sensors will also be included in each of the three observational packages. This will allow us to quantify the CARI emission impacts on T and RH at various distances downwind of the system. Previous studies have quantified how sea salt size varies with humidity (e.g. Zieger et al., 2017), and these growth factors will be combined with the measured dry aerosol sizes and ambient humidity to derive the ambient-RH aerosol size distribution.

Full-sized aerosol sizing instruments

While the miniSEMS and miniOPCs to be deployed on the scissor will measure most of the aerosols in the size range that will be generated by CARI, ambient aerosol of larger sizes may be present in the background atmosphere, and some number of large particles are also expected to be emitted by the spray system (Figure 5). The marine environment in particular can have a significant number of coarse-mode sea salt aerosols, so we need to study whether the generated aerosol plume significantly augments the ambient coarse mode aerosol population.

The background and perturbed aerosol size distribution will therefore also be measured using:

- Scanning Electrical Mobility Sizer (Model 2100 SEMS; Brechtel Mfg.) [5nm to 1.0μm]
- Optical Particle Sizer (Model 3330 OPS; TSI, Inc.) [0.3μm-10μm]
- Aerodynamic Particle Sizer (Model 3321 APS; TSI, Inc.) [0.5μm-20μm]

These instruments will be located on one of the scissor lifts; to start with, it will be located on the lift located about 100 m downwind of CARI, but can be moved to the other lifts for cross-calibration of the mSEMS and mOPC instruments.

The three instruments will be used to measure the aerosol size distribution and concentration in the background atmosphere and after introduction of the CARI sea salt plume in order to study how the generated aerosol size and concentration are affected by the background aerosol under different ambient aerosol and meteorological conditions. In addition, measurements of the background aerosol made with these instruments can be compared to that from the mSEMS and mOPC in order to cross-calibrate the miniaturized and full-sized instruments.

Plume remote sensing with Lidar

The in-situ measurements described above will allow characterization of the aerosol size distribution and concentration at distances of up to 200 m downstream of CARI, and study of how the size and concentration are affected by the ambient meteorology and background aerosol. Also of importance, due to its effect on aerosol dilution, and thus coagulation, and its relevance for transporting sprayed aerosol to cloud base, is understanding how the aerosol plume disperses in the atmosphere and, in particular, how effectively it is transported and mixes vertically.

Figure 7. *The miniaturized MicroPulse Lidar instrument built by Droplet Measurement Technologies is a widely-used instrument designed to measure tropospheric aerosols.*

The scanning mini Micro Pulse Lidar (miniMPL) (Droplet Measurement Technologies) (Figure 7) is designed to be easily portable and has low enough power requirements that it can be run off a generator if wall power is not available. As such, it can be mounted on a mobile platform and located at different distances downwind of CARI, based on the conditions during a given study period, and for measuring the plume at different points along its evolution. This instrument has a coaxial design with a manufacturer-calibrated overlap function, with full overlap at 150 m. The unit is eye-safe and comes with weather-proof housing. It provides fully programmable -10° to 90° elevation and -200° to +200° azimuth range scanning, so that the plume can be mapped in 3D, and not just directly above the instrument. The adjustable elevation angle will allow for measuring the lower reaches of the plume which, within the first 10 km of transport to be studied, is likely to be below the 150m range of the instrument's overlap region, by locating the instrument some lateral distance from the plume trajectory.

The miniMPL, as with all elastic-scatter lidars, measures the backscatter coefficient – i.e., the amount of light scattered in the backward direction. Measured or assumed atmospheric density profiles can be used to account for the contribution of air molecules to the backscatter coefficient. To convert the resulting aerosol backscatter coefficient to an aerosol light scattering coefficient or aerosol concentration (N_a, our desired quantity) requires knowledge of the aerosol lidar ratio, *S*, defined as the ratio of light scattered by aerosols in the backward (180°) direction to the total aerosol light extinction. Aerosol light extinction is the sum of aerosol light scattering and light absorption; for sea salt aerosol, light absorption is effectively zero so extinction equals scattering. Total light scattering and 180° back-scattered light – and therefore the lidar ratio, *S* – for spherical aerosols with a known size distribution and index of refraction can be calculated using Mie theory.

Parcel model calculations indicate that the shape of the dry size distribution of the aerosol measured by the lidar should be well-represented by that measured in-situ at the study site, as aerosol coagulation largely occurs in the plume within a few meters of the spray system. The index of refraction of pure dry sea salt aerosols has also been carefully quantified using laboratory measurements (Irshad et al., 2009). However, *S* must be calculated using the ambient – i.e., hydrated – aerosol size, not the dry size. Relative humidity as a function of altitude can be estimated from the surface to the scissor lift height on the main platform (Figure 6) and applying typical vertical lapse rates to infer R.H. at higher altitudes.

Previous studies have investigated the growth of sea salt aerosols of different sizes as a function of R.H. (e.g. Zieger et al., 2017), and their parameterizations can be used to calculate the ambient aerosol size distribution sensed by the lidar based on the dry size distributions and ambient R.H. Mixing rules can also be combined with knowledge of the indices of refraction for dry sea-salt and water to calculate the index of refraction of the hydrated aerosol. Finally, we note that the requirement that R.H. in the boundary layer at the study site remains above 60% assures that R.H. remains above the deliquescence point of sea salt aerosol (which is <60%; Zeng et al., 2013), allowing for an assumption of aerosol sphericity and therefore the use of Mie theory to calculate *S*.

In sum, the miniMPL will measure 3D distributions of aerosol backscatter, and therefore allow measurements of the plume spatial distribution. Using the procedure described above, 3D distributions of aerosol concentrations will be retrieved from the backscatter measurements, and uncertainties in inputs to the retrievals, such as in the value of *S*, will be used to quantify uncertainties in the retrieved concentrations. As discussed above, when conditions permit miniMPL measurements made preceding the sea salt plume release and following its dissipation will also be used to estimate the atmospheric mixing height and whether it changed during the studying a given plume, as a metric for characterizing the boundary layer and assessing its similarity to the marine boundary layer in marine stratocumulus regions.

Study Operations

A given study period will be 3-6 weeks duration, depending on the suitability of meteorological conditions for making observations. Studies will ideally be conducted three to four days per week, to allow for data processing and analysis and small engineering modifications on off days, with study days selected based on meteorological conditions. As noted above, conditions favoring observations include periods with winds consistent in direction and with relative humidities of >60%, though some observations will be made in periods with lower humidities in order to study the impact on the generated aerosol size. Typically, cooler morning temperatures will favor higher humidities, so it is expected that most studies will take place in the morning hours.

On a given day, CARI will generate aerosol in approximately 30 minute windows, alternating with periods to measure background aerosol and humidity. We expect there to be three to four cycles of aerosol generation on a given study day, with the meteorological in-situ instruments operating continuously across the entire study period on a given day. As dictated by shifting winds, changes in wind speed, and scientific priority for observing the plume at different distances downwind of CARI, the miniMPL may be moved between aerosol generation cycles and/or study days.

Modeling strategy

Both a parcel model and large-eddy simulations (LES) have informed this study design. The LES model used is the System for Atmospheric Modeling (SAM) version 6.10 (Khairoutdinov and Randall, 2003), modified to include representation of an injected Aitken mode aerosol (Wyant et al., 2022), such as that generated by CARI. Simulations with these models, for example, have provided estimates of the expected rate at which the sea salt droplets generated by CARI will come to equilibrium with the ambient air, the distance downstream of the instrument that coagulation is likely to significantly alter the size distribution, and the lateral and vertical mixing and transport rates of the plume under different meteorological conditions (see Figure 8). Numerical simulations will also be an integral part of the field study itself, both as a tool for studying the role of critical processes in how the aerosol plume evolves and for testing and improve model representation of aerosols in aerosol-cloud interaction studies where the aerosol orignates from a surface-based point source.

A particular focus will be on how near-field aerosol microphysical processes affect the generated sea salt aerosol size distribution. Aerosol microphysical processes in the very near field after injection occur at rates that depend nonlinearly on the aerosol concentration and size. Coagulation is dependent upon the dilution of the injected plume by the background air (Stuart et al., 2013). This dilution is controlled by turbulent mixing, which is a function of the buoyant and inertial stability of the environment and also will be impacted by the fan system in CARI, which is designed to maximize the flow across the nozzles and minimize particle concentration.

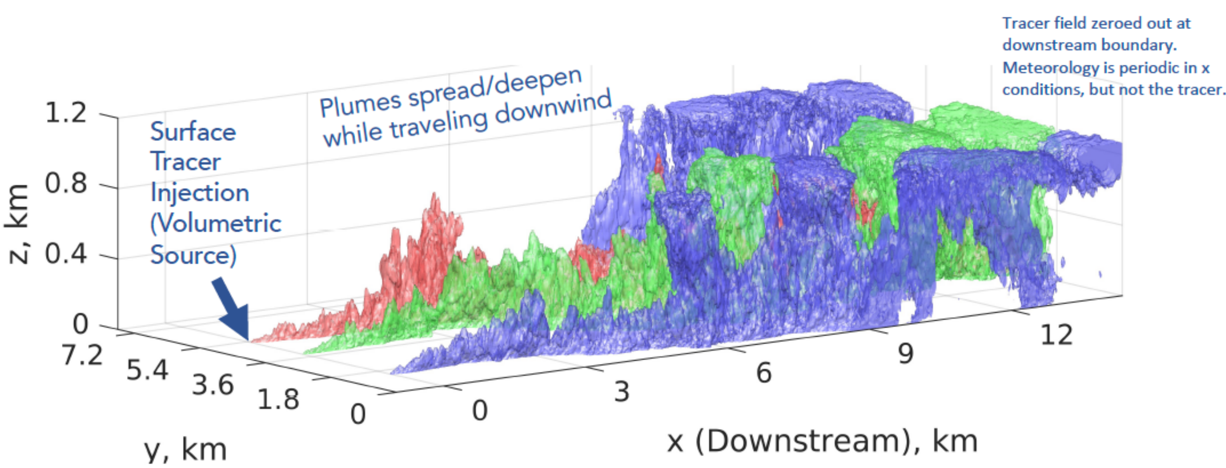

Figure 8. *High resolution LES simulations of tracer plume spreading downwind of an idealized spray system. The LES domain in this case is 15x7.5 km, with resolutions of $\Delta x= \Delta y=5m$, $\Delta z=5\text{-}25m$.*

The initial aerosol size distribution immediately after injection has been determined by testing individual nozzles in the laboratory, and by a few nozzles operating at a range of relative angles, but we need to be able to quantify how the presence of many nozzles injecting aerosols simultaneously and in close proximity influences the aerosol size distribution emerging downstream of the composite sprayer. The outdoor measurements will provide important constraints, but numerical modeling is needed to understand and ultimately mitigate any coagulation effects, if needed.

Computational fluid dynamics (CFD) simulations accounting for the CARI system geometry and operational parameters and ambient conditions will be used to study: a) how mixing affects droplet evaporation, collision and collescence in the first 10 m downwind of CARI and b) the fate of the generated droplets under different ambient conditions in the first ~100 m downwind of CARI.

LES will also be used to study the plume evolution over the entire study site. A size-resolved sectional aerosol model would form the base model, but this must be combined with a model that describes the turbulent mixing. The simplest turbulent models are Gaussian plume spreading models with spread/dilution rates parameterized as a function of the environmental stability (Pasquill 1961). More sophisticated variants of these involve multiple plume shells (e.g., Stuart et al., 2013).

A yet more realistic approach would be to embed the sectional aerosol scheme (e.g., the Model for Simulating Aerosol Interactions and Chemistry, MOSAIC, Zaveri et al., 2008) within a high-resolution eddy resolving model, which will then explicitly represent the aerosol dynamic processes within the turbulent boundary layer. Through collaboration with the Department of Energy's Pacific Northwest National Laboratory we are planning to collaborate on such simulations, likely within a newly developed Python-based large eddy simulation framework (PINACLES, Pressel et al., 2015). It will also be possible to take advantage of PINACLES' newly-developed capability to do runs with heterogeneous ground surfaces. The model plume spreading will be constrained using the observed plume structure at various distances downstream. We will likely use at least two LES models for our simulations: one with a detailed representation of the aerosol size distribution, and one with a much more simplified bulk aerosol scheme, with comparison between the two providing insights into the ability of a simple aerosol scheme to capture the near-field size distribution evolution.

A second potentially important effect of injecting seawater droplets is that they are relatively dilute compared with the concentration of salt in aerosols at equilibrium with typical near-surface relative humidities (typically ~70-80%). Thus, considerable water must be evaporated from the drops in the immediate near field of the spray system. This evaporation may produce negative buoyancy and has the potential to hinder the rate at which the injected aerosol plume is lofted toward cloud base. To fully understand these impacts requires high resolution computational fluid dynamics models that can include the effects of individual particles (or representative "super" droplets). Whether or not the LES simulations include either a sectional aerosol model or a representation that involves Lagrangian superdroplets is an open question that will be explored in test simulations. Key questions involve understanding if delayed lofting due to negative buoyancy results in additional particle losses (e.g., to dry deposition at the surface) or simply a delay in the time needed to loft particles to cloud base.

Laboratory testing shows that the aerosol size distribution produced by the single nozzle is rather broad. Despite having a modal diameter (of ~50 nm) in the size range that is effective for MCB, these tests also show that, while it is a small fraction of the total, a number of the injected salt mass is present in particles larger than 1-2 μm (Medrado et al., 2022; Figure 5). The presence of even a small number of supermicron (also known as "giant") cloud condensation nuclei (GCCN) can potentially lead to the following negative impacts for MCB efficacy:

- *Supersaturation suppression*: Coarse particles provide surface area that lowers the maximum supersaturation achieved in a given updraft and reduces the fraction of the smaller particles that can activate to form drops, reducing efficiency (see Ghan et al., 1998).
- *Precipitation nuclei*: Very large particles (dry diameter, D_{dry}, > 2 m) can lower the energy barrier to forming precipitation because they can initiate the collision-coalescence process (Houghton 1938; Feingold et al., 1999; Dziekan et al. 2021). Salt particles with D_{dry} > 2 μm swell to create droplets that are around 30 μm diameter at saturation (Hoffmann and Feingold, 2021). This leads to cloud water loss, offsetting or possibly overwhelming Twomey brightening.
- *Wasted energy cost*: These larger sea salt aerosols contain much mass that, if split up, would yield far more effective CCN. Given that laboratory tests currently show that 80-90% of the injected mass is in sizes D_{dry}>1 μm (Medrado et al., 2022), this potentially represents a significant waste of energy.

Lagrangian cloud parcel modeling using size-resolved aerosols and simulating cloud droplet activation can address the potential impact of different concentrations of GCCN on supersaturation and precipitation formation once the particles have reached cloud base. The size and concentration of GCCN in the ambient, background air and in the generated plume during the outdoor tests will be used as initial conditions for the microphysical modeling, which will then be used to drive cycles of nozzle improvement and testing with the goal of mitigating the production of supermicron particles (see Wood et al., 2022).

In addition to these process-focused studies, these observations will inform how to more realistically represent MCB aerosol injection in cloud-resolving modeling studies. Simulations will be conducted at higher resolution to study plume evolution in the near-field, using the same model the MCB Program team is using to simulate aerosol-cloud interactions under NOAA's ERB (Earth's Radiation Budget) program. In that study, the model representation of cloud evolution along trajectories from the stratocumulus to the cumulus regime in the NE Pacific is being systematically tested against observations, and the impact on cloud evolution is being quantified through a series of runs that include aerosol injections. Higher-resolution studies of the near-field evolution of a plume from a point source to scales matching the model resolution used in aerosol-cloud interactions will allow more accurate representation of the injected aerosol plume characteristics in those studies.

For this modeling component, Eulerian LES simulations of the observed aerosol and plume evolution will be conducted using the SAM model, and possibly also the PINACLES model. Higher-resolution, smaller-domain simulations will be run focusing on aerosol and plume evolution in the first 1 km and on plume mixing and transport within the first ~10 km after emission. The horizontal grid resolution of these runs will vary from <5 km for the former runs to 20 m for the later runs. Vertical grid resolution in the SAM model gradually varies from 5 m at 400-800 m altitude, where clouds form, to 15 m near the surface and to 70 m at the top (1.55 km) of the vertical domain.

Surface temperatures are specified in the model. For this coastal study, local data on sea surface temperature will be used to specify the surface temperature for the parts of the domain covered by ocean. Temperatures measured at the five weather stations in the ~100 m x 250 m area where CARI and the instruments are located will be used to interpolate the surface temperatures over that domain. Both the study site temperature measurements and publicly reported temperature data will be used to estimate surface temperatures over the rest of the model domain. For each period of plume observations, the model will also be initialized with atmospheric temperature, humidity and wind profiles generated using a combination of the meteorological observations made as part of the study (described above), merged with temperature, humidity and winds from twice daily nearby weather sondes and reanalysis data.

In the LES, aerosol will be injected into to the single model gridbox corresponding to CARI's location. Initially, the size of the injected aerosol will be based on the dry aerosol size distribution measured in the lab, then hydrating to ambient RH using standard sea salt aerosol size growth factors. The number of droplets will be scaled by simply linearly scaling from the number produced by a single nozzle to the number of nozzles used in the study. We fully expect this to be an iterative process (e.g. to account for any impacts on aerosol size or production efficiency) based on measurements made during the field studies. The results of the near-field CFD simulations described above will likely also be used to initialize the aerosol field in the LES model. The model will then be run in Eulerian mode to simulate the plume evolution. The LES model will be able to simulate RH variations in the plume, including the effects of initial droplet evaporation as the dilute seawater droplets adjust to their hydrated equilibrium sizes. It is possible that the RH may approach or even exceed saturation due to the excess water vapor and associated evaporative cooling. In this case, we may need to represent droplet activation in the early stages of the plume itself. It is unclear if this will occur, for what duration, and if this impacts the eventual aerosol size distribution downwind of the spray system.

The simulated aerosol size distribution and gradient in aerosol concentrations will be compared to that measured at multiple points in-situ in the first 200 m downwind of the spray system, as described above. The simulated plume spatial distribution will also be compared to that retrieved from the miniMPL further downstream (at a location varying from 1 to 10 km from CARI, depending on where the lidar is located for a given study) as a test of the model representation of aerosol transport (lofting and turbulence in particular). In cases when aerosol concentrations can be retrieved from the miniMPL, this would provide a further test of the simulated effects of dilution.

Scope of Studies of Generated Aerosol Evolution

Using the observations and model simulations described above, studies will be conducted of the generated sea salt aerosol plume under a range of meteorological and operational conditions, allowing us to meet the scientific goals of the study.

Varying meteorological conditions

Studies will target a range of meteorological conditions to test how sensitive the plume evolution is to, in particular, ambient boundary layer relative humidity, depth, stability and winds. Of most direct relevance to the utility of the spray system for the CACIE aerosol-cloud interaction study is determining the aerosol and plume evolution when conditions are representative of that in the remote marine boundary layer. Studying how sensitive the generated aerosol properties are to more widely varying conditions is also of interest since conditions in the remote marine environment may not always be "typical" and because this will provide information on the potential utility of the spray system for aerosol-cloud interaction studies in other locations.

During fair weather conditions, coastal sites generally have a diurnally-driven cycle of land-sea breezes, with onshore winds increasing in the afternoon. Driven by nighttime cooling and daytime solar heating, the boundary layer is generally shallower and more humid in the morning and deeper and drier in the afternoon. Cloudier days will have less surface-driven turbulence and convection than sunny days, when solar heating of the ground provides a source of surface lift that is not present in the offshore marine environment. This naturally allows for studying the generated aerosol plume under a range of conditions:

- In the morning on cloudy days with light (~1-5 m s^{-1}) winds of a consistent direction conditions will be optimized to study the generated aerosol plume in conditions most similar to that in marine stratocumulus regions. Slower wind speeds will additionally mean instruments located at fixed distances downwind of the spray system will observe the aerosol evolution over a longer time period after emission, since transport to these instruments will be slower.
- Comparison of the plume evolution in the morning with that later in the day, and on sunny versus cloudy days, will allow us to quantify the sensitivity of the generated aerosol size, concentration and vertical transport to ambient relative humidity, winds, turbulence and convection.

Varying background aerosol size and concentration

As local conditions allow, studies will be done to quantify how the size of the generated aerosol in particular, as well as its concentration, is affected by variations in the background aerosol concentration and size. At a coastal site, windy days in particular may produce significant coarse mode sea salt aerosol, as it would in the more remote marine environment, providing an opportunity to study whether the generated aerosol size distribution is altered through, for example, Brownian diffusion and coagulation with ambient coarse-mode aerosol.

Varying operational conditions

Studies will also be conducted of how the generated aerosol properties vary under different spray system operational conditions. Indoor studies of the nozzles incorporated into CARI have been conducted using a solution made from a synthetic seawater (sea-salt ASTM D1141-98 composition and DI water) to produce 3.2% to 3.5% salinity, which is typical of ocean water. Eventually the CARI system will include a filtration system so it can be run with ambient sea water. This filtration system will be designed to filter out any biological material and pollutants in the sea water, so we expect the aerosol generated with the sea salt standard solution and filtered sea water will be effectively identical. Nonetheless, to avoid any concerns of contaminants of any sort being sprayed in the area of the coastal study, the CARI system will initially be operated only using either fresh water or a sea salt standard solution of 3.2-3.5% salinity. Later studies may include using CARI with sea water from the open ocean, filtered on-site, to test whether the aerosol size and concentration differs when using the sea salt standard solution versus filtered ocean water.

The following studies assessing the effects of varying operational conditions will be run:

<u>Nozzle matrix scaling effects</u>. Studies will be done of the effects of scaling up from the 4-nozzle matrix tested indoors to the full CARI system, which currently includes a 60 nozzle-matrix. The team will first run tests using fresh water to assess the proper engineering behavior of the installed equipment (i.e., nozzle distribution system, compressed-air system and high-pressure pumps). Following successful operation of the system with fresh water only, the 60-nozzle system will be run using the sea salt standard solution. Measurements of the generated sea salt aerosol will be compared to those done in a laboratory of an individual nozzle and of up to 4 nozzles. Depending on the results using the 60-nozzle system, we anticipate scaling up to a 250-nozzle system. Each step in the transition will inform the engineering modifications required to accommodate all nozzles and guarantee that the final number, placement, and operation of each nozzle is optimized.

<u>Spray rate and gas-to-liquid ratio variations</u>*.* The CARI system operates by combining air and sea salt solution flows in an array of nozzles. The number of aerosols generated can be varied by changing a) the number of nozzles utilized in the spray system and b) the water and air flow rates. Our indoor studies show that maintaining a fixed ratio between the air and sea salt solution flows (the "gas to liquid ratio", GLR) will produce a particular aerosol size distribution. This ratio will be varied, and the effects on the generated aerosol size and concentration quantified, as both a test of how sensitive the generated aerosol is to maintaining a consistent GLR and as a test of the degree to which varying the GLR can be used to customize the generated aerosol size.

<u>Sprayer angle</u>. The design concept of the CARI system is based on that used for snow-making machines. These machines operate by having the array of nozzle installed on the front-end of a metallic case, generating a plume that is propelled forward by a fan installed on the back end. The machine allows for the rotation of the nozzle matrix from vertical to horizontal position. Studies will be done to evaluate how the sprayer orientation impacts the generated aerosol as well as the mixing of the generated aerosol into the boundary layer.

Reporting of findings

Following standard scientific practice, results from the study will be reported through peer-reviewed publications and talks and posters at scientific conferences. Data from the field study measurements will be made publicly available either following the acceptance of papers reporting the results or within one year of the end of the field study, depending on which occurs first.

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