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Drizzle Representation via Enclosed Atmosphere Measurements and Modeling (DREAM²)

September 2026

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Abstract

Improving the ability of models to simulate precipitation will require breakthroughs in representing several poorly constrained cloud and aerosol processes, including droplet collision-coalescence rates, ice nucleation, scavenging in mixed-phase clouds, and the in-cloud chemical processing of atmospheric aerosols. These complex processes are fundamentally difficult to measure in the free atmosphere and cannot be fully addressed by existing cloud chambers. DOE is exploring the idea of building a new laboratory facility to study these and other important atmospheric processes under representative yet controlled, reproducible, and sustained cloud conditions to improve large-scale simulations of weather and climate. This LDRD project consists of a series of six experimental, instrument development, and model development activities that will position PNNL for a leadership role in a future DOE cloud chamber facility. First, we will use a combination of instruments unique to PNNL—a single particle mass spectrometer coupled to a counterflow virtual impactor—to demonstrate experimental measurements of how cloud droplets grow via collision-coalescence, a key uncertainty in converting clouds to precipitation. Second, we will perform experiments examining the chemical processing of organic gasses in cloud droplets and perform simulations to guide and interpret the experimental results. This task will provide data on the relative importance of cloud-phase production of organic aerosol, a process that has been extensively modeled, but poorly constrained by experimental data. Third, we will develop a new holographic droplet sensing instrument to measure the cloud droplet size distributions inside cloud chambers with minimal disturbance to the chamber environmental conditions and flow. Fourth, we will develop a digital twin of a cloud chamber using a PNNL-developed model capable of representing turbulence and other small-scale processes in the chamber. The digital twin will enable us to reproduce and interpret the cloud droplet microphysics in chamber experiments. The fifth task consists of a series of modeling exercises demonstrating our ability to utilize cloud chamber observational data to improve the representation of cloud microphysical processes in models, thereby demonstrating a pathway for knowledge gained from chamber process studies to impact precipitation. Finally, we study how secondary organic aerosol coatings impact the ability of dust particles to nucleate ice. Collectively, these six tasks will demonstrate new and unique capabilities needed to maximize the scientific impact of cloud chamber research.

Summary

This report summarizes the results of six experimental and modeling projects aimed at increasing the scientific understanding of clouds, aerosols and the interactions between them. These projects developed new capabilities at PNNL that can be used in future cloud chambers to increase the scientific impact of these facilities. Ultimately, these findings will improve model representations of clouds, aerosol, and precipitation.

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Acronyms and Abbreviations

CPU	central processing unit
DOE	U.S. Department of Energy
DREAM ²	Drizzle Representation via Enclosed Atmosphere Measurements and Modeling
E3SM	Energy Exascale Earth System Model
EMPM-Chem	Explicit Mixing Parcel Model coupled to Chemistry
ePIV	event-based Particle Image Velocimetry
ERF	Energy Research and Forecasting
GPU	graphics processing unit
HOLODROPS	HOLOgraphic DROplet Sensor
IEPOX	isoprene epoxydiols
INC	ice nucleation chamber
INP	ice-nucleating particle
JEFE	Jacobian Estimation of Functional Error
LDRD	Laboratory Directed Research and Development
ML	machine learning
MTU	Michigan Technological University
OSSE	observation-system-simulation experiment
PCVI	pumped counterflow virtual impactor
PNNL	Pacific Northwest National Laboratory
SOA	secondary organic aerosol

Contents

Abstract.....	ii
Summary.....	iii
Acknowledgments.....	iv
Acronyms and Abbreviations	v
1.0 Introduction	2
2.0 Outcomes.....	3
2.1 Research Objective 1: Direct measurement of collision-coalescence.....	3
2.2 Research Objective 2: Investigate SOA cloud processing	4
2.3 Research Objective 3: Characterize cloud droplet size distributions.....	5
2.4 Research Objective 4: Develop a digital twin of the DOE chamber	6
2.5 Research Objective 5: Demonstrate an end-to-end strategy for utilizing chamber observations to improve climate models	7
2.6 Research Objective 6: Investigate the Effect of SOA coatings on Warm- Temperature Ice Nucleating Dust Aerosol.....	8

Figures

Figure 1.	a) Time series of miniSPLAT-measured size (d_{va}) of aerosol particles (AI) and cloud droplet residuals (PCVI) in the MTU Pi chamber. (b) Mobility-resolved d_{va} distributions of cloud droplet residuals after IEPOX-SOA aqueous processing, showing the dominant reacted particle population and the distinct coalesced ammonium sulfate peak.	3
Figure 2.	Cloud droplet size distributions measured during experiments in the MTU Pi chamber. The figure shows that IEPOX chemistry generated a cloud.....	4
Figure 3.	HOLODROPS images of commercial resolution targets demonstrating the instrument resolution.	5
Figure 4.	An illustration of ERF super droplet simulation of the Pi-chamber showing warm and cold air plumes (colored isosurfaces) and positions of selected super droplets (light blue dots). The inserts depict injected dry aerosol size distribution (green) and the mean haze/cloud droplet size distribution in the chamber (blue).	6
Figure 5.	ML-enabled digital-twin analysis identified informative sensor configurations for a tall convection-cloud chamber. Virtual sensors sampled synthetic chamber conditions generated under varying physical-process assumptions. The results show that the measurements are sensitive to collision–coalescence efficiency, with sensitivity varying by sensor location.	7
Figure 6.	Concurrent changes of ice activation and SOA coatings for thin and thinner coating experiments.	9

1.0 Introduction

Several of the largest uncertainties in predicting seasonal to decadal availability of water for energy production are associated with clouds. While considerable progress has been made in understanding cloud properties and processes, a number of important gaps remain. For example, we lack quantitative knowledge of critical parameters and processes that control the evolution of the size and number of cloud drops, including collision-coalescence rates, and the rates at which they convert to precipitation. We also lack tools for understanding and representing in-cloud chemical processing of secondary organic aerosols (SOA). These complex processes are fundamentally difficult to measure in the free atmosphere. This LDRD project consisted of a series of six experimental, instrument development, and model development activities that will enhance our understanding of cloud processes and the interactions between aerosol particles and clouds. Here, we briefly report the major findings from these projects.

2.0 Outcomes

2.1 Research Objective 1: Direct measurement of collision-coalescence

The formation of drizzle embryos, or drizzlets, i.e., droplets few tens of microns in size, is a poorly understood process in cloud and precipitation physics and is difficult to directly observe in real clouds. The effects of the collision-coalescence are usually inferred from measurements of the cloud droplet size distribution. However, other processes also contribute to changes in the droplet size distribution (e.g., evaporation and condensation due to fluctuations in water vapor supersaturation within turbulent eddies). Therefore, measuring the evolution of the size distribution by itself provides only indirect evidence of collisional growth. This research objective set out to develop and demonstrate a direct measurement of collision-coalescence in cloud droplets using a combination of the single-particle mass spectrometer, miniSPLAT, and a pumped counterflow virtual impactor (PCVI).

We first demonstrated that miniSPLAT was able to detect the minor component of a mixed particle population on timescales comparable to a cloud lifetime. In these experiments miniSPLAT successfully detected the minor component present at <1% a total concentration of only ~ 83 particles cm^{-3} . In the same experiments, coagulated particles were also detected and, from their combined size and composition, were determined to be composed entirely of the major component, NaNO_3 , demonstrating that miniSPLAT can identify individual coagulated particles. The measurements were then extended to deliquesced particles, which are a closer analogue of coalescing cloud droplets, where miniSPLAT successfully detected and quantified rare wet coagulation events. The most significant result was a demonstration of chemical sensitivity: the minor component of the particles contributed less than 5% of the volume of the coalesced particles, yet they were readily identified by miniSPLAT. The integrated PCVI/miniSPLAT system was deployed at the MTU Pi cloud chamber and operated during experiments on IEPOX SOA formation in cloud droplets as described below. In this experiment, ammonium sulfate seed particles were activated into cloud droplets and exposed to IEPOX, a reactive organic gas. In this experiment, miniSPLAT detected reactive uptake of IEPOX gas onto the ammonium sulfate particle population (Figure 1b, particles with $\rho=1.4$).

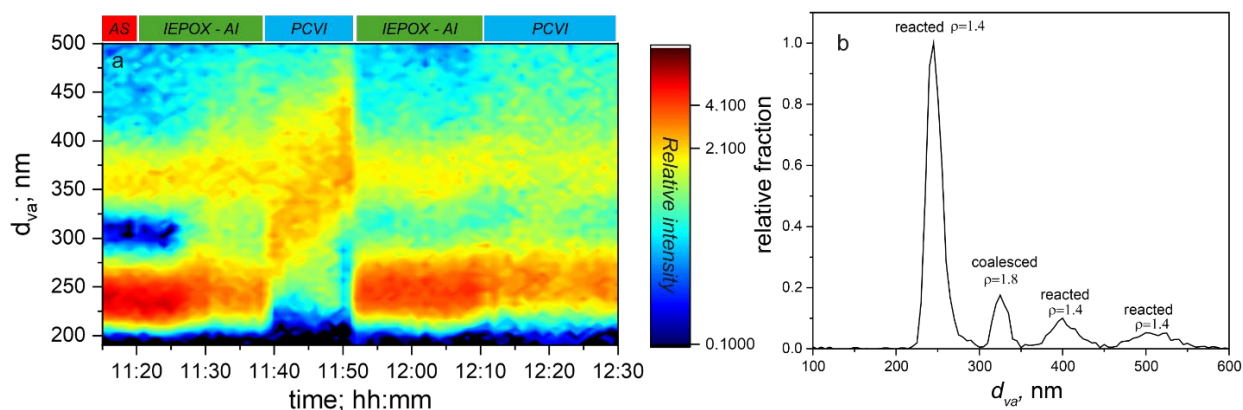
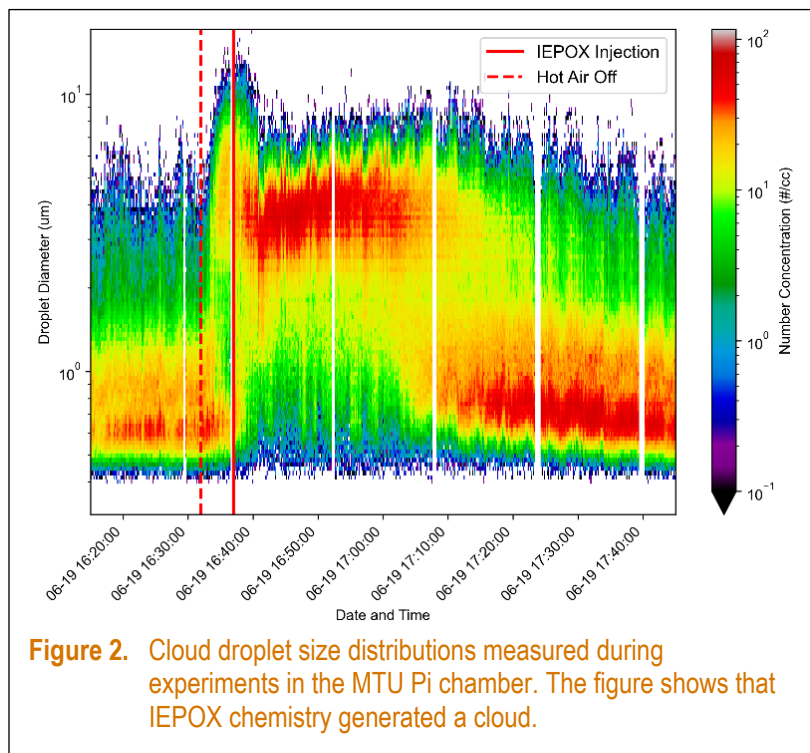


Figure 1. a) Time series of miniSPLAT-measured size (d_{va}) of aerosol particles (AI) and cloud droplet residuals (PCVI) in the MTU Pi chamber. (b) Mobility-resolved d_{va} distributions of cloud droplet residuals after IEPOX-SOA aqueous processing, showing the dominant reacted particle population and the distinct coalesced ammonium sulfate peak.

In addition, it detected a distinct secondary peak at $p = 1.8 \text{ g cm}^{-3}$ corresponding to coalesced ammonium sulfate residuals with no IEPOX. This demonstrated that coalesced droplets can be detected, sized, and chemically fingerprinted within an active cloud.

2.2 Research Objective 2: Investigate SOA cloud processing

Several field and modeling studies have suggested that cloud processing could be a significant and sometimes dominant source of secondary organic aerosol (SOA), which grows small aerosol particles to sizes capable of nucleating cloud droplets. However, relatively few studies have investigated the processing of SOA in authentic cloud droplets. Instead, most studies are



conducted in bulk solution and results are extrapolated to liquid water content characteristic of clouds. In previous work, we showed that chemical kinetics derived through these types of bulk solution measurements are too fast compared to the in-situ measurements of isoprene aqueous SOA formation in aerosol. However, measurements of SOA processing in actual cloud droplets have been lacking so far. Experiments that directly examine cloud processing of SOA vapors and particles are needed to determine the conditions under which cloud processing produces significant amounts of SOA or significantly changes SOA particle properties.

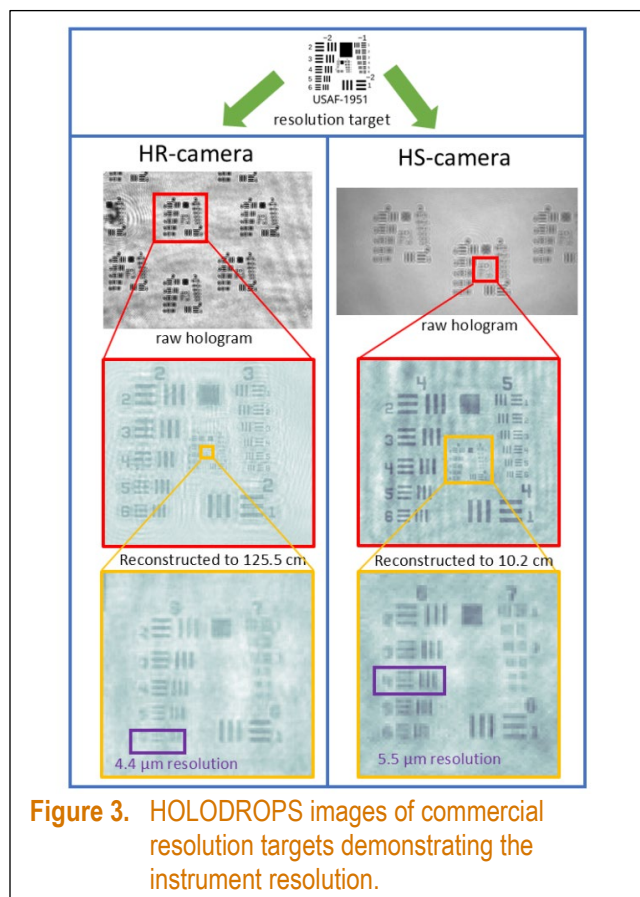
We conducted experiments in the Michigan Technological University cloud chamber in which unactivated aerosol (haze) and cloud droplets were exposed to IEPOX gases. The experiments reveal that IEPOX gas is converted to SOA on timescales of tens of minutes in the chamber. This gas-to-particle conversion is likely driven by cycling between cloud and aerosol particles. Measurements show that this chemical processing profoundly affected the cloud droplet size distribution, effectively forming a cloud from haze mode particles (Figure 2). This cloud mode persists for tens of minutes until IEPOX gas and SOA products are removed by deposition. IEPOX chemistry is important in many regions of the atmosphere. These experiments suggest aerosol chemistry and clouds are more tightly coupled than is represented in current earth system models.

We also developed the Explicit Mixing Parcel Model coupled to Chemistry (EMPM-Chem), a novel extension of the one-dimensional explicit mixing parcel model that incorporates droplet-resolved multiphase chemistry to simulate secondary organic aerosol (SOA) formation from isoprene epoxydiols (IEPOX) in individual haze particles and cloud droplets. EMPM-Chem resolves processes at millimeter scales that conventional models cannot efficiently represent because of their high computational cost. We showed that IEPOX dissolution is preferentially

weighted toward larger cloud droplets because of their greater water content. In contrast, conversion to IEPOX-SOA proceeds most rapidly in smaller, more acidic, and concentrated haze particles. This work establishes EMPM-Chem as a new capability for resolving droplet-scale chemical processes that govern SOA formation. It provides mechanistic insight and a modeling foundation for improving the representation of these processes in larger-scale atmospheric models.

2.3 Research Objective 3: Characterize cloud droplet size distributions

The study of atmospheric processes is intricately linked with the advancement of instrumentation, as cutting-edge tools are essential for unravelling the complexities of the atmosphere. Many properties and processes related to clouds in particular need improved observational data to fully understand the underlying processes and to improve predictions of cloud properties and precipitation. To understand, for example, why some clouds rain, and others do not, we need to measure the internal structure of the clouds and the droplet size distributions accurately, which can only be done with measurements by deploying the high precision instruments in a cloud chamber or a cloudy environment.



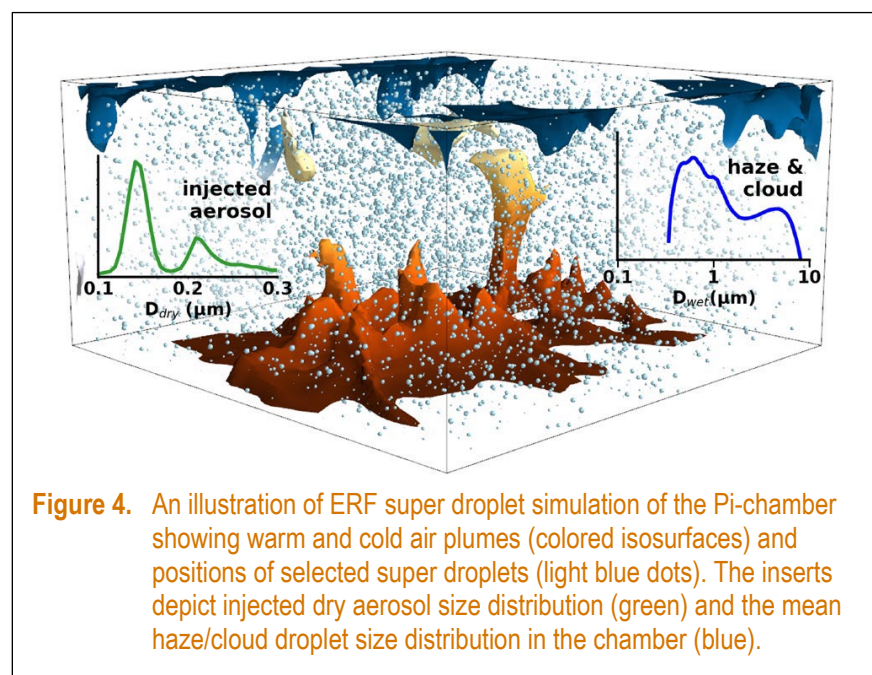
In this research objective, we develop and characterize an advanced holographic instrument — HOLODROPS (HOLOgraphic DROplet Sensor). The HOLODROPS operates in two configurations: (1) HOLODROPS, designed for long-term, high-precision measurements of cloud droplet size distributions, and (2) HOLODROPS-TRACK, designed for targeted, short-interval particle tracking to study turbulence and collision-coalescence processes. Unlike traditional optical scattering or imaging probes, both configurations can simultaneously capture the size, shape, and three-dimensional position of multiple droplets and ice crystals with diameters larger than $8.8\ \mu\text{m}$ within a sample volume, eliminating the need for multiple instruments and yielding more accurate and cohesive data.

The detailed three-dimensional particle tracking provided by HOLODROPS-TRACK requires significant computational resources for hologram reconstruction and data processing, limiting its use over extended measurement periods. To address this

challenge, we developed a complementary event-based Particle Image Velocimetry (ePIV) system that uses an event-based camera to assess key cloud motion features in real time with substantially lower data volume and processing. ePIV provides continuous monitoring of fluid flows to identify optimal measurement windows and informs targeted deployment of

HOLODROPS-TRACK, paving the road for efficient, high-resolution particle tracking when conditions are most scientifically valuable. We successfully characterized and validated all three instrument operational modes in the laboratory and have submitted a manuscript describing the results to a peer-reviewed journal (Figure 3). Additionally, we tested HOLODROPS outdoors during a drizzle event to demonstrate the prototype's adaptive operation. We also reconfigured a weather-proof HOLODROPS module for long-term outdoor operation.

2.4 Research Objective 4: Develop a digital twin of the DOE chamber



A digital twin is a virtual representation of an actual physical facility. It can simulate processes of interest in a laboratory setting and assist in interpreting observations from experiments.

Perhaps more importantly, the digital twin can be used to predict the behavior of the system under various operational scenarios, refine the controls to achieve desired regimes, and evaluate and optimize various observational methods. Although many factors contribute to the efficacy of the digital twin, including

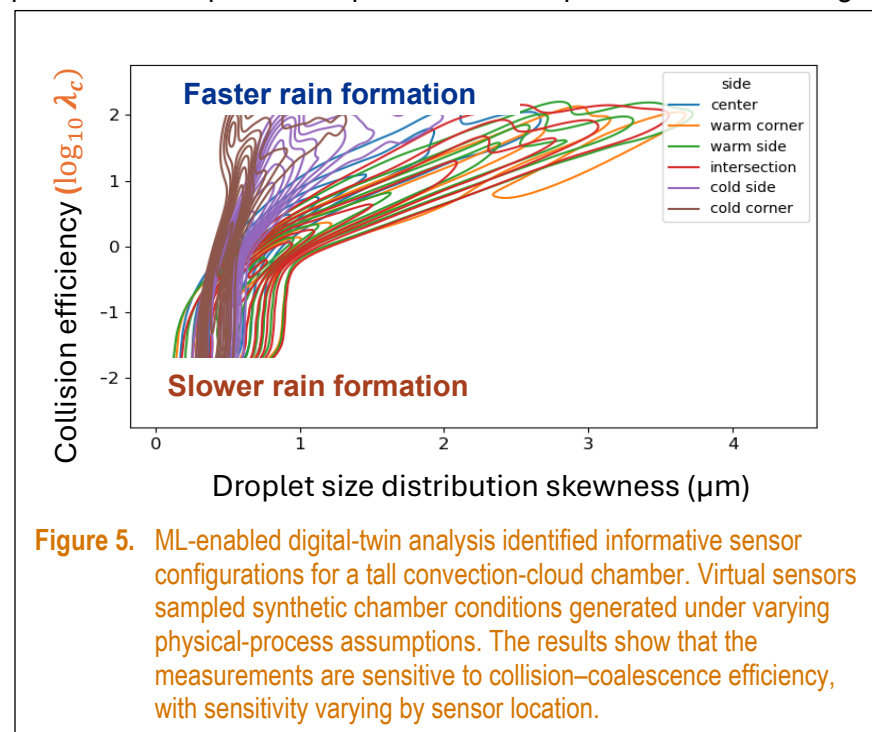
meaningful integration of models and observations, the key underpinning for a viable digital twin is a reliable model capable of integrating representations of all major processes in a convection cloud chamber. This objective aims to build such a model, using existing and newly developed components. We aim for the digital twin to become an essential tool for exploring design options for the proposed chamber and, once the chamber is built, to serve as an integral part of the operational facility. In this project, we will be performing simulations in two main configurations: the Pi-chamber to evaluate the performance of the model and the scaled-up tall chamber to explore conditions in the proposed DOE facility.

The Digital Twin effort has made substantial advances in model development, numerical simulations, and analysis capabilities for cloud chamber research. We adapted the Energy Research and Forecasting (ERF) atmospheric modeling code – built on a massively parallel, block-structured adaptive mesh refinement framework – to simulate the cloud chamber environment with excellent computational performance, achieving nearly an order-of-magnitude speedup on GPUs compared to the previous CPU-only implementation. Building on this foundation, we developed a versatile wall boundary condition treatment for the Pi chamber, using Direct Numerical Simulations as a benchmark, and demonstrated that tiled sidewall boundary conditions can significantly improve mixing uniformity in tall chambers (Thomas et al., 2025). The project also carried out a large ensemble of tall-chamber simulations for the Research Objective 5 to explore how instrument placement influences the extraction of physical insights, providing a proof of concept for digital-twin-guided experimental design (Wang et al,

2026). To faithfully represent the Pi-chamber conditions during the experiments in the Pi Chamber for the Research Objective 1, we employed a Gordon Bell award-winning particle-in-cell algorithm to model aerosol and cloud particles using Lagrangian “super-droplet” microphysics, conducting a suite of simulations that investigated haze and cloud droplet behavior under various sidewall boundary conditions and aerosol injection scenarios. To support efficient analysis workflows, we also developed and released pltview, a lightweight open-code visualization tool enabling rapid inspection and plotting of ERF output. Together, these accomplishments establish a robust computational foundation for integrating chamber experiments with advanced modeling and machine learning tools, forming a basis for the digital twin.

2.5 Research Objective 5: Demonstrate an end-to-end strategy for utilizing chamber observations to improve climate models

Cloud microphysics must ultimately be understood across a hierarchy of scales, from controlled laboratory studies and process-resolving simulations to parameterizations used in Earth system models. A central challenge in this area is equifinality: distinct combinations of uncertain process assumptions and parameters can produce similar emergent model behavior, making it



difficult to determine whether models are reproducing the right outcomes for the right physical reasons. This research objective addressed that challenge by developing and applying complementary machine learning, Bayesian inference, and numerical modeling approaches to connect and accelerate multiscale simulations of cloud processes, including cloud-chamber studies, high-resolution cloud-field simulations, and global simulations in the DOE's Energy Exascale Earth System Model (E3SM).

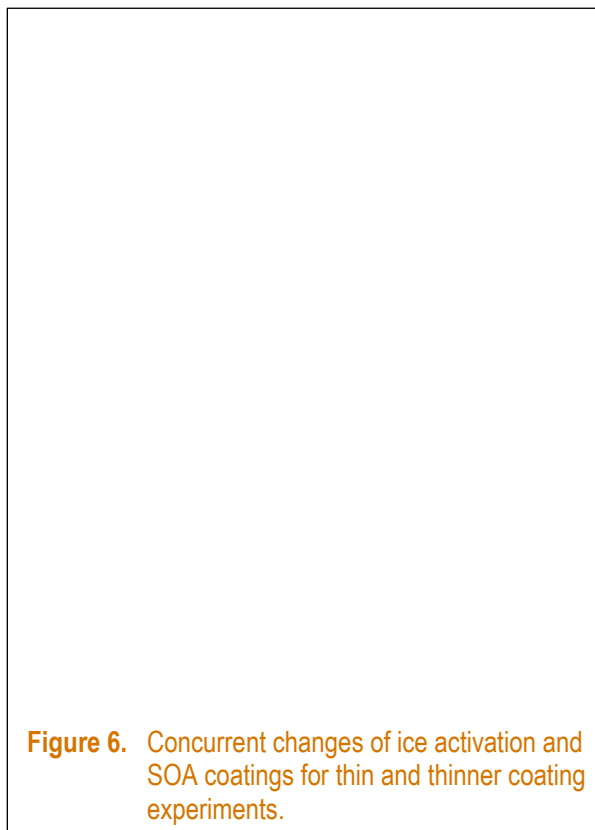
We developed and demonstrated an ML-enabled digital twin of the proposed DRACO tall convection-cloud chamber to evaluate how sensor placement affects the ability to infer key cloud microphysical process rates. By varying physical process rate assumptions and sampling simulated chamber states with virtual instruments, we showed that a sparse sensor network can provide useful constraints on collision-coalescence efficiency and can guide instrument placement prior to experimental deployment (Wang et al., 2026). In response to DOE feedback, we extended this observation-system-simulation experiment (OSSE) approach into an E3SM-centered Bayesian framework focused on marine stratocumulus observations from DOE field campaigns. Using an idealized perturbed-parameter experiment and Bayesian Markov chain

Monte Carlo inversion, we demonstrated that strategically selected top-down and bottom-up observational constraints can reduce aerosol-cloud parametric degeneracy, while showing that the resulting inference is sensitive to assumptions regarding observational uncertainty (Stanford et al., 2026 preprint). In parallel, we developed a machine-learning warm-rain emulator for E3SM to improve the computational efficiency, accuracy, and interpretability of warm-cloud microphysics in coarse-resolution Earth system models, with the underlying approach designed as a transferable framework that can be extended to additional process modules. To bridge between microscale and macroscale processes in realistic atmospheric regimes, we also generated an ensemble of high-resolution large-eddy simulations of northeast-Pacific marine stratocumulus and used feature identification, clustering, and complementary diagnostics to examine how turbulence, cloud organization, and meteorological conditions influence cloud structure, warm-rain susceptibility, and aerosol-cloud interactions. Finally, we released version 1.0.0 of the Jacobian Estimation of Functional Error (JEFE) software package, which provides an efficient implementation of collision-coalescence calculations for bin microphysics together with adjoint and tangent-linear capabilities to enable efficient sensitivity analysis. Together, these efforts advanced complementary tools and analyses for linking targeted observations, high-resolution simulations, machine learning, and efficient numerical methods to the study of multiscale cloud and turbulence processes, while supporting continued improvement of cloud and turbulence process representations in both Large Eddy Simulations and Earth system models.

2.6 Research Objective 6: Investigate the Effect of SOA coatings on Warm-Temperature Ice Nucleating Dust Aerosol

Mineral dust is one of the most abundant aerosol components by mass and among the most efficient ice-nucleating particles (INPs) in the atmosphere. Previous research has shown that secondary organic aerosol (SOA) coatings can suppress the deposition freezing efficiency of dust particles. The suppression is dependent on coating thickness and/or surface coverage as well as dust type, as mineral structure sets the baseline upon which coatings exert their influence. In this project, we investigated the deposition freezing (-40 to -42 °C) ability of K-feldspar dust particles with thin SOA coatings.

Polydisperse K-feldspar dust particles were first injected into a ~1 m³ custom-built FEP chamber, followed by injection of SOA precursors (α -pinene and H₂O₂). UVA blacklights surrounding the chamber initiated the OH-driven oxidation of α -pinene, producing SOA that coated the dust particles over time. The resulting thin-SOA-coated dust particles were size selected and continuously monitored using a single particle mass spectrometer (miniSPLAT) and an ice nucleation chamber (INC). Figure 6 shows the results for two experiments with thin and thinner SOA coatings, which are not differentiable in traditional particle sizing instruments and scanning electron microscopy images. However, miniSPLAT unambiguously showed a concurrent increase of vacuum aerodynamic diameter (d_{va}) and α -pinene SOA mass spectral peaks, indicating that SOA is smoothing the surfaces or/and filling voids of the initially non-spherical dust particles. In turn, INC results showed a significant suppression of ice activation fraction once a critical mass of coating was formed (~ 0.02 mass spectral abundance). The ice active site density calculated from the instrument data shows a reduction up to 2 orders of magnitude. Our results demonstrate that dust's ice-forming capacity is significantly reduced, even after a minimal SOA formation, implying aged dust particles are poor INPs.



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