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REDUCING UNCERTAINTY IN AEROSOL-CLOUD INTERACTIONS USING
REMOTE SENSING: THEORY, APPLICATIONS, AND CLIMATE EDUCATION

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REDUCING UNCERTAINTY IN AEROSOL-CLOUD INTERACTIONS USING
REMOTE SENSING: THEORY, APPLICATIONS, AND CLIMATE EDUCATION

A DISSERTATION APPROVED FOR THE
SCHOOL OF METEOROLOGY

BY THE COMMITTEE CONSISTING OF

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Acknowledgments

In August 2015 I moved to Norman to start at OU without friends, family, or any idea of what I was doing. Left to my own devices, I would have switched out of my intended meteorology major after only a few months because I was convinced that I wasn't smart enough. Eleven years later, at the end of writing an entire dissertation, I have so many people I'm grateful for who didn't let me discount myself and who carried me through the hills and valleys.

The first person I met in the School of Meteorology was Dr. Fred Carr. His first-year mentoring group for out-of-state meteorology freshmen was one of the first sources of community I had during the difficult transition from high school to college, and I vividly remember the meeting in his office where he convinced me to at least give it a little more time before I decided to change my major out of meteorology. Without that conversation, I might not be writing this paragraph and you might not be reading it. Upon deciding to continue, my next "I am not smart enough for this" crisis manifested itself as my showing up at Dr. Elinor Martin's office hours for Introduction to Meteorology 1 every week, and thank goodness it did. Dr. Martin was so patient with all of my questions, and as I got to know her better, she gave me my first opportunity to do research as an undergraduate student (always go to office hours, kids). I am so thankful for her and for that experience and how it taught me to fight with code and data for the first time and how to present and write about my work. Dr. Martin encouraged me to apply for the NOAA Hollings Scholarship, which also turned into something that changed my life and trajectory for the better. I'm so grateful for Mary Fairbanks and Doug Hilderbrand for giving me the opportunity to work at the NWS Office of Communications in Silver Spring, MD for a summer. It was at this point

that I became really interested in weather science and safety communication, which has continued to manifest itself in several ways over the past few years.

The end of undergrad was another situation in which I had no idea what I was doing. I had ideas of what parts of meteorology I was most interested in, but still not much direction. I can't really describe my decision to apply for grad school other than it just felt like something I was supposed to do and that my time at OU wasn't supposed to be over yet. I owe so much to my advisor, Dr. Jens Redemann, for taking me on as his first graduate student when I barely even knew what an aerosol was. Thank you for truly giving me a space where I've been able to learn and grow as a researcher, and for giving me the time to discover and pursue what I'm most interested in. Though I never thought I'd write these words, thank you for letting me struggle through the theoretical calculations paper, because (unfortunately) it did teach me a lot about CCN and interpreting aerosol optical properties. It was also through that low point that I learned a lot about myself and developed perseverance I may not have otherwise. Thank you also for not forcing me to be (too much of an) extrovert at conferences. Additionally, I would not have made it to this point without my co-advisor, Dr. Lan Gao. Thank you for your patience with me during the stage where I mostly just had MATLAB questions every other week, and for truly teaching me what it looks like to do good research and ask good questions. Some of the moments I've felt most proud are when I've caught myself anticipating the kinds of questions you'll ask me when I show you a new figure or run into an issue. I'll be forever grateful for the endless time, support, and encouragement that you both gave me to find research direction and goals for my future.

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If you're reading this section and getting tired, maybe go get a snack, because we're only about halfway through. For as many people who supported the science side of this work, even more supported me personally. First, to my roommate of seven (7!) years, Rachael Cross - thank you for putting up with me for that long. We survived three moves, a global pandemic, an (arguably) ill-fated half marathon, living next to some of the wildest neighbors, the threat of an eviction (iykyk), several of the happiest

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I hope by reading all of this, you also see the hand of God as I do when I think

about this entire crazy eleven-year trajectory and all of the people who have supported me through it. At so many important points I had no solid plans or direction, and yet God created an incredible story that isn't over yet.

“He draws up the drops of water, which distill as rain to the streams; the clouds pour down their moisture and abundant showers fall on mankind. Who can understand how he spreads out the clouds, how he thunders from his pavilion.” (Job 36:27-29)

Dedication

Dedicated to each of my grandparents, Charles and Patricia Lenhardt, and Warren and Jeanne Albers, with whom I did not have nearly enough time. Without their hard work, love, and sacrifices, our family would not be the same.

Selected Publications

Chapter 2 has been published in *Atmospheric Chemistry and Physics* (ACP; Lenhardt et al., 2025), and Chapter 3 is currently a pre-print under review in ACP (Lenhardt et al., 2026). Chapter 3 as written here reflects our responses to reviewer comments.

First-Author Publications:

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The following are co-authored publications discussed in Chapter 5:

Chang, I., Gao, L., Adebisi, A. A., Doherty, S. J., Painemal, D., Smith, W. L., Lenhardt, E. D., Fakoya, A. A., Flynn, C. J., Zheng, J., Yang, Z., Castellanos, P., Da Silva, A. M., Zhang, Z., Wood, R., Zuidema, P., Christopher, S. A., and Redemann, J.: Regional aerosol warming enhanced by the diurnal cycle of low cloud, *Nat. Geosci.*, 18, 702–708, <https://doi.org/10.1038/s41561-025-01740-1>, 2025.

- Gao, L., Redemann, J., Lenhardt, E. D., Chen, B., Xu, F., Ferrare, R., Hair, J., Nehrir, A., Hostetler, C., Shingler, T., Fenn, M., Cairns, B., Sorooshian, A., and Chang, I.: Below-Cloud CCN Concentrations Provide Better Constraints on the Twomey Effect Than Aerosol Optical Properties, *Geophysical Research Letters*, 53, e2026GL122 006, <https://doi.org/10.1029/2026GL122006>, 2026.
- Soloff, C., Crosbie, E. C., Diskin, G. S., Gao, L., Kirschler, S., Lenhardt, E. D., Moore, R. H., Redemann, J., Shook, M. A., Tang, S., Thornhill, K. L., Voigt, C., Wang, H., Winstead, E. L., Ziemba, L. D., and Sorooshian, A.: Cloud Condensation Nuclei Behavior and Closure Assessment for the Northwest Atlantic Ocean, *ACS EST Air*, p. acsestair.5c00151, <https://doi.org/10.1021/acsestair.5c00151>, 2025.
- Witthuhn, J., Deneke, H., Macke, A., Ritter, O., Redemann, J., Flynn, C. J., Fakoya, A. A., Lamkin, B. F., Lenhardt, E. D., Mitchell, L. T., West, E. K., Romps, D. M., "Oktem, R., and Kalesse-Los, H.: Characterisation of cloud shadow transition signatures using a dense pyranometer network, <https://doi.org/10.5194/egusphere-2025-5808>, 2025.

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Abstract

Understanding the vertical distribution of cloud condensation nuclei (CCN) concentrations (N_{CCN}) is crucial for reducing uncertainty in aerosol-cloud interactions (ACI) and effective radiative forcing, and yet has been an aspect missing from many ACI-focused studies. Many proxies, parameterizations, and relationships between N_{CCN} and aerosol optical properties (AOPs) have been developed and tested using widely available remote sensing observations. In Lenhardt et al. (2023), we developed a simple linear regression method to approximate vertically resolved N_{CCN} from High Spectral Resolution Lidar 2 (HSRL-2) backscatter (BSC) and extinction profiles (EXT). This method was reliable for observations of smoke at low ($\leq 50\%$) relative humidities (RH) from the NASA ORACLES (ObseRvations of Aerosols above CLouds and their intEr-actionS) campaign over the Southeast Atlantic (SEA), but is less reliable when applied to observations of mixed aerosol types in other regions.

Several uncertainties and limitations exist when developing methods to relate N_{CCN} and AOPs, generally related to high RH, environments with internal or external mixtures of several aerosol types, and differences in which parts of the aerosol size distribution are most relevant to N_{CCN} compared to AOPs. To understand the dominant governing factors of N_{CCN} —AOP relationships, and for which cases a simple linear approximation is and is not applicable, we use in situ observations of the aerosol size distribution and chemical composition to inform theoretical calculations of N_{CCN} and aerosol BSC at 532 nm. These observations come from the recent NASA ACTIVATE (Aerosol Cloud meTeorology Interactions oVer the western ATlantic Experiment) campaign in the Northwest Atlantic, where there is a diverse mixture of aerosols located primarily in the marine boundary layer (MBL). Estimates from random forest mod-

els indicate that, for smoke, marine, and urban aerosols, the aerosol size distribution, as parameterized using the effective radius (R_{eff}), is the most important predictor of the theoretical N_{CCN} —BSC relationship. We further investigate the physical impact of R_{eff} on the N_{CCN} :BSC ratio and find an exponential relationship between the parameters. We find that modeling N_{CCN} :BSC using this exponential R_{eff} relationship can explain about 68%–79% of the variance in the theoretical N_{CCN} —BSC relationship. These findings suggest that including information about aerosol size is critical for future studies in constraining N_{CCN} from AOPs.

While assessing the theoretical N_{CCN} —BSC relationship and its non-linear, multi-variate dependencies, (Redemann and Gao, 2024) developed a machine learning (ML) method to predict N_{CCN} from HSRL-2 observables and reanalysis data. Due to the high accuracy of this method to predict N_{CCN} within about 14% uncertainty, we use this reliable and vertically resolved product (ML-CCN) to investigate ACI in the SEA. Evidence of interactions between the biomass burning smoke plume and underlying marine stratocumulus cloud deck has been found using in situ data. Motivated by these findings and equipped with this new remote sensing-based N_{CCN} dataset, we further investigate these cloud top ACI and their dependence on lower tropospheric stability (LTS). Taking advantage of cloud edge HSRL-2 profiles, we assess the simultaneous impact of above- and below-cloud N_{CCN} on cloud top microphysical properties. We observe a decrease in cloud droplet effective radius (R_{eff}) and an increase in cloud droplet number concentration (N_d), both of which are associated with increasing above-cloud N_{CCN} . These relationships are strongly dependent on LTS, with ACI_{REF} decreasing from 0.161 to 0.042 (-73.9%) and ACI_{CDNC} decreasing from 0.452 to 0.116 (-74.3%) as LTS increases from 10 to 22 K. Additionally, we find that above-cloud N_{CCN} —cloud property relationships are similar for cloud edge and cloud center observations. The relationship between below-cloud N_{CCN} and cloud top properties is strongly dependent

on LTS, with ACI metrics increasing as LTS increases. This speaks to the dominance of above-cloud smoke entrainment as a modulator of stratocumulus cloud properties under unstable conditions, while below-cloud N_{CCN} nucleation dominates in stable environments. These findings demonstrate the importance of using vertically resolved N_{CCN} in ACI studies and establish a remote sensing-based analysis method with which future satellite studies can investigate ACI.

Improving the representation of N_{CCN} and ACI in climate models will help to better predict future climate warming in different emissions scenarios. However, climate warming cannot be viewed exclusively as a future issue, as anthropogenic emissions have already begun to impact Earth's weather and climate. Therefore, it is also important to take action now to safeguard the future of our environment. One such route to do so is by improving climate change communication (CCC) and educating students on climate science. As a practical step towards this goal, a three-day aerosol and cloud-focused set of modules was developed and implemented in four sections of the meteorology elective at Norman High School (NHS). Responses to pre- and post-surveys, as well as three reflection question worksheets, were collected to evaluate the effectiveness of the different lectures and activities. For 22 of the 24 survey statements, there was an overall shift towards more correct responses in the post-survey, and responses to the reflection questions provided context and possible explanations for why certain questions saw only a very small improvement or a shift towards decreased understanding. Student responses show the benefit of communicating complicated topics using hands-on activities and the need to balance how many topics are included in individual activities. Additionally, responses emphasized the importance of insight from the classroom teacher in terms of students' background knowledge and what words and phrasing may be confusing. The findings also demonstrated the confirmation of the primary hypothesis, which was that by focusing on teaching the mechanisms of aerosol

direct and indirect effects, students would be able to better deduce their possible long-term climate impacts.

Several studies that took place alongside the projects described here can help place and interpret our work in a broader scientific context. We discuss the benefits of using ML (Redemann and Gao, 2024) and physics-based approaches in tandem instead of in isolation, as well as a project that evaluated more details of N_{CCN} closure from the ACTIVATE observations than we investigated in Chapter 2 (Soloff et al., 2025). We discuss studies by Witthuhn et al. (2025) and Chang et al. (2025) that emphasize the importance of capturing spatial and temporal variations in aerosol and cloud properties for constraining global horizontal irradiance and the direct aerosol radiative effect, respectively. Lastly, we discuss recent work by Gao et al. (2026) that applies a very similar method as shown in Chapter 3 to observations of a cumulus cloud field. This study found more realistic ACI relationships using ML-predicted N_{CCN} when compared to AOPs, which again supports the importance of and need for vertically resolved N_{CCN} in ACI studies.

The work detailed here has several implications for future remote sensing of N_{CCN} and ACI from both aircraft and satellite platforms, as well as suggestions for outreach efforts and climate science education. Additionally, the work highlights many routes for potential future analysis that were outside the scope of these projects. We conclude by discussing several of these possible directions, such as investigating N_{CCN} —AOP relationships for dust and examining similar theoretical relationships for aerosol mixtures in different types of environments. The ACI study could be expanded to new cloud and environmental regimes, and more meteorological variables could be included to better understand and explain the observed relationships. There are several ways in which the climate education modules could be improved and iterated upon, including implementing them in more schools and classrooms, refining some wording issues in

the surveys and worksheets, and developing new hands-on activities for topics such as cloud albedo and precipitation susceptibility.

Chapter 1

Introduction

The atmospheric sciences aim to understand phenomena which are fully ungovernable. We try to better predict how atmospheric circulations will impact the weather tomorrow, severe storm chances next week, how much precipitation is likely in the coming season, and what our climate will look like several years and decades into the future. One particularly pressing issue is understanding how the climate will continue to warm in different emissions scenarios, which will directly impact life across the globe. Crucial to these predictions is an understanding of Earth's energy budget, or how energy moves into and out of the Earth system, and the various forcings that govern it. Greenhouse gases are the primary source of energy imbalance in the atmosphere, as they absorb longwave radiation emitted from the Earth that would otherwise escape to space (Forster et al., 2021). This energy imbalance has resulted in an overall warming effect on the atmosphere. Aerosols are one component of the atmosphere with the ability to cause cooling, particularly through their interactions with clouds. However, their impacts on Earth's energy balance remain associated with the highest uncertainty of all forcing agents (Forster et al., 2021) even after decades of research. The work encompassed here concerns this high uncertainty and aims to find observational approaches for improving the representation of aerosol concentrations and their impact on cloud properties.

1.1 Characteristics and effective radiative forcing of atmospheric aerosols

Atmospheric aerosols are defined as a collection of solid or liquid particles in suspension in a gas, while in the atmospheric sciences the term “aerosols” refers only to the particles, separate from the suspending atmosphere (Boucher, 2015). They are also defined independently from hydrometeors, such as cloud droplets, raindrops, ice crystals, snowflakes, fog, and graupel, which are treated as separate phenomena (Pöschl, 2005; Boucher, 2015). Common examples of atmospheric aerosols include smoke from wildfires, sea salt from marine sea spray, pollution from urban areas, dust from deserts, and ash from volcanic eruptions. In large concentrations, such as a dense smoke plume coming from a fire, aerosols can be visible in the atmosphere. However, at lower concentrations and typical atmospheric conditions aerosols are too small to be visible.

There are several ways to characterize aerosols, one being the method of their formation. Primary aerosols are emitted into the atmosphere as particles through combustion processes, such as biomass burning or vehicle emissions, or wind driven processes, such as the generation and re-suspension of sea salt and desert dust (Prather et al., 2008; Boucher, 2015). Secondary aerosols form in the atmosphere through condensation of precursor gases (Boucher, 2015). This process is often referred to as new particle formation (NPF), which is an important area of research, as the processes leading to particle nucleation and growth are uncertain and difficult to observe (Pöschl, 2005; Lee et al., 2019). Aerosols can also be characterized based on region and source of origin, where source of origin may differ from region of observation for cases of long-range transport (Boucher, 2015).

One of the most fundamental properties of atmospheric aerosols is their size. Aerosol

radii are generally between 1 nm - 10 μm (Prather et al., 2008), and most aerosols fall within the smaller end of this range. Aerosol size distributions are generally characterized by two modes, but up to five have been documented. In order of increasing size, these modes are referred to as the nucleation (or ultrafine) mode, Aitken mode, accumulation mode, coarse mode, and super-coarse mode (Boucher, 2015). Nucleation mode particles tend to have a short lifetime in the atmosphere due to their increased chemical reactivity and physical mobility (Prospero et al., 1983), and accumulation mode aerosols (approximately 0.05-0.5 μm in radius) are those that accumulate the most mass through coagulation and condensation (Boucher, 2015). Coarse mode and super-coarse mode particles usually have a radius greater than 0.5 μm and tend to be few in number but dominate the volume size distribution. Lognormal distributions are generally used to approximate the aerosol size distribution, where fine and coarse modes are described by their geometric mean radius or effective radius.

Chemical composition is another highly important aerosol property. The primary components in most atmospheric aerosols are sulfate, nitrate, ammonium, sea salt, mineral dust, organic compounds, and black or elemental carbon (Pöschl, 2005). The exact composition is highly dependent on the aerosol source, where the chemical make-up even among biomass burning aerosols (BBA) can vary significantly depending on the exact material that was burned (Fakoya et al., 2025). Aerosol chemistry can also be characterized by mixing state. An external mixture of aerosols describes chemically homogeneous particles that comprise a population of particles with varying chemical compositions, while an internal mixture describes different chemical species being mixed within individual particles (Boucher, 2015). Boucher (2015) describes external and internal mixtures as conceptual models, where reality lies somewhere between these options. Additionally, Prather et al. (2008) describes a specific example of the importance of mixing state, where internally mixed soot-sulfate mixtures are three times

as absorbing as separate soot and sulfate particles. The chemical composition of an aerosol impacts reactivity, water uptake, and optical properties (Prather et al., 2008). Water uptake is also referred to as hygroscopicity, which is the ability of an aerosol to attract and hold water molecules in a condensed phase (Boucher, 2015). This water uptake can cause substantial increases in particle diameter and ability to serve as cloud condensation nuclei (CCN; Pöschl, 2005).

While greenhouse gases trap longwave energy emitted by Earth’s surface and atmosphere, causing warming, aerosols primarily interact with incoming solar radiation by scattering or absorbing it (Schwartz, 1996), which can lead to cooling or warming, respectively. Aerosols can also cause a greenhouse effect through interactions with longwave radiation, but this requires large concentrations of aerosols at heights able to cause temperature differences between the surface and the aerosol layer (Boucher, 2015). Therefore, most work focuses on aerosol interactions with solar (shortwave) radiation. These interactions have been called aerosol direct effects, but most recently are being referred to as aerosol-radiation interactions (ARI).

Scattering and absorption of individual aerosols depend on their size and chemical composition, where larger aerosols generally scatter and absorb more radiation, and aerosols with a high fraction of black or brown carbon, for example, are more likely to be strong absorbers. Growth of hygroscopic aerosols in high relative humidity (RH) environments also increases their ability to scatter radiation. The combined effect of scattering and absorption is called extinction, and the computation of scattering, absorption, and extinction is described by Mie Theory (Sect. 1.3).

In practice, we are generally interested in the collective effect of a population of aerosols on solar radiation, in which the scattering and absorption coefficients of individual aerosols are integrated over the entire size distribution. ARI for a population of aerosols are described using various aerosol optical properties (AOPs). AOPs include

scattering, absorption, and extinction coefficients, but also consist of parameters such as aerosol optical depth (AOD), single scattering albedo (SSA), or Ångström Exponent (AE), each of which provides slightly different information (Boucher, 2015). For example, AOD is the integral of the aerosol extinction coefficient over a vertical column and therefore provides a general measure of aerosol loading. SSA is the ratio of scattering to extinction, which indicates the degree of absorption, and AE measures the spectral dependence of AOD or extinction, providing information about relative aerosol size.

When quantifying the net effect of ARI on Earth’s energy balance, both radiative effects and radiative forcing (RF) can be discussed. Radiative effects refer to the net change in Earth’s energy balance from all aerosols, including natural and anthropogenic, while radiative forcing refers specifically to the net change due to anthropogenic aerosols (Myhre et al., 2013; Boucher, 2015). Since anthropogenic emissions of sulfates, organics, and black carbon exceed natural emissions (Ramanathan, 2001), radiative forcing is most critical to constrain. In recent reports from the Intergovernmental Panel on Climate Change (IPCC), effective radiative forcing (ERF) has been considered the central definition for RF (Forster et al., 2021). While RF is defined as the “change in net downward radiative flux at the tropopause after allowing for stratospheric temperatures to readjust to radiative equilibrium,” ERF is defined as the change in the net top of atmosphere (TOA) downward radiative flux “after allowing for atmospheric temperatures, water vapor and clouds to adjust” (Myhre et al., 2013). In other words, the ERF includes the effects of the forcing agent and the rapid adjustments to the agent. The ERF of aerosol-radiation interactions (ERFari) is estimated to be approximately -0.3 W/m^2 (Forster et al., 2021), indicating a net cooling effect. While uncertainties in ERFari remain due to variations in aerosol composition, vertical distribution, and regional emissions, most uncertainty in the total aerosol ERF is associated with their indirect effects, or aerosol-cloud interactions (ACI). The most re-

cent assessment report (AR5) from the IPCC estimates a global ERF_{aci} of -1.0 W/m^2 and states that there is high confidence that ERF_{aci} contributes to 75-80% of the total aerosol ERF. ACI mechanisms and reasons for high uncertainty will be further explored in Section 1.4.

1.2 Cloud condensation nuclei (CCN) and Köhler Theory

Before exploring ACI mechanisms in more depth, it is important to characterize the subset of aerosols that form cloud droplets and impact cloud properties. Cloud droplets form through heterogeneous nucleation as water vapor condenses on atmospheric aerosols called CCN (Rogers and Yau, 1996), since homogeneous nucleation of water vapor to form cloud droplets requires ambient RH much larger than 100% and therefore is not the primary mechanism by which they form (Rogers and Yau, 1996; Wang, 2013). CCN form cloud droplets in warm phase (liquid) clouds, while aerosols that form ice particles in mixed and ice phase clouds are called ice nucleating particles (INP). Whether an aerosol can serve as a CCN generally depends on environmental supersaturation and the aerosol size and chemical composition, and the relationship between these factors is quantified by Köhler theory.

Köhler theory predicts CCN activity based on the mass of solute within a droplet as a function of particle size and equilibrium saturation ratio (Köhler, 1936). The Köhler equation accounts for the Raoult effect, which describes how the addition of a solute decreases the required equilibrium vapor pressure of water needed to form a droplet, and the Kelvin effect, which describes the increased equilibrium vapor pressure needed to form a curved droplet as opposed to a plane of water (Rogers and Yau, 1996). The Köhler curve depicts the equilibrium saturation ratio as a function of droplet size,

where the specific curve for an aerosol is dependent on solute mass and the initial starting size. Where the curve reaches a maximum defines the critical saturation ratio (S_{crit}) and critical diameter (D_{crit}). At environmental saturation ratios higher than S_{crit} , or for particle sizes larger than D_{crit} , droplets activate and continue to grow into cloud droplets without need for additional increases in the saturation ratio (Köhler, 1936; Rogers and Yau, 1996). For environmental saturation ratios lower than S_{crit} , or for particle sizes smaller than D_{crit} , droplets are called haze particles. An aerosol is considered an activated CCN when the droplet nucleated around it grows to the size defined by D_{crit} .

The Köhler equation is effective for modeling CCN activity of water-soluble inorganic compounds but does not account well for particles with lower hygroscopicity, such as many organics (McNeill et al., 2013). Therefore, several studies have developed modified versions of Köhler theory. Many of these techniques still require information about individual aerosol components that is generally unavailable from ambient measurements, and some require that all calculations be done for each individual chemical component. To overcome these limitations, Petters and Kreidenweis (2007) developed a Köhler theory modification that uses a single parameter to represent aerosol hygroscopicity, called κ -Köhler theory. This variation of Köhler theory is applied and further explained in Chapter 2.

1.3 Aerosol remote sensing and Mie theory

Observations of aerosols, including their size, chemical composition, and concentrations within the atmosphere are crucial for quantifying ARI and ACI, as well as their potential health impacts. Aerosol properties are often observed using in situ instrumentation located at ground-based stations or on aircraft during targeted campaigns. These ob-

servations are generally highly accurate, but are not available on spatial and temporal scales large enough to characterize global aerosol variability (Boucher, 2015). Since remote sensing instruments have increased spatiotemporal resolution, several techniques have been developed to use such platforms for aerosol-relevant observations. However, the retrieval of aerosol information from remote sensing observations is an ill-posed problem, as there is an insufficient amount of information to constrain all possible solutions (Kokhanovsky and Leeuw, 2009). Therefore, a priori information or assumptions are required to retrieve specific information about aerosol size, concentration, or composition, which introduces uncertainty. For this reason, in situ observations are often used to constrain retrievals and to develop parameterizations or proxies for translating remote sensing observations to aerosol properties (Rosenfeld et al., 2014). For the purpose of this work, the remainder of this section will focus specifically on remote sensing techniques for CCN concentration (N_{CCN}).

Passive remote sensing techniques are based on natural electromagnetic radiation, most commonly shortwave radiation from the sun or longwave radiation emitted by the Earth system (Boucher, 2015). Passive aerosol remote sensing observations measure AOD using ground-based sun photometers or satellite-based spectrometers. Since AOD scales with the total atmospheric aerosol concentration (N_A) and N_{CCN} is a fraction of N_A , many previous studies have used AOD as a proxy for N_{CCN} (Myhre et al., 2007; Andreae, 2009; Liu and Li, 2014; Jia et al., 2019). However, while AOD may be able to approximate N_{CCN} over large spatiotemporal extents (Stier, 2016), it cannot fully explain N_{CCN} variance (Andreae, 2009; Shinozuka et al., 2015; Stier, 2016; Choudhury and Tesche, 2022a,b). Furthermore, Stier (2016) found that the lowest correlations between N_{CCN} and AOD are found near marine stratocumulus decks, which are highly susceptible to aerosol perturbations (Boucher et al., 2014), and therefore are an area where accurate representations of N_{CCN} are critical. One commonly hypothesized

reason for the lack of correlation between N_{CCN} and AOD, as well as for other AOPs, is hygroscopic growth at high RH (Stier, 2016). This impact, as well as other governing factors of N_{CCN} —AOP relationships, are further addressed as the central question of Chapter 2.

One of the main reasons why AOD does not fully capture N_{CCN} variance is because passive remote sensing observations cannot resolve vertical distributions. Understanding the vertical placement of N_{CCN} relative to clouds is especially relevant for ACI, as cloud droplets are generally understood to nucleate near the cloud base. Therefore, many recent studies have relied on active remote sensing instrumentation, which emit artificial electromagnetic radiation and can resolve vertical distributions (Boucher, 2015). The two most common active remote sensing instruments are radar, which emits radio waves, and lidar, which emits primarily in the near-infrared or visible parts of the electromagnetic spectrum and is therefore better suited for observing aerosols (Weitkamp, 2005). Lidars emit short pulses of light with a laser, and the intensity of backscattered light is recorded and analyzed as a function of time after the laser pulse to estimate the altitude of the observation (Boucher, 2015).

Aerosol studies have frequently used the commonly available elastic-backscatter lidar, but this method requires an assumed lidar ratio to determine the particle extinction coefficient from the elastic-backscatter signal (Ansmann et al., 1992; Weitkamp, 2005). The aerosol lidar ratio is range-dependent, as it depends on the aerosol size distribution, shape, and chemical composition, making it nearly impossible to accurately estimate extinction profiles using this method (Klett, 1985; Weitkamp, 2005). In contrast, the development of the Raman (Mattis et al., 2004; Müller et al., 2007) and High Spectral Resolution Lidars (HSRL; Hair et al., 2008; Burton et al., 2012; Groß et al., 2013) has enabled more accurate and independent profiling of the extinction coefficient without the need for an assumed lidar ratio (Weitkamp, 2005). Further, observed lidar

ratios from these systems have enabled advances in aerosol profiling and typing (Mattis et al., 2003; Noh et al., 2007). Raman lidars separate aerosol and molecular signals by detecting molecular inelastic scattering, while the HSRL uses differences in the spectral distribution of light that is elastically backscattered by particles and air molecules (Weitkamp, 2005). While both methods have the advantage of independently determining aerosol backscatter and extinction coefficients, Raman lidars require powerful lasers and large telescopes to provide sufficient signal strength since Raman scattering is weak. Additionally, competition between this weak signal and sunlight makes daytime observations difficult. The HSRL method requires very narrow bandwidth filters and the laser linewidth must be smaller than the filter width, making HSRL systems more difficult to implement than Raman lidars. However, they provide much larger molecular signals and can use very narrow bandwidths to block noise from solar radiation (Weitkamp, 2005).

As previously mentioned, retrieving aerosol properties from remote sensing observations is an ill-posed problem with more unknowns than available information. Weitkamp (2005) describes three main methods by which studies have retrieved aerosol information from lidar observations. One common method combines lidar observations with measurements from additional instruments. This approach provides information that can be used to address the unknowns, but introduces additional error due to differences in measurement time, location, and technique. The second major approach is reconstructing AOPs retrieved from lidar using Mie-scattering calculations and assumed information about the aerosol size distribution and refractive index. The third method described is a mathematical inversion approach using the spectral information contained in lidar signals measured at different wavelengths.

These three approaches are also reflected in the numerous studies that have used AOPs to parameterize or retrieve N_{CCN} . Many studies have taken advantage of

aircraft-based observations (Ghan and Collins, 2004; Ghan et al., 2006; Kapustin et al., 2006; Shinozuka et al., 2009, 2015; Tsekeri et al., 2017; Haarig et al., 2019; Patel et al., 2024), sometimes in combination with ground-based lidar or in situ aerosol measurements (Ghan and Collins, 2004; Ghan et al., 2006). Other approaches have used entirely ground-based observations of AOPs (Jefferson, 2010; Liu and Li, 2014; Mamouri and Ansmann, 2016; Lv et al., 2018; Shen et al., 2019). More recently, studies have used active satellite remote sensing approaches, such as observations from the CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) lidar onboard the CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) satellite (Ansmann et al., 2021; Choudhury and Tesche, 2022a,b; Patel et al., 2024). These studies have used AOPs such as extinction (EXT), backscatter (BSC), scattering (SCA), AOD, aerosol index (AI), AE, backscatter fraction, and SSA in proxies and parameterizations to get to N_{CCN} . Most approaches either explicitly include RH in parameterizations or correct lidar signals from ambient to dry conditions to account for aerosol hygroscopic growth. Accounting for RH, aerosol chemical composition, and heterogeneities in the aerosol size distribution remain some of the biggest challenges in deriving N_{CCN} from AOPs.

Critical to understanding how N_{CCN} relates to AOPs is Mie theory. Mie theory describes how light is scattered by spherical particles that are approximately the same size as the wavelength of incident radiation (Mie, 1908). Aerosol scattering and extinction calculations depend on efficiency factors, which are a function of aerosol refractive index, radius, and the wavelength of incident radiation. The aerosol size distribution also impacts scattering and extinction, as larger aerosols have a greater impact on AOPs. On the other hand, aerosols that serve as N_{CCN} tend to fall at the smaller end of the aerosol size distribution. This discrepancy is another source of uncertainty when comparing AOPs and N_{CCN} . The use of Mie theory in relating AOPs to N_{CCN}

is discussed further in Chapter 2.

1.4 Aerosol-cloud interactions and constraints from observations

Accurate retrievals of N_{CCN} are necessary for assessing ACI, as the nucleation of CCN as cloud droplets contributes to changes in cloud macro- and microphysical properties. Cloud macrophysical properties describe cloud size, shape, and altitude, and include parameters such as liquid water path (LWP) and cloud optical thickness (COT). Cloud microphysical properties such as droplet number concentration (N_d) and cloud droplet effective radius (R_{eff}) describe properties of individual droplets (Kubar et al., 2009). Two primary mechanisms by which CCN impact cloud properties include the Twomey and Albrecht effects. Assuming a constant LWP, increases in N_{CCN} usually cause an increase in N_d and a decrease in R_{eff} , and this effect is called the cloud albedo or Twomey effect (Twomey, 1974). The presence of smaller cloud droplets has been hypothesized to extend cloud lifetime by reducing collision-coalescence rates and delaying or suppressing precipitation formation (Albrecht, 1989).

However, if all ACI were so straightforward and could be fully described by these two effects, ERFaci uncertainty would not remain so large. Rather, there are numerous pathways through which aerosols have been hypothesized to influence cloud properties, and much of the remaining uncertainty is due to the interconnected nature of cloud macro- and microphysical properties and challenges in their observational constraints. For example, the Twomey effect assumes a constant cloud LWP, which is not only impossible to constrain from observations, but there are also several hypothesized pathways through which aerosols may increase or decrease LWP (Gryspeerd et al., 2019). A decrease in the loss of cloud water to precipitation can cause clouds to

grow to higher vertical extents (Andreae, 2004; Rosenfeld, 2006; Myhre et al., 2007), eventually increasing LWP. Warm-cloud invigoration may also act to increase LWP. In N_d -limited clouds, an increase in N_d can increase cloud liquid water content and latent heat release, allowing clouds to reach larger vertical extents and thus increase LWP (Koren et al., 2014; Gryspeerdt et al., 2019). Aerosol concentration may also lead to decreases in LWP through sedimentation-entrainment (Ackerman et al., 2004; Bretherton et al., 2007) and evaporation-entrainment (Wang et al., 2003; Small et al., 2009) feedback processes, where increased evaporation and entrainment at cloud edges can decrease LWP if the ambient RH is low (Gryspeerdt et al., 2019). As put by Boucher (2015), the Twomey effect “does not occur in isolation.” Rather, the nucleation of a high concentration of small cloud droplets causes multiple rapid adjustments that interact with each other. Furthermore, local meteorology also influences the outcome of ACI, making it difficult to clearly separate aerosol and meteorological impacts on cloud properties (Stevens and Feingold, 2009; Gryspeerdt et al., 2014; Rosenfeld et al., 2014).

To better understand these processes and feedback loops, represent them in climate models, and estimate ERF_{aci} for current and future climate, more reliable, global observational methods are necessary to constrain ACI. In terms of aerosol observations, Rosenfeld et al. (2014) suggested that while satellite observations of aerosol-related quantities have enabled a global perspective of aerosol-cloud-rainfall feedbacks, satellite and ground-based observations of aerosol microphysical properties are missing on regional scales. More specifically, they argue that accurate measurements of N_{CCN} on a global scale are necessary to understand the impacts of ACI on local meteorology and Earth’s energy budget (Rosenfeld et al., 2014). Similarly, Ma et al. (2018a) found that satellite observations of cloud susceptibility were weakened by limitations in aerosol retrievals, as satellite observations of AOD were less accurate in clean environments where

clouds are generally more sensitive to aerosol perturbations. Three of five suggested remote sensing strategies from Seinfeld et al. (2016) for improving ERF_{aci} in climate models focus on aerosols, including improving measurements of aerosol microphysical and chemical properties, enhancing aerosol type discrimination, and developing multi-platform aerosol retrieval algorithms. Lastly, in a recent review of the current state of ACI understanding, one proposed pathway forward is improving satellite data and retrieval accuracy, as CCN retrievals are hampered by a lack of information content and retrieval biases, thus hindering our satellite-based understanding of ACI (Im et al., 2026).

Satellite retrievals of cloud properties from platforms such as MODIS (Moderate Resolution Imaging Spectroradiometer), SEVIRI (Spinning Enhanced Visible and Infrared Imager), and CloudSat are frequently used for ACI studies, and assumptions made in their retrieval processes still introduce bias and uncertainties. Remote sensing retrievals of cloud properties continue to need further improvements (Im et al., 2026), but this is not a focus of the work presented here. Rather, this work concentrates on improving remote sensing-based retrievals of N_{CCN} for use in ACI studies.

1.5 Southeast Atlantic marine stratocumulus cloud deck

One area in which ACI uncertainties propagate and exacerbate other issues is in climate model representation of low-level cloud feedbacks (Bony and Dufresne, 2005). Low clouds are often underestimated in their cloud cover and overestimated in their optical thickness, which has been referred to as the “too few, too bright” problem (Nam et al., 2012; Lin et al., 2014). Better representing these clouds in climate models is crucial, as they cover about one-fifth of Earth’s surface (Warren et al., 1988) and reflect more

solar radiation than they trap outgoing longwave radiation (Wood, 2012). Therefore, they have a net cooling effect that could compete with the greenhouse warming effect (Randall et al., 1984; Slingo, 1990). Not only is it important to accurately represent their radiative feedbacks for our current climate, but in warmer climate scenarios as well. Low clouds are hypothesized to be more horizontally and vertically extensive and optically thicker in a warmer climate (Boucher et al., 2014), which will impact their role in Earth’s energy budget (Wood, 2012).

One particularly interesting and important region for studying low-level stratocumulus clouds is over the Southeast Atlantic Ocean (SEA), where seasonal biomass burning from July through October is advected westward atop a semi-permanent deck of marine stratocumulus clouds. The stratocumulus cloud top heights in this region usually fall between 1-2 km, with the smoke plume generally located between 2.5-6 km (Redemann et al., 2021). The marine boundary layer (MBL) deepens offshore as sea surface temperatures (SST) warm, resulting in higher cloud top heights and BBA layers in closer proximity to the cloud top (Redemann et al., 2021). BBA are transported westward primarily by the southern African Easterly Jet (AEJ-S; Adebisi and Zuidema, 2016), with another characteristic feature of SEA circulation being the Benguela low-level jet (LLJ; Ryoo et al., 2021).

Due to the absorbing nature of these BBA and their proximity to the cloud deck, ARI and ACI are both of interest in the region. Smoke in the SEA has some of the highest optical depths of BBA globally (Redemann et al., 2021), and their ARI are dependent on concentration, composition, vertical distribution, and cloud albedo below the smoke plume (Doherty et al., 2022). Often these smoke aerosols are located above and separated from the cloud deck, but at different times and locations they are entrained into the MBL and interact directly with clouds (Adebisi et al., 2015; Zuidema et al., 2016; Redemann et al., 2021). BBA observed in this region are efficient

CCN (Che et al., 2020), and many have relatively large median diameters due to the coagulation of smoke particles (Ross et al., 2003). Additionally, BBA can impact regional climate and cloud properties via the semi-direct effect, as warming due to absorption acts to reduce cloud cover (Hansen et al., 1997). The semi-direct effect has been observed to decrease cloud cover in the SEA (Zhang and Zuidema, 2019), while models have shown both increases and decreases in cloud cover due to the semi-direct effect (Doherty et al., 2022).

Several airborne campaigns observed the SEA from 2016 to 2018, creating multiple data sets with which to study the radiative and cloud-relevant effects of BBA and the stratocumulus cloud deck. These include the CLARIFY (Clouds and Aerosol Radiative Impacts and Forcing) campaign based out of Ascension Island in 2017, the AEROCLO-sA (Aerosols, Radiation and Clouds in southern Africa) project based out of Walvis Bay, Namibia, in 2017, the Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) mobile facility LASIC (Layered Atlantic Smoke Interactions with Clouds) deployment to Ascension Island in 2016-2017, and the ORACLES (Observations of Aerosols above CLouds and their intEractionS) project that was based out of Walvis Bay, Namibia in 2016 and São Tomé and Príncipe in 2017-2018. In Chapter 3 we investigate ACI in the SEA using observations from the ORACLES campaign. One of the goals of the ORACLES campaign was to develop a remote sensing testbed for future satellite studies, and the work in Chapter 3 contributes to this goal by using remotely sensed aerosol and cloud products to assess ACI.

1.6 Climate change communication: challenges and paths forward

Interactions between aerosols, solar radiation, and clouds have significant implications for Earth’s future climate. It is crucial to better understand, represent, and predict future impacts of these interactions in climate models due to their high ERF uncertainty, but climate warming has already been observed. Therefore, improving predictions of future climate while enacting change in how we interact with the Earth need to occur simultaneously, as climate warming is not an issue fully confined to the future.

In “Bending the Curve: Climate Change Solutions,” authors present ten solutions, or actions, that together can “bend the curve and avoid dangerous warming of the planet” (Ramanathan et al., 2019). These ten solutions are split into six clusters, including science pathways, societal transformation, governance, markets and regulations, technology measures, and ecosystem management. They emphasize that solutions to climate warming will require new collaborative efforts — not only between scientists, but amongst engineers, civic, business, and religious leaders, and community members. Many of their proposed solutions focus on emissions reductions, international action, clean energy incentives, promotion of new technologies, and restoring ecosystems. However, they also argue that innovations in technology and clean energy will not be effective if people are unwilling to restructure their lives to use new infrastructure. Similarly, policy proposals are likely to succeed only if people are willing to support them. Much climate-related work, if not well-translated to the public, may fall short of the long-lasting impacts we hope for. Therefore, as an introduction to the project presented in Chapter 4, this section focuses on the societal transformation cluster, specifically the solution of fostering “a global culture of climate action

through coordinated public communication and education at local to global scales” (Ramanathan et al., 2019).

In the book “Psychology and Climate Change” by Markowitz and Guckian (2018), Chapter 3 discusses challenges, insights, and opportunities related to climate change communication (CCC). They mention the significant heterogeneity amongst Americans in terms of individuals’ level of engagement with climate change, and argue that disagreements infrequently concern “the facts,” but pertain to the societal and economic implications of managing climate change. Additionally, they suggest that simply inundating people with more information is unlikely to significantly shift opinions. Similarly, Pearce et al. (2015) reviews several efforts in CCC and emphasizes the importance of two-way dialogue and focusing not only on the scientific consensus about climate change but also being willing to discuss disagreements. Further, they discuss old models of CCC that have assumed the public is lacking in scientific knowledge and that scientists need only to find more colloquial language with which to communicate (Pearce et al., 2015). Rather, they present a different model from Felt and Wynne (2007) that allows for communication and engagement and trusts people to deliberate, discuss, and deduce solutions without making them a target of “interventions.” Moser (2010), summarizing findings from Bak (2001), Schultz (2002), and Sturgis and Allum (2004), phrases it this way:

“...it is far too simplistic to assume that individuals merely lack education, information, or understanding of climate change, and if these knowledge gaps could be filled and lay individuals could be forced to interpret the findings in a particular way, they would automatically act to reduce their energy consumption and carbon footprint.”

There are several difficulties when it comes to effective CCC. For example, the im-

pacts tend to be diffuse and primarily future-oriented, making it difficult to highlight the urgency and immediacy of the issue (Moser, 2010; Markowitz and Guckian, 2018). Another source of tension in CCC is how to deal with uncertainty. While reports, such as those from the IPCC, identify differing levels of uncertainty associated with different conclusions, there are contrasting views on how to relay uncertainty when communicating with the public without undermining trust (Pearce et al., 2015). Perhaps the most significant obstacle of CCC is the extreme politicization and polarization of the issue that has existed particularly in American culture (Markowitz and Guckian, 2018) and has been exacerbated through social media platforms and online discussions in recent years (Pearce et al., 2015).

Many studies have identified challenges in effectively communicating about climate change, but several others have provided recommendations for improvement. For example, Markowitz and Guckian (2018) focus on knowing the audience and catering information based on their unique background knowledge. Chapman et al. (2017) reiterate that authentic, honest, and audience-catered communication is preferable to trying to force any emotional response. This view is also reinforced by Pearce et al. (2015), where authors contend that there is not a single message that will be successful for all audiences and contexts. For example, they find that audiences skeptical of anthropogenic climate change are more likely to support changes in environmental actions if they have clear economic benefit or are cast in the light of consideration for others. Additionally, focusing on hopeful messages and potential solutions can be more effective for skeptical audiences. Other recommendations include finding better ways of addressing uncertainty and complexity and of discussing relatively gradual processes or periods of stability. Lastly, Markowitz and Guckian (2018) also highlight the need for continued experimentation and evaluation-oriented work to be done by researchers to answer the question: *How are climate change communication efforts influencing*

audiences?

This work will conclude with a project aimed at educating high school students about aerosols and clouds as drivers of climate change. A more in-depth discussion of studies that have targeted climate change education at the K-12 level will be discussed in Chapter 4. Pre- and post-surveys were developed as an evaluation-focused component of the project that aim to answer the previous question from Markowitz and Guckian (2018).

1.7 Previous work and motivation

Chapters 2 and 3 of this work investigate remote sensing constraints for resolving the vertical distribution of N_{CCN} from lidar observations and applying vertically resolved N_{CCN} to ACI analyses. Our initial efforts to create a vertically resolved N_{CCN} product involved directly relating HSRL-2 observables to collocated in situ N_{CCN} using observations from the NASA ORACLES campaign. Using a linear regression approach, we found that directly relating HSRL-2 BSC and EXT coefficients to in situ N_{CCN} was a reasonable approximation for observations of smoke under relatively dry ambient conditions, with $RH \leq 40\text{-}50\%$ (Fig. 1.1; Lenhardt et al., 2023). Other necessary requirements included tight spatiotemporal collocation between both data sets and CCN counter supersaturation between 0.2-0.4%. In cases of a long time delay between remote sensing and in situ observations, the correlation began to weaken. This being the case for a relatively homogeneous smoke layer emphasized that even small changes in aerosol properties over time and space can change how N_{CCN} relates to AOPs. It also highlighted the benefit of two spatially coincident aircraft in studies combining in situ and remote sensing observations.

The final CCN—BSC 532 nm regression equation was used to derive a vertically

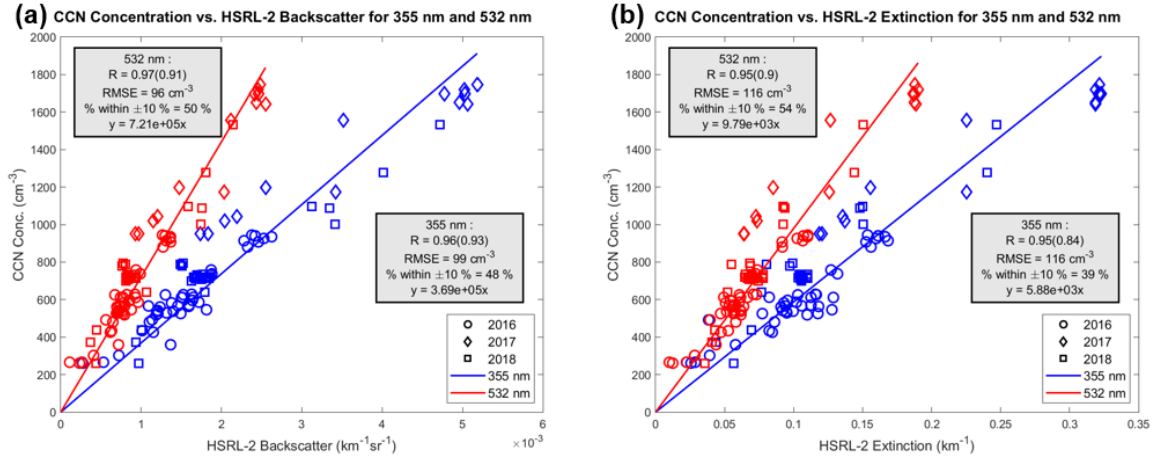


Figure 1.1: Figure 3 from Lenhardt et al. (2023) showing the derived linear relationships between in situ CCN concentration and HSRL-2 (a) backscatter (BSC) and (b) extinction (EXT) coefficients at 355 and 532 nm from observations across all three ORACLES deployments.

resolved N_{CCN} curtain that we compared to output from the WRF-CAM5 model (Weather Research and Forecasting Model coupled with physics packages from the Community Atmosphere Model version 5). An example of this comparison is shown in Fig. 1.2 for August 15, 2017, where the BSC 532 nm-derived N_{CCN} curtain depicts a much less dense smoke plume located at a lower altitude than in WRF-CAM5. There are several factors that can impact this comparison and cause errors in model representation of N_{CCN} . However, fully explaining and/or improving model discrepancies is outside the scope of this work. Rather, we presented this comparison to emphasize the importance of accurately constraining the vertical distribution of N_{CCN} and demonstrated this lidar-derived N_{CCN} data set as a method for improving model representations of vertically resolved N_{CCN} .

After developing this linear approximation method using the ORACLES data, we applied the same approach to observations from the Cloud, Aerosol and Monsoon Processes Philippines Experiment (CAMP²Ex) and the Aerosol Cloud meTeorology Interactions oVer the western ATLantic Experiment (ACTIVATE) campaign with a

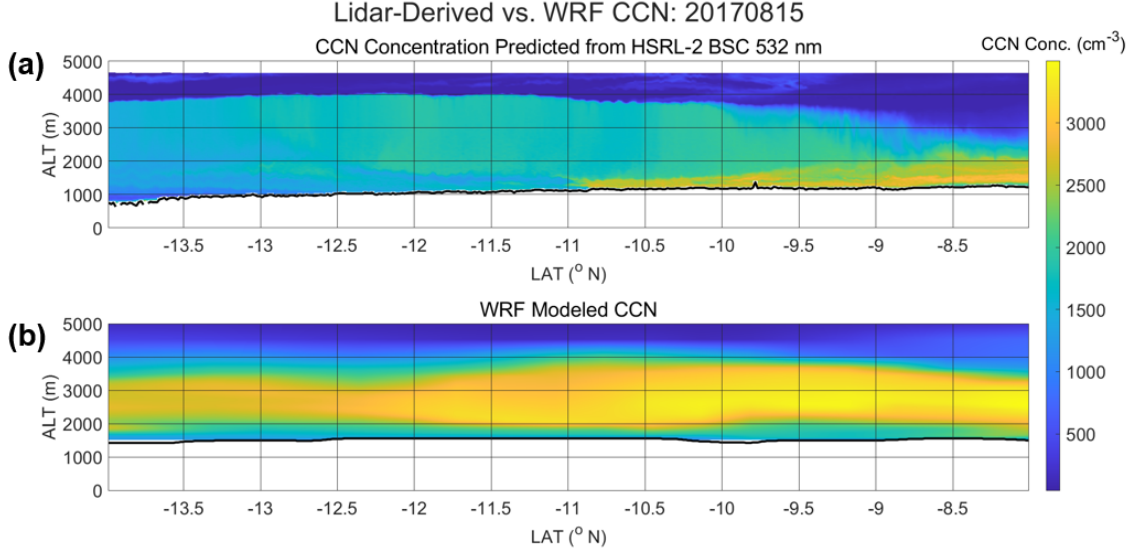


Figure 1.2: Figure 9 from Lenhardt et al. (2023) showing the vertical curtain of N_{CCN} for 20170815 from (a) our HSRL-2-derived method and (b) WRF-CAM5 output.

goal of deriving similar relationships for additional aerosol types. Here we encountered new complexities that adjusted the direction of our work. As previously mentioned, in the ORACLES analysis, correlation between N_{CCN} and BSC or EXT weakened when observations were collocated using a longer time gap. Since all CAMP²Ex observations were made using a single aircraft, there was generally a longer time gap between HSRL-2 and N_{CCN} observations, meaning that there were fewer collocated data points available before any RH or supersaturation filtering. While the time criteria for ORACLES ranged from ± 0.1 - 0.3 hr, data collocation for CAMP²Ex had to be done at ± 0.5 hr. Additionally, while ORACLES observations could reasonably be assumed to be smoke based on altitude, for CAMP²Ex and ACTIVATE we relied on the HSRL-2 Aerosol ID product to separate different aerosol types. If the full set of HSRL-2 observables is not available, an Aerosol ID is usually not determined, which limited the CAMP²Ex data set further. With the remaining collocated data available, the linear relationships between N_{CCN} , BSC, and EXT were relatively strong. However, due to

the limited number of data points, filtering out observations of smoke at high RH did not provide as clear of a solution for hygroscopic growth as it had for ORACLES. Therefore, at this point the hypothesized reasons for weaker linear relationships in CAMP²Ex were increased uncertainty due to fewer data points and a longer averaging time for collocation.

Due to spatially coordinated flights using two aircraft during ACTIVATE, the availability of collocated data points was not an issue when applying our methodology to ACTIVATE observations. However, despite having a larger data set available than for CAMP²Ex, the linear approach still did not prove successful. Rather, we found non-linear relationships with scatter not fully attributable to hygroscopic growth at high RH. Examples of these relationships for different aerosol types and comparisons between campaigns are shown in Fig. 1.3-1.5. Figure 1.3 compares results from ORACLES, CAMP²Ex, and ACTIVATE for observations of smoke aerosols made at ambient RH < 60% and at altitudes above 1500 m, as these were the overlapping conditions at which smoke was observed for all campaigns. All relationships are relatively strong, but weakening of the relationship for ORACLES observations is apparent as RH approaches 60%. Additionally, since most ACTIVATE observations were made in the MBL, the 63 points in the smoke comparison represents only a small fraction of the available data. In Figures 1.4 and 1.5, observations of marine and urban aerosols are shown, respectively, for observations at all RH and all altitudes. Here we found relatively strong linear relationships in the CAMP²Ex data and significant scatter in the ACTIVATE data for both high BSC—low CCN and low BSC—high CCN scenarios.

At this point, we concluded that a linear approximation for deriving N_{CCN} from HSRL-2 BSC and EXT was only appropriate for observations of smoke at low ambient RH in the ORACLES region. Therefore, we did not continue trying to improve and develop similar linear relationships for other observed data sets and aerosol types.

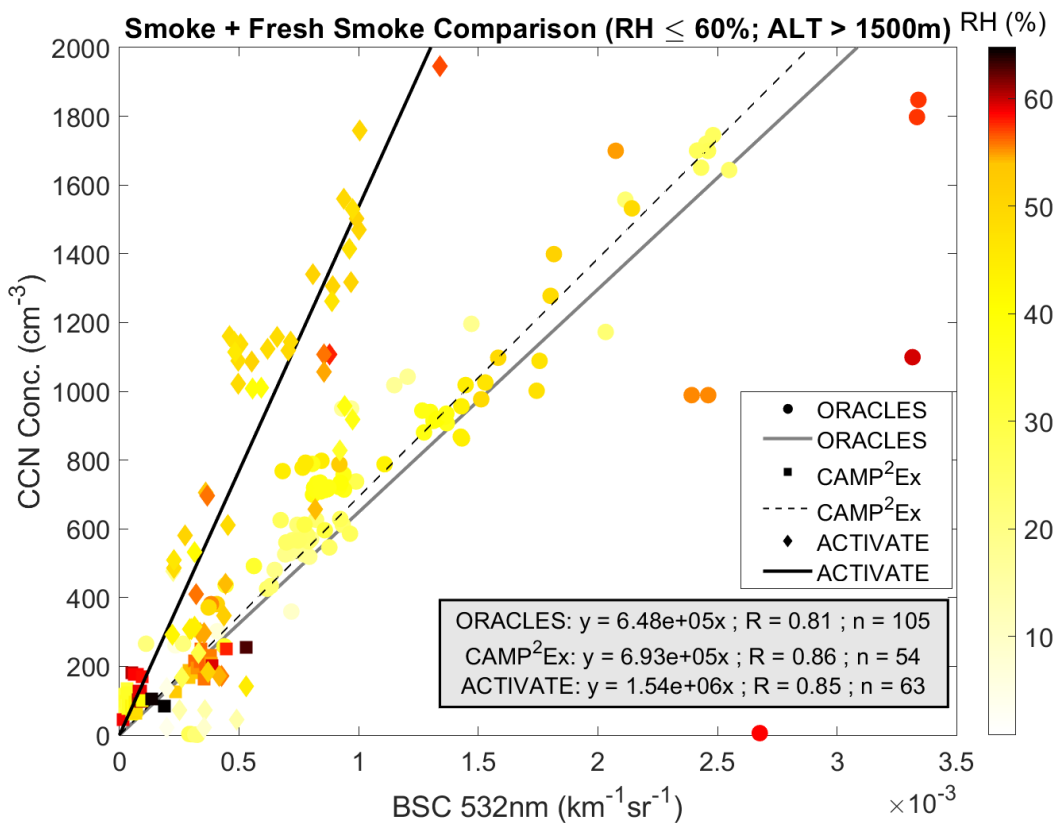


Figure 1.3: Linear relationships between N_{CCN} and BSC 532 nm for ORACLES, CAMP²Ex, and ACTIVATE for observations of smoke made at ambient RH < 60% and altitudes > 1500 m.

Rather, this finding serves as the motivation for the work outlined in Chapter 2: *What specifically governs the N_{CCN} —BSC relationship and determines whether or not it can be approximated linearly?* In Chapter 3, we look at the applicability of using these remotely sensed vertical curtains of N_{CCN} for assessing ACI in the SEA to answer the following questions: *How can N_{CCN} profiles in cloud gaps be used to assess simultaneous impacts of above- and below-cloud N_{CCN} on cloud properties?* and *Do remote sensing observations produce the same ACI results as in situ data?* Chapter 4 documents work translating some of the key concepts from Chapters 1-3 into high school-level classroom activities, assessing the applicability and success of the developed lectures and activities for communicating climate concepts within a meteorology elective course.

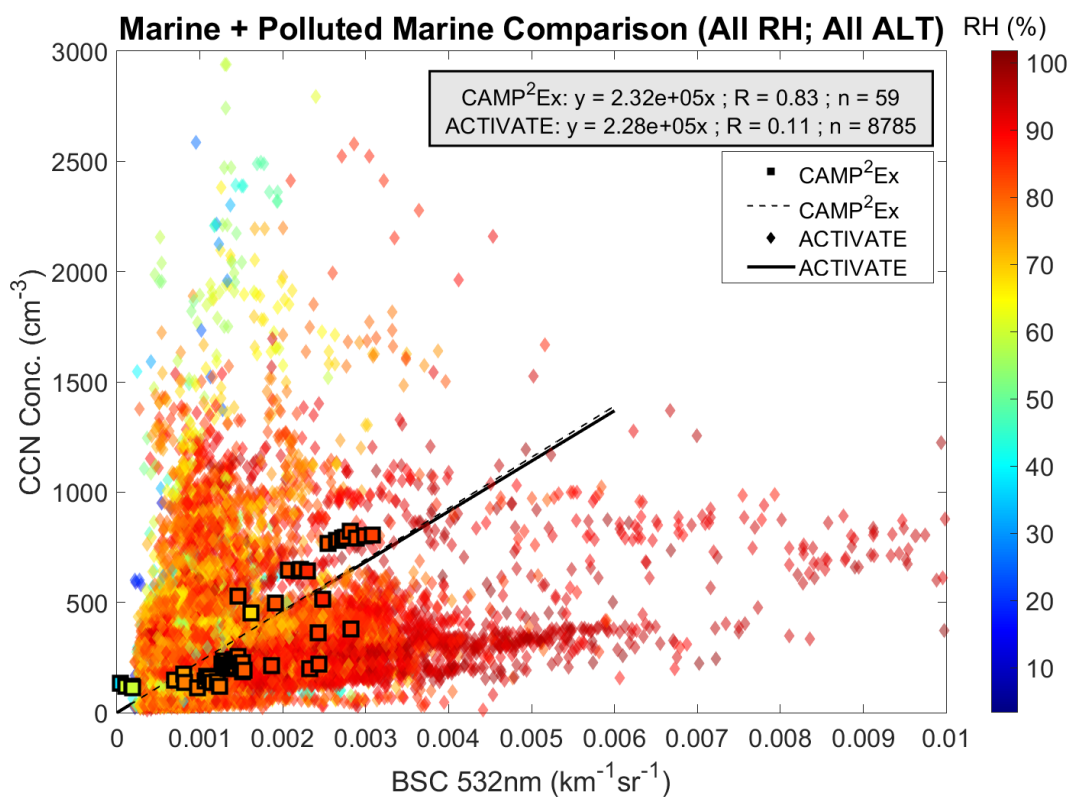


Figure 1.4: Linear relationships between N_{CCN} and BSC 532 nm for observations of marine and polluted marine aerosols in CAMP²Ex and ACTIVATE. Observations are from all ambient RH and all altitudes.

Lastly, Chapter 5 will provide a discussion of the key implications of this work in the context of collaborative efforts and related projects, and Chapter 6 will present the main conclusions from Chapters 2-4.

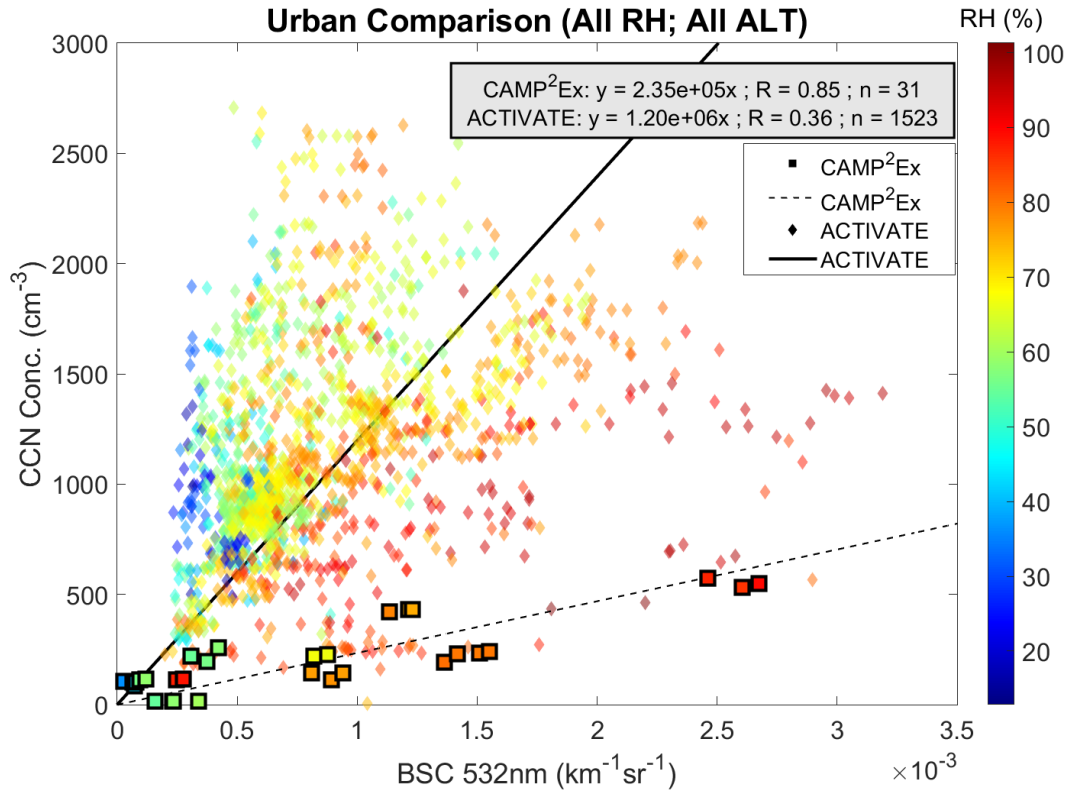


Figure 1.5: Linear relationships between N_{CCN} and BSC 532 nm for observations of urban aerosols in CAMP²Ex and ACTIVATE. Observations are from all ambient RH and all altitudes.

Chapter 2

Aerosol effective radius governs the relationship between cloud condensation nuclei (CCN) concentration and aerosol backscatter

2.1 Introduction

Natural and anthropogenic atmospheric aerosols and their interactions with clouds and radiation have a significant role in climate change and uncertainties in future climate predictions. Specifically, the highest uncertainties compared to other climate forcings are attributed to effective radiative forcing due to interactions between aerosols and clouds (ERF_{aci} ; Forster et al., 2021). Much of the uncertainty of aerosol-cloud interactions (ACI) is due to limited process-level understanding (Boucher et al., 2014) and observing methods. For example, there is limited ability for passive satellite instruments to retrieve cloud and aerosol properties simultaneously in the same environment. Hygroscopic aerosol growth in high relative humidity (RH) environments can also com-

plicate observations (Rosenfeld et al., 2014). Additionally, varying observational scales and meteorological conditions may buffer the responses of clouds to aerosol perturbations (Stevens and Feingold, 2009).

Untangling the impact of ACI from such observational complications requires information on the distribution of those aerosols that interact with clouds by nucleating cloud droplets, i.e., cloud condensation nuclei (CCN). More specifically, knowledge of the vertical distribution of CCN concentration relative to clouds is needed to properly assess and understand ACI. The main challenge in understanding the vertical distribution of CCN lies in the sparsity of in situ observations. Ground-based observations are useful in terms of the length of available observations, but they lack vertical extent. Alternatively, aircraft-based observations can provide observations of CCN closer to cloud base over shorter campaign periods, but these observations are expensive and less frequently available. Therefore, many studies have developed parameterizations, proxies, and retrieval methods to determine CCN from more commonly available remotely sensed observations of aerosol optical properties (AOPs).

One of the most widely used proxies for CCN concentration is aerosol optical depth (AOD), a column-integrated measure of aerosol extinction (EXT). While AOD may approximate CCN concentration over large spatiotemporal extents (Stier, 2016), it often cannot fully explain CCN variance (Andreae, 2009; Shinozuka et al., 2015; Stier, 2016; Choudhury and Tesche, 2022a,b), lacks any information about the vertical distribution of CCN, and is subject to effects of aerosol swelling and cloud contamination (Rosenfeld et al., 2016; Patel et al., 2024). Several studies have related CCN to a combination of other AOPs from lidar and satellite such as aerosol extinction, scattering and backscattering coefficients, backscatter fraction, or the ratio of backscattering to total scattering, single scattering albedo (SSA), scattering Ångström exponent, and aerosol index (AI), which is the product of Ångström exponent and extinction (Ghan

and Collins, 2004; Ghan et al., 2006; Kapustin et al., 2006; Shinozuka et al., 2009; Jefferson, 2010; Liu and Li, 2014; Shinozuka et al., 2015; Mamouri and Ansmann, 2016; Stier, 2016; Tsekeri et al., 2017; Lv et al., 2018; Haarig et al., 2019; Shen et al., 2019; Choudhury and Tesche, 2022a,b; Lenhardt et al., 2023; Patel et al., 2024; Redemann and Gao, 2024). Among such approaches, AOPs can provide constraints for CCN but several underlying uncertainties and limitations exist.

One fundamental issue in relating CCN concentration to AOPs is that particles that act as CCN are generally smaller than particles that have a more significant impact on AOPs when measured at visible wavelengths. Most CCN fall in the Aitken and accumulation modes of the aerosol size distribution, and studies have shown that changes in the aerosol size distribution are the primary drivers of changes in the CCN spectrum (Dusek et al., 2006; Miao et al., 2015; Perkins et al., 2022). In terms of AOPs, many are dominated by coarse mode particles (Shinozuka et al., 2015) and optical measurements tend to be insensitive to small particles that activate as CCN (Jefferson, 2010), causing further uncertainty in correlating both measurements. Another common issue in relating CCN to AOPs is hygroscopic growth of aerosols at high ambient RH. Hygroscopic growth increases aerosol size, thus increasing their light scattering. However, the lack of a corresponding increase in CCN concentration (Shinozuka et al., 2015) causes CCN—AOP relationships to change rapidly at high RH (Liu and Li, 2014; Shinozuka et al., 2015; Stier, 2016; Wang et al., 2025). Since CCN are of particular interest in humid environments near cloud base, this issue can become problematic for ACI applicability. Additionally, aerosol chemical composition influences both CCN concentration and AOPs and their relationship. Some studies have found that CCN—AOP relationships are more uncertain for observations of marine aerosols (Liu and Li, 2014; Shen et al., 2019; Choudhury et al., 2025), which may be related to their more dominant coarse mode and tendency for marine aerosol shapes to be non-spherical

(Fitzgerald, 1991; Von Hoyningen-Huene and Posse, 1997; Bi et al., 2018). In summary, the three most common sources of potential error when relating CCN to AOPs are related to high ambient RH, the shape of the aerosol size distribution, and aerosol chemical composition.

While each of these sources of uncertainty and potentially weak correlation have been noted by numerous studies, many that investigate underlying causes of error focus on each source individually. Additionally, many studies that take a modeling or calculation-based approach to investigating CCN and/or AOPs often use idealized, generated, or average size distributions as a starting point (Li et al., 2015; Lowe et al., 2016; Shen et al., 2019) or vary individual observed size distributions in concentration but not the functional shape (Chuang et al., 2000). While this approach avoids the uncertainties inherent to in situ aerosol size distribution observations, it also does not capture the full range of variability seen in observed size distributions.

In this study we investigate the collective impact of ambient RH, aerosol size distribution, and aerosol chemical composition on CCN—AOP relationships using a broad range of actual observed aerosol properties. Specifically, we follow and expand on Lenhardt et al. (2023), hereafter L23, by applying the same methodology to multiple aerosol types under a variety of ambient RH conditions observed during the Aerosol Cloud meTeorology Interactions oVer the western ATlantic Experiment (ACTIVATE) campaign (Sect. 2.2). L23 focused on optimizing a linear regression model between in situ CCN concentration and aerosol extinction and backscatter from the High Spectral Resolution 2 (HSRL-2) for observations of smoke in mostly dry ($RH \leq 50\%$) conditions during the ObseRvations of Aerosols above CLouds and their intEractionS (ORACLES) campaign. In this study, we perform an L23-motivated analysis and follow it with a more in-depth investigation of the underlying factors that govern how CCN concentration and backscatter at 532 nm (BSC) are related to understand which one may

be the most important predictor. To achieve this, we perform observation-informed theoretical calculations of CCN ($\text{CCN}_{\text{theory}}$) and aerosol BSC ($\text{BSC}_{\text{theory}}$) using in situ observed aerosol size distribution and chemical composition as inputs to both the κ -Köhler and Mie theories (Sect. 2.3 and 2.4). Throughout the study, all observations and calculations of BSC will be at 532 nm. Requisite input data for both calculations comes from ACTIVATE in situ observations, meaning that we do not assume average values for the hygroscopicity parameter or use a singular idealized, representative aerosol size distribution. This approach allows us to capture the observation-informed variability in aerosol size and composition to investigate how such variability impacts the theoretical relationship between CCN and BSC.

2.2 Field Deployment Background and Motivation for the Present Study

The National Aeronautics and Space Administration (NASA) ACTIVATE campaign took place between February 2020 and June 2022 across six deployments over the northwest Atlantic Ocean and generated a unique in situ and remote sensing data set relevant for investigating aerosol-cloud-meteorology interactions. Unlike subtropical regions frequently chosen for ACI-related campaigns, the northwest Atlantic features numerous cloud types, including warm and mixed phase cumulus, that are less well-understood than stratocumulus cloud decks (Sorooshian et al., 2023). Additionally, observations over different seasons allow for analysis of a wide range of aerosol and meteorological conditions. Data were collected using coordinated flights of the NASA Langley Research Center HU-25 Falcon for in situ measurements and King Air aircraft for remote sensing observations. In this study, we collocate in situ and remote sensing observations from both aircraft. The study region and locations of these collocated

data points broken down by aerosol type are shown in Fig. 2.1.

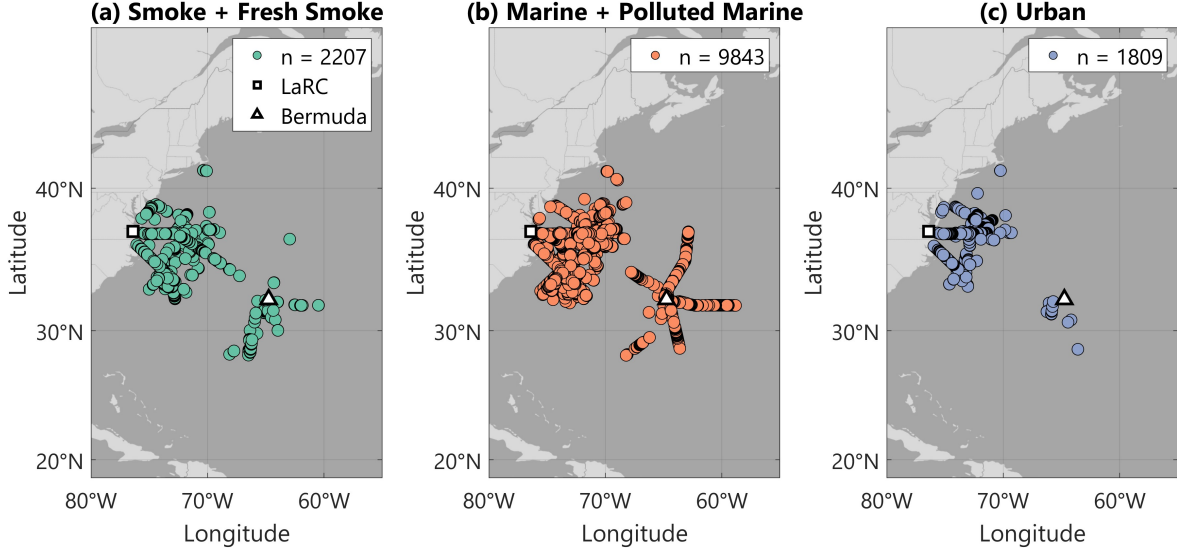


Figure 2.1: Maps showing the ACTIVATE study area and locations of collocated data points for observations of (a) smoke and fresh smoke, (b) marine and polluted marine, and (c) urban aerosols. Langley Research Center (LaRC) in Hampton, Virginia, and Bermuda, the two major bases of operations, are also shown.

During the ACTIVATE campaign, the HU-25 Falcon aircraft conducted profiling flights within, above, and below boundary layer clouds while collecting in situ observations, and the spatially coordinated King Air flew above the Falcon (~ 9 km) conducting remote sensing observations and launching dropsondes (Sorooshian et al., 2023). Following the methodology of L23, our first step is a direct comparison between observed CCN concentration (CCN_{obs}) and BSC at 532 nm (BSC_{obs}), the instrumentation for which are described in Section 2.3.1. Due to the different spatiotemporal resolutions of the in situ and remote sensing data sets, we first collocate both data sets to enable a one-to-one comparison. Fortunately, ACTIVATE prioritized systematic and spatially coordinated flights between both aircraft, with approximately 73% of the cumulative data set having the two aircraft within 6 km and 5 minutes of one another (Schlosser et al., 2024). Therefore, collocation between both aircraft results in many collocated data points for remaining analyses. Our collocation process uses three independent

collocation criteria to find in situ measurements that fall within a set amount of time ($dt = \pm 0.1h$) from when an HSRL-2 profile was measured, within a set horizontal distance ($dd = \pm 0.01^\circ$, or $\pm \sim 1.1$ km) from the profile, and within set vertical bins ($dh = 45$ m). After these criteria have been applied, in situ observations that meet all three criteria are averaged to enable a one-to-one comparison with HSRL-2 BSC. For more details on our sensitivity testing method to determine appropriate values for dt , dd , and dh , and a schematic describing the collocation process, see L23.

We analyze the correlation between collocated CCN_{obs} and BSC_{obs} separated by aerosol type (Fig. 2.2), indicated by the HSRL-2 Aerosol ID product (Sect. 2.3.1.2). In this study, we combine smoke with fresh smoke (SFS) and marine with polluted marine (MPM) due to similarity in their optical properties. We also consider the urban/pollution (URB) aerosol type. Following L23, we fit all relationships using a bisector regression to account for both variables being measured with observational uncertainty. Additionally, we show the coefficient of determination (R^2), a measure of the proportion of variation in CCN_{obs} that is explained by variation in BSC_{obs} , root mean square error (RMSE), a measure of the average difference between linear regression predicted CCN and CCN_{obs} , and number of data points (n). Observations are limited to a small supersaturation range of 0.36-0.38% and marker colors correspond to ambient RH. One of our primary findings in L23 was that the correlation between CCN_{obs} and BSC_{obs} was strongest at low ambient RH ($\leq 50\%$). Therefore, we show separate statistics and regression lines for all observations and the subset observed at $RH \leq 50\%$.

R^2 values for all aerosol types across the full RH spectrum range from 0.0014-0.14, and for $RH \leq 50\%$ range from 0.0023-0.038, suggesting that there is no aerosol type for which variations in CCN_{obs} are well-explained by changes in BSC_{obs} . For all RHs, R^2 is strongest for URB, while smoke has the highest R^2 under limited RH

conditions. For SFS and URB, R^2 decreases when limiting the data set to low RH, contrary to the findings of L23. In the case of the SFS and MPM analyses, RMSE increases when limiting the data set to low RH. Overall, RMSE varies from 342-592 cm^{-3} , and these values are significantly higher than the median CCN_{obs} uncertainty of approximately 150 cm^{-3} for this data set (assuming a relative uncertainty of 10% as reported in the data). Additionally, we see the impact of hygroscopic growth most clearly in the MPM results, where several observations made at $\text{RH} > 80\%$ show increased BSC_{obs} associated with nearly constant, and low, CCN_{obs} values. This aerosol type is primarily influenced by sea salt, one of the most hygroscopic aerosols with a high growth factor and kappa that can range from 0.91-1.33 (Petters and Kreidenweis, 2007). If we consider, as an example, the subset of SFS CCN_{obs} with BSC_{obs} between 0.0006-0.0008 $\text{km}^{-1}\text{sr}^{-1}$ and RH between 80-90%, both small ranges that capture the peak of observed conditions for SFS aerosols, CCN_{obs} ranges from 25 to 2128 cm^{-3} . While this range captures the maximum observed variability, similar magnitudes can also be seen for MPM and URB aerosols within similar small ranges of BSC_{obs} and RH.

Unlike in L23, the direct relationship between CCN_{obs} and BSC_{obs} in the ACTIVATE data cannot be represented well using a linear approximation. We find that even when limiting the data set to observations made at low ambient RH that the correlation is weak and scatter around the regression line is high. Additionally, we find that within individual aerosol types and for small ranges of BSC_{obs} and ambient RH that the magnitude of CCN_{obs} can vary by nearly two orders of magnitude. Another difference between this analysis and ORACLES results is the relatively low frequency of observations made in low RH environments. While more than half of the smoke plume observations in ORACLES were made at low RH, only about 2-10% of the observations in Fig. 2.2 were observed at low RH since the HU-25 Falcon primar-

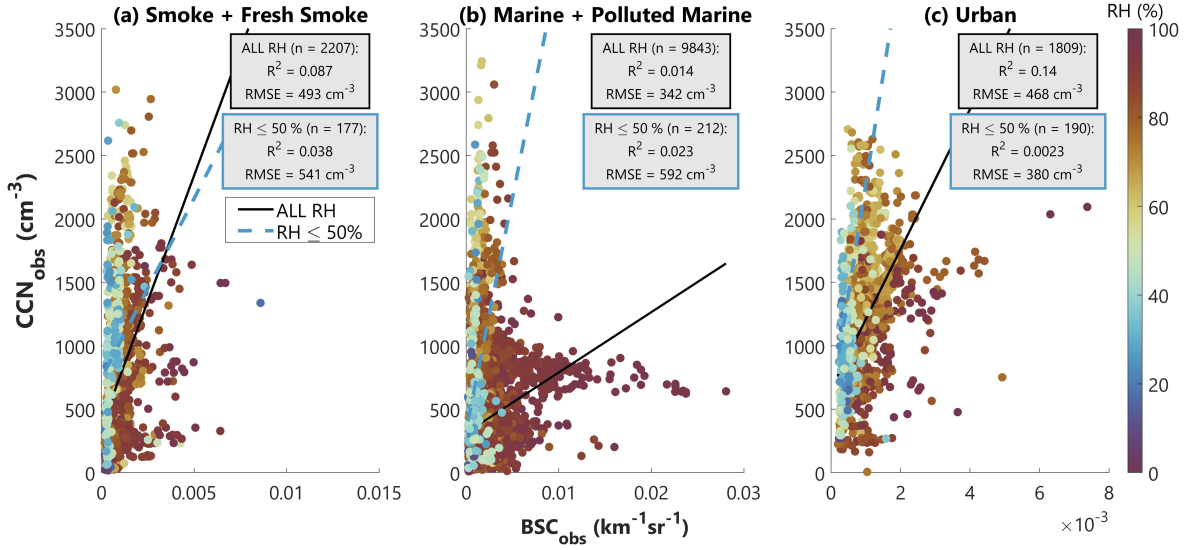


Figure 2.2: Bisector regression of CCN_{obs} vs. BSC_{obs} at 532 nm for (a) smoke and fresh smoke, (b) marine and polluted marine, and (c) urban aerosols. This combined data set covers all years of ACTIVATE and represents 76 flight days. Supersaturation for these observations ranges between 0.36-0.38%. The number of collocated data points (n) is given, as well as the R^2 value and root mean square error (RMSE). Statistics are given for the full data set (black-outlined box), as well as the subset of data observed at $RH \leq 50\%$ (blue-outlined box). The solid black line of best fit applies to the full data set, and the dashed blue line of best fit applies to the low RH subset of data.

ily sampled in the marine boundary layer (MBL) during ACTIVATE. This observed non-linearity between CCN_{obs} and BSC_{obs} in the ACTIVATE data (Fig. 2.2) serves as motivation for the rest of this study – unlike L23, we do not try to optimize a linear relationship between CCN_{obs} and BSC_{obs} . Rather, we perform calculations of CCN_{theory} and BSC_{theory} based on actual observations of aerosol size distribution and chemical composition to understand this observed non-linearity and determine which factors dominate in governing the CCN_{theory} — BSC_{theory} relationship. The goal of this theoretical investigation is to use observations from ACTIVATE as a basis to determine what additional information is most important in constraining CCN concentration from remotely sensed AOPs such as lidar aerosol backscatter.

2.3 Data & Theoretical Calculation Methods

The four primary ACTIVATE data sets used in this study are described in Sect. 2.3.1 and summarized in Table 1. Our methodologies for calculations of $\text{CCN}_{\text{theory}}$ and $\text{BSC}_{\text{theory}}$ are outlined in Sect. 2.3.2 and 2.3.3, respectively. Lastly, we describe pre-analysis data filtering steps in Sect. 2.3.4.

2.3.1 Instrumentation

2.3.1.1 Droplet Measurement Technologies (DMT) CCN Counter

The Droplet Measurement Technologies (DMT) CCN counter measures in situ CCN concentration at multiple levels of water vapor supersaturation (S) and can be run in constant S or scanning S modes (Moore and Nenes, 2009), with most observations from ACTIVATE made at approximately $S = 0.37\%$. This instrument is designed as a continuous-flow streamwise thermal-gradient chamber (CFSTGC; Roberts and Nenes, 2005) where a quasi-uniform supersaturation is generated in the center of a cylindrical flow chamber as heat and water vapor are continuously transported from wetted walls under a temperature gradient. Supersaturation levels vary based on the instrument pressure, flow rate, and imposed column temperature gradient. The continuous-flow feature enables quick (1 Hz frequency) sampling (Roberts and Nenes, 2005), which is important for airborne observations in quickly evolving environments. At the end of the growth chamber, aerosols that activated into droplets with a radius greater than $0.5\ \mu\text{m}$ are counted as CCN. The uncertainty reported for CCN concentration is $\pm 10\%$, with a supersaturation uncertainty of $\pm 0.04\%$ (Rose et al., 2008).

2.3.1.2 High Spectral Resolution Lidar 2 (HSRL-2)

The NASA Langley Research Center HSRL-2 measures aerosol backscatter and depolarization at 355, 532, and 1064 nm and aerosol extinction via the HSRL technique at 355 and 532 nm (Shipley et al., 1983; Hair et al., 2008; Burton et al., 2018). Using the spectral distribution of the return signal, the HSRL measurement technique enables separation of aerosol and molecular backscatter signals, which in turn allows independent, accurate retrieval of aerosol backscatter and extinction profiles without reliance on external assumptions such as the value of the lidar ratio, as is common for basic elastic backscatter lidars (Hair et al., 2008). In this study, we focus on particulate backscatter at 532 nm. The 532 nm wavelength is more frequently available in the data set, and results from L23 suggested that when directly relating CCN concentration with HSRL-2 backscatter and extinction at 355 and 532 nm, there was no substantial difference in performance between either product or wavelength. Additionally, BSC at 532 nm is broadly applicable to existing ground-based and spaceborne lidars and may also be more applicable to observations from a Raman lidar potentially included in the future NASA Atmosphere Observing System (AOS) mission. Uncertainty in the HSRL-2 observables depends on factors such as contrast ratio and aerosol loading, but uncertainties within 5% can be achieved under certain conditions (Burton et al., 2018).

Additionally, since we are interested in the impact of different aerosol types on the CCN – BSC relationship, we also use the HSRL-2 Aerosol ID variable from the observed data set. This Aerosol ID is a qualitative indication of aerosol type from a classification scheme based on HSRL-2 measurements of aerosol intensive parameters including lidar ratio at 532 nm, 1064-to-532 nm backscatter color ratio, depolarization at 532 nm, and depolarization spectral ratio (Burton et al., 2012). The method categorizes eight particle types, which include ice, dusty mix, marine, urban/pollution,

smoke, fresh smoke, polluted marine, and dust. As in Sect. 2, we combine smoke with fresh smoke (SFS) and marine with polluted marine (MPM) due to similarity in their optical properties. We also consider the urban/pollution (URB) aerosol type. These three aerosol types are the most frequently available in the ACTIVATE data. We do not consider observations categorized as ice, dusty mix, or dust in this study. Optically thin ice is infrequently detected by the HSRL-2 in ACTIVATE and does not designate an aerosol type relevant for CCN activation. Aerosols characterized as dust or dusty mix are also infrequently observed, making up only about 9% of the data points with a valid Aerosol ID, which does not permit a statistically relevant consideration of dust-related aerosol types. Implications regarding the applicability of this analysis for dust contributions to aerosol mixtures will be discussed in Sect. 2.5.3.

2.3.1.3 Scanning Mobility Particle Sizer (SMPS) and Laser Aerosol Spectrometer (LAS)

In situ aerosol size distributions come from a combination of the Scanning Mobility Particle Sizer (SMPS) and Laser Aerosol Spectrometer (LAS), both part of the Langley Aerosol Research Group Experiment (LARGE) instrument suite. The uncertainty reported for data from the SMPS and LAS in ACTIVATE is 20% (Sorooshian et al., 2023).

The SMPS uses a soft X-ray aerosol charger (TSI model 3088) to impart an aerosol sample with a known charge distribution and classifies the electric mobility of charged particles with a nano-column differential mobility analyzer (DMA; TSI Model 3085). The particle concentration of aerosols between 0.003-0.089 μm midpoint diameter is then measured using an ultrafine condensation particle counter (CPC; TSI Model 3776; Moore et al., 2017). The resultant size-resolved particle number size distribution is reported as $dN/d\log D_p$ at standard temperature and pressure (STP; 0°C and 1013.25

mb) with 45 second time resolution. Size-dependent corrections have been applied based on laboratory calibration that result in excellent closure with total number concentrations measured by an independent CPC (Sorooshian et al., 2023).

The LAS measures the particle number size distribution ($dN/d\log D_p$) of aerosols with midpoint diameters between 0.1-3.5 μm using an optical method where light intensity scattered from a laser is used to measure particle size (TSI Model 3340; Moore et al., 2021). Unlike less sophisticated optical instruments, a wide-angle scattering technique allows for a monotonic response to the intensity of light scattering to resolve Mie scatter sizing issues. Additionally, an intracavity helium-neon laser design allows for higher light scattering sensitivity at lower laser power. The LAS is calibrated with monodisperse ammonium sulfate particles owing to a refractive index ($n = 1.52$) close to many ambient aerosols (Shingler et al., 2016). Concentrations are reported at STP and with 1 Hz time response. The combination of SMPS and LAS measurements provides a continuous size distribution.

2.3.1.4 Aerodyne High-Resolution Time-of-Flight Aerosol Mass Spectrometer (AMS)

The Aerodyne high-resolution time-of-flight (HR-ToF) aerosol mass spectrometer (AMS) measures submicron, non-refractory composition, including mass concentrations of sulfate, nitrate, ammonium chloride, and organic aerosols, as well as several mass spectral markers (DeCarlo et al., 2008; Sorooshian et al., 2023). The AMS uses an aerodynamic lens to focus particles into a narrow beam within a vacuum chamber, and particles are then impacted onto a 600°C vaporizer. This results in flash vaporization and ionization of non-refractory aerosol components. Ion extraction then allows for the generation of a complete mass spectrum (Jimenez et al., 2003; Drewnick et al., 2005). Refractory components including black carbon, sea salts, and crustal species are not measured

efficiently by the AMS (Jimenez et al., 2003; Cai et al., 2018). Additionally, AMS measurements apply to aerosols with an aerodynamic diameter of approximately 60-600 nm, where transmission efficiencies can be nearly 100% (Jimenez et al., 2003). Although this size range does not cover the full aerosol size distribution, it covers sizes that make up the majority of CCN, so uncertainty due to particle sizes covered by the AMS is small. The AMS was operated at 1 Hz in FastMS mode (i.e., 25 s open, 5 s closed) and averaged to 30 s resolution for the data archive. The uncertainty of AMS observations measured during ACTIVATE is reported to be up to 50% based on processing assumptions related to collection efficiency.

2.3.2 κ -Köhler Theory

The activation of aerosols into cloud droplets is described by Köhler theory, in which the water vapor supersaturation in stable equilibrium with a condensed water droplet is a function of the particle radius. For a constant water vapor supersaturation, particles larger than a critical diameter will experience uncontrolled water condensation and growth to form a cloud droplet (Köhler, 1936). For calculations of CCN_{theory} in this study, we use κ -Köhler theory, which uses a single, bulk hygroscopicity parameter kappa (κ) to represent the relative hygroscopicities of individual aerosol components (Petters and Kreidenweis, 2007). Using this methodology, the critical diameter (D_{crit}) of activation can be calculated with Eq. 2.1,

$$D_{crit} = \left(\frac{4A^3}{27\kappa \ln^2 S_c} \right)^{1/3}, \quad (2.1)$$

where S_c is the specified instrument supersaturation during ACTIVATE, and A is defined in Eq. 2.2,

Instrument	Variables	Resolution (tempo- ral/vertical)	Measurement Type	Aircraft
DMT Cloud Condensation Nuclei (CCN) Counter	CCN concentration at given super- saturation (S)	1 s	In situ	HU-25 Falcon
High Spectral Resolution Lidar 2 (HSRL-2)	Aerosol backscatter coefficient (532 nm), Aerosol ID	10 s / 15 m	Remote sensing	King Air
Scanning Mobility Particle Sizer (SMPS)	Aerosol size distribution (diameter = 0.003–0.1 μm)	45 s	In situ	HU-25 Falcon
Laser Aerosol Spectrometer (LAS)	Aerosol size distribution (diameter = 0.1–3.5 μm)	1 s	In situ	HU-25 Falcon
Aerodyne High Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF- AMS)	Non-refractory chemically resolved mass concentration	30 s	In situ	HU-25 Falcon
Diode Laser Hygrometer (DLH)	Ambient relative humidity (RH)	1 s	In situ	HU-25 Falcon

Table 2.1: List of instruments and data sets used in this study, including their respective resolution, measurement type, and aircraft location.

$$A = \frac{4\sigma_{s/a}M_w}{RT\rho_w}, \quad (2.2)$$

where $\sigma_{s/a}$ is droplet surface tension, which is assumed to be a constant equivalent to

that of pure water (0.0728 N m^{-1} ; Petters and Kreidenweis, 2007), M_w is the molecular weight of water ($18.01528 \text{ g mol}^{-1}$), R is the universal gas constant ($8.3145 \text{ J mol}^{-1} \text{ K}^{-1}$), T is temperature (298.15 K), and ρ_w is the density of water (1000 kg m^{-3}).

2.3.2.1 Kappa Calculations from AMS Data

As shown in Eq. 2.1, D_{crit} depends on a bulk kappa value representing aerosol chemical composition, and we calculate it using AMS observations and the Zdanovskii-Stokes-Robinson (ZSR) mixing rule (Zdanovskii, 1948; Stokes and Robinson, 1966), given in Eq. 2.3,

$$\kappa = \sum_i \epsilon_i \kappa_i, \quad (2.3)$$

where ϵ_i represents the volume fraction of each chemical component and κ_i is the hygroscopicity value of each component. This set of calculations is done using the collocation-averaged AMS data associated with each collocated data point (Sect. 2.7) and a histogram of calculated kappa values for all three aerosol types is given in Fig. 2.3. Additionally, we show a literature-average kappa range for each aerosol type, along with the standard deviation for each end of the range. These values are calculated using six literature values per aerosol type, including SFS (Carrico et al., 2008; Petters et al., 2009; Cerully et al., 2011; Engelhart et al., 2012; Bougiatioti et al., 2016; Gomez et al., 2018; Twohy et al., 2021), MPM (Andreae and Rosenfeld, 2008; Pringle et al., 2010; Gaston et al., 2018; Quinn et al., 2019; Miyazaki et al., 2020; Gong et al., 2023), and URB (Andreae and Rosenfeld, 2008; Pringle et al., 2010; Hung et al., 2014; Kim et al., 2017; Cai et al., 2018; Zamora et al., 2019). Note that Cerully et al. (2011) and Engelhart et al. (2012) provide the same range for SFS, but this value is only counted once in the average. Overall, calculations tend to agree well with those seen in

the literature. Typical kappa values for marine and polluted marine aerosols can vary widely depending on the amount of pollution in a region or if observations are made in cleaner, more remote areas.

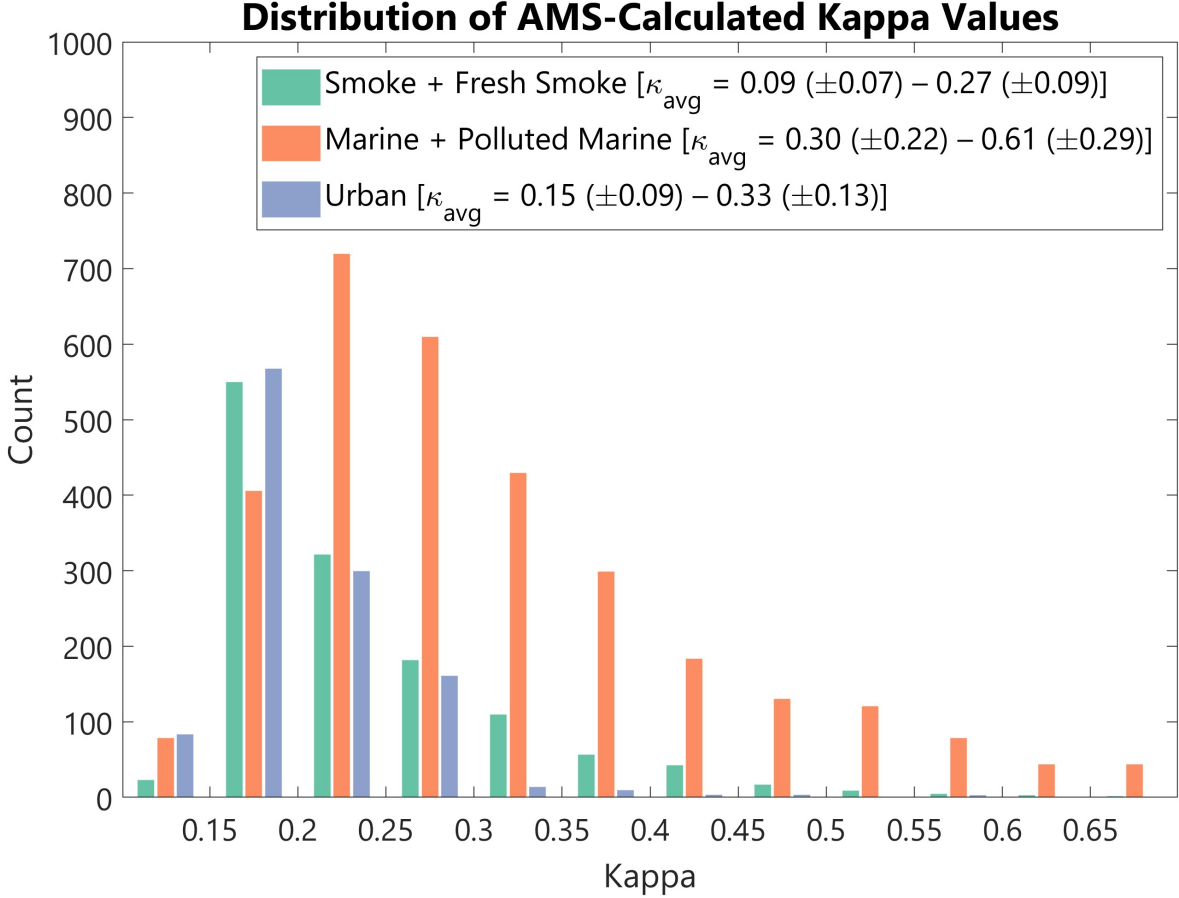


Figure 2.3: Distribution of kappa values calculated using the methodology in Section 2.3.2.1 for each aerosol type. Literature average ranges, with standard deviation, for each aerosol type are given in parentheses.

2.3.2.2 Critical Diameter (D_{crit}) and CCN_{theory} calculation

After calculating kappa following the methodology in Sect. 2.3.2.1, we use Eq. 2.1 to calculate D_{crit} and calculate CCN_{theory} using Eq. 2.4,

$$CCN_{theory} = \int_{D_{crit}}^{D_{max}} \frac{dN}{d \log D_p} d \log D_p, \quad (2.4)$$

where D_{max} signifies the diameter of the largest bin from the combined SMPS and LAS number size distribution (Schmale et al., 2018; Patel et al., 2024), $dN/d\log D_p$ represents the number concentration of aerosols in each bin of the combined size distribution, and S is the CCN counter supersaturation. For direct comparisons between CCN_{theory} and CCN_{obs} , we use the exact CCN counter supersaturation value reported for each collocated data point within a small range of 0.36-0.38% (Sect. 2.4.1). For analyses using only CCN_{theory} without a comparison to CCN_{obs} , we use a constant 0.37% supersaturation (Sect. 2.4.2-2.4.3).

2.3.3 Mie Calculations

The properties of light scattered by atmospheric aerosols are described by Mie theory, where aerosols are assumed to be homogeneous, spherical, and have a diameter approximately equal to the wavelength of incident radiation (Mie, 1908). For our calculations of BSC_{theory} , we calculate size-resolved particle backscattering efficiencies (Q_{bsc}) using the Mie scattering program by Bohren and Huffman (1998) implemented in the libRadtran library of radiative transfer routines and programs (Emde et al., 2016). The two inputs needed to calculate Q_{bsc} are particle size, complex refractive index, and wavelength. To correspond to BSC_{obs} , we only use a wavelength of 532 nm. We use typical refractive index values as retrieved by the Aerosol Robotic Network (AERONET) for different aerosol types to inform our refractive index selection (Dubovik et al., 2002), with exact values given in Table 2.4.

For particle size input we use the SMPS and LAS size distribution bin diameters. However, here we must account for a significant difference in how in situ and HSRL-2 observations are made. With these BSC_{theory} calculations, we want to model ambient BSC_{obs} from the HSRL-2 to understand the relationship with in situ CCN_{obs} . However, since in situ instruments dry ambient air before collecting measurements, we need to

account for the change in particle diameters due to water uptake at ambient RH conditions, since particle size has a significant impact on the magnitude of light scattered. Calculations made to account for changes in particle diameter and refractive index due to hygroscopic growth are outlined in Section 2.8. After these adjustments, humidified bin diameters (D_{wet}) and refractive index components (m_{wet} and n_{wet}) are the final inputs into the Mie scattering calculations run in libRadtran. The size-resolved Q_{bsc} values returned from these calculations are used to calculate BSC_{theory} at 532 nm from the full aerosol size distribution, as shown in Eq. 2.5,

$$BSC_{theory} = \int_{r_1}^{r_n} \pi r_{wet}^2 Q_{bsc} n(r_{wet}) dr_{wet}, \quad (2.5)$$

where r_{wet} is each humidified bin radius, $n(r_{wet})dr_{wet}$ represents the aerosol number concentration in each bin, and r_n represents the largest bin in the SMPS and LAS combined and humidified size distribution. This set of calculations is done using the collocation-averaged size distribution data associated with each collocated data point.

2.3.4 Data filtering

All input data for κ -Köhler and Mie calculations come from the collocated data set used for the observational analysis in Sect. 2.2. Each collocated data point contains an average value of CCN_{obs} and BSC_{obs} , as well as an average combined SMPS and LAS size distribution and set of AMS observations. Therefore, to enable a direct comparison between CCN_{theory} and CCN_{obs} , as well as between BSC_{theory} and BSC_{obs} , this collocated data set is used throughout the entirety of the study. In this section, we describe several filtering steps that are performed to minimize potential errors in the subsequent analyses. Some are motivated by the observational methodology taken in L23, and others are specific to the CCN_{theory} and BSC_{theory} calculations. All steps

are summarized in Fig. 2.4.

We begin with the filtering criteria applied to data in the CCN_{obs} — BSC_{obs} relationships shown in Sect. 2.2. Since these data points are the basis for the rest of the analysis, each of these filtering steps also applies to data used for calculating $\text{CCN}_{\text{theory}}$ and $\text{BSC}_{\text{theory}}$. As we are analyzing these relationships by aerosol type, we start by removing observations from the collocated data set where an HSRL-2 Aerosol ID is not determined, which typically occurs if the full set of HSRL-2 observables is not available. This step removed 51% of the collocated data set. Additionally, we remove any points where the collocation method averages across varying Aerosol IDs to avoid introducing additional uncertainty into the aerosol type. Similarly, we remove points where the standard deviation is greater than the mean of the CCN concentration that falls within our collocation criteria to avoid potential errors due to large variability or gradients in aerosol concentration. Lastly, we remove any collocated points where fewer than two samples comprise the average. This is done to reduce potential noise in the data set, especially for the in situ size distribution data that have a critical role in both sets of theoretical calculations. In general, collocated data points represent an average of 10 observations from the 1 s merged in situ data files, some of which have a lower original resolution (Table 2.1). Each of these filtering steps is applied to all of the data in this study, and the impact of each step on the total number of collocated data points is shown in Fig. 2.4.

In L23, we found that the correlation between CCN concentration and HSRL-2 observations was strongest for supersaturations greater than 0.25%. Additionally, since CCN depends strongly on supersaturation, we limit observations to a small range of supersaturation values to reduce additional variability. Therefore, for analyses that are strictly observational (Sect. 2.2) or that compare theoretical calculations with observations (Sect. 2.4.1), we limit our collocated data set to a supersaturation range

of 0.36%–0.38%. This range was chosen due to the fact that a supersaturation of 0.37% is the most frequent value used during ACTIVATE. This step is only applied to analyses that include observations because, for calculations of CCN_{theory} , we apply a constant supersaturation of 0.37% to any observed size distribution. That is, we do not unnecessarily limit the data used for theoretical calculations by filtering according to CCN counter supersaturation.

The last data-filtering step serves as a check of CCN_{theory} calculations. In addition to using Eq. 2.1 to calculate D_{crit} , as described in Sect. 2.3.2, we also use an estimation method to validate κ -Köhler calculated values. This method integrates the combined SMPS and LAS number size distribution from the largest to smallest bin diameters until the difference between the summed aerosol concentration and observed CCN concentration reaches a minimum. We refer to the bin diameter where this difference reaches a minimum as the estimated D_{crit} ($D_{crit,est}$). We compare these values to the κ -Köhler calculated values ($D_{crit,calc}$) and require that $D_{crit,calc}$ values fall within $\pm 20\%$ of the $D_{crit,est}$ values. This step ensures that our calculated CCN_{theory} values will closely match CCN_{obs} values and removes size distributions that may have higher noise or several bins with missing concentrations. The threshold of $\pm 20\%$ is chosen to correspond to the SMPS and LAS reported uncertainty that impacts the accuracy of the $D_{crit,est}$ value. This step applies to all analyses involving calculations of CCN_{theory} and BSC_{theory} (Sect. 2.4).

As seen in Fig. 2.4, some of these steps remove a significant amount of data from the analysis. While the amount of data removed was taken into consideration at each step, all steps were taken as a precaution against introducing anomalous variation and uncertainty into the analysis. The application of slightly different combinations of filtering steps to the analyses in Sect. 2.4 was done intentionally to allow for as much data as possible to be included in each step. Therefore, while the D_{crit} agree-

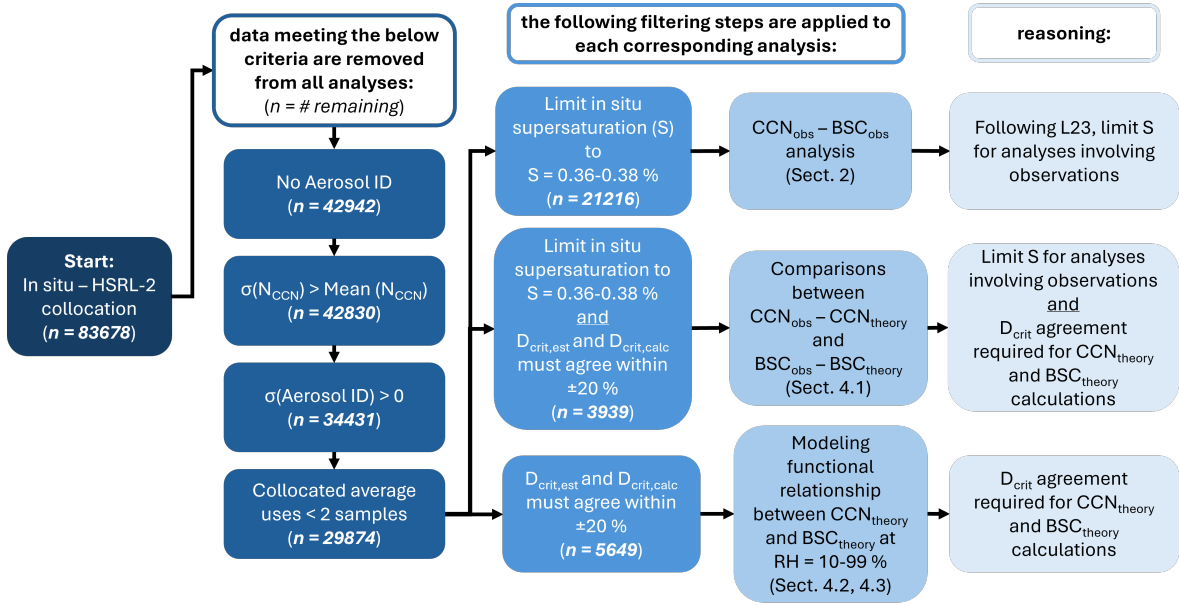


Figure 2.4: Flowchart describing the data-filtering steps applied to data for all analyses and filtering steps applied to data for specific analyses. The number of points remaining after each step (n) is given in parentheses. Therefore, approximately 25% of the original number of collocated samples remain for the observational analysis in Sect. 2.2, 5% remain for analyses comparing observations and theoretical values in Sect. 4.1, and 7% remain for purely theoretical analyses in Sect. 2.4.2 and 2.4.3.

ment filtering step is applied everywhere where we calculate $\text{CCN}_{\text{theory}}$ and $\text{BSC}_{\text{theory}}$, the CCN counter supersaturation filter is only applied where it needs to be used to control the supersaturation dependence of CCN_{obs} . Since the goal of this study is to understand the relationship between CCN_{obs} — BSC_{obs} through the lens of the theoretical calculations, removal of extraneous noise and variability from the input data allows for analyses to more accurately determine the true underlying factors governing the $\text{CCN}_{\text{theory}}$ — $\text{BSC}_{\text{theory}}$ relationship. We discuss a comparison between observed and theoretically calculated CCN and BSC in Sect. 2.4.1, but a detailed discussion of closure for these variables is beyond the scope of this study.

2.4 Results

2.4.1 CCN and BSC observations vs. calculations

We first calculate CCN_{theory} and BSC_{theory} under observed ambient conditions and compare calculations to observations. CCN_{theory} is calculated for all data using the corresponding CCN counter supersaturation, and BSC_{theory} is calculated from humidified aerosol size distributions using the corresponding observed RH value. Since this step involves theoretical calculations and comparison with observations, we limit the data set to observations made at CCN counter supersaturation between 0.36%–0.38% and apply the D_{crit} agreement filtering step (Fig. 2.4).

The comparison between CCN_{obs} and CCN_{theory} is given in Fig. 2.5. We show results requiring a D_{crit} agreement outlined in gray, while calculations without this requirement are plotted in the background. Results for calculations not requiring D_{crit} agreement are shown to demonstrate how this requirement impacts the data set. Results of a linear regression between CCN_{obs} and CCN_{theory} for data requiring the D_{crit} agreement show that, for all aerosol types, R^2 ranges from 0.91 to 0.94, and RMSE ranges from 87 to 133 cm^{-3} . These RMSE values are very close to the approximate median value of CCN uncertainty of 150 cm^{-3} . Data are generally clustered very close to the 1:1 line for all aerosol types, and the lines of best fit also fall close to the 1:1 line. Overall, this analysis gives us confidence that our methodology accurately calculates CCN_{theory} as a necessary precursor for the correlation analysis with BSC_{theory} .

The same statistics are shown for our comparison between BSC_{obs} and BSC_{theory} at 532 nm in Fig. 2.6, where marker colors correspond to RH to show the impact of hygroscopic growth on calculated BSC_{theory} . Here, our R^2 values range from 0.45 to 0.75, and RMSE ranges from 2.7×10^{-4} to $1.6 \times 10^{-3} \text{ km}^{-1}\text{sr}^{-1}$. We find that the performance

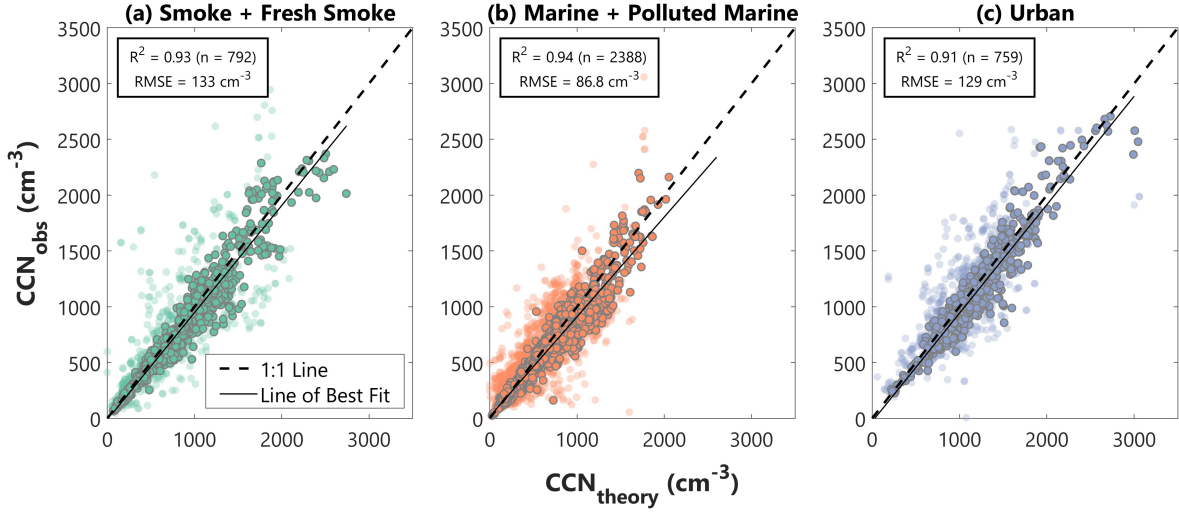


Figure 2.5: CCN_{obs} vs. CCN_{theory} for (a) smoke and fresh smoke, (b) marine and polluted marine, and (c) urban aerosols. The 1:1 lines are dashed, and the lines of best fit for the linear regressions between both variables are solid. Markers outlined in gray denote results for calculations requiring a certain level of D_{crit} agreement. Results for calculations not requiring a D_{crit} agreement are shown in the background with lighter transparency to demonstrate how this requirement impacts the data set.

of our calculations does not appear to systematically decline for observations made at high RH, providing confidence in our humidification calculation methods. The R^2 and RMSE values indicate a weaker correlation between observations and calculations than for CCN, but data remain primarily clustered around the 1:1 line. While the use of the D_{crit} filtering step for this analysis and subsequent removal of size distributions with higher noise or missing concentrations does benefit BSC_{theory} calculations, this does not force a degree of agreement between BSC_{obs} and BSC_{theory} in the same way as it does for agreement between CCN_{obs} and CCN_{theory} . Additionally, CCN_{obs} and the inputs for the CCN_{theory} calculation all come from in situ observations, while the BSC_{theory} calculation uses in situ observations as input but is compared to BSC_{obs} from remote sensing instrumentation on a separate platform. Varying resolutions and collocation averaging between in situ and HSRL-2 observations may cause discrepancies between BSC_{obs} and BSC_{theory} . Other discrepancies in the BSC_{theory} calculation may

come from approximations including the Mie theory assumption of spherical particles and our use of literature-averaged refractive index values for different aerosol types. As with the CCN comparison (Fig. 2.5), this analysis also gives us confidence that our methodology results in BSC_{theory} values of a similar magnitude as BSC_{obs} .

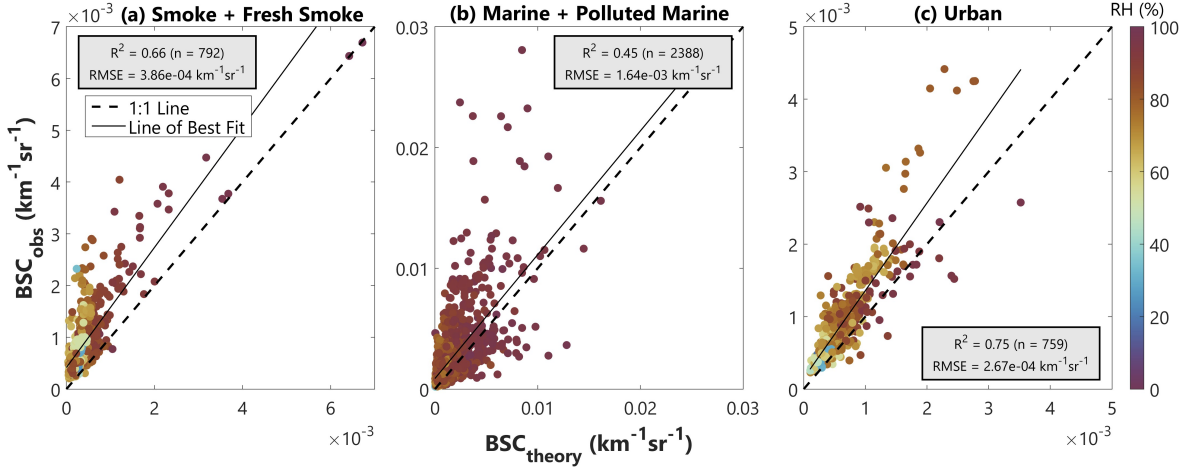


Figure 2.6: BSC_{obs} vs. BSC_{theory} at 532 nm for (a) smoke and fresh smoke, (b) marine and polluted marine, and (c) urban aerosols. The 1:1 lines are dashed, and the lines of best fit for the linear regressions between both variables are solid. Marker colors correspond to ambient RH that was observed by each BSC_{obs} and applied to calculate each corresponding BSC_{theory} .

2.4.2 Estimating predictor importance

In investigating the CCN_{obs} — BSC_{obs} relationship for different aerosol types, we determined that a linear regression is not an appropriate model for the ACTIVATE data (Fig. 2.2). Additionally, we have shown reasonable agreement between CCN_{obs} and CCN_{theory} and between BSC_{obs} and BSC_{theory} at ambient conditions. Next, we use the theoretical calculations to investigate and interpret the causes of scatter and non-linearity in the CCN_{obs} — BSC_{obs} relationship. Analyses in this and the next section use CCN_{theory} calculated at a constant supersaturation of 0.37%.

Recall that the three main factors influencing how CCN concentration relates to

AOPs are ambient RH, the shape of the aerosol size distribution, and aerosol chemical composition. Due to the highly interconnected nature of these factors and their relationships with CCN and BSC, we use random forest (RF) models to determine the relative importance of each factor in controlling the $\text{CCN}_{\text{theory}}$ — $\text{BSC}_{\text{theory}}$ relationship. A random forest is an ensemble of decision trees where each tree is created using the best split from a randomly selected subset of predictors. The final prediction comes from a majority vote among individual trees (Breiman, 2001; Hu et al., 2017). This method was chosen due to its high accuracy, generalization capability, ability to handle non-linear relationships between features, and ability to provide estimates of predictor importance. Another benefit of this method is the ability to consider all input variables collectively as opposed to investigating or perturbing individual input variables one at a time. For each model, we use 200 ensemble learning cycles and specify that all predictor variables are used at each node to ensure that each tree uses all predictor variables. Additionally, 10-fold cross-validation is used during training to prevent overfitting by any single model. The final predictor importance is determined by averaging the importance estimates across the 10 models, and the standard deviation is used to reflect the variations in the final calculated predictor importance estimates. Additionally, we do not separate our data into training and testing subsets because our purpose is not to train and refine a model that predicts $\text{CCN}_{\text{theory}}$ or the $\text{CCN}_{\text{theory}}$ — $\text{BSC}_{\text{theory}}$ relationship. Redemann and Gao (2024) provide a well-tested machine learning method with which CCN concentration is predicted from several HSRL-2 and reanalysis input variables. Rather, here, we use RF predictor importance as a tool to help investigate the impact that ambient RH, aerosol size distribution, and aerosol chemical composition each have on the $\text{CCN}_{\text{theory}}$ — $\text{BSC}_{\text{theory}}$ relationship.

We use a combination of observed effective radius (R_{eff}), geometric mean radius (GMR), RH, and kappa as predictors of the $\text{CCN}_{\text{theory}}$: $\text{BSC}_{\text{theory}}$ ratio in our RF mod-

els. Effective radius is the ratio of the third and second moments of the aerosol size distribution, sometimes called the area-weighted mean radius. This makes it useful for optical measurements as the energy removed from light by an aerosol is proportional to its area. Effective radius is calculated using Eq. 2.6:

$$R_{eff} = \frac{\int_0^\infty \pi r_{wet}^3 n(r_{wet}) dr_{wet}}{\int_0^\infty \pi r_{wet}^2 n(r_{wet}) dr_{wet}}, \quad (2.6)$$

where r_{wet} is the humidified particle radius, and $n(r_{wet})dr_{wet}$ is the aerosol concentration within each bin of the humidified size distribution. The geometric mean radius is the mean of the humidified aerosol size distribution in log space, as given by Eq. 2.7:

$$GMR = \exp \left(\frac{\int_0^\infty \ln r_{wet} n(r_{wet}) dr_{wet}}{N_0} \right), \quad (2.7)$$

where N_0 is the total number of particles in the size distribution. It is important to note that the predictors used in this analysis are not fully independent. For example, RH impacts R_{eff} depending on the corresponding kappa value, meaning that the influence of RH on the CCN_{theory} — BSC_{theory} relationship may be captured through R_{eff} . However, we include both parameters separately to investigate if one of these variables is more important than the other in constraining the CCN_{theory} — BSC_{theory} relationship. Additionally, both R_{eff} and GMR capture the shape of the size distribution and can be related through functional relationships. We use R_{eff} and GMR separately because of their different information content. The weighting of R_{eff} toward larger particles increases its relevance for AOPs, while GMR tends to fall within the fine mode of the size distribution closer to D_{crit} and aerosol sizes relevant for CCN activation. Therefore, based on this combination of input variables, we train the RF models to predict the ratio of $CCN_{theory} \cdot BSC_{theory}$.

First, we train a model for all aerosol types combined. Here, the Aerosol ID from

our collocated in situ and remote sensing data set is added as an additional predictor to test the dependence of $\text{CCN}_{\text{theory}}:\text{BSC}_{\text{theory}}$ on lidar-indicated aerosol type. Average relative predictor importance estimates across all 10 folds are shown in Fig. 2.7a, with a standard deviation designated for each average. Overall, R_{eff} is determined to be the most important predictor of $\text{CCN}_{\text{theory}}:\text{BSC}_{\text{theory}}$, followed by RH. Aerosol ID is the third most important predictor, and GMR and kappa are approximately equal as the fourth and fifth most important predictors, respectively.

Next, we train three individual models that predict $\text{CCN}_{\text{theory}}:\text{BSC}_{\text{theory}}$ for each individual aerosol type as separated by Aerosol ID and again average the relative predictor importance estimates across all 10 folds (Fig. 2.7b). We find that, after separating aerosol types, R_{eff} remains the most important predictor of $\text{CCN}_{\text{theory}}:\text{BSC}_{\text{theory}}$ for all aerosol types. Kappa ranking least important for each aerosol type indicates that separating aerosol types using the Aerosol ID adequately constrains the impact of aerosol chemical composition on the $\text{CCN}_{\text{theory}}-\text{BSC}_{\text{theory}}$ relationship. These separate models also indicate that RH is the second most important predictor for all aerosol types. The relatively low importance of Aerosol ID and kappa in these models is expected considering the fact that $\text{BSC}_{\text{theory}}$ is primarily determined by aerosol size and that CCN activation is also more sensitive to size than to aerosol chemical composition (Dusek et al., 2006).

2.4.3 Modeling $\text{CCN}_{\text{theory}}:\text{BSC}_{\text{theory}}$ using effective radius

Based on the RF predictor importance estimate indication that R_{eff} is the most important predictor for the $\text{CCN}_{\text{theory}}-\text{BSC}_{\text{theory}}$ relationship compared to RH, kappa, and GMR (Fig. 2.7), we now investigate the physical relationship between R_{eff} and the

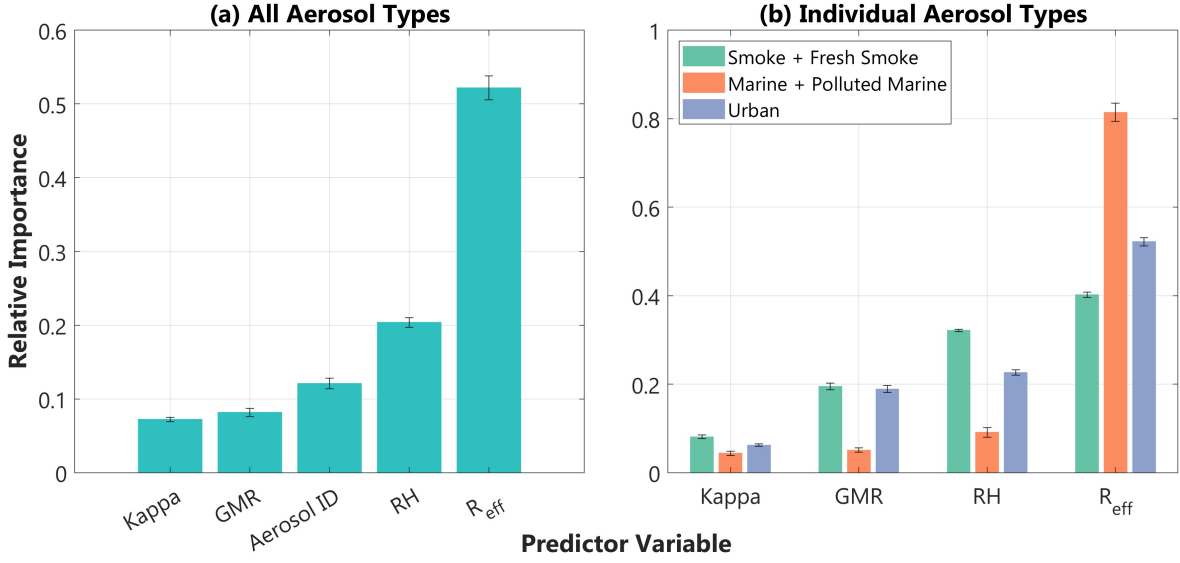


Figure 2.7: Average random forest predictor importance estimates across 10-fold cross-validation for (a) the model run for all three aerosol types combined and (b) individual models run for the three different aerosol types. Each model predicted the $CCN_{theory}:BSC_{theory}$ ratio based on the observed input variables listed on each x axis. All importance estimates are relative. Error bars designate standard deviation across the 10-fold cross-validation.

$CCN_{theory}:BSC_{theory}$ ratio. We focus on R_{eff} to further explore and understand the RF indication of its high importance compared to the other predictors and to understand how much variance in $CCN_{theory}:BSC_{theory}$ can be explained by R_{eff} alone.

We start by humidifying each dry aerosol size distribution at 10% RH increments from 10% to 99% and calculating CCN_{theory} , BSC_{theory} , and R_{eff} from each humidified size distribution. This process allows us to model all variables for a wide range of plausible environmental RH values that are not constrained to observed ambient conditions and to form a more comprehensive understanding of the underlying physical relationship between R_{eff} and $CCN_{theory}:BSC_{theory}$. When comparing $CCN_{theory}:BSC_{theory}$ and R_{eff} , we fit two term exponential curves for each aerosol type to represent the relationship (Fig. 2.8). A two-term exponential was chosen for each aerosol type due to a slightly higher R^2 , lower RMSE, and better visual fit to the larger R_{eff} values than

a one-term exponential fit. For each aerosol type, we provide the R^2 and RMSE (Fig. 2.8), and fit coefficients are provided in Table 2.2.

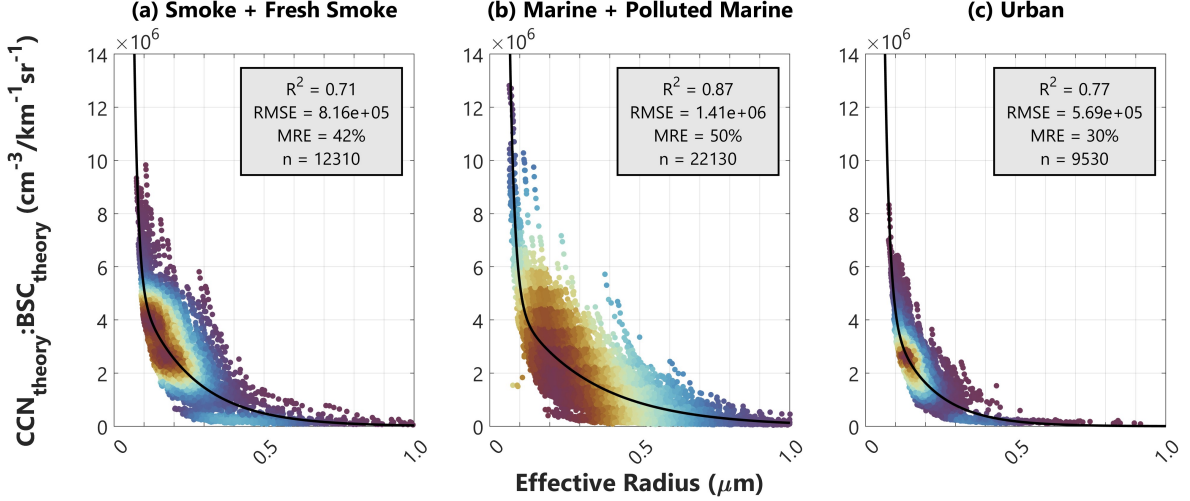


Figure 2.8: $CCN_{theory}:BSC_{theory}$ vs. R_{eff} for (a) smoke and fresh smoke, (b) marine and polluted marine, and (c) urban aerosols. Marker colors correspond to the density of surrounding points, with red shades indicating high density and blue shades indicating lower density. The black line represents the two-term exponential curve fit for each aerosol type. The R^2 , RMSE, mean relative error (MRE), and number of data points (n) for each exponential fit are also provided.

Next, we use each of these two-term exponential fits to calculate $CCN:BSC$ from values of R_{eff} in our ambient collocated data set and compare to $CCN_{theory}:BSC_{theory}$. Here, we refer to $CCN:BSC$ modeled using the two-term R_{eff} exponential fits as $(CCN:BSC)_{model}$ to capture the fact that the ratio itself is modeled using R_{eff} and not each term individually and to distinguish it from $CCN_{theory}:BSC_{theory}$. This comparison is shown in Fig. 2.9, where we find that, overall, most data are clustered around the 1:1 line for each aerosol type. We see that RMSE and mean relative error (MRE) are lowest for the URB category and highest for MPM. Additionally, SFS and URB have many data points at or slightly below the 1:1 line, and a majority of $(CCN:BSC)_{model}$ ratios have magnitudes of about 2×10^6 to $4 \times 10^6 \text{ cm}^{-3} \text{ per km}^{-1} \text{ sr}^{-1}$, while most values for MPM are less than $2 \times 10^6 \text{ cm}^{-3} \text{ per km}^{-1} \text{ sr}^{-1}$. The R^2

values for all aerosol types range between 0.68-0.79.

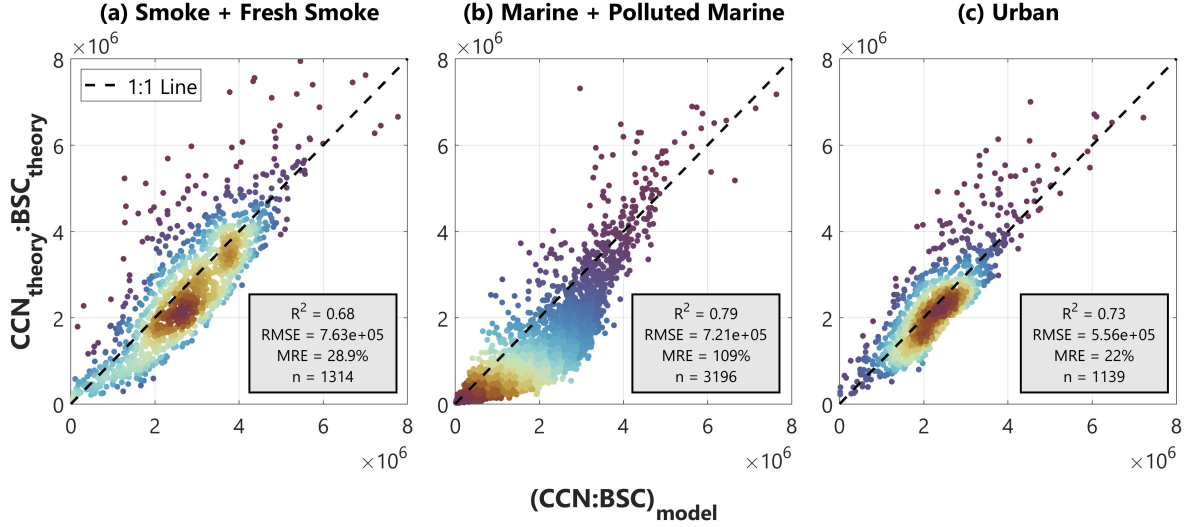


Figure 2.9: Comparison of $CCN_{theory}:BSC_{theory}$ to $(CCN:BSC)_{model}$ for (a) smoke and fresh smoke, (b) marine and polluted marine, and (c) urban aerosols. $(CCN:BSC)_{model}$ values come from the two-term exponential fits shown in Fig. 2.8 and defined in Table 2.2. The units for both axes are cm^{-3} per $km^{-1} sr^{-1}$. Marker colors correspond to the density of surrounding points, with red shades indicating high density and blue shades indicating lower density. The dashed line on each panel is the 1:1 line. The R^2 , RMSE, MRE, and number of data points (n) for each exponential fit are also provided

	a_1 (cm^{-3}/km^{-1} sr^{-1})	b_1 (μm^{-1})	a_2 (cm^{-3}/km^{-1} sr^{-1})	b_2 (μm^{-1})
Smoke + fresh smoke	1.417E09 (26.4%)	-75.64 (4.2%)	7.627E06 (1.4%)	-5.512 (1.4%)
Marine + polluted marine	6.134E08 (1.3%)	-62.01 (0.6%)	6.028E06 (1.1%)	-3.846 (1.4%)
Urban	7.032E08 (21.3%)	-65.73 (3.9%)	6.919E06 (1.4%)	-7.231 (1.9%)

Table 2.2: Coefficients for the two-term exponential curves fit to each aerosol type to model the $CCN_{theory}:BSC_{theory}-R_{eff}$ relationship. All fit equations take the form of $y = a_1 \exp(b_1 x) + a_2 \exp(b_2 x)$, where y corresponds to the $CCN_{theory}:BSC_{theory}$ ratio in cm^{-3} per $km^{-1} sr^{-1}$, and x corresponds to R_{eff} in μm . Relative uncertainties for each coefficient are given in parentheses, estimated using 95% confidence bounds.

2.5 Discussion

Several recent studies have used lidar-observed aerosol optical properties to develop physics-based or ML (machine learning)-based parameterizations and retrieval methods for the CCN concentration of different aerosol types (Mamouri and Ansmann, 2016; Lv et al., 2018; Haarig et al., 2019; Choudhury and Tesche, 2022a; Patel et al., 2024; Redemann and Gao, 2024). In this study, we have included in situ observed aerosol size and chemical composition information to determine which factors most strongly govern the $\text{CCN}_{\text{theory}}\text{---BSC}_{\text{theory}}$ relationship. Therefore, this analysis provides a broad theoretical context in which relationships between observed CCN and aerosol optical properties can be interpreted. In this section, we discuss the physical interpretation of the relationships found, implications for future remote sensing techniques, and a summary of the sources of uncertainty and limitations of the study.

2.5.1 Physical relationships

Based on a set of predictors for the $\text{CCN}_{\text{theory}}\text{:BSC}_{\text{theory}}$ relationship, including R_{eff} , GMR, kappa, and RH, RF predictor importance estimates indicated that R_{eff} was the most important predictor for all three aerosol types of interest in this study. Therefore, we investigated further the relationship between $\text{CCN}_{\text{theory}}\text{:BSC}_{\text{theory}}$ and R_{eff} for a wide range of plausible environmental RH conditions and found a two-term exponential relationship. In further understanding this pattern, it is important to recall that R_{eff} is influenced more significantly by coarse-mode particles than by fine-mode particles. As the coarse-mode number concentration increases, we expect $\text{BSC}_{\text{theory}}$ to increase more compared to $\text{CCN}_{\text{theory}}$, thus decreasing the $\text{CCN}_{\text{theory}}\text{:BSC}_{\text{theory}}$ ratio. This finding is similar to that of Shen et al. (2019), where an exponential relationship was found between the CCN:AOP ratio and the geometric mean diameter of generated

lognormal unimodal size distributions. Additionally, here, the exponential fits show a steeper decrease in $\text{CCN}_{theory}:\text{BSC}_{theory}$ with R_{eff} for MPM aerosols compared to other aerosol types (Table 2.2). Since MPM aerosols are expected to have a more significant coarse-mode contribution, it appears that the effect of BSC_{theory} increasing more than CCN_{theory} with R_{eff} is more pronounced for this aerosol type. As previously mentioned, RH also has an impact on R_{eff} that depends on kappa. The indication that R_{eff} is the most important predictor suggests that understanding the $\text{CCN}_{theory}:\text{BSC}_{theory}$ relationship as based on ACTIVATE observations is not as straightforward as simply constraining RH, as could be done in L23. Rather, the impact of RH on the aerosol size distribution is more important in determining how CCN_{theory} and BSC_{theory} are related.

Based on the R^2 values of 0.68-0.79 in our comparison of $\text{CCN}_{theory}:\text{BSC}_{theory}$ and $(\text{CCN}:\text{BSC})_{model}$ (Fig. 2.9), we find that modeling $\text{CCN}_{theory}:\text{BSC}_{theory}$ using two-term exponential R_{eff} relationships can explain approximately 68%-79% of the variance in the CCN_{theory} — BSC_{theory} relationship. While we previously hypothesized in L23 that aerosol hygroscopic growth at high ambient RH may be the leading cause of variability when relating CCN_{obs} and BSC_{obs} , here, we find that most variability is attributable to differences in R_{eff} . Furthermore, this analysis also speaks to inherent differences in how CCN_{theory} relates to BSC_{theory} for different aerosol types. We find that, when using a large set of actually observed aerosol size distributions as the input into theoretical calculations, there is a significant difference in the range of possible R_{eff} values for each aerosol type (Fig. 2.8). For example, many MPM observations span a wide range of R_{eff} between approximately 0.1-0.5 μm , while SFS and URB, even at wide range of possible ambient RH, primarily see R_{eff} values limited to a small range between 0.1-0.2 μm . Additionally, when we look at the magnitude of $\text{CCN}_{theory}:\text{BSC}_{theory}$ values for each aerosol type, MPM tends to have much lower values than SFS and URB (Fig.

2.9). Higher R_{eff} values, in addition to a higher likelihood for hygroscopic growth in humid marine environments, act to increase BSC_{theory} more than CCN_{theory} , thus decreasing the $CCN_{theory}:BSC_{theory}$ ratio more than for other aerosol types.

Here, we present three $CCN_{theory}:BSC_{theory}-R_{eff}$ exponential fits as a methodology for explaining variance in the $CCN_{theory}-BSC_{theory}$ relationship. The exact functional forms presented in Fig. 2.8 are most appropriate for ACTIVATE observations, and the coefficients would likely need to be adjusted before applying to other data sets. While we expect the general exponential pattern to hold for other data sets, any differences in observed aerosol size distribution or chemical composition would likely change the exact fit coefficients.

2.5.2 Implications for remote sensing techniques

This study indicates several important considerations for future work constraining CCN concentrations from remote sensing observations and future spaceborne lidar data sets. Most importantly, given our finding that particle size, as parameterized by R_{eff} , is the most important predictor in determining the $CCN_{theory}-BSC_{theory}$ relationship, two key points are suggested. First, a simple linear approximation with BSC_{obs} will not constrain CCN_{obs} well in most cases in the ACTIVATE data set. Many previous studies have suggested that the relationship between CCN concentration and various AOPs is often non-linear, specifically for AOD. Considering this background, the results presented here suggest that variations in aerosol size distribution may be a leading cause of non-linearity when using AOD as a proxy for CCN concentration. Seemingly in contrast with the results presented here, in L23, we investigated the relationship between CCN concentration and aerosol index (AI), an indicator of particle size, and found little to no difference between CCN—AI and the CCN—EXT or CCN—BSC relationships. Therefore, for observations of smoke at low RH over the ORACLES region,

we concluded that there was a very small variation in aerosol size in the observations. With minimal differences in aerosol size and with most smoke plume observations being made at low ambient RH, conditions permitted a simple linear approximation to relate CCN_{obs} and BSC_{obs} . On the contrary, the larger data set from the ACTIVATE campaign is characterized not only by a variety of aerosol types but also by a wider range of aerosol size distributions and a higher fraction of observations made at high ambient RH in the MBL, all of which contribute to increased non-linearity between CCN_{obs} and BSC_{obs} .

Related to this non-linearity, a second key point from this analysis is that, in most cases, efforts to constrain CCN concentration using AOPs need to include a measure of the aerosol size distribution to accurately represent variability in the relationship. Here, we have taken advantage of the availability of in situ aerosol size distributions and represented them using R_{eff} . However, to constrain CCN concentration solely from spaceborne lidar observations, our findings suggest that either satellite retrievals of R_{eff} would need to be collocated with lidar observations or a different lidar-derived indicator of aerosol size would need to be used. For example, AI can be calculated using two wavelengths of aerosol extinction from lidar, and other multi-wavelength parameters such as the lidar ratio or backscatter color ratio contain information about aerosol size that could be tested in place of R_{eff} for future methods based solely on a spaceborne lidar system. Additionally, R_{eff} retrievals from the recently launched SPEXone multi-angle polarimeter on board the NASA Plankton, Aerosol, Cloud, and ocean Ecosystem (PACE) mission (Hasekamp et al., 2019) are another option for quantifying aerosol size in CCN concentration estimates.

Lastly, when predicting $CCN_{theory}:BSC_{theory}$ for all aerosol types combined, the RF predictor importance estimates indicated that aerosol type, as represented by the HSRL-2 Aerosol ID, is the third most important predictor (Fig. 2.7a). Since the

Aerosol ID product categorizes aerosol types based on HSRL-2 optical properties, such as BSC, this may explain why Aerosol ID is estimated to be a more important predictor of $\text{CCN}_{theory}:\text{BSC}_{theory}$ than kappa in terms of aerosol type and chemical composition. This finding, in addition to the qualitative differences seen in the impact of high RH between aerosol types (Fig. 2.4), suggests that, while Aerosol ID is not the most important predictor, separately analyzing the CCN—BSC relationship for different aerosol types provides insight into physical differences in CCN—AOP relationships between aerosol types.

2.5.3 Sources of uncertainty and limitations

There are several assumptions underlying both κ -Köhler and Mie theories in addition to uncertainties associated with the observations used as input. Individual instrument uncertainties are discussed in Sect. 2.2.1, and calculation assumptions are discussed in Sect. 2.2.3 and 2.2.4. Here, we acknowledge the primary sources of uncertainty underlying this analysis and the limitations in its applicability.

First, the most significant sources of uncertainty come from uncertainty associated with in situ observations. For example, we use AMS observations to calculate a bulk kappa value needed for κ -Köhler calculations. While we find that our calculated values are generally close to those found in the literature for all three aerosol types (Fig. 2.2), there are a variety of factors that may cause discrepancies. For example, the fraction of mass observed at sizes close to D_{crit} is generally small, meaning that AMS sensitivity to chemical composition at relevant CCN sizes can be limited. Additionally, κ -Köhler theory assumes that chemical composition is fixed across all aerosol sizes (Petters and Kreidenweis, 2007), which may cause discrepancies between CCN_{obs} and CCN_{theory} . Additionally, Kim et al. (2017) found that CCN closure using AMS-calculated kappa values was less accurate than when using kappa calculated from humidified tandem

differential mobility analyzer (HTDMA) observations.

We also consider observational uncertainty associated with in situ size distributions that impact both CCN_{theory} and BSC_{theory} calculations. For example, when considering the comparison between BSC_{obs} and BSC_{theory} , we see the lowest R^2 for the MPM comparison (Fig. 2.6b), for which we present two possible causes. First, marine aerosols have a greater tendency compared to smoke and urban aerosols to be non-spherical in shape, as was observed over Barbados by Haarig et al. (2017) and as has been discussed for the ACTIVATE data set by Ferrare et al. (2023), while Mie theory assumes that particles are spherical (Von Hoyningen-Huene and Posse, 1997; Bi et al., 2018). Second, in situ aerosol size distributions tend to under-represent coarse-mode aerosols due to inefficient sampling at large sizes (McMurry, 2000; Ryder et al., 2018; Kangasluoma et al., 2020). Since marine aerosols tend to have a dominant coarse mode that contributes significantly to light scattering and since this coarse mode is likely to be underrepresented by the in situ size distributions used as input into Mie calculations, this may be another cause of the discrepancy between BSC_{obs} and BSC_{theory} . Lastly, aerosols may be undersized due to the loss of volatile aerosol components that occurs during the heating and drying of in situ observations during inlet transmission (Shrestha et al., 2018; Sandvik et al., 2019), and this may be another source of uncertainty in BSC_{theory} and CCN_{theory} calculations. However, overall, Figs. 2.5 and 2.6 provide confidence that the combination of uncertainties in the size distributions and other input variables does not prohibit reasonable agreement between CCN_{obs} and CCN_{theory} or between BSC_{obs} and BSC_{theory} . Therefore, while uncertainties in the in situ data are likely to cause errors in our theoretical calculations, the intermediate comparison between observations and calculations provides confidence that these uncertainties do not undermine the validity of this study.

Lastly, there are a few important considerations for the applicability and limita-

tions of this study. While the ACTIVATE campaign collected one of the most complete airborne data sets in terms of the range of aerosol types and meteorological conditions, our findings are limited to the campaign study area and the encountered aerosol mixtures; they have not been tested on other data sets. For example, since we are unable to include dust in the analysis due to observational constraints, our results cannot speak to differences in the CCN_{theory} — BSC_{theory} relationship for aerosol mixtures with large proportions of dust. We would expect the results shown here to differ for observations of dust due, in part, to its hydrophobic nature and large, generally non-spherical sizes and shapes that are not easily represented using Mie theory. Recent studies have started using lidar products to better model and understand dust aerosol optical properties (Saito and Yang, 2021; Haarig et al., 2022), but more work is needed to understand the relationship between dust optical properties and its ability to activate as CCN. Additionally, as previously mentioned, we would also expect the general exponential relationship between CCN_{theory} : BSC_{theory} — R_{eff} to hold for other non-dust data sets, but the exact fit coefficients would likely need to be adjusted.

2.6 Conclusion

To improve our understanding of CCN distributions, many techniques have developed proxies and parameterizations using remotely sensed AOPs. Such strategies often provide a good constraint for CCN, but challenges remain due to factors such as aerosol hygroscopic growth and variations in the aerosol size distribution. In this study, we investigate the dominant governing factors of the CCN_{theory} — BSC_{theory} relationship at 532 nm for different aerosol types using observation-informed theoretical calculations and find that R_{eff} is the most important predictor for smoke, marine, and urban aerosols.

This dependence of CCN_{theory} — BSC_{theory} on the aerosol size distribution explains why, as expected, a linear approximation is generally not an appropriate method for representing the relationship well. Rather, this approach only works in limited, specific cases. For example, when analyzing the CCN_{obs} — BSC_{obs} relationship for observations of smoke at low ambient RH with a narrow range of aerosol sizes in ORACLES, a linear regression performed well. However, in cases such as ACTIVATE, where (i) most observations are made at high ambient RH, (ii) there is a variety of aerosol types present, and (iii) there exists a wider range of observed aerosol size distributions, this approach is not possible. Through these observation-informed analyses, we have provided a theoretical framework for understanding the impact of different governing factors on the CCN_{obs} — BSC_{obs} relationship and the relative importance of the size distribution compared to chemical composition and hygroscopic growth at high ambient RH.

Our findings suggest a few key takeaways for future studies using spaceborne remote sensing instrumentation, such as CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation) or other future spaceborne lidar observations, to retrieve CCN concentrations at cloud-relevant altitudes. Most importantly, we found through using a wide range of in situ-observed size distributions that R_{eff} captures well the strong dependence of the CCN_{theory} — BSC_{theory} relationship on the aerosol size distribution for non-dust aerosol mixtures. That is, for areas with a wide variety of observed size distributions, CCN cannot be estimated well from BSC without including aerosol size. Therefore, future remote sensing methods based on estimating N_{CCN} from particulate backscatter would require a lidar capable of providing R_{eff} , a backscatter lidar in combination with a polarimeter, or collocated satellite retrievals of R_{eff} . Overall, we found that there is great benefit in using a wide variety of in situ observed aerosol size distributions as input for CCN_{theory} and BSC_{theory} calculations to

understand, in detail, how the size distribution impacts the relationship between CCN and AOPs.

2.7 Appendix A

The AMS-measured ion concentrations of NH_4^+ , SO_4^{2-} , and NO_3^- must first be converted into volume fractions as required by Eq. 2.3. For this conversion, we first use the simplified ion pairing scheme developed by Gysel et al. (2007) to calculate the number of moles (n) of ammonium nitrate (NH_4NO_3), sulfuric acid (H_2SO_4), ammonium bisulfate (NH_4HSO_4), and ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$), as outlined in Eqs. 2.8:2.12:

$$n_{\text{NH}_4\text{NO}_3} = n_{\text{NO}_3^-}, \quad (2.8)$$

$$n_{\text{H}_2\text{SO}_4} = \max(0, n_{\text{SO}_4^{2-}} - n_{\text{NH}_4^+} + n_{\text{NO}_3^-}), \quad (2.9)$$

$$n_{\text{NH}_4\text{HSO}_4} = \min(2n_{\text{SO}_4^{2-}} - n_{\text{NH}_4^+} + n_{\text{NO}_3^-}, n_{\text{NH}_4^+} - n_{\text{NO}_3^-}), \quad (2.10)$$

$$n_{(\text{NH}_4)_2\text{HSO}_4} = \max(n_{\text{NH}_4^+} - n_{\text{NO}_3^-} - n_{\text{SO}_4^{2-}}, 0), \quad (2.11)$$

$$n_{\text{HNO}_3} = 0, \quad (2.12)$$

where the number of moles of NH_4^+ , SO_4^{2-} , and NO_3^- is calculated using their AMS-observed ion concentrations and molar mass values. Next, the number of moles of ammonium nitrate, sulfuric acid, ammonium bisulfate, and ammonium sulfate is

converted into units of mass. After this step, their dry densities, as given in Table A1 (Gysel et al., 2007; Kuang et al., 2020), are used to convert each mass into a volume. During this step, the AMS-measured concentration of organics is also converted into a volume. The five resultant volumes are summed, and the total volume is used to calculate the volume fraction (ϵ_i) of each component. Following this step, the individual volume fractions and κ_i values given in Table 2.3 (Cai et al., 2018; Kuang et al., 2020) are used in Eq. 2.3 to calculate a bulk kappa.

Compound	NH ₄ NO ₃	H ₂ SO ₄	NH ₄ HSO ₄	(NH ₄) ₂ SO ₄	Organics
Density ρ (kgm ⁻³)	1720	1830	1780	1769	1400
κ_i	0.58	0.90	0.56	0.48	0.10

Table 2.3: Density and hygroscopicity constants of individual chemical components used to calculate bulk hygroscopicity value.

2.8 Appendix B

The change in particle diameter is described using a hygroscopic growth factor $g(\text{RH})$, as defined in Eq. 2.13:

$$g(\text{RH}) = \frac{D_{\text{wet}}(\text{RH})}{D_{\text{dry}}}, \quad (2.13)$$

Here, D_{dry} is the dry particle diameter from the SMPS and LAS-observed size distribution, and D_{wet} is the adjusted particle diameter at a given RH. To calculate D_{wet} , we follow the methodology of Zieger et al. (2013), who note that the RH dependence of Eq. 2.13 can be parameterized using a relationship introduced by Petters and Kreidenweis (2007), as given in Eq. 2.14:

$$g(a_w) = \left(1 + \kappa \frac{a_w}{1 - a_w}\right)^{1/3}, \quad (2.14)$$

where a_w is water activity, and κ is the bulk hygroscopicity parameter as calculated in Sect. 2.3.2.1. If the Kelvin effect can be neglected, a_w can be replaced with RH. Since the Kelvin term of the Köhler equation is small for large particles ($D > 80$ nm), we make this replacement moving forward since particles larger than 80 nm contribute most to BSC compared to smaller particles. Therefore, we calculate humidified aerosol sizes using Eq. 2.15:

$$D_{\text{wet}}(\text{RH}) = D_{\text{dry}} \left(1 + \kappa \frac{\text{RH}}{1 - \text{RH}} \right)^{1/3}. \quad (2.15)$$

Additionally, the change in the refractive index due to hygroscopic growth is calculated using Eqs. 2.16 and 2.17 for the real (m_{wet}) and imaginary (n_{wet}) components, respectively:

$$m_{\text{wet}}(\text{RH}) = \frac{m_{\text{dry}} + m_{\text{H}_2\text{O}} (g(\text{RH})^3 - 1)}{g(\text{RH})^3}, \quad (2.16)$$

$$n_{\text{wet}}(\text{RH}) = \frac{n_{\text{dry}} + n_{\text{H}_2\text{O}} (g(\text{RH})^3 - 1)}{g(\text{RH})^3}. \quad (2.17)$$

Here, m_{dry} and n_{dry} are the dry real and imaginary refractive indices for each aerosol type, as given in Table 2.4 and informed by Dubovik et al. (2002). Additionally, $m_{\text{H}_2\text{O}}$ and $n_{\text{H}_2\text{O}}$ are the real (1.33) and imaginary (0) refractive indices for water.

Aerosol type	Real component (m_{dry})	Imaginary component (n_{dry})
Smoke	1.505	2.005E – 02
Fresh smoke	1.425	2.005E – 02
Marine	1.389	1.005E – 03
Polluted marine	1.407	5.050E – 04
Urban (URB)	1.475	5.500E – 03
Smoke + fresh smoke (SFS)	1.465	2.005E – 02
Marine + polluted marine (MPM)	1.398	7.550E – 04

Table 2.4: Dry refractive indices for each aerosol type. The two bottom rows represent the two combined aerosol types used in this study. Their refractive indices are calculated using an average of both components from both aerosol types (i.e., the real and imaginary components for SFS are an average of the real and imaginary components for smoke and fresh smoke).

Chapter 3

Evidence of cloud sensitivity to above-cloud CCN as a function of environmental stability in the Southeast Atlantic based on remote sensing observations

3.1 Introduction

Amidst several identified climate forcings that have and will continue to determine current and future climate warming, the highest uncertainty remains linked to aerosol-cloud interactions (ACI; Forster et al., 2021). Clouds play a significant role in the climate system by regulating the atmosphere’s radiative budget and surface precipitation. To improve their accuracy in climate model projections, the impact of cloud condensation nuclei (CCN) and ice nucleating particles on cloud properties must be better understood and quantified (Seinfeld et al., 2016). Recent improvements in our understanding of cloud processes have suggested that in a warming climate, clouds may act to amplify warming instead of suppressing it (Forster et al., 2021). The strong

dependence of our future climate on clouds and aerosols motivates continued efforts to reduce uncertainty associated with their interactions.

Increases in CCN concentration (N_{CCN}) at a fixed cloud liquid water path (LWP) are generally understood to reduce cloud droplet effective radius (R_{eff}) while increasing cloud droplet number concentration (N_d), thereby increasing the reflectance of clouds (Twomey, 1974)). The nucleation of more numerous small droplets may delay or suppress the formation of raindrops by reducing collision-coalescence rates, thereby extending cloud lifetime (Albrecht, 1989) due to precipitation suppression and susceptibility to aerosols (e.g., Gupta et al., 2022b). Delayed precipitation formation can allow the cloud layer to reach higher cloud top heights (Andreae, 2004; Rosenfeld, 2007; Myhre et al., 2007), and the consequent increase in cloud geometric thickness can buffer the initial precipitation suppression via an increase in LWP. The numerous pathways and outcomes of ACI are interlinked, complicating our understanding of their overall effect on Earth’s radiative budget because of the uncertainty associated with the sign and magnitude of their effective radiative forcing (Myhre et al., 2007; Forster et al., 2021). Additionally, local meteorology strongly governs cloud properties, making it difficult to disentangle aerosol effects from the effects of dynamics and meteorological regimes (McFarquhar, 2004; Lohmann et al., 2006; Mauger and Norris, 2007; Stevens and Feingold, 2009; Gryspeerdt et al., 2014; Rosenfeld et al., 2014; Zhang et al., 2016; Malavelle et al., 2017; Douglas and L’Ecuyer, 2019). Previous studies have constrained the impact of meteorology or climatological regimes by developing regime-based approaches to constrain ACI as a function of cloud top pressure, cloud optical depth, vertical pressure velocity, lower tropospheric stability (LTS), estimated inversion strength (EIS), precipitation rate, and updraft velocity (Wood and Bretherton, 2006; Gryspeerdt and Stier, 2012; Gryspeerdt et al., 2014; Zhang et al., 2016; Chen et al., 2018; Zhao et al., 2025).

Many studies investigate evidence of ACI using retrievals of cloud microphysical properties from passive satellite instruments such as the Moderate Resolution Imaging Spectroradiometer (MODIS; Myhre et al., 2007, Alam et al., 2010, Goren and Rosenfeld, 2012, Chen et al., 2015, McCoy et al., 2017, Pan et al., 2018, Painemal et al., 2020, Gryspeerdt et al., 2022, Gupta et al., 2022a) and the Spinning Enhanced Visible and Infrared Imager (SEVIRI; Goren and Rosenfeld, 2012, Alexandri et al., 2024) or from active sensors such as CloudSat (Pan et al., 2018; Douglas and L’Ecuyer, 2019). Cloud retrievals from these platforms have been tested and evaluated against airborne in situ observations (Roebeling et al., 2008; Painemal and Zuidema, 2011; Min et al., 2012; King et al., 2013; Gupta et al., 2022a; Wang et al., 2024; Painemal et al., 2025). However, their aerosol observations are limited to columnar products such as aerosol optical depth (AOD), which may serve as an adequate N_{CCN} proxy over large spatiotemporal extents (Stier, 2016) but cannot fully explain N_{CCN} variance and lacks any information about the vertical distribution of N_{CCN} (Andreae, 2009; Shinozuka et al., 2015; Stier, 2016; Choudhury and Tesche, 2022a,b). Additionally, aerosol retrievals from passive remote sensing platforms are limited to clear-sky conditions and are further limited spatially by cloud contamination and near-cloud aerosol humidification effects, meaning that their aerosol products are often limited to those 15 km away from cloud edge (Christensen et al., 2017). Enhanced near-cloud reflectance due to three-dimensional scattering can also occur at sunlit cloud edges (Várnai and Marshak, 2009; Várnai and Marshak, 2011), which does not impact active lidar observations. These limitations often make passive aerosol retrievals insufficient for studying ACI.

The vertical distribution of N_{CCN} is especially relevant for ACI since N_{CCN} is of greater interest near the cloud base, where most cloud droplets nucleate. Therefore, many satellite-based studies have incorporated observations from the satellite-based Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) to add information about

aerosol vertical distributions to ACI analyses (Várnai and Marshak, 2012; Costantino and Bréon, 2013; Pan et al., 2018; Douglas and L’Ecuyer, 2019; Painemal et al., 2020; Alexandri et al., 2024; Li et al., 2025). CALIOP observations allow for estimates of aerosol layer heights relative to cloud and therefore provide more reliable ACI estimates than vertically integrated products. For example, Pan et al. (2018) found a weakening of ACI relationships with increasing cloud base height in South Asia, which was associated with the vertical distribution of aerosols. Painemal et al. (2020) and Li et al. (2025) found strong correlations between droplet number concentration and extinction measured below cloud top, compared to less meaningful relationships seen using AOD. Studies using CALIOP to incorporate vertical aerosol distributions into ACI analyses have facilitated continued efforts in improving vertically resolved aerosol and N_{CCN} information to constrain ACI uncertainties. Other studies have investigated ACI using ground-based (e.g., Lihavainen et al., 2010, Modini et al., 2015, Dedrick et al., 2025) or airborne in situ observations (e.g., Twohy et al., 2005, Modini et al., 2015, Diamond et al., 2018, Kacarab et al., 2020, Gupta et al., 2021, D’Alessandro et al., 2023, Zheng et al., 2024). Airborne platforms allow for higher spatial resolution than satellite observations and greater spatial coverage than ground-based in situ observations.

Marine stratocumulus clouds cover approximately one-third of global oceans (Warren et al., 1988) and have a strong impact on the shortwave radiation budget. ACI and the radiative properties of stratocumulus clouds are regulated in part by cloud top entrainment, which can cause droplet evaporation and thinning of clouds depending on the moisture content of free tropospheric air (Wood, 2012; Mellado, 2017). One region of particular interest for studying these clouds is the Southeast Atlantic Ocean (SEA), where seasonal biomass burning aerosols (BBA) are emitted from Southern Africa and lofted atop a semi-permanent deck of marine stratocumulus clouds (Redemann et al.,

2021). The absorbing nature of these aerosols has implications for the direct aerosol radiative effect (DARE; Doherty et al., 2022, Chang et al., 2025), and the entrainment of BBA into both the cloud layer and the underlying marine boundary layer (BL) has implications for cloud microphysical properties (Kaufman et al., 2003; Ross et al., 2003; Adebiyi et al., 2015; Zuidema et al., 2016). Diamond et al. (2018) found a strong relationship between in situ N_d and below-cloud BBA in the SEA region. Subsequently, Gupta et al. (2021) found in situ-based evidence of ACI that were dependent on not just below-cloud aerosols, but also the vertical separation between cloud tops and the above-cloud BBA plume. While considerable focus is placed on interactions between below-cloud N_{CCN} and cloud microphysics, recent observational and modeling studies address the possibility of cloud droplet nucleation at cloud-environment interfaces above cloud base (Sun et al., 2012; Hernández Pardo et al., 2019; Oh et al., 2023; Sterzinger and Igel, 2024).

The overarching objective of this study is to investigate relationships between stratocumulus cloud properties and N_{CCN} using a remote sensing-based approach. Motivated by the results of Gupta et al. (2021) for the NASA ObseRvations of Aerosols above CLouds and their intEractionS (ORACLES) campaign (Redemann et al., 2021), we expand on this study using remotely-sensed cloud microphysical properties and a new method for retrieving above- and below-cloud N_{CCN} from High Spectral Resolution Lidar 2 (HSRL-2) observables (Redemann and Gao, 2024; Sect. 3.2.1.1). The use of vertically resolved N_{CCN} from this method allows us to look at ACI as a function of N_{CCN} within 100 m above cloud top and to investigate the simultaneous impact of above- and below-cloud N_{CCN} on cloud top microphysical properties using lidar profiles adjacent to the cloud edge. Specifically, the research discussed here addresses the following questions:

1. Can remote sensing retrievals replicate the relationships between above-cloud

N_{CCN} and cloud top microphysical properties identified from in situ measurements for clouds impacted by smoke aerosols?

2. How do above-cloud ACI vary based on meteorological conditions?
3. Can the impacts of below-cloud N_{CCN} be constrained using retrievals from cloud edge lidar profiles? If so, are above- or below-cloud N_{CCN} more strongly related to cloud top microphysical properties?

3.2 Data & Methods

The NASA ORACLES campaign was comprised of three deployments in September 2016, August 2017, and October 2018. Deployments were based out of Walvis Bay, Namibia, in September 2016 and São Tomé and Príncipe in August 2017 and October 2018, and observations were made using a combination of remote sensing and in situ instruments located on the NASA P-3 (2016-2018) and the ER-2 (2016 only) aircraft. In this study, results are based on N_{CCN} and cloud microphysical properties retrieved by remote sensing instrumentation. However, we also use two in situ data sets to provide context and prerequisite information for the analysis. Therefore, the remainder of this section is organized as follows. The primary remote sensing-based data sets used in this study are described in Sect. 3.2.1, in situ data sets are described in Sect. 3.2.2, and all data sets are summarized in Table 3.1. Our method for calculating N_{CCN} autocorrelation and using the results to inform data collocation, in addition to the calculation of LTS, are given in Sect. 3.2.3.

Instrument	Variables	Resolution (temporal / vertical)	Uncertainty	Measurement Type	Aircraft
High Spectral Resolution Lidar 2 (HSRL-2)	ML- retrieved CCN con- centration at super- saturation ($S = 0.4\%$) (cm^{-3}), Cloud top height (CTH; m)	10 s / 15 m	ML-CCN: Within 15%	Remote sensing	ER-2 (2016), P-3 (2017– 2018)
Research Scanning Polarimeter (RSP)	Cloud droplet effective radius (R_{eff} ; μm), Cloud optical thickness (COT)	10 s / 200 m	10% for both	Remote sensing	ER-2 (2016), P-3 (2017– 2018)
Cloud con- densation nuclei (CCN) counter (DMT CCN-100)	CCN number concentra- tion at different supersatu- rations (cm^{-3})	1 s	10%	In situ	P-3
CAS/CDP	Cloud droplet number concentra- tion (cm^{-3})	1 s	20%	In situ	P-3

Continued on next page

Instrument	Variables	Resolution (temporal / vertical)	Uncertainty	Measurement Type	Aircraft
King hot wire	Bulk liquid water content (g/m ³)	1 s	5%	In situ	P-3

Table 3.1: List of instruments and data sets used in this study, including their respective resolution, measurement type, and aircraft location.

3.2.1 Remote sensing-based data sets

3.2.1.1 ML-CCN retrievals from HSRL-2

Redemann and Gao (2024) recently developed a machine learning (ML) methodology for retrieving N_{CCN} (ML-CCN) from lidar observables, a method which has significant implications for future spaceborne retrievals of N_{CCN} from systems such as the Atmospheric Lidar (ATLID) on the EarthCARE satellite (Wehr et al., 2023). The ML algorithm was trained using HSRL-2 observables, reanalysis data (temperature and relative humidity), and in situ N_{CCN} from several recent suborbital field campaigns and can predict N_{CCN} with mean relative errors (MRE) of about 15%. In addition to creating a larger spatial coverage of N_{CCN} data than is available from the in situ measurements, another benefit is the retrieval of the vertical distribution of N_{CCN} possible with this method. This allows us to assess the impact of above- and below-cloud N_{CCN} simultaneously from lidar profiles adjacent to cloud edges (Sect. 3.2.3). Additionally, since relative humidity (RH) is one of the predictors used to train the model, the effect of aerosol swelling at high RH on lidar observables is considered to predict N_{CCN} under dry conditions (Redemann and Gao, 2024). While Redemann and Gao (2024) use the full suite of HSRL-2 observables to predict N_{CCN} , this study uses the same

methodology to predict N_{CCN} at a supersaturation of 0.4% with a model that uses backscatter coefficients at 532 and 1064 nm and depolarization ratio at 532 and 1064 nm as input. This combination of observables was chosen to maximize the number of available retrievals, as extinction coefficients and all observables at 355 nm are less frequently available at below-cloud altitudes in the ORACLES observations. Compared to the model trained with the full set of HSRL-2 observables (15% MRE), this adjusted model has an MRE of 19.9%. This adjustment to improve data availability only results in a modest increase in the MRE of the N_{CCN} prediction.

3.2.1.2 Research Scanning Polarimeter

Retrievals of cloud top microphysical properties in this study come from the Research Scanning Polarimeter (RSP), which measures total and polarized reflectance at nine spectral channels (Cairns et al., 1999; Alexandrov et al., 2012a). The RSP is generally oriented to scan along the aircraft track at 0.8° intervals, such that observational data from each scan contains about 150 instantaneous Earth viewing measurements (Alexandrov et al., 2012a). R_{eff} retrievals use polarized reflectance in the scattering angle range between 135° and 165° , where the rain- or cloud-bow structure is observed. Since the rainbow is created by single scattering of light by cloud droplets, the structure is characteristic of droplet sizes at cloud top and within a unit optical depth into the cloud layer, or approximately 50 m (Alexandrov et al., 2018). The RSP R_{eff} retrieval is insensitive to spatial inhomogeneities and three-dimensional radiative transfer effects (Alexandrov et al., 2012b), a significant advantage over satellite retrievals, such as those from MODIS, which tend to underestimate COT and R_{eff} in cases with above-cloud absorbing aerosols if not accounted for in the algorithm (Meyer et al., 2013, 2025). The RSP also retrieves cloud optical thickness (COT) using 865 nm unpolarized observations with a 1D radiative transfer model and constraining results

using the polarimetric R_{eff} (Alexandrov et al., 2012a; Painemal et al., 2025). While R_{eff} and COT are retrieved quantities and subject to retrieval assumptions and radiative sensitivities, RSP retrievals have been compared and evaluated against in situ observations, demonstrating their accuracy and potential for satellite-based retrievals of this kind, with R_{eff} agreements generally better than $1 \mu\text{m}$ and effective variance (V_{eff}) agreements better than 0.02 (Alexandrov et al., 2018; Painemal et al., 2021; Fu et al., 2022; Painemal et al., 2025).

From RSP retrievals of R_{eff} and cloud COT, we also calculate N_d and LWP. N_d is calculated using the method from Painemal and Zuidema (2011) given in Eq. 3.1:

$$N_d = 1.4067 \times 10^{-6} \frac{\text{COT}^{1/2}}{R_{eff}^{5/2}}, \quad (3.1)$$

and LWP is calculated using the method from Wood and Bretherton (2006), as given in Eq. 3.2:

$$LWP = \frac{5}{9} \rho_w \text{COT} R_{eff}, \quad (3.2)$$

where ρ_w is the density of water. The calculation of N_d assumes a value of $2.0 \text{ g/m}^3/\text{km}$ for the condensation rate of water vapor with height, which is an average based on observations from the Southeast Pacific of offshore clouds with nearly adiabatic LWC profiles and coastal clouds with decreased LWC at cloud top due to increased entrainment, and thus less adiabatic observed profiles (Painemal and Zuidema, 2010). The relative uncertainties for R_{eff} (Alexandrov et al., 2012a,b) and COT (Nakajima and King, 1990) are both assumed to be 10%, and these values are used in Gaussian error propagation calculations to calculate uncertainties for N_d and LWP, as given in Eqs. 3.3 and 3.4, respectively:

$$\delta(N_d) = N_d \left[\frac{1}{4} \left(\frac{\delta(COT)}{COT} \right)^2 + \frac{25}{4} \left(\frac{\delta(R_{eff})}{R_{eff}} \right)^2 \right]^{1/2}, \quad (3.3)$$

$$\delta(LWP) = LWP \left[\left(\frac{\delta(COT)}{COT} \right)^2 + \left(\frac{\delta(R_{eff})}{R_{eff}} \right)^2 \right]^{1/2}, \quad (3.4)$$

where δ represents the error of each variable.

3.2.2 In situ data sets

3.2.2.1 CCN Counter

The Georgia Institute of Technology (GIT) Droplet Measurement Technologies (DMT) CCN counter (CCN-100) measures in situ N_{CCN} at various water vapor supersaturation (Kacarab et al., 2020), which for ORACLES ranged between 0.1% and 0.4% (Redemann et al., 2021). The instrument is designed as a continuous-flow streamwise thermal-gradient chamber, where the continuous transport of heat and water vapor from wetted walls is subject to a temperature gradient, which generates quasi-uniform supersaturation in the center of a cylindrical flow chamber (Roberts and Nenes, 2005). Aerosols that activate into droplets with a radius greater than $0.5 \mu\text{m}$ at the end of the chamber are counted as CCN. The continuous-flow feature allows for quick (1 Hz) sampling, which is important for airborne sampling that often encounters rapidly changing environments (Roberts and Nenes, 2005). N_{CCN} uncertainty is reported as $\pm 10\%$ at high signal-to-noise ratios, while supersaturation uncertainty is given as $\pm 0.04\%$ (Rose et al., 2008). In this study, we use in situ-measured CCN for an autocorrelation analysis to assess over what distances we may reasonably extrapolate cloud edge N_{CCN} values further towards cloud center from the edge (Sect. 3.2.3-3.2.4).

3.2.2.2 In situ cloud probes and cloud base height estimates

To estimate cloud base height, we follow the methodology of Gupta et al. (2021) who derived a relationship between in situ measured cloud top height (Z_T) and cloud base height (Z_B) for six research flights in the 2016 deployment based on the highest and lowest altitudes at which in situ Nd is greater than 10 cm^{-3} and bulk liquid water content (LWC) is greater than 0.05 g/m^3 within sawtooth profiles. We follow their methodology using observations from cloud probes located on the P-3 aircraft during all three ORACLES deployments. Using the resultant statistical relationships between Z_B and Z_T , we estimate cloud base from HSRL-2 cloud top heights (CTH). This method and the resulting cloud base height distribution are described in Section 3.6.

3.2.3 Collocation methodology and calculations

3.2.3.1 Horizontal N_{CCN} autocorrelation

As will be described in more detail in Sect. 3.3.2, we leverage the vertical resolution of the ML-CCN retrievals to assess the simultaneous impact of both above- and below-cloud N_{CCN} on cloud top microphysical properties by sub-selecting clear-sky profiles that are directly adjacent to profiles that detect a cloud. We assume that N_{CCN} retrieved from the clear profile is approximately equal to N_{CCN} observed below the adjacent cloud. To test the validity of this assumption, we perform an in situ-based autocorrelation analysis to assess the self-consistency of N_{CCN} observed below cloud base heights.

Use of the autocorrelation metric for assessing variability of aerosol properties across various spatial scales has been documented by studies such as Anderson et al. (2003); Heintzenberg et al. (2004); Redemann et al. (2006); Shinozuka and Redemann (2011); Chau et al. (2021); LeBlanc et al. (2022); Perkins et al. (2022) and Franco et al. (2024).

Here, we focus on the horizontal variability of N_{CCN} isolated from any effects of vertical variability since we are interested in extrapolating below-cloud N_{CCN} at a constant altitude from the nearest clear-sky profile. Therefore, the in situ data used to calculate autocorrelation come from constant altitude flight paths where observations do not vary more than ± 25 m in altitude from the first measurement and where data are collected for at least 30 seconds. This minimum time criterion is required to eliminate flight segments where the aircraft may have been slowly ascending or descending through an aerosol layer. Constant altitude flights where the aircraft turns and re-samples an area are split into separate segments. To display autocorrelation as a function of horizontal distance, we convert the original temporal increments to distance using an estimated P-3 aircraft speed of 150 m/s.

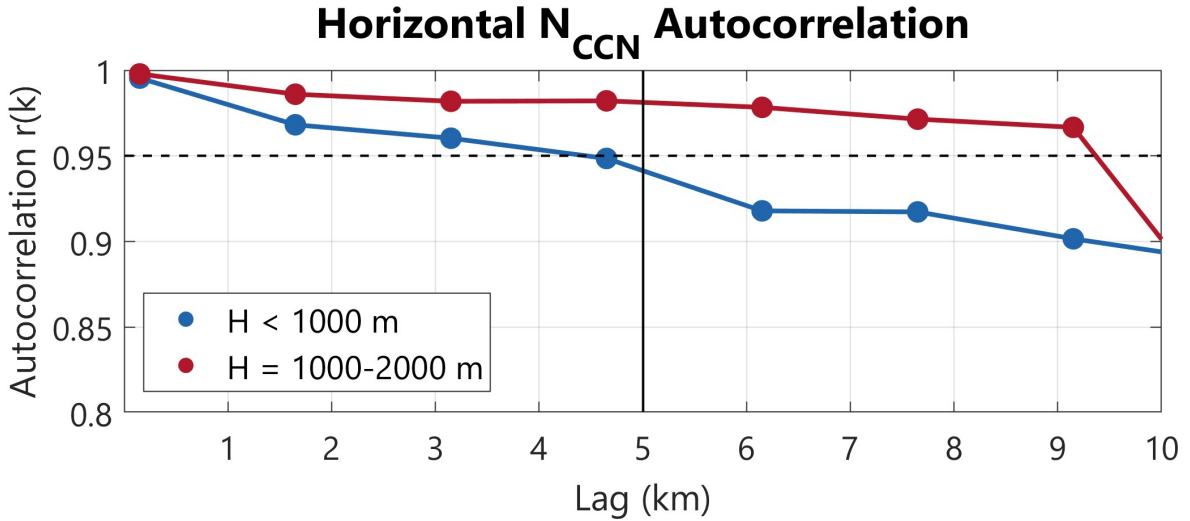


Figure 3.1: The autocorrelation coefficient of in situ N_{CCN} observed in horizontal flight legs is given as a function of lag distance. The dark blue line represents observations from flight legs at $H < 1000$ m, and the red line represents observations from flight legs between $H = 1000$ - 2000 m. The black dashed line depicts an autocorrelation coefficient of 0.95, and the solid vertical line depicts the 5 km over which we assume that below-cloud N_{CCN} is approximately constant.

Data from all constant altitude flight legs are combined and autocorrelation coefficients are calculated at 10 second lag increments (k) between all data pairs y_j and

y_{j+k} using Eq. 3.5:

$$r(k) = \frac{\sum_j^N (y_j - \bar{y}_{+k})(y_{j+k} - \bar{y}_{-k})}{(N-1)\sigma_{+k}\sigma_{-k}}, \quad (3.5)$$

where $r(k)$ is the autocorrelation coefficient at a given lag, N indicates the number of lagged pairs, y is N_{CCN} , $\bar{y}_{+k}$ and σ_{+k} represent the mean and standard deviation, respectively, of data points located $+k$ away from another data point, and $\bar{y}_{-k}$ and σ_{-k} are the corresponding quantities for data points located $-k$ away from another data point (Anderson et al., 2003).

Since we are interested in the self-consistency of N_{CCN} below cloud base, we calculate autocorrelation for constant altitude flight legs flown at altitudes below 1000 m to capture the range of most cloud base heights observed in this analysis (Fig. 3.12b). We limit in situ N_{CCN} to those with a supersaturation between 0.2-0.4%. This range captures the supersaturation at which most in situ N_{CCN} observations were made and eliminates significant variation due to changes in supersaturation. Autocorrelation results for all three deployments are given in Fig. 3.1. We find that assuming that N_{CCN} retrieved from a clear-sky lidar profile is approximately equal to N_{CCN} below cloud base for the adjacent cloudy profile is reasonable, considering the horizontal resolution of these HSRL-2 based profiles is 2 km, at which the autocorrelation coefficient is around 0.96. Additionally, we find that N_{CCN} autocorrelation remains close to 0.95, as shown by the dashed line, over a horizontal distance of approximately 5 km.

We also examine N_{CCN} variability above cloud by calculating autocorrelation for constant altitude flight legs flown at altitudes between 1000-2000 m to capture a broad above-cloud range that does not overlap with most cloud base height altitudes (Fig. 3.12b). We find that over a horizontal range of 5 km the above-cloud autocorrelation coefficient is slightly higher than for below-cloud observations, with it remaining ap-

proximately constant around 0.97. Since autocorrelation patterns are similar for above- and below-cloud N_{CCN} , this suggests that across a horizontal range of 5 km, possible impacts of entrainment of BBA into the BL on below-cloud N_{CCN} homogeneity are reflected in the below-cloud N_{CCN} analysis. That is, we do not see rapid decreases in above-cloud N_{CCN} autocorrelation indicative of entrainment mixing that are not reflected in the below-cloud N_{CCN} autocorrelation trends. Therefore, it appears that the BL was often well-mixed and N_{CCN} did not vary significantly across short distances.

This information will be used when collocating both remote sensing-based data sets by assuming that below-cloud N_{CCN} is constant over 5 km (Sect. 3.2.3.2) to increase the number of data points in the analysis. However, this assumption comes with a few caveats. First, the constant altitude flight legs below 1000 m include both clear-sky and below-cloud observations. In a separate analysis, we distinguished clear-sky observations from below-cloud observations using changes in downward solar radiation and found that effects of cloud processing result in decreased N_{CCN} autocorrelation in below-cloud observations when compared to clear-sky observations. However, the autocorrelation coefficient remained, on average, above 0.9 at lags between 0 and 5 km (not shown), suggesting reasonable correlation for both below-cloud and clear-sky observations. While not a focus of this study, we plan to expand on these differences between clear-sky and below-cloud N_{CCN} autocorrelation in a future analysis. Additionally, flight segments observed below-cloud assume that the cloud was non-precipitating. Therefore, the precipitation sink of N_{CCN} below-cloud is not accounted for here and we would expect below-cloud N_{CCN} autocorrelation to be lower if it were included. However, LWP in the following analyses is limited to 80 g/m², and for analyses where we consider below-cloud N_{CCN} it generally falls below 40 g/m². Therefore, it is unlikely that precipitation is a major below-cloud N_{CCN} sink within 5 km of cloud edge in these cases. These caveats should be considered when interpreting results involving below-

cloud N_{CCN} , but we consider the autocorrelation trends in Fig. 3.1 to be a general approximation of below-cloud N_{CCN} homogeneity across all ORACLES observations and contend that limiting the constant N_{CCN} assumption to 5 km is reasonable and unlikely to introduce significant errors.

3.2.3.2 Data collocation and case classifications

In this study, we define three main subsets of data, all requiring that the aircraft was flying above 2500 m and that cloud top heights fall below 2000 m to exclude potential mid- or high-level clouds. The collocation process for these subsets is shown in Fig. 3.2. To assess the simultaneous impact of above- and below-cloud N_{CCN} on cloud top microphysical properties, we define a subset of the data as cloud edge (CE) cases. These cases are found by first identifying clear-sky, cloud-adjacent ML-CCN profiles. That is, we find clear-sky profiles such as profile 1 in Fig. 3.2 that are adjacent (2 km) to a profile where the HSRL-2 detects a cloud. We average the N_{CCN} retrieved from profile 1 within a 500 m layer below the cloud base height determined by profile 2 and thereby determine below-cloud N_{CCN} . We use a 500 m layer to represent overall BL conditions in the below-cloud region. We find coincident RSP retrievals within 10 seconds of the profile 2 measurement, and these are also averaged, resulting in a 2 km RSP average corresponding to the 2 km horizontal resolution of the ML-CCN profile. Additionally, we define above-cloud N_{CCN} as an average of the ML-CCN in profile 2 within 100 m of the cloud top, following the vertical spacing criteria used by Gupta et al. (2021). Therefore, the below-cloud N_{CCN} retrieved from profile 1 is then paired with the cloud top RSP average and above-cloud N_{CCN} associated with profile 2. Additionally, since the in situ autocorrelation analysis indicates that within the BL N_{CCN} maintains an autocorrelation coefficient greater than 0.95 over a horizontal range of 5 km, and thus should remain approximately constant, we extend the same below-cloud N_{CCN} value

from profile 1 to additional profiles within 5 km. That is, the below-cloud N_{CCN} value from profile 1 is applied to profiles 2-4, and this approach allows us to approximately triple the number of CE cases in the remainder of the analysis. While profiles 2-4 have the same below-cloud N_{CCN} , the above-cloud N_{CCN} is determined independently for each profile using observations made within 100 m of cloud top height.

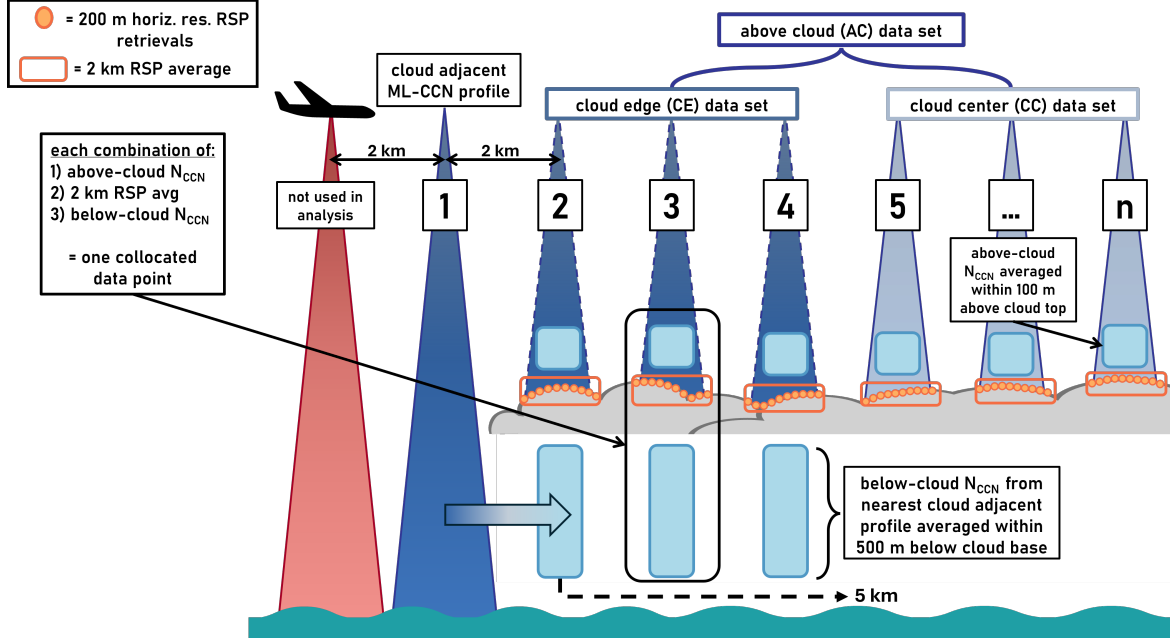


Figure 3.2: Collocation schematic depicting above cloud (AC), cloud edge (CE), and cloud center (CC) cases and the collocation of ML-CCN profiles with RSP retrievals. Profiles 2 through n are included in the AC data set (Sect. 3.3 and 3.3.1). Profile 1 is the clear-sky, cloud-adjacent ML-CCN profile used to determine below-cloud N_{CCN} for adjacent cloudy profiles. Profiles 2-4 are cloudy ML-CCN profiles within 5 km of a cloud edge for which above-cloud N_{CCN} is determined from the layer 100 m above cloud top and below-cloud N_{CCN} is extrapolated from profile 1, making up the CE cases (Sect. 3.3.2). Profiles 5 through n are not within 5 km of a cloud edge and thus are designated as CC cases (Sect. 3.3.3). Profile n represents the variable nature of this collocation process, that is that individual cloud transects have differing numbers of observations that fall within the AC and CC cases. The orange outlined circles represent each 200 m horizontal resolution RSP retrieval, and the orange boxes around them represent 2 km averages to correspond to the 2 km horizontal resolution of the ML-CCN profiles. The red profile represents a ML-CCN profile not adjacent (2 km) to a cloud.

For ML-CCN profiles further than 5 km from cloud edge that are associated with an HSRL-2 detected cloud top and have coincident RSP retrievals within 10 seconds, we pair 2 km RSP averages with above-cloud N_{CCN} and call these cloud center (CC) cases, which are represented by profiles 5 through n in Fig. 3.2, representing the variable nature of the number of cases that may fall in this data set depending on the extent of any given cloud transect. Such cases are only used to assess impacts of above-cloud N_{CCN} , as we do not pair any below-cloud N_{CCN} value with profiles further than 5 km from cloud edge. For analyses in Sect. 3.3.1 where we focus on relationships between cloud top microphysical properties and above-cloud N_{CCN} , the CE and CC cases are combined into a subset called the above cloud (AC) data set. In Sect. 3.3.2 we compare the impact of above- and below-cloud N_{CCN} , meaning that only data from the CE data set is considered. In Sect. 3.3.3 we investigate the dependence of ACI on proximity to cloud edge by comparing above-cloud observations from CE and CC data sets separately.

In summary, for the example given in Fig. 3.2, the paired above-cloud N_{CCN} and RSP measurements from profiles 2 through n would all be included in the AC data set. The above-cloud N_{CCN} , RSP measurements, and below-cloud N_{CCN} in profiles 2-4 are included in the CE data set. Profiles 5 through n do not fall within 5 km of a cloud edge and therefore are included in the CC data set but not the CE data set. Note that these subsets represent a combination of observations from all cloud transects across all three ORACLES deployments and are not in any way grouped by individual cloud profiles. After applying this collocation methodology to observations from all three ORACLES deployments, the location of all cases is given in Fig. 3.3. Here we find that CE cases are spatially as well-distributed as AC cases. That is, the locations of both cases are very similar, and while the number of CE cases is smaller, they are not limited to any certain geographic area that is not also represented in the AC data set.

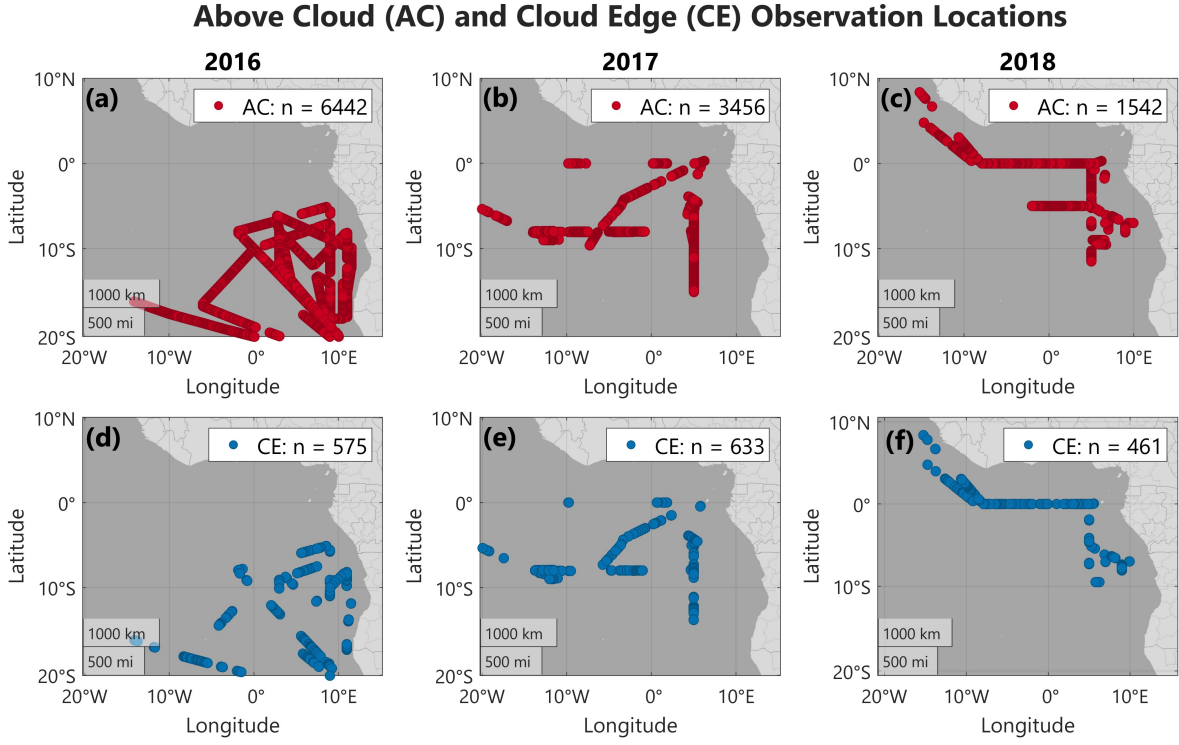


Figure 3.3: Map depicting the location of (a-c) AC and (d-f) CE cases for 2016 (a, d), 2017 (b, e), and 2018 (c-f) deployments. The number of cases is given in the legend.

3.2.3.3 Lower tropospheric stability calculations

To constrain and assess the impact of environmental stability on ACI, we calculate lower tropospheric stability (LTS) using temperature profiles from Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA-2) data that have been interpolated to HSRL-2 spatiotemporal resolution (Global Modeling And Assimilation Office and Pawson, 2015). LTS is calculated using Eq. 3.6:

$$LTS = \theta_{800} - \theta_{1000}, \quad (3.6)$$

where θ is potential temperature at 800 hPa and 1000 hPa. Though LTS is often calculated using potential temperature at 700 hPa, we use 800 hPa to focus on the BL portion relevant for these low clouds and to avoid the aerosol layer around 700 hPa

(Adebiyi et al., 2015; Ryoo et al., 2021). While several approaches have been used to constrain meteorology in previous ACI studies, LTS is a commonly used metric to constrain the impact of stability on clouds (Matsui et al., 2006; Mauger and Norris, 2007; Gryspeerd et al., 2014; Zhang et al., 2016; Jia et al., 2019; Murray-Watson and Gryspeerd, 2022; Zhao et al., 2025).

3.3 Results

We start by investigating the relationships between above-cloud N_{CCN} , R_{eff} , N_d , and LWP for AC cases across all three ORACLES deployments (Fig. 3.4). For this analysis we remove data from any cloudy profiles that had a clear-sky profile on both sides. Such broken cloud regimes are more likely to be impacted by entrainment mixing and evaporation, and here our focus is on determining aerosol effects on cloud properties. This step reduces the AC data set from 11440 to 11231 profiles. Additionally, we limit observations to those with $LWP \leq 80 \text{ g/m}^2$. This range accounts for most of the ORACLES low cloud observations, limits scatter from a small number of higher LWP values likely formed under different meteorological conditions, and reduces the number of precipitating clouds. This step reduces the AC data set from 11231 to 9223 profiles.

For Fig. 3.4 and those like it in the remainder of Sect. 3.3, shading on the figures show probability distributions of where individual data points fall, while the scatter points represent the median of each cloud property within ten N_{CCN} bins determined such that each bin contains an equal number of data points. The error bars give the median uncertainty of each cloud property within that bin. We calculate ACI metrics to quantify the R_{eff} (ACI_{REF}) and N_d (ACI_{CDNC}) response to increasing N_{CCN} following the method described in McComiskey et al. (2009), given in Eqs. 3.7 and 3.8:

$$ACI_{REFF} = - \left. \frac{\partial \ln(R_{eff})}{\partial \ln(N_{CCN})} \right|_{LWP}, \quad (3.7)$$

$$ACI_{CDNC} = \frac{d \ln(N_d)}{d \ln(N_{CCN})}. \quad (3.8)$$

Here, Eq. 3.7 describes the fractional change in cloud droplet R_{eff} in response to fractional changes in N_{CCN} at a constant LWP and Eq. 3.8 captures the fractional change in cloud droplet N_d when there is a fractional change in N_{CCN} . Such ACI metrics describe microphysical responses and are critical components in cloud radiative forcing calculations (McComiskey et al., 2009; Ghan et al., 2016). ACI_{REFF} and ACI_{CDNC} correspond to the slope of linear regressions calculated for log-transformed R_{eff} , N_d , and N_{CCN} , and here these regressions are calculated for all data points (not the bin averages). The ACI_{REFF} calculation is done within 20 g/m² LWP bins, and the final value is a sample size-weighted average of the individual values calculated for each small LWP range. The error associated with these metrics are calculated as the standard error (SE) of the slopes via Eq. 3.9:

$$SE = \sqrt{\frac{\sum_{i=1}^n (y_i - \hat{y}_i)^2 (n-2)}{\sum_{i=1}^n (x_i - \hat{x}_i)^2}}. \quad (3.9)$$

where x represents $\log(N_{CCN})$ and y represents the logarithm of each respective cloud property. These linear regressions calculated for log-transformed data are also used to plot the corresponding fit lines for R_{eff} and N_d relationships by transforming the fit equation back into linear space. These fit lines correspond to the full set of data represented by the probability distributions, not the individual bin averages.

From the combination of data from all three ORACLES deployments, we see clear signals of an above-cloud aerosol influence on the underlying cloud top microphysical properties. As N_{CCN} within the 100 m layer above cloud top increases, there is a de-

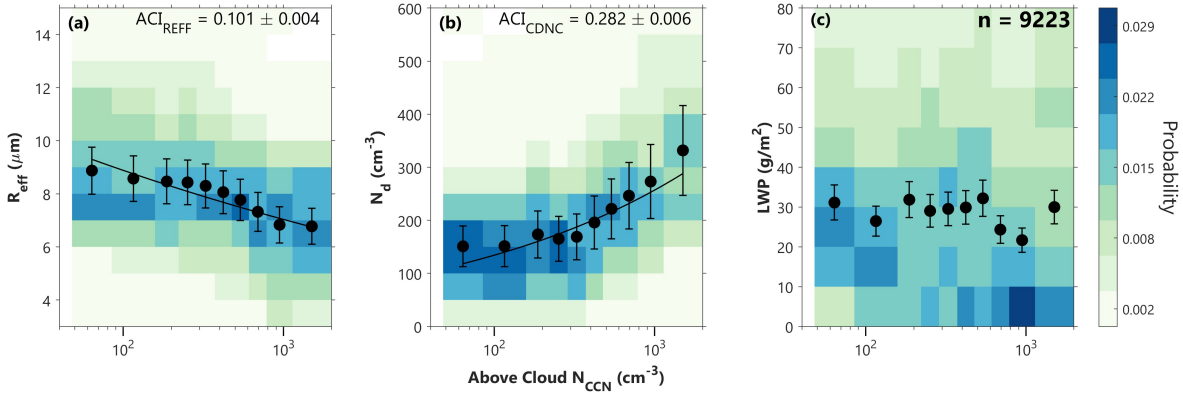


Figure 3.4: Relationship between above-cloud N_{CCN} and (a) R_{eff} , (b), N_d , and (c) LWP across all three ORACLES deployments. Shading in the background represents the probability of where the 9223 individual data points from this AC data set fall within each panel. Data are separated into 10 bins such that each bin contains an equal number of data points. Each scatter point represents the median cloud property within each individual N_{CCN} bin. The error bars represent the median uncertainty of the respective cloud property within that bin. ACI metrics and their SE ($\pm$) are given for R_{eff} and N_d . The fit lines correspond to the slope determined by each ACI metric.

creasing trend in R_{eff} (Fig. 3.4a) and an increasing trend in N_d (Fig. 3.4b). ACI_{REFF} is 0.101 and ACI_{CDNC} is 0.282, where the former falls within the 0.04-0.17 range observed for continental stratus clouds by Kim et al. (2008) and the latter falls towards the lower end of the 0.18-0.69 range given for stratocumulus clouds by McComiskey et al. (2009). While LWP is limited to 0-80 g/m², we still see some fluctuations in the LWP with changes in N_{CCN} (Fig. 3.4c), which is an inherent constraint when using observed data. This analysis also allows us to characterize the average properties of low-level stratocumulus clouds in the SEA, as we see that most clouds have R_{eff} values between approximately 6-10 μ m, N_d values between 50-400 cm⁻³, and an LWP between 10-40 g/m².

3.3.1 ACI sensitivity to environmental stability

In this study, we use a k-means clustering approach (Anderberg, 1973) to separate observations of aerosol and cloud properties into different regimes, as has been done in

previous studies (Gryspeerdt and Stier, 2012; Gryspeerdt et al., 2014; Di Bernardino et al., 2022). This methodology was tested with several combinations of variables including HSRL-2 CTH, RSP-retrieved COT, R_{eff} , N_d , LWP, and MERRA-2 calculated LTS and EIS. All variables were range-standardized before clustering (Milligan and Cooper, 1988), and standard Euclidean distance metrics are used. Various combinations of these variables were compared using Calinski-Harabasz criterion values to determine which combination resulted in the highest between-cluster variance to within-cluster variance ratio (Calinski and Harabasz, 1974). We determined the final number of clusters by maximizing climatological and geographic distinctions among clusters while preserving adequate sample sizes within each to perform statistically meaningful ACI analyses. These testing steps showed that the clusters determined using only LTS had the highest Calinski-Harabasz criterion values, and we determined that using four clusters resulted in adequate sample sizes between clusters for further analysis.

Previous studies have determined that atmospheric stability is often correlated with stratiform cloud amount (Klein and Hartmann, 1993), and LTS specifically has been used to differentiate between low cloud types in subsidence regimes, such as the SEA (Zhang et al., 2016). Additionally, Painemal et al. (2014) found stability to be a strong control on cloud microphysical properties over the SEA, where R_{eff} and LWP both decrease with LTS for observations north of 5°S, a pattern that is not observed south of 5°S where LTS values are higher. Over the Eastern China Ocean, Zhao et al. (2025) recently found that in high LTS environments, weak entrainment was observed, thus suppressing the negative effects of entrainment observed in environments with low to moderate LTS. While EIS has been suggested to be a better, regime independent predictor of stratus cloud amount than LTS (Wood and Bretherton, 2006), our analysis indicated that using LTS for k-means clustering resulted in higher Calinski-Harabasz

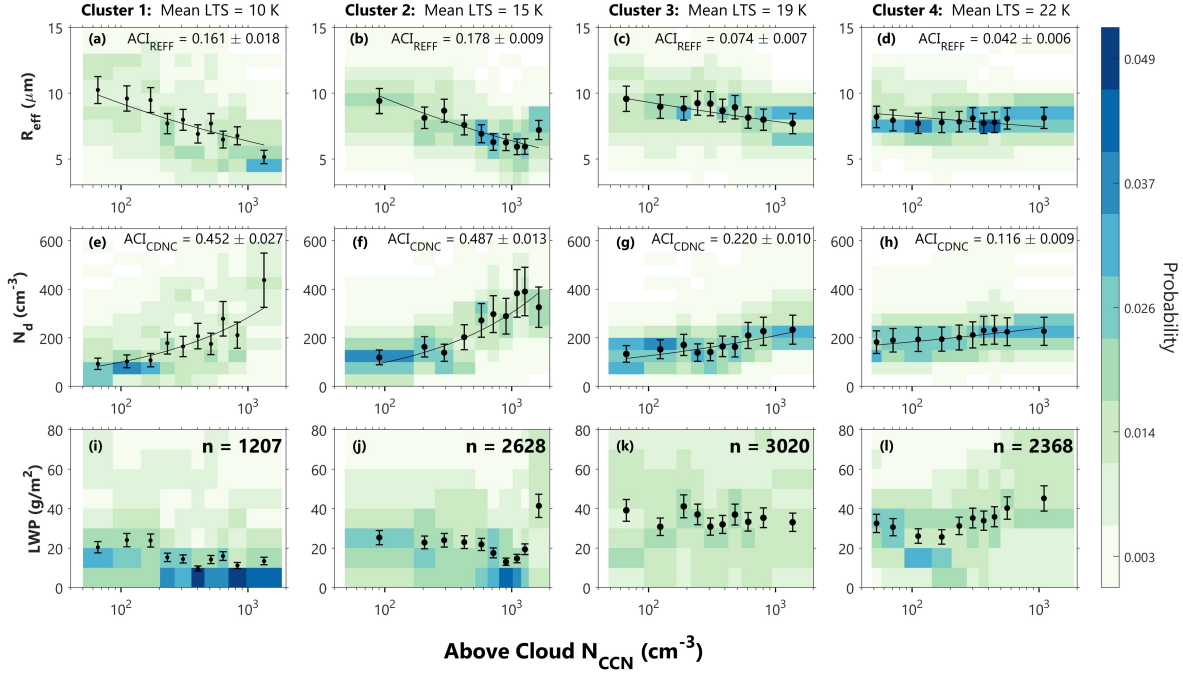


Figure 3.5: Relationship between above-cloud N_{CCN} and (a-d) R_{eff} , (e-h) N_d , and (i-l) LWP across LTS clusters determined using k-means clustering. The shading, scatter points, ACI metrics, and fit lines are calculated and displayed in the same way as Fig. 3.4.

criterion values. This could be due to increased uncertainty in the MERRA-2 relative humidity profiles used in the EIS calculation.

This k-means clustering analysis is applied to the same AC data set as shown in Fig. 3.4, with results in Fig. 3.5 given as a function of LTS. Mean LTS ranges from 10 K in cluster 1 to 22 K in cluster 4. As LTS increases, cloud sensitivity to increasing above-cloud N_{CCN} generally decreases, seen as the fit lines become more flat, though ACI metrics increase slightly from cluster 1 to cluster 2 before proceeding to decrease for clusters 3 and 4. This suggests that less stable environments experience increased entrainment mixing, where smoke aerosols located above cloud top are entrained into the cloud and nucleate as cloud droplets. LWP decreases slightly with increasing above-cloud N_{CCN} for lowest LTS (cluster 1) and increases with increasing above-cloud N_{CCN} for highest LTS (cluster 4), while clusters 2 and 3 show mostly constant LWP with an

increase for the highest N_{CCN} bin in cluster 2. Most observations in cluster 1 are in the northwestern part of the ORACLES region, and observations in cluster 4 are in the southeastern part of the region, closer to the coast (Fig. 3.6). This corresponds well with average sea surface temperatures (SST) in the SEA, where values generally decrease from northwest to southeast (Ryoo et al., 2021).

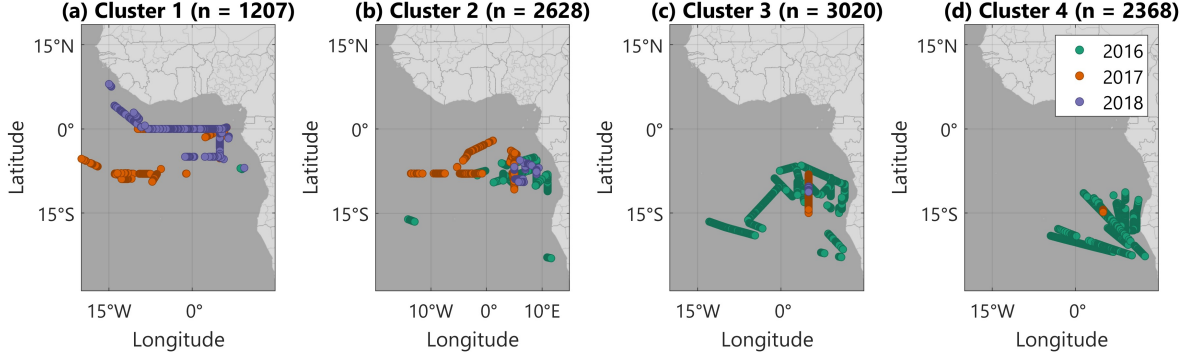


Figure 3.6: Map showing location of observations in (a) cluster 1, (b) cluster 2, (c) cluster 3, and (d) cluster 4. Average LTS in each cluster increases from left to right. The color of each point corresponds to the deployment year of the observation.

Lastly, we show mean values of cloud properties and above-cloud N_{CCN} within each cluster in Table 2. As LTS increases from cluster 1 to 4, average CTH decreases, indicating that a more stable BL is associated with stronger inversions that restrict cloud vertical development (Costantino and Bréon, 2013). Additionally, cluster 2, where the most dense part of the smoke plume was often observed in 2017, has the highest average N_d , smallest average R_{eff} , and highest average above-cloud N_{CCN} , consistent with in situ analyses (Gupta et al., 2022b). Cluster 3 has the lowest average N_d , highest average R_{eff} , and highest average LWP, with values similar to cluster 4. Variability in above-cloud N_{CCN} is high for each cluster, and findings from Lenhardt et al. (2023) suggest that CCN observed across all three campaigns represent a wide range of smoke ages and a small range of aerosol sizes.

	Cluster 1 ($n = 1207$)	Cluster 2 ($n = 2628$)	Cluster 3 ($n = 3020$)	Cluster 4 ($n = 2368$)
LTS (K)	10.3 ± 1.3	14.9 ± 1.1	18.5 ± 1.1	22.4 ± 1.3
R_{eff} (μm)	8.3 ± 3.5	7.5 ± 2.4	8.8 ± 2.2	8.0 ± 1.5
N_d (cm^{-3})	245.4 ± 214.2	273.8 ± 167.4	184.1 ± 91.0	218.2 ± 90.2
LWP (g m^{-2})	24.0 ± 21.0	26.2 ± 19.8	36.9 ± 21.4	34.6 ± 19.1
COT	4.8 ± 3.7	5.9 ± 3.9	7.2 ± 3.6	7.4 ± 3.6
CTH (m)	1507.7 ± 387.4	1091.4 ± 249.1	1042.4 ± 171.3	715.0 ± 197.5
Above-Cloud N_{CCN} (cm^{-3})	454.1 ± 363.3	718.6 ± 465.9	458.8 ± 387.7	315.1 ± 242.0

Table 3.2: Mean $\pm$ standard deviation of cloud and environmental properties for each cluster.

3.3.2 ACI sensitivity to above- and below-cloud N_{CCN}

Next, we use the CE data set to investigate the relationships between above- and below-cloud N_{CCN} and R_{eff} , N_d , and LWP. Here, we again remove cases of cloudy profiles that had a clear profile on both sides to avoid conflating the aerosol effect with effects due to lateral entrainment of ambient air into the cloud layer. This step reduces the CE data set from 1669 to 1269 profiles. Additionally, limiting observations to those with $\text{LWP} \leq 80 \text{ g/m}^2$ reduces the CE data set from 1269 to 1249 profiles. Limiting cases to those observed at cloud edge inherently constrains cloud properties, which will be further discussed in Sect. 3.3.3.

The above-cloud relationships at cloud edge are similar to what was seen from the full AC data set in Fig. 4 and 5. In comparison, the below-cloud N_{CCN} analyses show a weaker relationship to the cloud top microphysical properties, with ACI metrics that are smaller in magnitude compared to the above-cloud metrics. The range of average N_{CCN} in the 500 m below-cloud is of similar magnitude as N_{CCN} in 100 m above cloud, with fewer clean cases below-cloud than above cloud, which is likely evidence of entrainment mixing of BBA from the smoke plume into the BL. Additionally, one

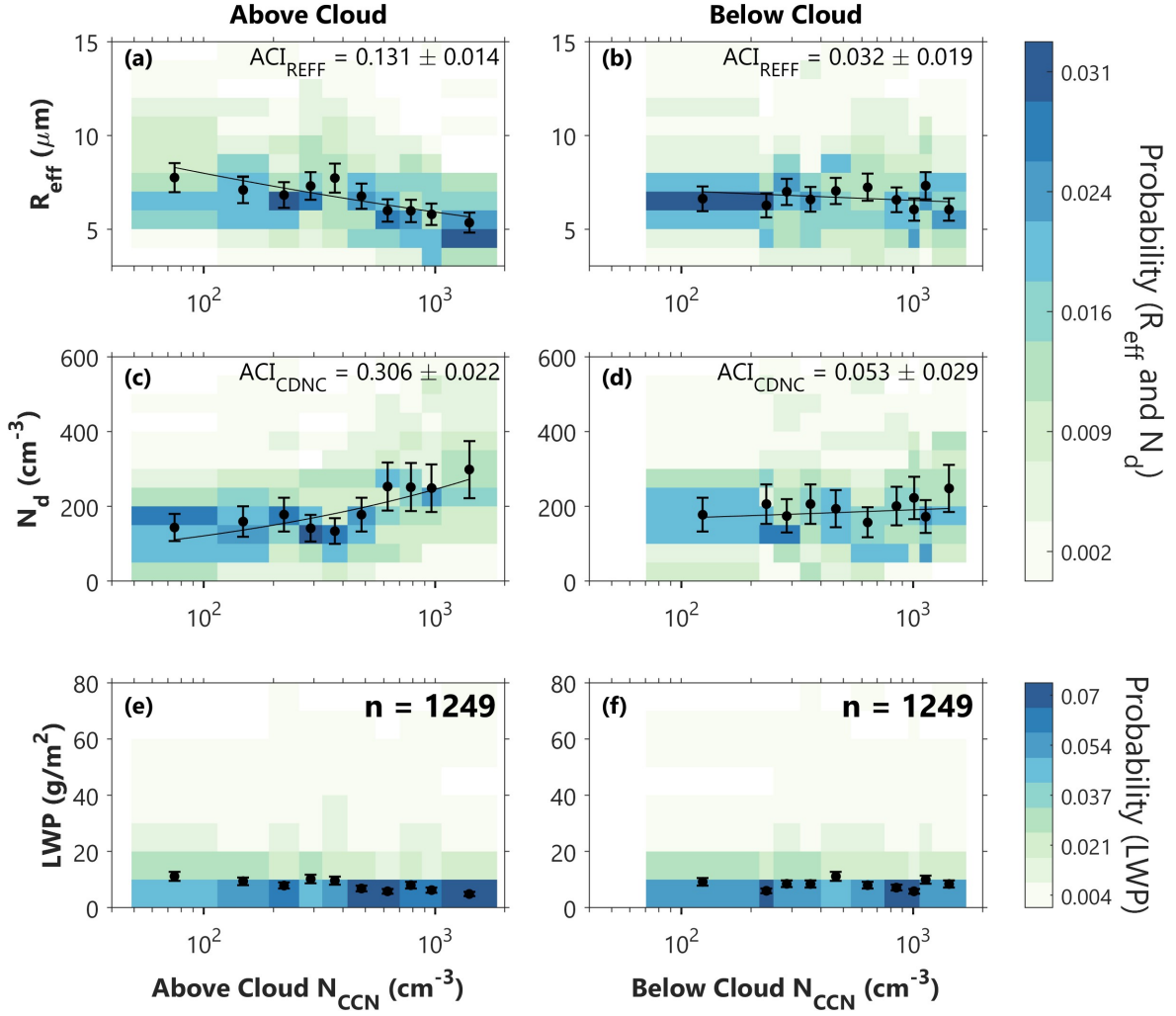


Figure 3.7: Results of from CE data set ($n = 1249$) show the relationship between (a, c, e) above- and (b, d, f) below-cloud N_{CCN} and (a-b) R_{eff} , (c-d), N_d , and (e-f) LWP. The shading, scatter points, ACI metrics, and fit lines are calculated and displayed in the same way as Fig. 3.4.

major difference from Fig. 3.4 and 3.5 is that the range of observed LWP values is much smaller at cloud edge. This will be discussed further in the next section, but a lower LWP does not seem to prohibit observations of above-cloud ACI at cloud edge using this methodology.

We also assess the impact of BL aerosol loading on above-cloud ACI relationships by separating the analysis into two cases. Gupta et al. (2021) found that sensitivity

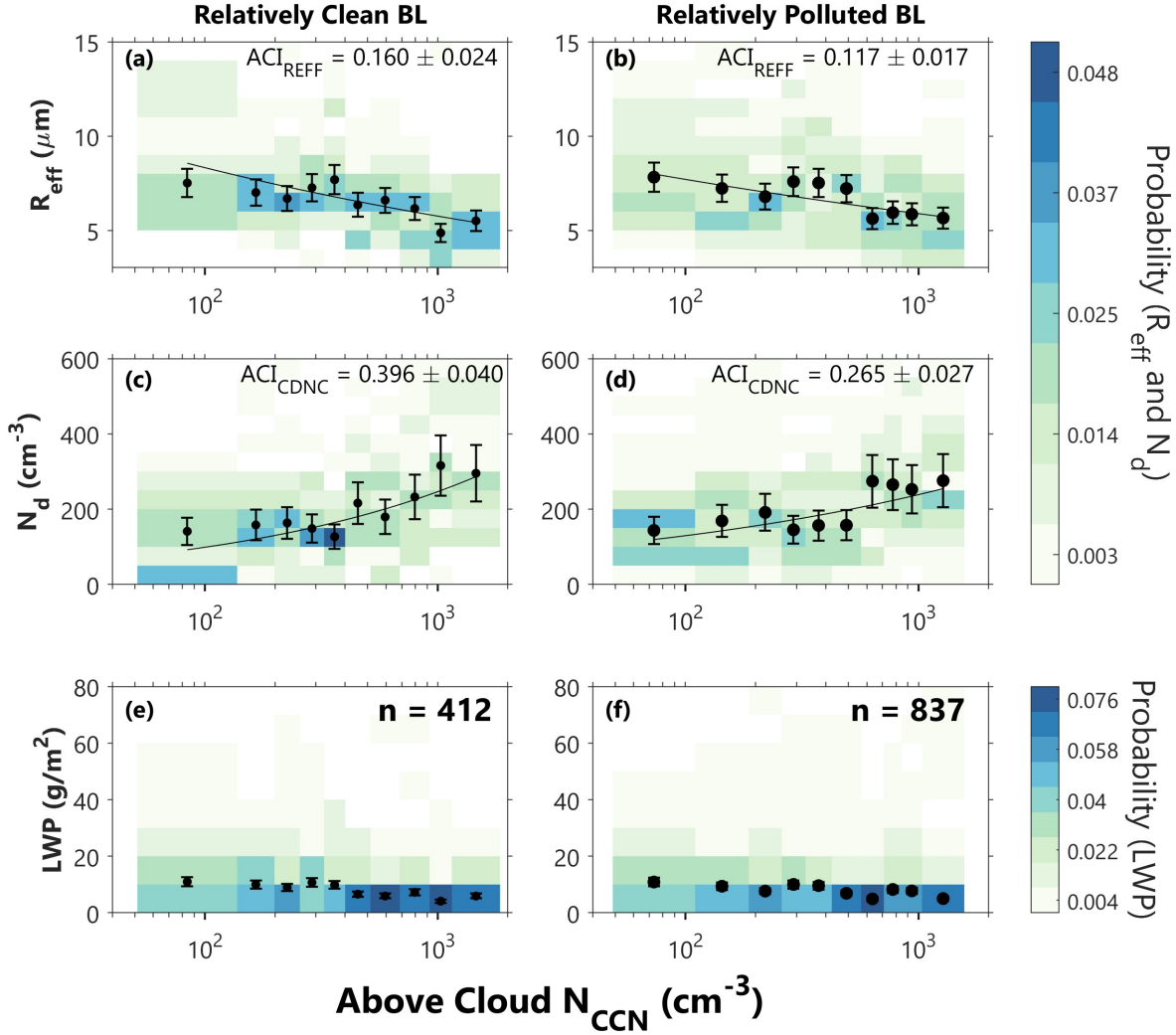


Figure 3.8: Results of from CE data set ($n = 1249$) show the relationship between above-cloud N_{CCN} and (a-b) R_{eff} , (c-d), N_d , and (e-f) LWP for cases with a relatively clean BL (below-cloud $N_{CCN} \leq 350 \text{ cm}^{-3}$) and a relatively polluted BL (below-cloud $N_{CCN} > 350 \text{ cm}^{-3}$). The shading, scatter points, ACI metrics, and fit lines are calculated and displayed in the same way as Fig. 3.4.

to above-cloud aerosol existed regardless of the BL aerosol loading conditions using an aerosol concentration threshold of 350 cm^{-3} . Therefore, in Fig. 3.8 we show the data from panels a, c, and e of Fig. 3.7 separated into two cases based on each point's corresponding below-cloud N_{CCN} . Here we are again able to corroborate the results of Gupta et al. (2021). We find that regardless of BL aerosol loading, relationships

between above-cloud N_{CCN} and cloud properties are evident. However, we also see that above-cloud ACI_{REF} and ACI_{CDNC} are slightly higher when the below-cloud region is relatively clean. When the below-cloud region is more polluted, above-cloud ACI_{REF} and ACI_{CDNC} are slightly decreased. This dampening of the above-cloud impact for polluted below-cloud regions suggests an impact of below-cloud N_{CCN} on cloud top microphysical properties that is lower in magnitude than the above-cloud N_{CCN} impact but non-zero. Additionally, an investigation of the relationship between above- and below-cloud N_{CCN} did not show significant correlation between the two, and we do not hypothesize that the observed correlation between above-cloud N_{CCN} and cloud properties is highly biased by a co-variability with below-cloud N_{CCN} .

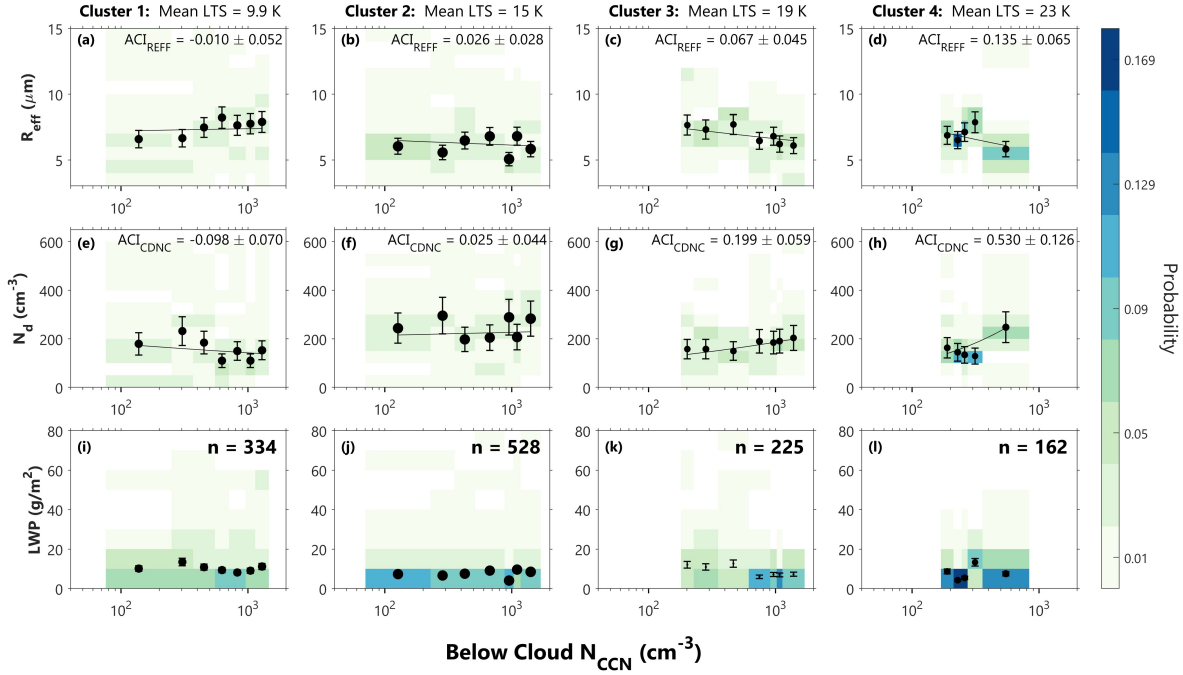


Figure 3.9: Below-cloud observations from Fig. 3.7 are assigned to clusters determined in Fig. 3.5. Results of this clustering show the relationship between below-cloud N_{CCN} and (a-d) R_{eff} , (e-h) N_d , and (i-l) LWP. Average LTS in each cluster increases from left to right. The shading, scatter points, ACI metrics, and fit lines are calculated and displayed in the same way as Fig. 3.4.

Lastly, we investigate the LTS dependence of the below-cloud N_{CCN} relationship to

cloud top microphysical properties using a similar approach as in Fig. 3.5. Data from the CE data set are assigned to the existing clusters determined using the AC data set so that both analyses are directly comparable. These results are shown in Fig. 3.9, where we observe from the fit lines and ACI metrics a trend inverse of that observed for the above-cloud N_{CCN} . That is, as LTS increases, the sensitivity of cloud properties to increasing below-cloud N_{CCN} increases. This suggests that in less stable environments, entrainment mixing of BBA into the cloud layer is the dominant process responsible for the microphysical changes to stratocumulus cloud properties. However, in more stable environments, where vertical mixing is suppressed and low-level cloud cover increases, the lack of such entrainment mixing results in a stronger response of cloud properties to increasing below-cloud N_{CCN} . Another important finding from this analysis is that most observations in the CE data set fall into the first two clusters characterized by low LTS. Therefore, when considering the total impact of below-cloud N_{CCN} across all clusters, as done in Fig. 3.7, low LTS cases dominate the analysis, explaining why the relationship between cloud properties and below-cloud N_{CCN} appears to be weak and nearly negligible.

3.3.3 ACI sensitivity to proximity to cloud edge

Lastly, we investigate ACI sensitivity to proximity to cloud edge. In the previous two sections we investigated the entire AC data set, which includes observations made anywhere there is a cloud, and the CE data set, which is limited to observations made when a cloudy profile is directly adjacent to a clear profile. Additionally, we have seen that observations in the CE data set are characterized by a lower mean LWP (Fig. 3.7 and 3.8) compared to those in the AC data set (Fig. 3.4 and 3.5). Here we investigate the differences between cloud edge and cloud center observations, so we no longer exclude cases where a cloudy profile has clear profiles on both sides. Rather,

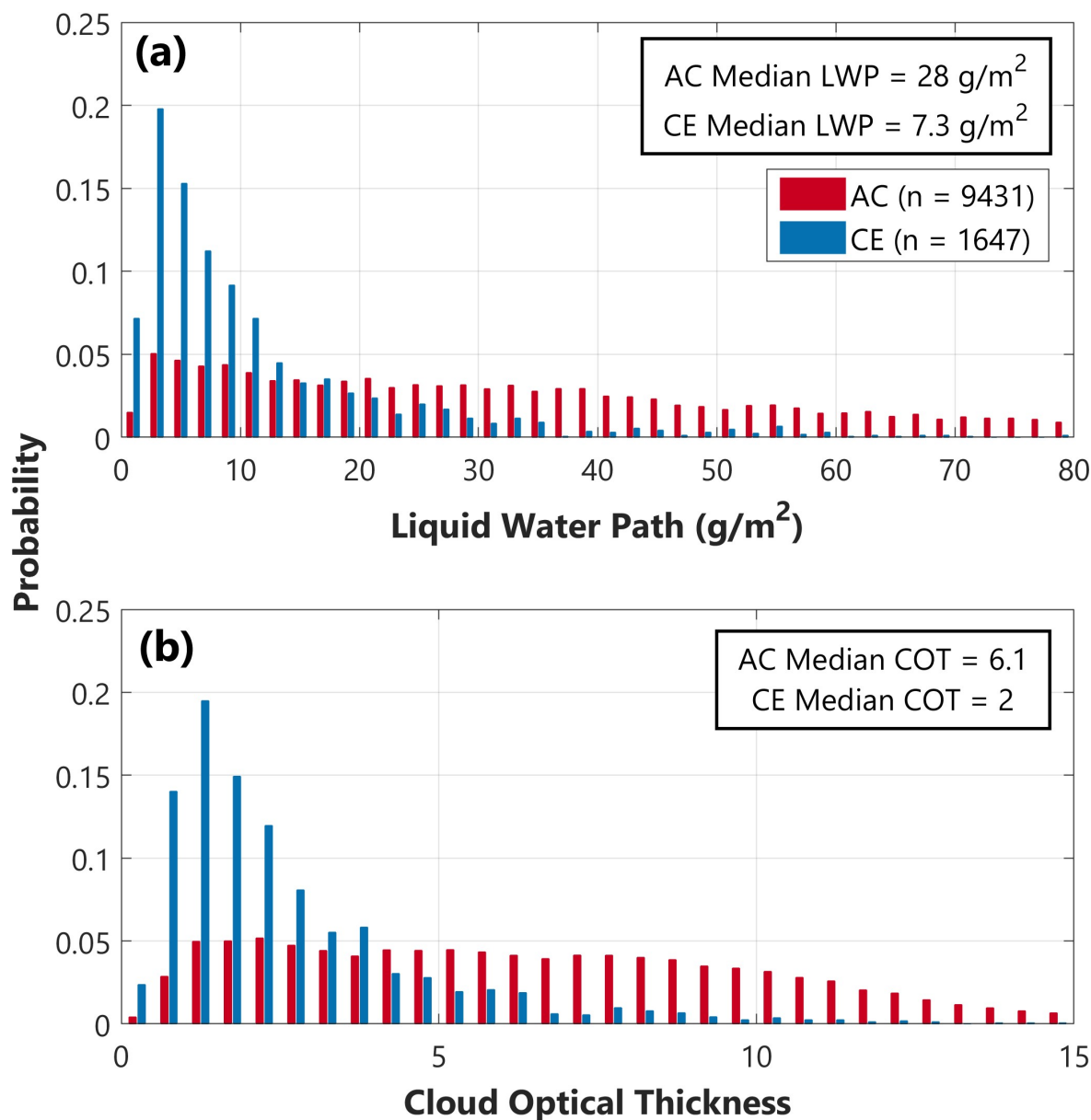


Figure 3.10: Distributions of (a) LWP and (b) COT for all AC data points (red) and CE data points (blue). Median values for both variables and both subsets of data are also shown.

we include them to retain all cloud edge observations in the analysis. The following analyses are still limited to observations made at $LWP \leq 80 \text{ g/m}^2$.

First, we examine differences in LWP and COT between the full AC data set and the CE data set (Fig. 3.10). This figure reiterates the differences in LWP for observations

made at cloud edge, as also seen in Fig. 3.7 and 3.8. Cloud edge observations have a median LWP of 7.3 g/m² while the entire AC data set has a median LWP of 28 g/m². We see similar differences in COT, where the median COT at cloud edge is 2.0 compared to a median value of 6.1 for the full AC data set. Observations made for the full AC data set also have a much broader distribution in both LWP and COT than those made at cloud edge.

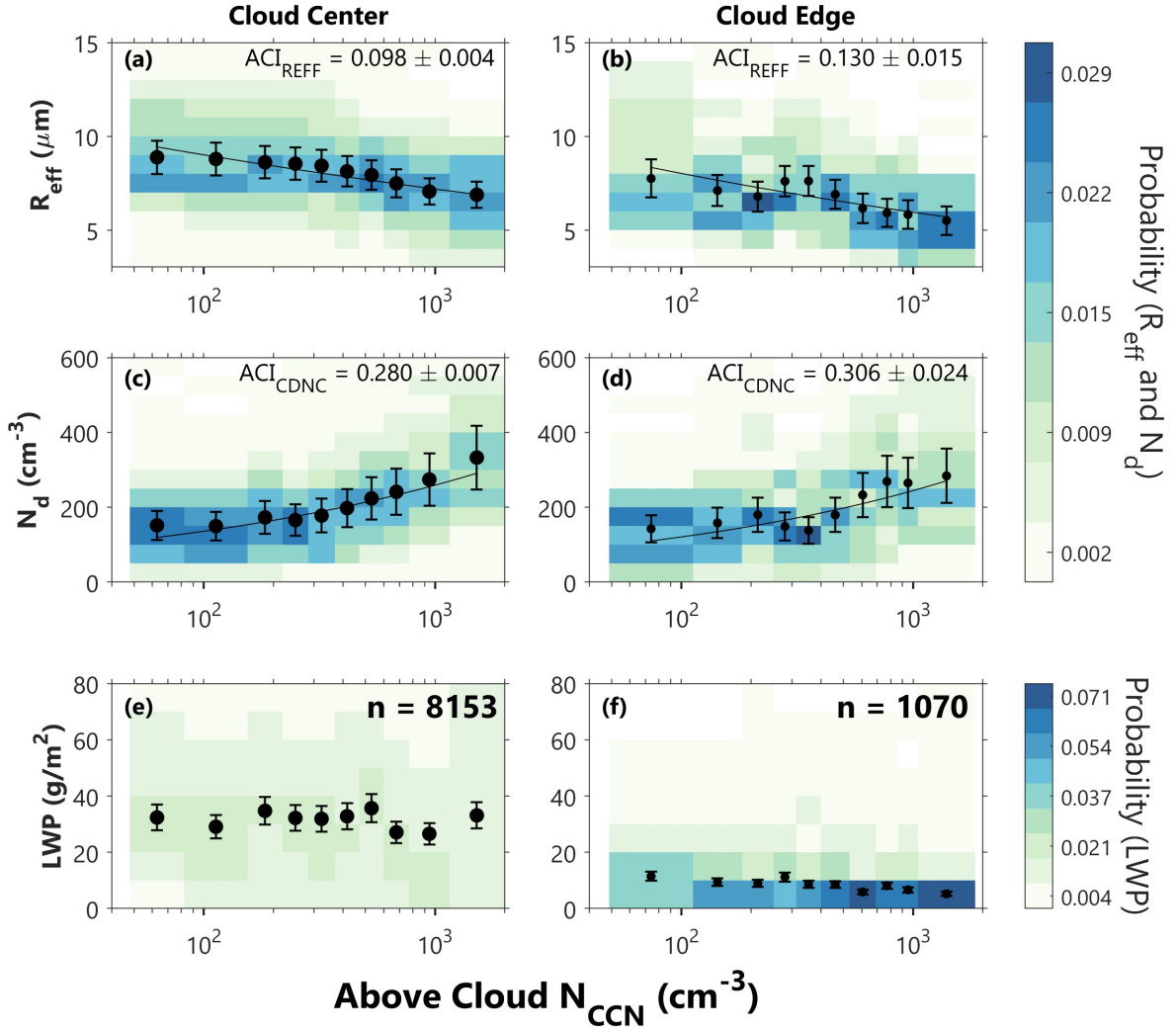


Figure 3.11: Results from cloud center and CE data sets show the relationship between above-cloud N_{CCN} and (a-b) R_{eff} , (c-d), N_d , and (e-f) LWP. The shading, scatter points, ACI metrics, and fit lines are calculated and displayed in the same way as Fig. 3.4.

To investigate if lower LWP and COT at cloud edge are associated with any differences in ACI_{REF} and ACI_{CDNC} , we compare results between CE and CC cases (Fig. 3.11). Like the comparison between cases with relatively clean and polluted BL conditions (Fig. 3.8), we see that regardless of whether observations are made closer to the center of a cloud or at a cloud edge, the relationships between above-cloud N_{CCN} and cloud top R_{eff} and N_d are similar, with ACI metrics being slightly higher for the cloud edge observations than cloud center observations. This suggests that while increased mixing and entrainment of smoke aerosols into the cloud and nucleation of additional cloud droplets is observed for both cases, it may be occurring more frequently at cloud edges. Furthermore, while we do not have below-cloud N_{CCN} for CC cases using this methodology, the similarity in above-cloud N_{CCN} relationships at cloud center and cloud edge could imply that below-cloud N_{CCN} relationships may also be similar in these two regimes. However, further investigation of below-cloud N_{CCN} relationships towards cloud center would likely require modeling efforts or dependence on in situ observations and is beyond the scope of this study.

3.4 Discussion

In our remote sensing-based analysis of ACI between BBA and the underlying stratocumulus cloud deck in the SEA, we have investigated the relationships between above- and below-cloud N_{CCN} and cloud top microphysical properties as a function of environmental stability, below-cloud aerosol loading, and proximity of observations to the cloud edge. In this section we will further discuss differences in the above- and below-cloud N_{CCN} relationships (Sect. 3.4.1), cases of a slight reversal in expected cloud responses (Sect. 3.4.2), and implications for future remote sensing of ACI (Sect. 3.4.3).

3.4.1 Above- vs. below-cloud N_{CCN} relationships

Overall, the relationships analyzed here follow the major findings of Gupta et al. (2021), around which this study was formulated. That is, for cases where the smoke plume is in contact with the cloud top, there is evidence of these CCN impacting cloud top microphysical properties. For all cases, increases in above-cloud N_{CCN} were associated with increases in N_d and decreases in R_{eff} . Cases of contact between cloud top and the BBA plume are associated with greater entrainment mixing (Diamond et al., 2018; Gupta et al., 2021), and our finding here reiterates those suggesting that entrainment of BBA that serve as CCN can result in nucleation of cloud droplets near the cloud top. These relationships were evident regardless of BL aerosol loading, with a slightly dampened impact of above-cloud N_{CCN} in cases where the BL was relatively polluted.

Beyond this corroboration of in situ findings, a major focus of this study was the dependence of aerosol—cloud relationships on environmental stability. Based on a k-means clustering analysis using LTS as the sole clustering variable, we stratified observations of the cloud deck with collocated above-cloud N_{CCN} retrievals into four clusters which turn out to be geographically distinct, with LTS increasing from the northwestern part of the SEA toward the southeastern part closest to the African coast. This increase in LTS aligns well with observed decreases in SST and CTH. Using this method, we find that environmental stability is an important governing factor in determining the sensitivity of cloud properties to increases in above-cloud N_{CCN} . As the lower-tropospheric layer becomes more stable, cloud sensitivity to increasing above-cloud N_{CCN} decreases until there is almost no response (cluster 4; Fig. 3.5d, h). Less stable environments promote greater vertical growth of the cloud layer and mixing led by cloud top entrainment instability (e.g., Mellado, 2017; Gupta et al., 2021). These environments thereby support the modulation of cloud top properties by aerosols

from the overlying smoke plume that are entrained into the cloud layer. The percent differences in ACI metrics between cluster 4 and cluster 1 are -73.9% for ACI_{REF} and -74.3% for ACI_{CDNC} , again indicating that ACIs depend strongly on environmental conditions. While we see from Fig. 3.4 that above-cloud ACIs are evident across the full data set, stratifying the data by LTS demonstrates the significant role that climatological regimes with different environmental stability play in determining how clouds respond to increases in above-cloud N_{CCN} . For example, comparing the full data set (Fig. 3.4a,b) to the cluster of data with the lowest mean LTS (Fig. 3.5a,e) ACI_{REF} increases from 0.093 to 0.161 (73.1%) and ACI_{CDNC} increases from 0.275 to 0.452 (64.4%). Therefore, stratifying data by environmental stability has a large impact on the magnitude, and arguably the accuracy, of the ACI metrics.

Further, we find that constraining the below-cloud N_{CCN} —cloud property relationship using LTS elucidates ACI relationships that are not evident when considering the CE data set as a whole. Observations shown in Fig. 3.7 suggest that at and within 5 km of cloud edges, below-cloud N_{CCN} have a nearly negligible impact on cloud top microphysical properties, which by itself is a physically implausible result. However, upon assigning CE observations to the clusters determined in Fig. 3.5, we find that cloud sensitivity to increasing below-cloud N_{CCN} has the opposite dependence on LTS as for above-cloud N_{CCN} (Fig. 3.9). That is, in more stable environments (cluster 4), decreased entrainment of smoke aerosols into the cloud layer results in higher ACI metrics for below-cloud N_{CCN} than above-cloud N_{CCN} . Since high LTS promotes increased cloud fraction, selecting profiles at cloud edges with which to assess the simultaneous impact of above- and below-cloud N_{CCN} preferentially results in a subset of data with lower average LTS. Therefore, when considering all CE cases together, the stronger impact of below-cloud N_{CCN} for high LTS cases is masked. This finding speaks again to the importance of constraining environmental stability when assessing ACI and iden-

tifies a limitation of our methodology. Additionally, these stability-related findings corroborate those from other ORACLES ACI-focused studies. Using ORACLES 2016 in situ observations, Diamond et al. (2018) found a weaker relationship between cloud properties and above-cloud BBA compared to the below-cloud effect. Since cloud-focused in situ flight legs often target optically thick and continuous cloud segments, it is likely that these observations are characterized by a higher LTS than most of our CE cases. Moreover, a majority of the 2016 observations in this study are categorized by a high average LTS (Fig. 3.6), where we, like Diamond et al. (2018), also find stronger below-cloud ACI relationships than those observed above-cloud. Kacarab et al. (2020) discussed the sensitivity of ACI to velocity-limited and aerosol-limited regimes in the ORACLES 2017 observations, and this analysis indirectly suggests a dependence on up-draft velocity via environmental stability. Future work exploring differences in aerosol properties, cloud properties, and other meteorological variables within each of these clusters could further assess and constrain ACI in this region.

3.4.2 Reversals in expected patterns

There are a few small scale variations evident in the bin medians that are not significantly reflected in the fit lines and therefore are not a primary focus of this analysis. These patterns could be an artifact of retrieval uncertainties. However, here we will discuss a few alternative hypotheses based on previous literature. For example, in Fig. 3.5b and 3.5f, we see a slight increase in R_{eff} and decrease in N_d for the highest concentration N_{CCN} bin. Considering this pattern only occurs at $N_{CCN} > 1000 \text{ cm}^{-3}$, indicating significant contact between cloud top and the smoke plume, one hypothesis for this pattern could be the aerosol semi-direct effect, which describes how absorbing aerosols may act to change cloud properties (Hansen et al., 1997). A study by Koch and Del Genio (2010) that focused on the proximity of absorbing aerosol to different

cloud types found that stratocumulus clouds with absorbing aerosol near the cloud top are likely to experience cloud reduction due to the semi-direct effect. Other studies have found a positive correlation between high aerosol loading and cloud droplet size in highly polluted environments without directly characterizing it as the semi-direct effect (Tang et al., 2014; Ma et al., 2018b; Jose et al., 2020; Khatri et al., 2022). One commonly proposed pathway is that the aerosol radiative effect reduces moisture content and increases competition for water vapor, causing smaller droplets to evaporate, which could explain the decrease in N_d and increase in R_{eff} . This pattern would also align well with the findings of Kacarab et al. (2020), where ORACLES 2017 in situ observations showed no response in N_d to increasing aerosol concentration $> 800 \text{ cm}^{-3}$ when water vapor was limited.

Alternatively, these patterns could indicate that at $N_{CCN} > 1000 \text{ cm}^{-3}$ the environment is saturated by such a large number of small droplets that they begin to coalesce and form precipitation, thus forming fewer droplets that are larger in size. Such a decrease in precipitation susceptibility has been associated with an increase in LWP (Sorooshian et al., 2009), which is also associated with increasing R_{eff} and decreasing N_d here (Fig. 3.5j). However, untangling the response of LWP to aerosols compared to meteorology is difficult, with many proposed pathways by which LWP may increase or decrease in response to increased aerosol loading (Gryspeerdt et al., 2019). More detailed process modeling would likely be needed to fully understand and untangle the relationships and cause-and-effect pathways between above-cloud N_{CCN} and cloud top R_{eff} , N_d , and LWP for these cases of increased above-cloud aerosol loading, which is outside the scope of this study. Therefore, we hypothesize that the reversal of R_{eff} and N_d trends at high above-cloud N_{CCN} could be indicative of a semi-direct effect due to BBA absorption or the beginning of collision-coalescence due to a highly saturated environment.

A similar reversal in the expected response to increasing N_{CCN} is visible in cluster 4 of Fig. 3.9, though in this case it occurs at low N_{CCN} , and not within the highest concentration N_{CCN} bin. This increase in R_{eff} and decrease in N_d occurs for N_{CCN} bins between approximately $170\text{-}360\text{ cm}^{-3}$, representing relatively clean BL conditions. Therefore, it is unlikely that these patterns are attributable to the semi-direct effect of above-cloud BBA. Rather, this may be a case in which low N_{CCN} near cloud base creates a low concentration of larger droplets that is maintained by the collision-coalescence process (Saleeby and Cotton, 2005) before N_{CCN} increases above 400 cm^{-3} . Populations of large droplets at cloud base have been observed in clean aerosol regions for convective clouds over the Amazon by Braga et al. (2017), and this effect has been hypothesized to be attributable to the presence of giant CCN (GCCN) by this and other studies (Yin et al., 2000; Saleeby and Cotton, 2005). It is likely that the below-cloud N_{CCN} population in this region includes sea salt particles, which are an aerosol type more likely to reach such sizes to be classified as GCCN. However, this remains a hypothesis to explain the increase in R_{eff} and decrease in N_d at low below-cloud N_{CCN} as the exact composition and size of below-cloud N_{CCN} is outside the scope of this analysis.

3.4.3 Implications for remote sensing techniques

The major implication of these results confirming those of an in situ-based study (Gupta et al., 2021) is that, with the right considerations regarding environmental stability, ACI can reliably be estimated using only these remote sensing-based observations. We make use of well-collocated HSRL-2 and RSP observations from the ORACLES campaign to investigate relationships between cloud microphysical properties and vertically resolved N_{CCN} both above- and below-cloud. This strategy not only corroborates in situ-based findings using a larger amount of data than is available from

in situ observations, but it also demonstrates a methodology that can be used with current and future satellite-based observations.

As previously mentioned, the vertical distribution of N_{CCN} is especially relevant for understanding ACI. For the ORACLES region, where we have the unique scenario of an optically thick and strongly absorbing BBA plume overlying marine stratocumulus clouds, we found that changes in above-cloud N_{CCN} are more strongly related to changes in cloud top microphysical properties under unstable conditions, while changes in the below-cloud N_{CCN} have a more significant impact on cloud properties under stable conditions. However, without vertically resolved N_{CCN} from the ML-CCN method, this above- and below-cloud N_{CCN} distinction would not have been possible. Additionally, the high accuracy of the ML-CCN method has an advantage over other active spaceborne sensors such as CALIOP that have routinely misplaced the vertical extent of aerosol plumes and thus misrepresented ACI (Rajapakshe et al., 2017). Another benefit of the ML-CCN method is that it allows us to use data in close proximity (2 km) to cloud edge without concerns about cloud edge humidification effects since environmental relative humidity is considered in the model training.

A similar methodology as that shown here could be done in other regions using ATLID and Multi Spectral Imager (MSI) observations from the recently launched EarthCARE satellite (Wehr et al., 2023) in combination with other satellite-based cloud retrievals. While higher uncertainty and lower signal-to-noise may be associated with ATLID N_{CCN} retrievals, the overall framework presented here remains valid for spaceborne remote sensing of ACI. However, one important limitation inherent to this method is that the selection of cloud edge cases for assessing the simultaneous impact of above- and below-cloud N_{CCN} may preferentially create a subset of primarily low LTS observations, and this needs to be considered when interpreting results. Consequently, the dependence of ACI on LTS speaks to the need to constrain future satellite

observations by a stability-related parameter such as LTS to accurately represent the impacts of different climatological regimes on ACI metrics, which may also impact their parameterization in models.

3.5 Conclusion

One key component missing from several studies of ACI is the vertical distribution of N_{CCN} relative to cloud height, which in regions with significant tropospheric aerosol loading is critical to understanding how and where aerosols are nucleating as cloud droplets and impacting cloud microphysical properties. Here we use a fully remote sensing-based data set to investigate ACI over the SEA using HSRL-2-based, ML-predicted N_{CCN} curtains and cloud microphysical properties retrieved from the RSP. To assess the simultaneous above- and below-cloud impact of N_{CCN} on cloud properties, we infer below-cloud N_{CCN} values from cloud-edge adjacent profiles. Previous in situ-based studies have found evidence of ACI between smoke plume BBA and the underlying stratocumulus cloud deck. Therefore, the major goals of this study were to investigate these cloud top ACI in more detail and to determine whether such relationships could be observed using only remote sensing data.

We found that our results align well with those of the in situ-based study (Gupta et al., 2021). That is, we see a decrease in R_{eff} and increase in N_d when the BBA concentrations are significant within 100 m of the cloud top, and this finding is independent of horizontal proximity to cloud edge and the magnitude of BL aerosol loading. Additionally, to constrain the impact of environmental stability we cluster the above-cloud and cloud edge data sets by LTS, finding that cloud sensitivity to increasing above- (below-)cloud N_{CCN} decreases (increases) as LTS increases. Therefore, it appears that entrainment of above-cloud BBA into the stratocumulus cloud layer is a major con-

trol of cloud top microphysical properties under relatively unstable conditions, while the below-cloud N_{CCN} effect on cloud properties is stronger under more stable conditions. Therefore, both above- and below-cloud N_{CCN} effects are highly dependent on environmental stability.

A major implication of this work is the ability to assess ACI using remote sensing-based observations, a method that can be applied to current and future spaceborne observations. We have demonstrated the benefit of the Redemann and Gao (2024) ML-CCN product in that it can be used to separate the impact of above- and below-cloud N_{CCN} on cloud properties in the same column. Additionally, we have used autocorrelation analyses to characterize the variability of N_{CCN} in the SEA BL, which provides a tool to extrapolate clear-sky N_{CCN} retrievals to nearby clouds. We concede that this extrapolation carries its own uncertainty, but it is superior to the inherent spatial homogeneity assumptions in ACI studies that derive ACI metrics from large-scale averaged satellite retrievals of aerosol and cloud properties. Each of these methodologies can be applied to future studies in different regions and cloud types to further work toward reducing uncertainty associated with the radiative impacts of ACI.

3.6 Appendix A

As in Gupta et al. (2021), relationships between cloud top height (Z_T) and cloud base height (Z_B) are developed from in situ data including the Cloud and Aerosol Spectrometer (CAS) on the Cloud, Aerosol and Precipitation Spectrometer (CAPS; Baumgardner et al., 2001), two Cloud Droplet Probes (CDP; Lance et al., 2010), and a King hot-wire (King et al., 1978). The King hot-wire was used to determine bulk liquid water content (LWC). CAS and the CDP measured the cloud droplet size distribution, and the full size distribution spectrum covering diameters between 3-50

μm was determined using the probe most consistent with the King hot-wire LWC (Gupta et al., 2022b). These observations are used to find the highest (cloud top) and lowest (cloud base) altitudes within individual sawtooth profiles at which in situ cloud N_d is greater than 10 cm^{-3} and bulk liquid water content (LWC) is greater than 0.05 g/m^3 . Our resulting linear relationships between in situ Z_T and Z_B are shown in Figure 3.12a. We then estimate cloud base height from HSRL-2 cloud top heights using these statistical relationships developed from in situ observations. When this method is applied to each lidar profile observed at a cloud edge (Sect. 3.2.4), we find that most cloud base heights fall between about 400-750 m (Fig. 3.12b).

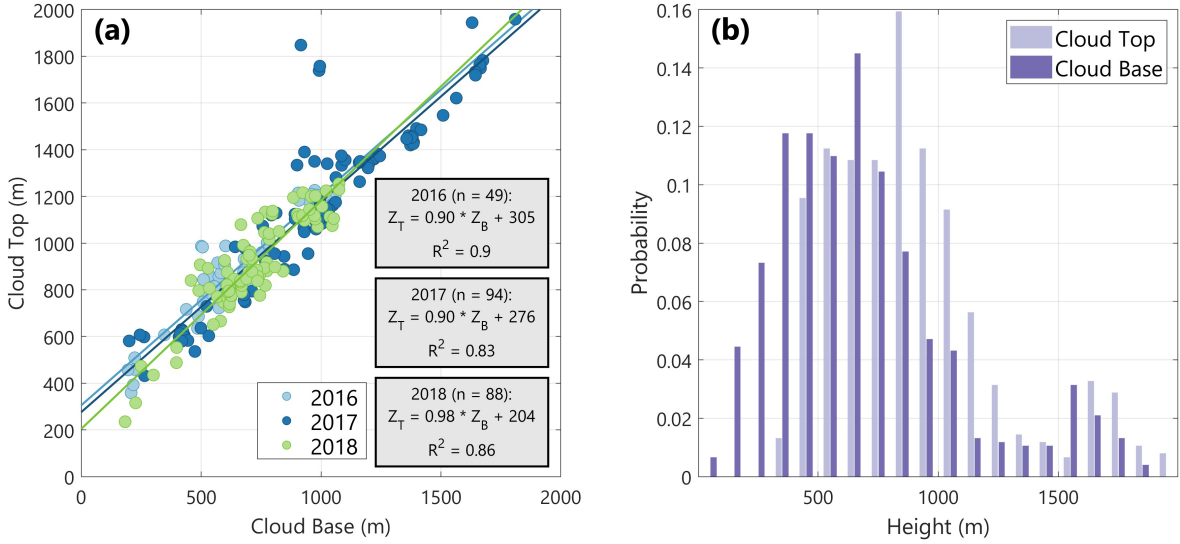


Figure 3.12: (a) In situ derived relationship between cloud top height (Z_T) and cloud base height (Z_B) based on the methodology of Gupta et al. (2021). These relationships are applied to HSRL-2 cloud top heights measured at cloud edge for all three deployment years of ORACLES. The distributions of these HSRL-2 measured cloud top heights and resultant calculated cloud base heights are given in panel (b)

Chapter 4

Design, Implementation, and Evaluation of an Interactive Aerosol, Cloud, and Climate Science Module for High School Students

4.1 Introduction

The sixth, and most recent, assessment report (AR6) from the Intergovernmental Panel on Climate Change (IPCC) stated that compared to AR5, our advances in observing Earth's energy budget indicate that Earth is warming everywhere (Forster et al., 2021). While anthropogenic climate warming has been a widely accepted reality in the scientific community for decades, politicization of the issue has caused division and skepticism amongst the public (McCright and Dunlap, 2011) that continues to impede progress in implementing solutions. To change attitudes, shift cultural norms, and ultimately impact lasting change for our environment, it is imperative that we educate and communicate effectively on the realities of our changing climate.

Several previous studies have examined methods and tactics for communicating climate science and the implications of climate change, providing guidance for approaches that may benefit, and not hinder, effective communication and understanding. For example, since climate change is too great an issue for any single discipline to solve alone, one important strategy is productively collaborating across disciplinary fields and with general audiences (Stevenson et al., 2017; Ramanathan et al., 2019). While meteorologists and climate scientists have detailed understanding of radiative climate forcings and the mechanisms that cause warming, they are unlikely to be specialists in aspects such as policy and economic impacts. Therefore, climate research efforts may ultimately be insufficient if time is not also spent actively seeking multi-faceted solutions to addressing our warming climate.

In addition to cross-disciplinary collaboration, another critical aspect is communication of climate science with broader audiences. Ramanathan et al. (2019) emphasize the importance of welcoming people into climate action as “active participants in solutions,” while also highlighting that simply providing people with information and focusing on education alone is unlikely to produce new attitudes and behaviors. Rather, one key finding suggests that effective climate communication customizes information to various audiences and communities by discussing what is at stake for them specifically (Monroe et al., 2019; Shwom et al., 2017; Ramanathan et al., 2019; Staffen et al., 2025). For example, one common type of climate change imagery depicts melting ice caps and detrimental effects on polar bears. However, for someone living in the land-locked Midwestern United States, these examples are not particularly relevant or relatable, and may make climate warming feel like an obscure and distant issue.

While there are many pathways for improving climate literacy and public trust in climate science, the focus in this study will be implementation of climate science education in the K-12 setting. Regardless of the dominant public opinion, climate

warming will continue to impact Earth moving into the future, so it is important that future generations are educated on the urgency of the situation. Stevenson et al. (2017) argues that one of the most effective ways to develop capacity for addressing the climate crisis is by integrating climate change education into formal education systems. However, since the United States does not have a uniform set of science standards, the extent of climate change content in school curricula varies by state and grade level (Beaver et al., 2024). Dupigny-Giroux (2010) found that weather and climate principles are usually found in geography, social studies, or Earth science courses, and that while some students may learn about weather at the elementary level, others may not experience an earth or environmental science course until high school or undergraduate courses. Through their analysis of K-12 climate change standards in the United States, Beaver et al. (2024) found that most standards are found in Earth and Space Science (ESS) courses at the high school level, many of which are not mandatory for all students.

In addition to disparities in terms of whether or not climate change is included in K-12 curricula, there are several other barriers to teaching climate science in classrooms. These barriers tend to fall into three main categories. One is related to external perceptions, such as political or administrative influence (McNeal et al., 2017), concern about possible negative responses from parents (Monroe et al., 2019), or apprehension about possible controversy (Sullivan et al., 2014) or a perceived decrease in their credibility as a consequence of choosing to teach climate science (Monroe et al., 2019). Another major barrier is a lack of teacher professional development for climate change education (Hestness et al., 2011; Sullivan et al., 2014). The conceptual difficulty and interdisciplinary nature of climate science may discourage teachers from broaching the topic if they are not comfortable or well-versed in the concepts (Boon, 2016; Monroe et al., 2019). Lastly, teachers may face difficulties in finding valid information and

resources online (Stallard et al., 2026), or in dealing with a lack of alignment between educational standards and curriculum (Wise, 2010; Sullivan et al., 2014). These complications may result in extra effort needed to adapt and modify resources for specific learning outcomes (Stallard et al., 2026).

While these and other obstacles continue to hinder climate science education in K-12 schools, many have also found success through innovative approaches. A systematic review of climate change education strategies by Monroe et al. (2019) found that two themes common to successful environmental education include focusing on personally relevant and meaningful information and using active and engaging teaching methods. Additionally, they discuss four themes specific to issues such as climate change that result in effective learning, which include engaging in deliberate discussions, interacting with scientists, addressing misconceptions, and implementing school or community projects. As previously mentioned, for climate science communication to be meaningful for the public, within or outside of a school setting, local and relevant examples are important. Monroe et al. (2019) refers to this as making “the distant, global, and nebulous threat of climate change personally relevant.” They emphasize the effectiveness of engaging learners through hands-on labs or in small group, worksheet-based discussions, or by involving the use of data and technology (Monroe et al., 2019; Staffen et al., 2025). They also discuss collaborations with scientists and climate experts, and the development of partnerships to enhance teacher professional development. Collaborations between local K-12 schools and universities via programs that teach about climate science have also been demonstrated to be successful in studies such as Hallar et al. (2011), Dupigny-Giroux et al. (2012), and Sullivan et al. (2014).

This chapter discusses the design, implementation, and evaluation of a three-day series of interactive modules developed to communicate key components of climate science in a high school setting through a focus on atmospheric aerosols and clouds. The

extent of content related to clouds in most K-12 ESS curricula tends to be limited to the role of cloud formation in the water cycle and knowledge of different cloud types, while information about atmospheric aerosols and cloud microphysical properties are generally outside the scope of curricula. Therefore, key concepts related to cloud condensation nuclei (CCN) concentrations (N_{CCN}) and aerosol-cloud interactions (ACI) from Chapters 2 and 3 were translated into high school level classroom modules to communicate the role that aerosols and clouds play in Earth's climate system. The following modules were designed to communicate:

1. The fundamentals of atmospheric aerosols (natural and man-made particulates such as wildfire smoke, desert dust, and pollution) and their interactions with clouds and Earth's energy balance.
2. How desert dust aerosols impact weather in the state of Oklahoma, using observations from the March 14, 2025 dust event.

The primary hypothesis is that by teaching the fundamental mechanisms of atmospheric aerosols and their interactions with solar radiation and clouds, student understanding of aerosols and clouds as drivers of climate change will improve.

It is important to clarify here what this project is not intended to do. First, the goal in outlining the classroom visit and developing the associated lecture slides and activities was not to incorporate every literature-recommended tactic for addressing climate change in K-12 classrooms. The goal also was not to replace the need for and benefit of established classroom teachers integrating climate science into their curricula. Additionally, as time did not allow for multiple iterations of these materials, the goal was not to perfect the survey statements, worksheets, or activities. Rather, the goal of this project was to encapsulate key concepts about N_{CCN} and ACI from Chapters 2 and 3 in a set of lectures and activities to be implemented and tested as a tool in

a high school classroom setting. Through implementing these lectures and activities, the goal was then to determine if they were successful in improving learning, and to decide which details should be changed for future application. As will be further discussed in Sect. 4.7, future work should continue to iterate on all components of this study, and it would be mutually beneficial to continue to implement them in classrooms and maintain a collaborative relationship between the OU School of Meteorology and Norman Public Schools.

4.2 Learning objectives

Learning objectives were written for each module following guidelines from Kennedy (2006) and using the cognitive framework of Bloom’s Taxonomy. Bloom’s Taxonomy describes how a student builds upon former learning to develop more complex means of understanding using six levels: knowledge (1), comprehension (2), application (3), analysis (4), synthesis (5), and evaluation (6). As the three classroom modules for this project would be implemented in only three days of the course (Section 4.5), most learning objectives were written to fall into the first two levels, knowledge and comprehension. A fourth group of higher-level learning objectives were also included that relate to aerosol and cloud impacts on climate. This group of learning objectives, labeled as “climate connection,” were not associated with an independent set of lecture slides and activities. Rather, these objectives were included to test the primary hypothesis of the work and determine if the three modules enabled students to understand the implications of aerosols and clouds on a larger scale. All learning objectives and their associated Bloom’s taxonomy level are shown in Fig. 4.1.

(a) Module 1 Learning Objectives (1) Recognize types and sources of different atmospheric aerosols (1) Describe the difference between aerosol direct and indirect effects (2) Explain how atmospheric aerosols can negatively impact human health (2) Compare the magnitude of direct effects from different aerosol types (3) Determine what aerosol characteristics facilitate and hinder cloud droplet nucleation	(b) Module 2 Learning Objectives (1) Name the key components necessary for cloud formation (1) Recognize cloud droplet size and number concentration as two different cloud characteristics (2) Relate aerosol concentrations to cloud properties (2) Interpret how the number and size of cloud droplets in a cloud impact their interactions with the sun's energy (3) Demonstrate the possible outcomes of changing cloud properties at the introduction of more aerosols (3) Interpret how changes in cloud properties impact the development of precipitation
(c) Module 3 Learning Objectives (1) Recognize types of measurement systems used to observe clouds and aerosols (1) Explain how the presence of dust aerosols over Oklahoma impacts local conditions and measurements (2) Compare and contrast various weather observations on a sunny day, a completely cloudy day, and during a dust transport event (3) Explain a specific example of how atmospheric aerosols have impacted weather in Oklahoma (3) Discover how to visualize and interpret data.	(d) Climate Connection Learning Objectives (4) Attribute possible long term climate effects to emissions of atmospheric aerosols (4) Attribute possible long term climate effects to aerosol-induced changes in cloud properties (4) Deduce potential long-term changes in climate due to frequent presence of atmospheric aerosols.

Figure 4.1: Learning objectives for (a), (b), (c) the three main modules and (d) how these topics relate to climate. The number in parentheses in front of each learning objective denotes the corresponding Bloom's taxonomy level.

4.3 Data collection

4.3.1 Survey design

Pre- and post-assessments are generally recognized as a legitimate tool, though they have at times been criticized as not being a dependable method for assessing learning (Hufford, 2010). One example of success using this method is seen in Ivanov et al. (2018), where Likert-scale questionnaires were implemented as an educational tool for a university level mathematics course. As a result of including the questionnaires, average exam scores increased and most students reported a positive impact of the surveys. Due to the short nature of this classroom visit and given that these topics were outside the scope of the normal classroom curriculum, a pre- and post-survey were determined to be the most effective way to assess the impacts of these activities

on student learning.

The pre- and post-surveys contained identical questions and were administered before and after the three-day classroom visit. Each question was directly linked to a specific learning objective (Fig. 4.1), and responses were based on a 5-point Likert scale (*strongly disagree, disagree, neither agree nor disagree, agree, strongly agree*). A Likert scale was used to assess how the degree of student understanding or misunderstanding changed from before the activities to after their implementation in the classroom, while providing a wider range of response options than a true/false quiz. While some studies suggest that 7-point Likert scales are the best option in terms of correlation with observed significance levels, others suggest that 5-point Likert scales can be less confusing and increase response rates (Taherdoost, 2019). Since students were unlikely to be familiar with the topics addressed in these statements, a 5-point Likert scale was chosen to simplify their experience taking the survey. The surveys included a combination of true (T), false (F), and neutral (N) statements, where the neutral statements were included to assess student confidence in specific skills or knowledge and therefore are not designated as true or false. All survey statements are listed below and categorized into individual modules, with the designation of T, F, or N in parentheses.

Module 1 Survey Statements

M1.1. Desert dust is the only natural aerosol. (F)

M1.2. Aerosol direct effects describe their interactions with each other. (F)

M1.3. Aerosol indirect effects describe their interactions with clouds. (T)

M1.4. The largest atmospheric particles are most likely to negatively impact human health. (F)

M1.5. Wildfire smoke aerosols are more likely to absorb the sun's energy than sea salt aerosols. (T)

M1.6. All types of atmospheric aerosols have the same ability to form cloud droplets. (F)

Module 2 Survey Statements

M2.1. Water vapor and atmospheric cooling are the only two ingredients necessary for cloud formation. (F)

M2.2. Clouds can be characterized by the number of individual droplets and droplet sizes. (T)

M2.3. An increase in atmospheric aerosols generally causes an increase in both the number and size of cloud droplets. (F)

M2.4. The amount of the sun's energy scattered by a cloud changes when the number of cloud droplets changes. (T)

M2.5. If the amount of liquid in a cloud stays the same, the number of cloud droplets and their size can both increase when more aerosols are introduced. (F)

M2.6. Aerosol interactions with clouds can impact whether or not precipitation can form. (T)

Module 3 Survey Statements

M3.1. Clouds and aerosols can only be measured from instruments on the ground. (F)

M3.2. Dust aerosols scatter the sun's energy more than they absorb it, so a large amount of dust aerosols will decrease the amount of the sun's energy that reaches the surface. (T)

- M3.3. A completely cloudy day and a day with a lot of dust aerosols in the atmosphere have no similarities in terms of what weather is like at the ground. (F)
- M3.4. Atmospheric aerosols transported from far away can reach Oklahoma and cause changes in weather. (T)
- M3.5. I am confident that I know how to access and visualize weather observations made in Oklahoma. (N)

Climate Connection Survey Statements

- M4.1. Greenhouse gases are the only important component of the atmosphere that impacts climate. (F)
- M4.2. All types of atmospheric aerosols absorb energy from the sun and cause warming. (F)
- M4.3. Aerosol interactions with the sun's energy can have long-term effects on climate. (T)
- M4.4. I am confident that I could list at least one way that aerosol interactions with the sun can impact climate. (N)
- M4.5. Aerosol interactions with clouds can have long-term effects on climate. (T)
- M4.6. I am confident that I could list at least one way that aerosol interactions with clouds can impact climate. (N)
- M4.7. Frequent atmospheric aerosol events in Oklahoma could cause long-term changes in climate. (T)

4.3.2 Reflection question worksheets

In addition to quantitative data collection from pre- and post-survey responses, three reflection question worksheets were developed alongside the activities in each module. These allowed for collection of open-ended responses to assess how students were thinking about the topics addressed in each activity. The specific questions from each worksheet are discussed further in Section 4.4.

4.3.3 Institutional Review Board approval

The pre- and post-surveys and all reflection question worksheets were first approved by the Norman Public Schools (NPS) Executive Director and Administrative Assistant of Teaching and Learning. They were then also approved by the University of Oklahoma (OU) Institutional Review Board (IRB; IRB number 19536, approved on 04/06/2026). Responses used in the remainder of this chapter received informed consent from both students and their parents. All students were included in the classroom lectures and activities, but participation in the surveys and reflection question worksheets was not mandatory.

4.4 Modules and activities

The lecture content and activities for this three-day classroom visit were divided into three modules. This section gives an overview of the contents of each set of lecture slides and describes the activity and its corresponding reflection question worksheet. The concentration of this section is on activity development and how individual aspects targeted different learning goals.

4.4.1 Module 1: Introduction to aerosols

Lecture Slides

To introduce students to aerosols and N_{CCN} , the lecture slides for the first day included a definition of atmospheric aerosols, a few common examples, and a discussion of why they are important. Different ways of classifying aerosols were explained, including the difference between primary and secondary aerosols, natural and anthropogenic sources, and describing aerosol size using fine and coarse modes. A discussion of aerosol importance included their human health impacts, interactions with the sun's energy, and role in cloud formation, ending with a definition of CCN.

Activity

The Module 1 activity was a board game called Life Cycle of an Aerosol (Fig. 4.2). The motivation behind this game was to convey that different types of aerosols behave differently in terms of their direct interactions with solar radiation and their ability to nucleate as CCN. In this game, each student plays as a different aerosol type (wildfire smoke, urban pollution, desert dust, marine sea salt), with a maximum of two students per aerosol type and eight students per game board. The goal of the game is to be the first player to make it to the cloud zone and to increase in size enough to nucleate as a CCN. Each aerosol type starts at the same size (10 nm) and has a specific size on the back of their player card (Fig. 4.3) that is needed to nucleate as a CCN. These sizes are critical diameter values estimated via κ -Köhler theory and the average kappa values calculated in Chapter 2. Additionally, each aerosol type has scattering and absorption “points” on the back of their player cards. These values are tallied each time a player lands on a game board space with a sun symbol. At the end of the game, these differences in each aerosol type's critical diameter and scattering and absorption

points are designed to communicate that different aerosols have different properties. For example, marine sea salt has the smallest size required to nucleate as a CCN due to its high hygroscopicity, while desert dust must reach the largest size due to its low hygroscopicity. Though partially dependent on how many times each player lands on a sun symbol space, wildfire smoke will usually end up with the most absorption points, while marine sea salt will usually have the most scattering points.



Figure 4.2: Life Cycle of an Aerosol board game, with player cards and weather cards.

As players move around the board from their starting space to the cloud zone, board spaces with exclamation points correspond to weather cards (Fig. 4.4), which describe how aerosols are transported via air movement (a windy day, a cold front, etc.) or wet deposition (rainfall). These cards can move a player forward or backward

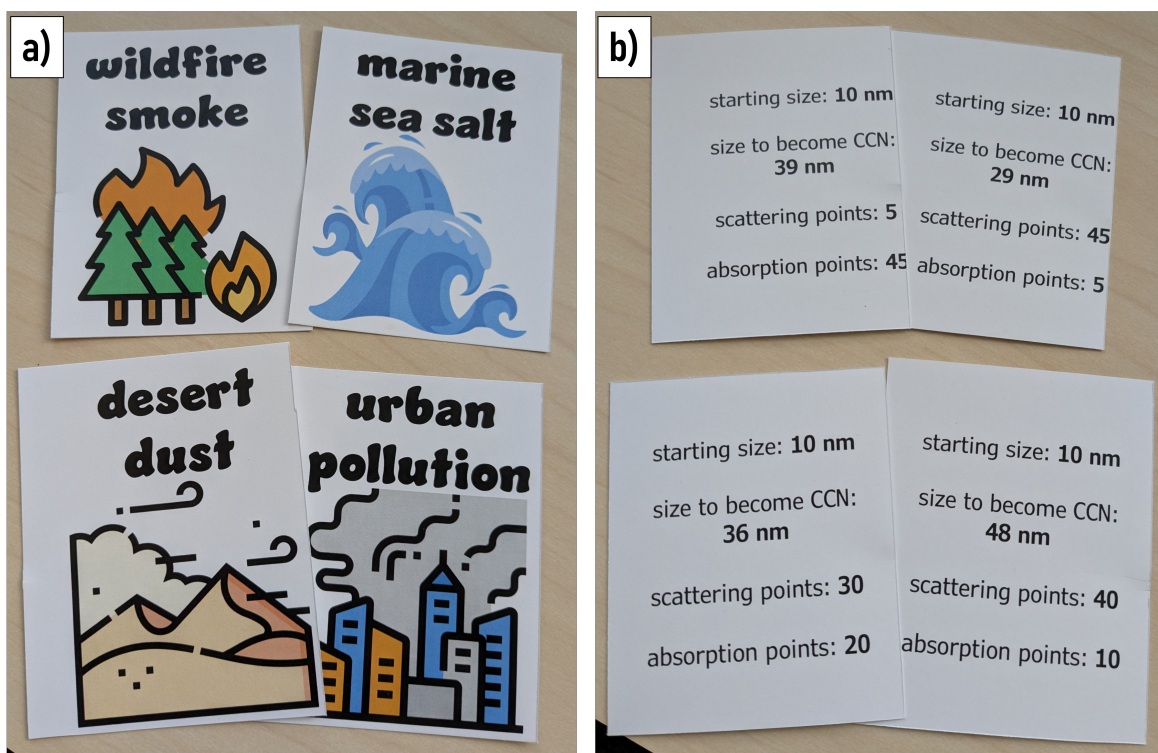


Figure 4.3: (a) Front and (b) back of player cards for Life Cycle of an Aerosol board game. Cards in panel (b) correspond to the same order as shown in panel (a).

or cause them to lose a turn. Once a player makes it to the cloud zone, they have five rolls of the dice to increase from their starting size to the goal size printed on the back of their player card. The numbers from each roll are added to their starting size. If a player does not reach the goal size within five rolls, they move back two spaces and will try again on their next turn, this time starting from their final size after the first attempt. For example, if a player increased from 10 nm to 24 nm on the first attempt, they would start increasing their size from 24 nm on the second attempt.

Reflection Question Worksheet

The reflection questions for this activity included the following:

1. What aerosol properties do you think might cause some to become cloud droplets easier than others?



Figure 4.4: Example of weather cards.

2. Based on the final count of scattering and absorption points, which aerosol type is most likely to cause cooling? Which is most likely to cause warming?
3. What aerosol properties do you think might cause some aerosols to mostly absorb the sun's energy and other aerosols to mostly scatter the sun's energy?

4.4.2 Module 2: Aerosol-cloud interactions and cloud radiative effects

Activity

For Module 2, the activity preceded the lecture slides so that students would discover and understand the relationship between aerosol concentration and the number and size of cloud droplets on their own, before discussing it in the lecture slides. This

activity was called Building a Cloud, and students were given 0.33 oz of individually packaged air-dry clay and 6 blue beads, where the clay represented the amount of available water vapor and the beads represented CCN. Their first task was to create a cloud out of individual droplets using the entirety of the clay and all beads, making sure that all droplets were approximately the same size (Fig. 4.5). At the end of this step, students were instructed to measure the diameter of each droplet and calculate an average.

In the next step, students were given an additional 12 beads and instructed to adjust their cloud to incorporate these additional CCN using the same amount of water vapor. This step was designed with the intention of students discovering and understanding on their own the relationship between CCN, available water vapor, and cloud droplet number concentration and size. To incorporate the additional 12 CCN, the clay needed to be redistributed, with each bead forming a smaller droplet than in the first step (Fig. 4.6). Because cloud microphysical properties can be unintuitive, this hands-on activity was aimed at making these relationships more tangible and visible. At the end of this step, students were again instructed to calculate and record the average drop diameter. While transitioning into the lecture slides, the change in average diameter from the first to second step was also discussed.

Lecture Slides

After the Building a Cloud activity, the lecture slides first included a review of the necessary components for cloud formation, and a discussion of the various ways that clouds can be characterized. Specifically, definitions of both macrophysical and microphysical properties were discussed, as macrophysical properties such as size, altitude, and type are more commonly recognized, while microphysical properties are generally not taught at a high school level. Next, there was a discussion of how aerosols impact



Figure 4.5: First cloud created using air-dry clay and 6 beads. Each resultant cloud droplet is approximately 1 inch in diameter.

cloud properties, beginning with the relationship between number and size of cloud droplets, as demonstrated in the activity. This included a definition of the Twomey effect (Twomey, 1974) and a conversation of how the number and size of cloud droplets influences cloud albedo, cloud lifetime, and the formation of precipitation. Lastly, the slides discussed the role of clouds in Earth's energy budget by highlighting that clouds

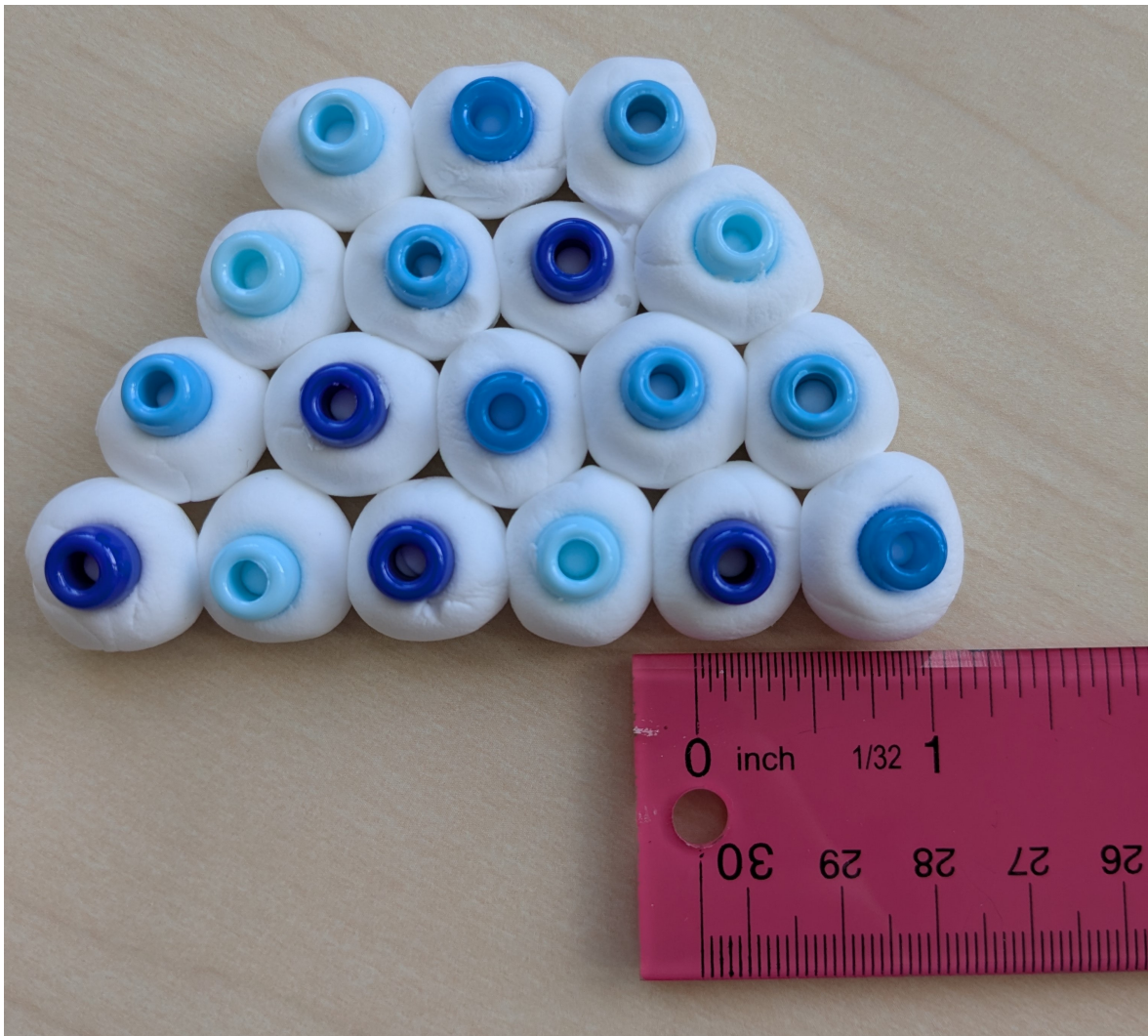


Figure 4.6: Second cloud created using air-dry clay and 18 total beads. Each resultant cloud droplet is approximately 0.5 inch in diameter.

can both reflect incoming solar radiation and trap outgoing longwave radiation. While clouds generally reflect more energy than they trap in the atmosphere, climate predictions estimate that clouds are most likely to cause warming in the future (Forster et al., 2021).

Reflection Questions

The reflection questions for this activity included the following:

1. With a set amount of water and the addition of more aerosols, what is the balance between number of cloud droplets and their size?
2. How can a large number of aerosols prevent a cloud from forming rain?
3. The sun's energy is reflected from individual droplets in a cloud. Which scenario (low vs. high aerosol concentration) had more droplets for this reflection?
 - (a) Do you think this reflection would cause cooling or warming?
4. Can you think of any other ways that clouds could cause cooling or warming?

4.4.3 Module 3: Observations and Oklahoma-specific example

Lecture Slides

Module 3 lecture slides focused on observations of clouds and aerosols and highlighted a few Oklahoma-specific observational networks. This lecture began with a discussion of how measurements can be made via satellites, aircraft, or ground-based stations, and then described the differences between in situ and remote sensing techniques. A few examples were given for how such observations can be viewed online, specifically looking at aerosol optical depth (AOD), cloud cover, and aerosol type from National Aeronautics and Space Administration (NASA) Worldview, in addition to air quality metrics from the National Oceanic and Atmospheric Administration (NOAA) and National Weather Service (NWS) Air Quality Forecast Guidance Viewer and the AirNow Fire and Smoke Map. Lastly, a brief overview of the AErosol RObotic NETwork (AERONET) station on the roof of the National Weather Center (NWC), the Atmospheric Radiation Measurements (ARM) Southern Great Plains (SGP) site, and the Oklahoma Mesonet network was given to introduce the Module 3 activity.

Activity

The Module 3 activity was a coding and data visualization experiment using observations from March 14, 2025, when a significant dust storm impacted Oklahoma alongside wind gusts near 80 mph and wildfires that forced evacuations in central parts of the state. Since previous studies have emphasized the importance of communicating climate impacts with locally relevant examples (Shwom et al., 2017; Monroe et al., 2019; Ramanathan et al., 2019; Staffen et al., 2025), this recent event was chosen to demonstrate how aerosol events have impacted Oklahoma specifically, and how Oklahoma Mesonet observations can be used to visualize aerosol impacts.



Figure 4.7: Interface of the Google Colab environment.

To allow students to easily visualize Mesonet observations from the March 14 event, a Google Colab notebook was developed. Google Colab allows anyone with a Google account to write and execute Python scripts from a web browser and enables easy sharing (Fig. 4.7). The notebook was developed such that all file processing and plotting scripts were pre-written, so that the main task for students was to run different sections of the code and produce figures. Additionally, in the final two sections of the code, students were encouraged to make slight adjustments to the code to plot different variables, only needing to edit variable names and axis labels. This approach was taken so that the focus of the activity would be on data interpretation and understanding

the science itself, rather than on writing the code. This was also done to accommodate students with varying degrees of coding experience and prevent significant time spent debugging code during the activity.

To demonstrate the impact of the March 14 dust event through Mesonet observations, the code first produced a figure comparing solar radiation at the surface between March 14 and March 11 (Fig. 4.8). March 11 was a sunny, clear-sky day in Oklahoma, and was included so that students would have a comparison point to determine how observations were different during the dust event. After this, the same plotting code was repeated and students were encouraged to create figures of other variables (air temperature, wind speed, relative humidity, etc.) to compare similarities and differences between these two days.

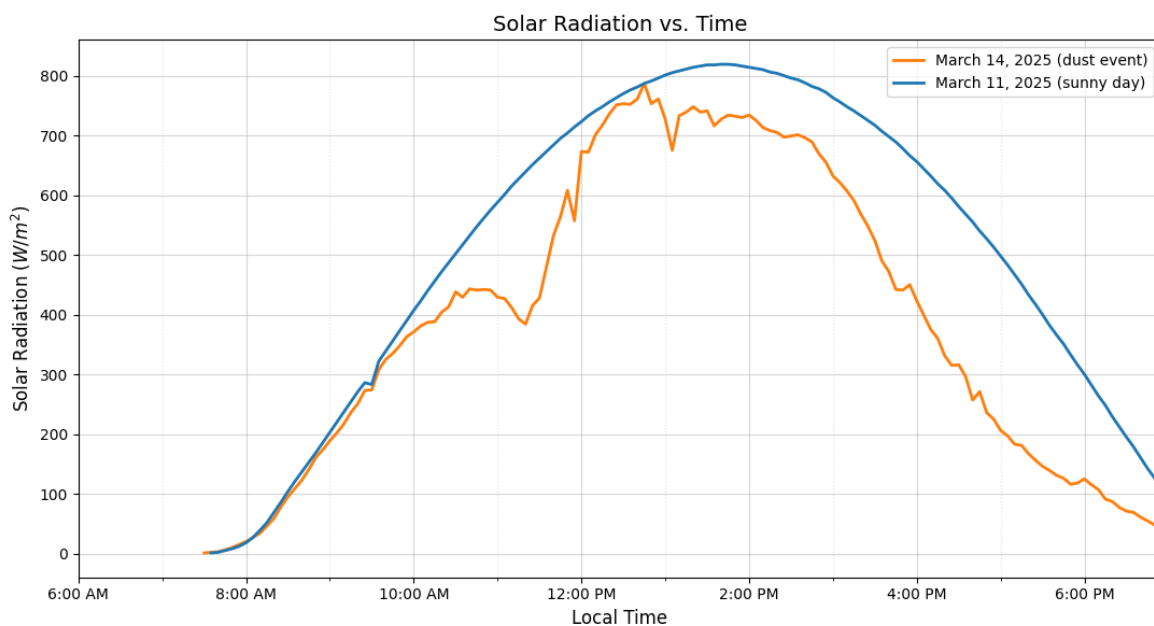


Figure 4.8: Time series of solar radiation observed by the Norman, OK Mesonet station from March 14, 2025 (dust event) in orange and March 11, 2025 (sunny day) in blue.

The March 14 and March 11 comparison emphasizes the impact of dust aerosols on observed solar radiation at the surface, reinforcing the learning objectives for Module 1

(Fig. 4.1). To incorporate Module 2 learning objectives and the role of clouds in Earth's energy budget, the code creates a final solar radiation time series, adding observations from March 8, 2025, which was a consistently overcast day across Oklahoma (Fig. 4.9). Students were then asked to create a figure comparing the air temperature time series between these three days (Fig. 4.10). The goal of this section was to emphasize that both aerosols and clouds impact solar radiation and air temperature at the surface, and they do so at different magnitudes. While the dust event was significant and resulted in substantial decreases in solar radiation, the overcast day reduced solar radiation and air temperature even further. These days were chosen within the span of a week to eliminate significant seasonal variation so that the main meteorological differences were attributable to the presence of dust aerosols and a widespread cloud layer.

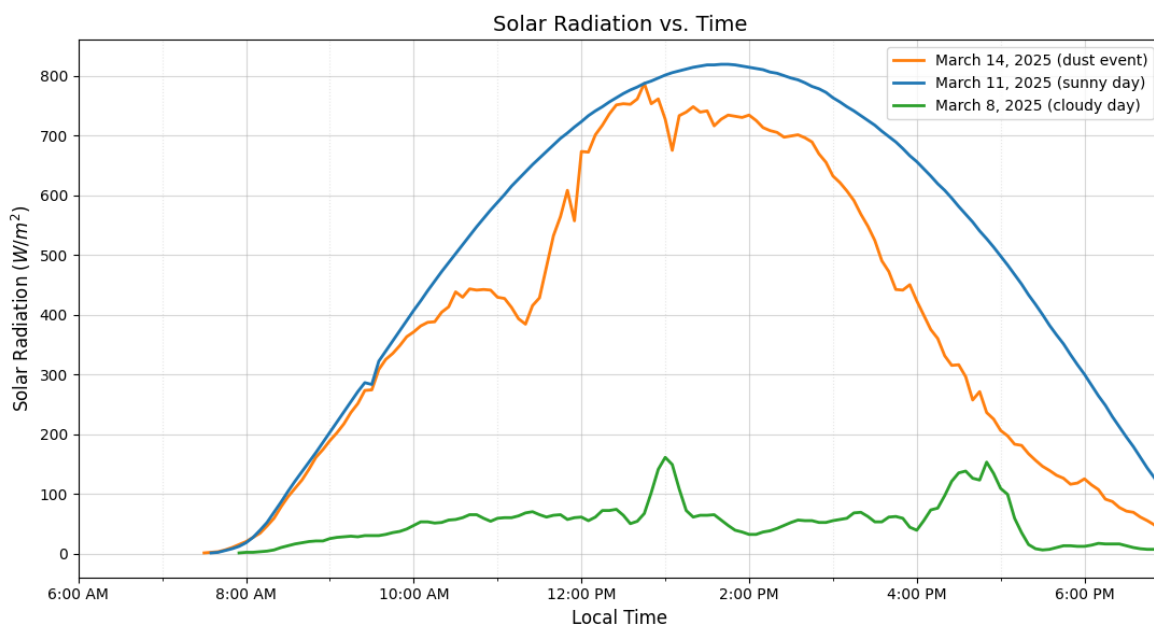


Figure 4.9: Time series of solar radiation observed by the Norman, OK Mesonet station from March 14, 2025 (dust event) in orange, March 11, 2025 (sunny day) in blue, and March 8, 2025 (cloud day) in green.

Reflection Question Worksheet

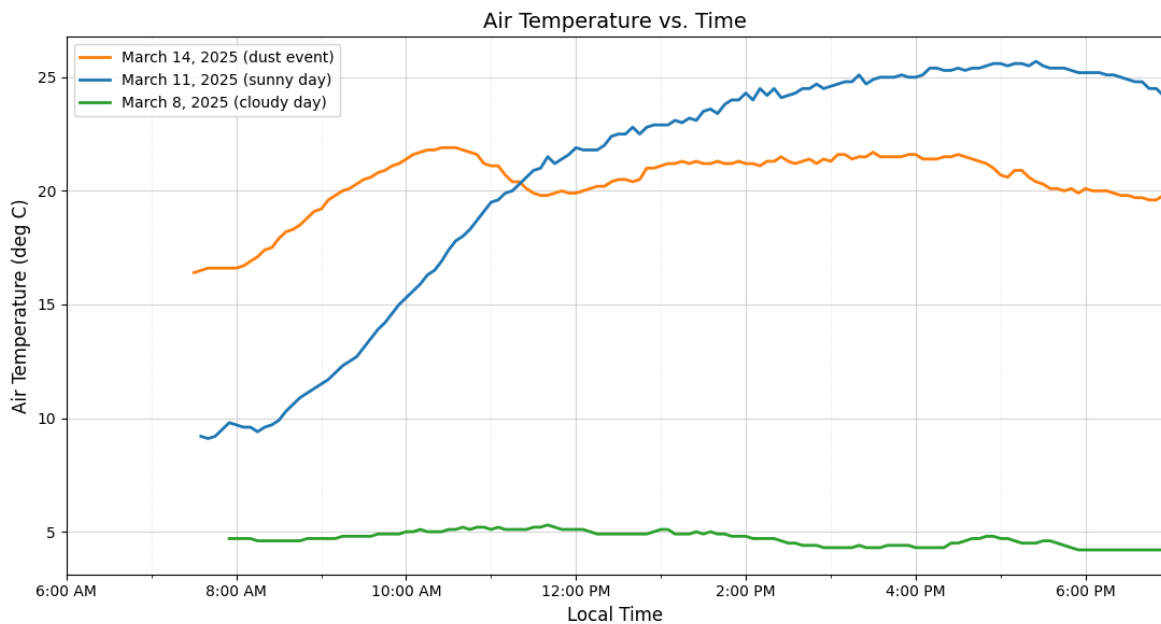


Figure 4.10: Time series of air temperature observed by the Norman, OK Mesonet station from March 14, 2025 (dust event) in orange, March 11, 2025 (sunny day) in blue, and March 8, 2025 (cloud day) in green.

The reflection questions for this activity included the following:

1. How did solar radiation measured at the ground differ during the March 14 dust event compared to the sunny day on March 11?
2. Based on the time series of solar radiation, at what time do you think the most concentrated amount of dust came through Norman?
3. Strong winds associated with this dust event also caused wildfires. What are some possible impacts of having large amounts of both smoke and dust aerosols in the atmosphere at the same time?
4. Between the dust event (March 14), the sunny day (March 11), and the cloudy day (March 8), which resulted in the least amount of solar radiation reaching the ground?

- (a) How does air temperature compare between these three days?

4.5 Participants and classroom implementation

The classroom visit took place at Norman High School (NHS) in the four sections of the Meteorology elective course from April 28-30, 2026. There were 98 students enrolled across these four sections, with most students in 11th and 12th grade, and a few in 10th grade.

4.5.1 Reception of activities

Before analyzing pre- and post-survey results and reflection question responses, I want to provide context by discussing a few differences between the description of the activities (Sect. 4.4) and the reality of how they were implemented and received in the classroom. One of the main goals of this project was to determine which aspects worked well, and which should be adjusted for future application. While some suggestions and considerations will be discussed here, additional suggestions for future improvement, based on the assessment of survey results and reflection question responses, will be discussed in Sect. 4.7.

First, the Life Cycle of an Aerosol board game and Building a Cloud activity both required more time to explain the rules and directions than originally anticipated. Students generally had enough time to complete both activities, but allotting more time for explanations and instructions would have been beneficial. This also resulted in less time for students to complete the reflection question worksheets at the end of each class. With the Life Cycle of an Aerosol board game, the tallying of scattering and absorption “points” was sometimes confusing for students, as well as how to keep track of increasing their aerosol size within the cloud zone. It may be helpful in future

iterations to simplify these aspects of the game or find a better way to describe them. The weather cards were intended to communicate how aerosols can be transported in the atmosphere or removed via wet and dry deposition. However, due to confusion with other aspects of the game and for the sake of time, this meaning of the weather cards was not discussed with the students or explained in detail.

Additionally, while I ensured before the classroom visit that all students had laptop access and Google accounts, they were unable to access Google Colab on their school-issued laptops for the doing activity, as it is considered an 18+ service. Therefore, I worked through the different steps that I had intended for the students to complete with my laptop connected to the classroom projector. This meant that students completed the reflection question worksheets as I went through each question one at a time. Therefore, responses for the Day 3 reflection questions may have more similar responses between students than for Days 1 and 2.

4.6 Survey results and reflection worksheet answers

Survey and reflection question responses were analyzed for students who consented to be part of the study, took both the pre- and post-surveys, and received parental permission to be part of the study. While approximately 75 students took both surveys and around 50 parents gave permission for their child's data to be used, all three of these qualifications were met for 36 students. Therefore, the results in this section represent responses from 36 students, approximately one-third of the total enrollment of the course. It is also important to note that 3 students only partially completed either the pre- or post-survey, and varying numbers of students completed and turned in a reflection question worksheet on any given day. Therefore, for any given analysis in this section, the maximum number of students represented is 36. When the number of

responses is lower for the survey analyses, this will be mentioned in the figure captions.

4.6.1 Module 1 Results

The distribution of pre- and post-survey results for Module 1 is shown in Fig. 4.11. This figure, and those like it in Sections 4.6.2-4.6.4, show the percent of students who selected each answer option, such that the sum of all blocks is equal to 100%. The “neither agree nor disagree” block is shown as centered on zero, such that the total percentage of students who selected this option is reflected as the sum of the portions on either side of the zero line. In these figures the survey statements are re-ordered and grouped into true/neutral on the top and false on the bottom, separated via the dashed line. This is done to enable easier understanding and interpretation of the results. Additionally, results from the pre-survey are shown as the top bar for each statement, while results from the post-survey are shown directly below in the hatched bar.

When looking at the shift in responses between the pre- and post-surveys for Module 1, we find a general improvement, or shift towards the right answers, for all statements except M1.3 and M1.3. Although not a major change, responses shift slightly towards disagree for the true statement of “aerosol indirect effects describe their interactions with clouds.” The statement that “all atmospheric aerosols have the same ability to form cloud droplets” saw fewer neutral responses in the post-survey, but the total distribution of responses did not shift significantly towards either disagree or agree. The misunderstanding of the aerosol indirect effect is due to the fact that when reviewing the lecture slides to look for any phrasing that seemed too jargon-heavy, I removed “direct effects” and “indirect effects,” having forgotten that two survey questions referenced these terms. The lack of a shift in responses to the cloud droplet nucleation statement may indicate that this was not adequately communicated by the Life Cycle of an

Aerosol board game, or perhaps that the wording of the statement was unclear.

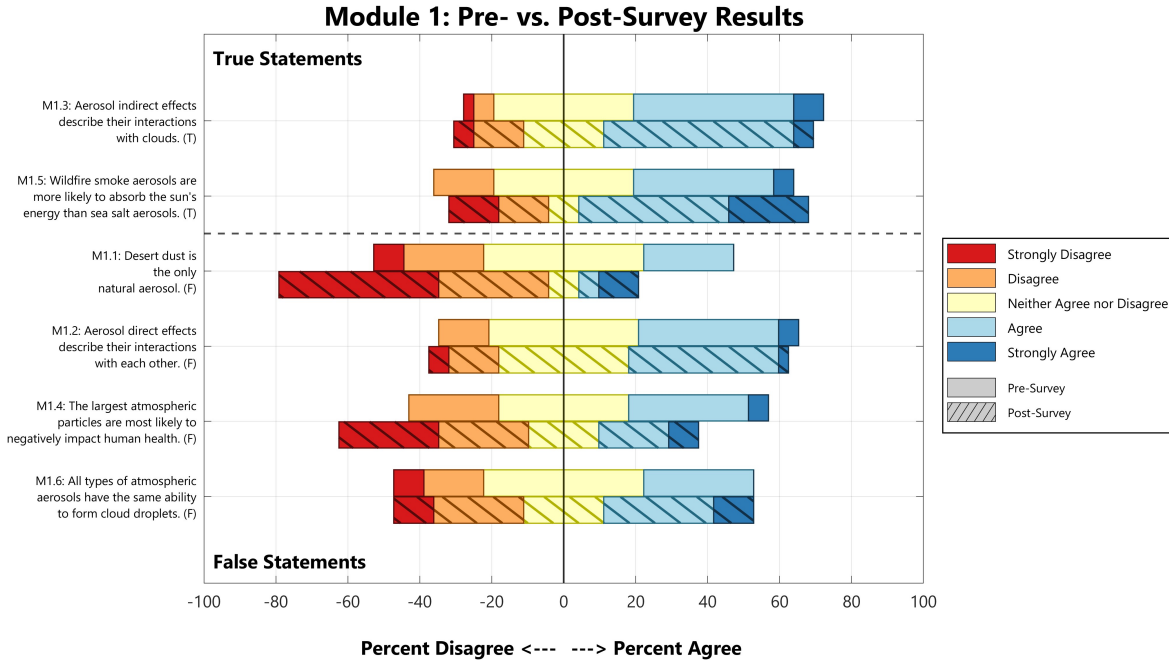


Figure 4.11: Distribution of responses from the pre- (solid bars) and post-surveys (hatched bars) for Module 1 survey statements. The horizontal dashed line separates true from false statements.

In addition to the general survey trends shown in Fig. 4.11, we can also look at trends on an individual level. The chart in Fig. 4.12 shows the frequency of various pre- and post-survey response combinations for each Module 1 survey statement. The top three rows show the frequency of students with the same response in the pre- and post-survey, split into correct, neutral, and wrong responses. If a statement was true, then a response of “agree” or “strongly agree” is counted as correct, while a response of “disagree” or “strongly disagree” is counted as wrong, and vice versa for false statements. The middle three rows show a count of students whose answers became more correct in the post-survey, indicating improved understanding, and the bottom three rows show a count of students whose answers were less correct in the post-survey, indicating an apparent decrease in understanding. For each statement, the green shading indicates the most frequent response combination, while the purple

shading indicates the second most frequent response combination.

As alluded to in Fig. 4.11, students most frequently responded to M1.2 incorrectly or neutrally in both surveys, while 10 students answered more correctly in the post-survey and 8 answered less correctly. There was no overall shift in responses seen in Fig. 4.11 for M1.6, with many students responding with the same answer in both surveys and several whose responses shifted from neutral to incorrect. Possible reasons for the misunderstanding associated with this statement will be further discussed in Sect. 4.7. Statements M1.1 and M1.4 had many students who answered neutrally or incorrectly in the pre-survey but correctly in the post-survey, showing mostly improved understanding. Statements M1.3 and M1.5 are primarily characterized by students who answered correctly in both surveys, with several who shifted from neutral to correct answers in the post-survey.

		M1.1	M1.2	M1.3	M1.4	M1.5	M1.6
no change in answer	correct → correct	7 (19.4%)	3 (8.3%)	13 (36.1%)	4 (11.1%)	12 (33.3%)	5 (13.9%)
	neutral → neutral	2 (5.6%)	7 (19.4%)	3 (8.3%)	4 (11.1%)	1 (2.8%)	5 (13.9%)
	wrong → wrong	1 (2.8%)	8 (22.2%)	1 (2.8%)	4 (11.1%)	2 (5.6%)	5 (13.9%)
understanding ↑	neutral → correct	12 (33.3%)	2 (5.6%)	7 (19.4%)	7 (19.4%)	8 (22.2%)	4 (11.1%)
	wrong → correct	8 (22.2%)	2 (5.6%)	1 (2.8%)	8 (22.2%)	3 (8.3%)	4 (11.1%)
	wrong → neutral	0 (0%)	6 (16.7%)	1 (2.8%)	2 (5.6%)	1 (2.8%)	2 (5.6%)
understanding ↓	correct → wrong	3 (8.3%)	2 (5.6%)	2 (5.6%)	4 (11.1%)	3 (8.3%)	3 (8.3%)
	correct → neutral	1 (2.8%)	0 (0%)	4 (11.1%)	1 (2.8%)	1 (2.8%)	1 (2.8%)
	neutral → wrong	2 (5.6%)	6 (16.7%)	4 (11.1%)	2 (5.6%)	5 (13.9%)	7 (19.4%)

Figure 4.12: Frequency of various pre- and post-survey response combinations for individual students for each Module 1 survey statement. These percentages are calculated from 36 complete survey responses. Boxes shaded with green indicate the most frequent response combination for the corresponding question, while purple shading indicates the second most frequent response combination.

Common themes from the Module 1 reflection questions and a few selected quotes

are shown in Fig. 4.13. One pattern that initially stands out is the number of students who gave an example of an aerosol type when asked about the kinds of properties that impact cloud droplet nucleation and absorption/scattering. While several other students did mention aerosol size and composition, the term “property” was less familiar to others. Another interesting trend was the references to absorption and scattering in the first question regarding aerosol ability to form cloud droplets. It may be that the inclusion of both aerosol direct and indirect effect concepts in the board game was more confusing than it was beneficial. Even still, the selected quotes highlight examples of students understanding these concepts at a level beyond what was discussed during the class period. Other examples of properties listed as impacting cloud droplet nucleation included surface area, quantity, temperature, and “if they like water or not.” For the question regarding properties that impact whether an aerosol primarily scatters or absorbs the sun’s energy, one student hypothesized about the types of bonds in molecules, another mentioned aerosol location and composition (anthropogenically-sourced), while another mentioned the color and hue of the aerosol.

4.6.2 Module 2 Results

The distribution of pre- and post-survey results for Module 2 is shown in Fig. 4.14. Here we find that there is a general shift in post-survey responses towards the correct answers for all statements. The biggest differences between pre- and post-survey results occur for M2.1, M2.3, and M2.5. That is, students seemed to best understand the ingredients necessary for cloud formation, as well as the relationship between cloud droplet number concentration and size when more aerosols are introduced. The smallest shifts towards more correct responses are seen for M2.2 and M2.4. Though there was improved understanding of how an increase in cloud droplet number concentration results in smaller droplets, the statement of “clouds can be characterized by the num-

Question	Common Themes	Selected Quotes
What aerosol properties do you think might cause some to become cloud droplets easier than others?	- Listed a type of aerosol: 8 - Listed something about aerosol size: 14 - Listed something about composition: 2 - Listed something about absorption/scattering: 6	<i>"Surface area, quantity, temperature, etc."</i> <i>"If they like water or not"</i>
Based on the final count of scattering and absorption points, which aerosol type is most likely to cause cooling? Which is most likely to cause warming?	- Smoke = warming (24); sea salt = cooling (21) - Listed one aerosol type but didn't directly tie to questions: 6	
What aerosol properties do you think might cause some aerosols to mostly absorb the sun's energy and other aerosols to mostly scatter the sun's energy?	- Listed a type of aerosol: 8 - Said something about aerosol size: 5 - Said something about composition or color or albedo: 14 - Mentioned something about which types absorb/scatter more: 8	<i>"Maybe the types of bonds in specific molecules?"</i> <i>"Where they are located at, if they are human made?"</i> <i>"Color and hue of the aerosol"</i>

Figure 4.13: Common themes and selected quotes from Module 1 reflection question answers. The number after each of the most common themes indicates the number of students whose response fits into a given theme.

ber of individual droplets and droplet sizes" did not see a shift of the same magnitude between the pre- and post-survey. There was also only a small improvement in understanding of the statement "the amount of the sun's energy scattered by a cloud changes when the number of cloud droplets changes." While scattering of solar radiation by cloud droplets was discussed after the Building a Cloud activity, there was not a direct connection in the activity to this concept. Jars of small and large white-colored beads were used to demonstrate and discuss differences in brightness for different cloud droplet sizes, so perhaps a statement related to how brightness is different for large and small cloud droplets may have been better understood.

In terms of the trends of individual responses between both surveys, Fig. 4.15 shows that for M2.1, M2.3, and M2.5, many students who responded incorrectly or neutrally in the pre-survey responded correctly in the post-survey. M2.2, M2.4, and M2.6 had many students who answered correctly in both surveys, which could be another reason

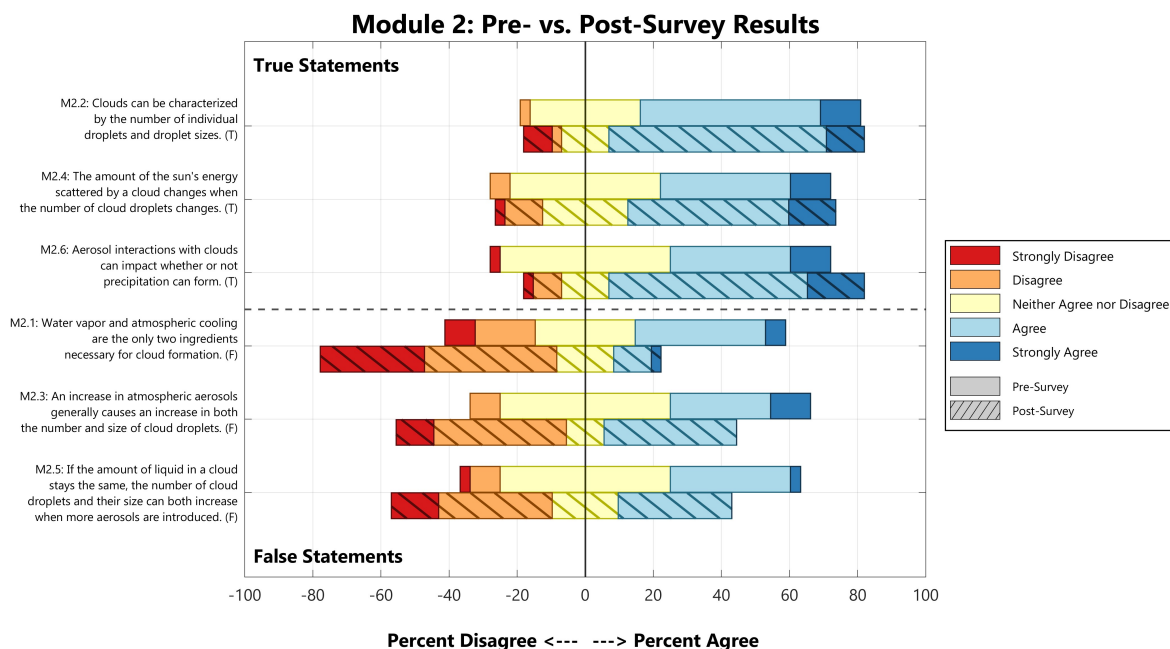


Figure 4.14: Distribution of responses from the pre- (solid bars) and post-surveys (hatched bars) for Module 2 survey statements. The horizontal dashed line separates true from false statements.

why the shift is minimal when comparing results between both surveys. Interestingly, though responses for M2.3 generally shift more towards the correct response in the post-survey, 20.6% of students who initially responded neutrally had an incorrect answer in the post-survey. Lastly, Fig. 4.14 shows a noticeable decrease in the number of neutral responses for all statements in the post-survey, and Fig. 4.15 shows that while some of these neutral responses stay neutral or become incorrect in the post-survey, in most cases these neutral responses become correct in the post-survey.

Common themes and a few selected quotes from two of the Module 2 reflection questions are given in Fig. 4.16. The other three questions on this worksheet had more straightforward answers that most students answered correctly, while the questions shown here were a bit more open-ended. Something discussed as part of the Building a Cloud activity and in the Module 2 lecture slides was the cloud lifetime effect, where an increase in aerosol concentration can form a large number of small cloud droplets

		M2.1	M2.2	M2.3	M2.4	M2.5	M2.6
no change in answer	correct → correct	8 (23.5%)	17 (50%)	2 (5.9%)	12 (35.3%)	2 (5.9%)	15 (44.1%)
	neutral → neutral	1 (2.9%)	3 (8.8%)	2 (5.9%)	6 (17.6%)	4 (11.8%)	5 (14.7%)
	wrong → wrong	3 (8.8%)	0 (0%)	5 (14.7%)	2 (2.9%)	5 (14.7%)	0 (0%)
understanding ↑	neutral → correct	7 (20.6%)	8 (23.5%)	8 (23.5%)	8 (23.5%)	9 (26.5%)	10 (29.4%)
	wrong → correct	9 (26.5%)	1 (2.9%)	7 (20.6%)	0 (0%)	5 (14.7%)	1 (2.9%)
	wrong → neutral	3 (8.8%)	0 (0%)	2 (5.9%)	1 (2.9%)	3 (8.8%)	0 (0%)
understanding ↓	correct → wrong	0 (0%)	3 (8.8%)	1 (2.9%)	3 (8.8%)	2 (5.9%)	1 (2.9%)
	correct → neutral	1 (2.9%)	2 (5.9%)	0 (0%)	2 (5.9%)	0 (0%)	0 (0%)
	neutral → wrong	2 (5.9%)	0 (0%)	7 (20.6%)	1 (2.9%)	4 (11.8%)	2 (5.9%)

Figure 4.15: Frequency of various pre- and post-survey response combinations for individual students for each Module 2 survey statement. These percentages are calculated from 34 complete responses. Boxes shaded with green indicate the most frequent response combination for the corresponding question, while purple shading indicates the second most frequent response combination.

Question	Common Themes	Selected Quotes
How can a large number of aerosols prevent a cloud from forming rain?	- Said something about water being taken/absorbed: 5 - Mentioned droplets getting smaller but did not elaborate further: 5 - Small droplets not heavy enough to fall as rain: 7 - Mentioned something about warming/cooling/absorbing sunlight: 5	<i>"It absorbs all of the rain drops"</i> <i>"Because there's too much energy and can't form droplets"</i> <i>"The formation of smaller clouds prevent rain"</i>
Can you think of any other ways that clouds could cause cooling or warming?	- Mentioned that rain causes cooling: 5 - Mentioned something about albedo/sun reflectance: 3 - Mentioned something about how clouds covering the sun causes cooling: 3	<i>"They can cause warming by reflecting IR rays back into earths energy budget"</i> <i>"Storm clouds cause cooling because they're darker and absorb more warmth/sunlight"</i>

Figure 4.16: Common themes and selected quotes from Module 2 reflection question answers. The number after each of the most common themes indicates the number of students whose response fits into a given theme.

which do not form precipitation as easily, enabling a longer cloud lifetime. Responses to the question related to this were mixed. While several students correctly mentioned

something about the small size of the cloud droplets, or how smaller droplets would not be heavy enough to form precipitation, others mentioned some impact of warming, cooling, or sunlight being absorbed. This may be another example where different concepts related to the same activity were confused. Another common theme in the responses was a mention of water drops being absorbed or “taken,” without much elaboration. Another question asked if students could think of any other ways that clouds can cause cooling or warming, to which several students mentioned cooling that happens as a result of precipitation. Others mentioned cloud albedo, as discussed during the lecture, and a few mentioned that clouds covering the sun causes cooling at the surface.

4.6.3 Module 3 Results

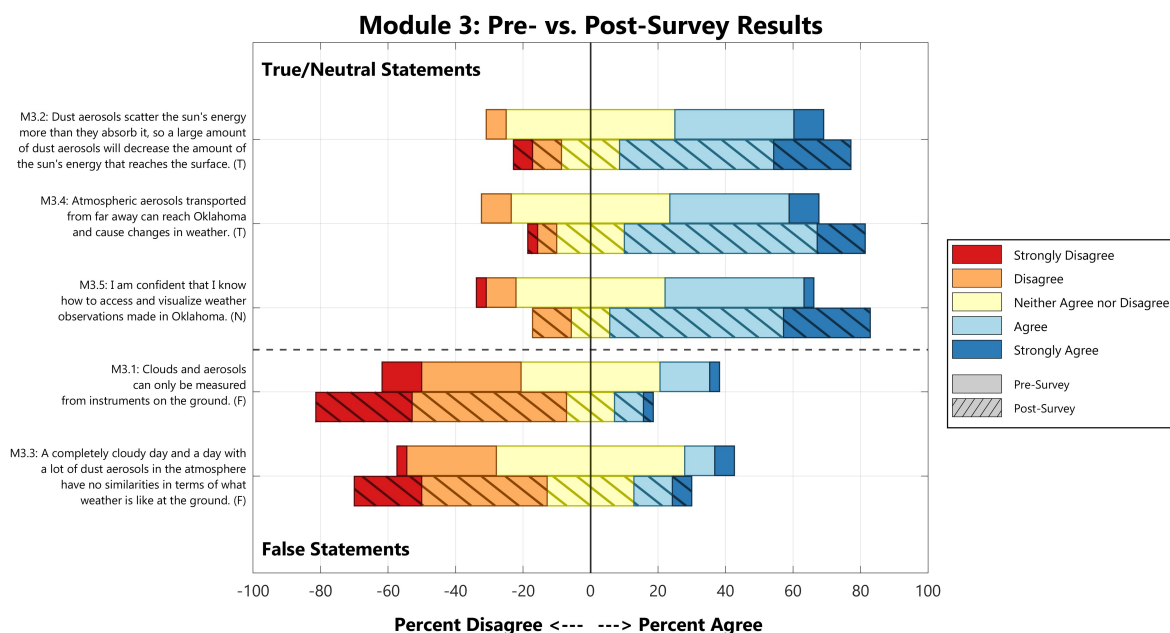


Figure 4.17: Distribution of responses from the pre- (solid bars) and post-surveys (hatched bars) for Module 3 survey statements. The horizontal dashed line separates true from false statements.

The distribution of pre- and post-survey results for Module 3 is shown in Fig. 4.17.

Here again we find a general shift towards more correct answers for all statements. Post-survey responses for M3.3, M3.4, and M3.5 appear to shift by similar magnitudes, while M3.1 has the largest change and M3.2 has the smallest. M3.2 is the longest statement, so it may be that the wording was more confusing for students compared to the other statements. Regardless, the comparable magnitude in improvement across each of these statements may also speak to an added benefit in having gone through the activity for this module together rather than individually due to technology issues.

		M3.1	M3.2	M3.3	M3.4	M3.5
no change in answer	correct → correct	13 (38.2%)	10 (29.4%)	7 (20.6%)	12 (35.3%)	14 (41.2%)
	neutral → neutral	4 (11.8%)	3 (8.8%)	7 (20.6%)	5 (14.7%)	3 (8.8%)
	wrong → wrong	1 (2.9%)	0 (0%)	1 (2.9%)	1 (2.9%)	1 (2.9%)
understanding ↑	neutral → correct	9 (26.5%)	12 (35.3%)	10 (29.4%)	11 (32.4%)	9 (26.5%)
	wrong → correct	4 (11.8%)	1 (2.9%)	2 (5.9%)	2 (5.9%)	3 (8.8%)
	wrong → neutral	1 (2.9%)	1 (2.9%)	2 (5.9%)	0 (0%)	0 (0%)
understanding ↓	correct → wrong	1 (2.9%)	3 (8.8%)	3 (8.8%)	2 (5.9%)	0 (0%)
	correct → neutral	0 (0%)	2 (5.9%)	0 (0%)	1 (2.9%)	1 (2.9%)
	neutral → wrong	1 (2.9%)	2 (5.9%)	2 (5.9%)	0 (0%)	3 (8.8%)

Figure 4.18: Frequency of various pre- and post-survey response combinations for individual students for each Module 3 survey statement. These percentages are calculated from 34 complete responses. Boxes shaded with green indicate the most frequent response combination for the corresponding question, while purple shading indicates the second most frequent response combination.

Unlike Modules 1 and 2, the individual survey trends for Module 3 show that cases of students shifting towards a less correct response from the pre- to post-survey were much less frequent. Rather, for M3.1, M3.2, and M3.4, the top two most frequent cases were of students either answering correctly in both surveys or answering neutrally in the pre-survey and correctly in the post-survey. Only in M3.3 was there also a relatively high percentage of students who answered neutrally in both surveys. In M3.5, which

asked students to agree or disagree that they were confident in knowing how to access and visualize weather observations made in Oklahoma, answers of “agree” and “strongly agree” are counted as “correct,” while “disagree” and “strongly disagree” are counted as “wrong.” Though this statement is not factually true or false, ideally more students would agree with the sentiment in the post-survey compared to the pre-survey. In these responses we see that several students indicated confidence in both surveys, while several also shifted from a neutral response to higher confidence at the end. While only the top two most frequent survey response shifts are highlighted in Fig. 4.18 and other figures like it, it is also worth acknowledging that even though many students saw improvement, seven students answered less correctly on M3.2 and five answered less correctly on M3.3. Therefore, there are still places where the lecture material and activities were not successful for all students, which should not be overlooked.

Question	Common Themes	Selected Quotes
How did solar radiation measured at the ground differ during the March 14 dust event compared to the sunny day on March 11?	- Solar radiation was lower during the dust event: 25 - Solar radiation was higher during the dust event 2	<i>“The dust blocked the radiation so less got through which made it colder”</i> <i>“The solar radiation was more stable compared to the dust”</i>
Strong winds associated with this dust event also caused wildfires. What are some possible impacts of having large amounts of both smoke and dust aerosols in the atmosphere at the same time?	- Bad air quality (14)/trouble breathing (2) - Decreased visibility: 4 - Dry/less humid: 5 - Scattering/less sunlight: 6 - Fog/smog: 2 - Wind/faster transport of aerosols: 10	<i>“The wind was stronger which helped start the wildfires and help it spread faster”</i> <i>“Plant life starts to decrease, soil in the ground starts to dry up and overflow”</i> <i>“Humidity, wind, and how dry it is, high AOD, tornados, bad storms”</i>

Figure 4.19: Common themes and selected quotes from Module 3 reflection question answers. The number after each of the most common themes indicates the number of students whose response fits into a given theme.

Similar to Module 2, three questions on this worksheet had more straightforward answers that most students answered correctly, especially since the questions were

discussed as a class. Common themes and a few selected quotes from the two slightly more open-ended questions from the Module 3 worksheet are given in Fig. 4.19. First, 25 students who answered the question regarding the impact of the dust event on solar radiation measured at the surface answered correctly, while only 2 answered incorrectly. One interesting response to this question, and something that multiple students pointed out during the class session, was that the solar radiation time series was more stable (the line was smoother) for the sunny day than the dust event. While I had planned to focus only on the magnitude differences, it was encouraging to see that students noticed other differences in the observations between these days. When asked about possible impacts resulting from having significant amounts of smoke and dust aerosols in the atmosphere at the same time, the most common answer was about decreased air quality. Other common themes in the responses were decreased visibility, increased scattering and less sunlight reaching the surface, and the possible formation of fog/smog. Others mentioned less humid conditions and strong winds transporting aerosols. These were referenced when discussing the driving factors and characteristics of this event, and are not direct impacts of high aerosol loading. This may be another scenario in which different wording of the question could have improved student understanding.

4.6.4 Module 4 Results

The distribution of pre- and post-survey results for Module 4 (Fig 4.20) again shows shifts towards more correct answers and/or higher confidence in knowledge for all statements in the post-survey. As a reminder, while some of these statements may have been discussed or alluded to during the other modules (M4.1 and M4.2), there was no separate lecture or activity devoted to discussing this set of statements. Rather, one of the goals of this survey section was to see if discussing the mechanics of aerosol direct and indirect effects and their influence on temperature would enable students to

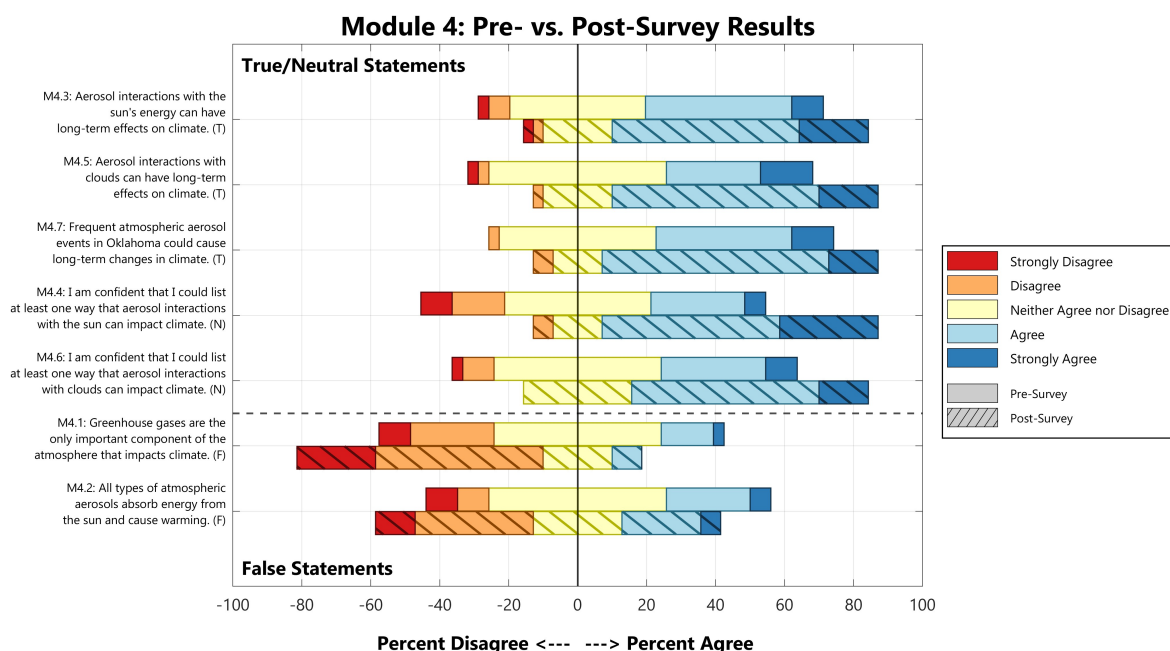


Figure 4.20: Distribution of responses from the pre- (solid bars) and post-surveys (hatched bars) for Module 4 survey statements. The horizontal dashed line separates true from false statements.

correctly hypothesize about long-term climate effects due to aerosol and cloud processes (Fig. 4.1).

The largest shifts in responses appear to be for M4.1 and M4.4. Looking at M4.3 and M4.4 together, it appears that in the pre-survey students generally agreed that aerosol interactions with the sun's energy could have long-term effects on climate, but fewer students were confident that they could list a way in which this could occur. In the post-survey, the distributions for these questions are more similar, emphasizing that more students agreed that they could list a way in which aerosol interactions with the sun's energy could impact climate. The same can be said for M4.5 and M4.6, but to a slightly lesser extent, implying that students understood the climate impacts of aerosol direct effects better than indirect effects.

Individual survey trends for Module 4 are shown in Fig. 4.21, and they are generally similar to those in Module 3 (Fig. 4.18). For most questions, the most common case

		M4.1	M4.2	M4.3	M4.4	M4.5	M4.6	M4.7
no change in answer	correct → correct	11 (33.3%)	3 (9.1%)	15 (45.5%)	11 (33.3%)	13 (39.4%)	11 (33.3%)	16 (48.5%)
	neutral → neutral	6 (18.2%)	6 (18.2%)	5 (15.2%)	4 (12.1%)	7 (21.1%)	9 (27.3%)	4 (12.1%)
	wrong → wrong	2 (6.1%)	5 (15.2%)	0 (0%)	1 (3.1%)	0 (0%)	0 (0%)	1 (3.0%)
understanding ↑	neutral → correct	10 (30.3%)	8 (24.2%)	8 (24.2%)	9 (27.3%)	10 (30.3%)	7 (21.2%)	10 (30.3%)
	wrong → correct	3 (9.1%)	3 (9.1%)	3 (9.1%)	7 (21.2%)	2 (6.1%)	4 (12.1%)	0 (0%)
	wrong → neutral	1 (3.1%)	2 (6.1%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)
understanding ↓	correct → wrong	0 (0%)	2 (6.1%)	0 (0%)	0 (0%)	1 (3.0%)	0 (0%)	0 (0%)
	correct → neutral	0 (0%)	1 (3.1%)	2 (6.1%)	0 (0%)	0 (0%)	2 (6.1%)	1 (3.0%)
	neutral → wrong	0 (0%)	3 (9.1%)	0 (0%)	1 (3.1%)	0 (0%)	0 (0%)	1 (3.1%)

Figure 4.21: Frequency of various pre- and post-survey response combinations for individual students for each Module 4 survey statement. These percentages are calculated from 33 complete responses. Boxes shaded with green indicate the most frequent response combination for the corresponding question, while purple shading indicates the second most frequent response combination.

was that students answered correctly (or confidently) in both the pre- and post-survey. In most cases, the second most common occurrence was that students who initially answered neutrally responded more correctly (or confidently) in the post-survey. For M4.2 and M4.6, the second most common occurrence was that students answered neutrally in both the pre- and post-survey.

4.7 Reflection and discussion

While some logistics that would benefit from future improvement were evident during the classroom visit, the survey results also give insight into which aspects were successful and which would most benefit from changes. First, the combination of aerosol direct and indirect effect concepts in the Module 1 board game may have been too much for one activity. Given the number of clarifying questions and the time needed to explain the game, it might have been more straightforward without the direct effect

aspect (the counting of scattering and absorption points). This is also observed in the reflection question responses, where several students mentioned absorption and scattering when asked about properties relevant to CCN nucleation. A similar pattern was found in Module 2, when students mentioned the reflection or absorption of sunlight in response to the question regarding how aerosols can impact precipitation susceptibility. Discussing both the impact of aerosols on cloud albedo and cloud lifetime/precipitation formation may have been too many details for one lecture, and focusing on only one of these topics may have made the lesson easier to understand.

Another possible issue with the board game may be reflected in the lack of improvement for the survey statement that “all types of atmospheric aerosols have the same ability to form cloud droplets.” Since the game was played until all students reached the cloud zone and grew to their goal size to nucleate as a CCN, it might have come across that all aerosol types had the same ability to *eventually* form a cloud droplet. If this question had been worded in a way that instead targeted how quickly or easily some aerosol types were able to nucleate as CCN compared to others, the distribution of responses in the post-survey may have looked different.

In spite of these potential issues with the board game, survey responses in Module 2 speak to the overall benefit of using hands-on activities to communicate complicated topics. More specifically, the biggest shifts towards more correct answers in Module 2 were seen for M2.1, M2.3, and M2.5, statements that had to do with necessary ingredients for cloud formation and the relationship between number and size of cloud droplets (Fig. 4.14). These concepts were directly targeted through the Building a Cloud activity. On the other hand, the statements regarding cloud albedo and precipitation formation saw less of an improvement in the post-survey responses. These concepts were discussed as part of the post-activity lecture, but were not directly incorporated in the activity itself. Therefore, this serves as an example of the benefit of hands-on

activities in communicating science concepts, as opposed to only verbally mentioning them.

While some of the lack of improvement in post-survey responses is likely attributable to flaws in the activities and lecture slides, some may also be a result of the wording of the survey statements themselves. While I tried to remove all scientific jargon from the survey statements and reflection questions, there were still areas where wording became an issue. For example, after the first day, I learned from the classroom teacher that the word “property” is not familiar or well-understood for many students. This explains why in the two Module 1 reflection questions that ask about aerosol properties, there were several students who listed a type of aerosol as opposed to something about the size or composition. I suspect that something similar may have happened with the word “characterized” in M2.2. While students were generally correct in their responses to M2.3 and M2.5 regarding the relationship between cloud droplet size and number concentration, there was a nearly negligible shift in responses for the statement that “clouds can be characterized by the number of individual droplets and droplet sizes.” Since the other questions show a general understanding of the relationship between number and size of cloud droplets, perhaps referring to these as characteristics of clouds was not well-understood. This is a key example of the importance of climate change communication, and science communication in general, being a collaborative effort. While climate education-focused studies have found it beneficial to involve scientists in the classroom, a scientist, or any outside speaker, is not going to know the students well enough to be able to predict what will and will not be understandable. This should serve to emphasize the importance of treating such outreach activities as a collaborative effort with the teachers already involved and familiar with the students and their background knowledge.

Although students were unable to do the coding activity in Module 3 individually, as

initially planned, there are generally fewer instances of students answering less correctly on the post-survey statements for Module 3 compared to Modules 1 and 2. While I expected to see more correct answers on the Module 3 reflection question worksheet due to going through the activity as a class, it seems there were also differences in the surveys that were done individually after the classroom visit. This could also indicate that these statements had fewer wording issues than those of other modules. Regardless, there may have been added benefit in guiding the class through the coding activity than if students had completed it individually. If these activities are repeated in the future and a platform can be found that would allow students to interact with the coding activity on their individual laptops, it would be interesting to compare survey results to what was seen here.

Lastly, an assessment of the Module 4 survey responses seems to suggest that student understanding of aerosol interactions with solar radiation was greater than their understanding of aerosol interactions with clouds. The statements with the largest shifts towards improved understanding or confidence in the post-survey are M4.1, “greenhouse gases are the only important component of the atmosphere that impacts climate,” and M4.2, “I am confident that I could list at least one way that aerosol interactions with the sun can impact climate.” On the other hand, the statements with many students who responded neutrally in both surveys are those regarding how aerosol interactions with clouds impact climate and whether all types of atmospheric aerosols can absorb the sun’s energy and cause warming. The small shift in the latter could again be attributable to confusion caused by the board game. However, the small shift in the ACI-related question corroborates the fewer correct answers in ACI-related questions within Modules 1 and 2. Therefore, the area that might need the most improvement in future iterations of these activities are those related to teaching about aerosol impacts on clouds, and specifically the climate relevance of ACI.

While there are several areas where the activities and lessons developed and implemented here can and should be improved in future iterations, responses to 22 out of 24 survey statements saw some degree of improved understanding between the pre- and post-survey when considering responses from 36 students. In many cases, student responses improved or remained the same between surveys, and cases of less correct answers in the post-survey were relatively infrequent. For these three modules only taking place over three class sessions, student responses and improvements generally exceeded expectations. Furthermore, as seen in the results from the Module 4 survey statements (Fig. 4.20 and 4.21), the hypothesis that primarily teaching the mechanisms of aerosol direct and indirect effects would lead to improved understanding of aerosol and cloud impacts on climate was seen to be true. Again, while the concepts in some of the Module 4 survey statements may have been alluded to, the activities and lectures did not include discussion of long-term effects on climate, as I wanted to see if students could make hypotheses and deductions about that on their own. While many students responded correctly about climate implications in the pre-survey, there was a large decrease in neutral responses in the post-survey, most of which became correct answers. Module 4 also featured the lowest percentages of students who answered less correctly (or confidently) in the post-survey compared to the other modules. Therefore, it seems that most students were able to independently connect the concepts taught across the three days to broader climate implications.

4.8 Conclusion

As we move into a warming future, multi-faceted and interdisciplinary approaches are necessary to predict the extent of warming in climate models, to enact policy and change to eradicate worst-case emissions scenarios, and to communicate and educate

the public on climate change and the importance of taking care of our planet. Many tactics have been tested for effectively communicating about climate change to various audiences, which is especially difficult when climate has become a divisive and politicized topic. This project developed a small-scale, community-based approach to educate high school students about the mechanisms by which atmospheric aerosols and clouds interact with the sun's energy and with each other, with a primary hypothesis that by focusing on the scientific mechanisms, student understanding of their potential broader impact on climate would improve.

A set of three modules was developed and implemented over the course of three days within the meteorology elective course at Norman High School. Each module contained a set of lecture slides, a hands-on or data-based activity, and a set of reflection questions. The first day focused on defining and explaining the importance of atmospheric aerosols and their interactions with solar radiation, the second day discussed aerosol interactions with clouds and modulation of cloud properties, and the third day used local observations from a recent dust storm over Oklahoma to give a regionally-relevant example of aerosol impacts on weather. To test the effectiveness of these modules in teaching the defined learning objectives, pre- and post- Likert-response surveys were also distributed to students. The results in this chapter represent survey responses from a maximum of 36 students, which is a little over one-third of the total enrollment of the course. Answers to the reflection questions were also collected and help provide context alongside the survey responses.

When looking at overall trends, student responses improved from the pre- to post-survey for 22 of the 24 statements. When looking at individual student patterns between the pre- and post-surveys, the most frequent pathways were correct responses in both surveys, a neutral response in the pre-survey and correct response in the post-survey, or an incorrect response in the pre-survey and correct response in the pre-survey.

Therefore, students were generally either correct in both surveys or their understanding improved between the pre- and the post-survey. While this is true for several of the survey statements, there were also some where a large percentage of students showed decreased understanding from the pre- to post-survey. For many of these cases, it can be hypothesized that the reason was an issue with the activity, the inclusion of too many topics at once, discrepancies with wording in the survey compared to how the material was taught, or the wording of survey statements and reflection questions being confusing.

Another goal of this project was to determine suggestions for future iterations based on what did go well and what could have been improved. Patterns in the Module 2 survey and reflection question responses suggest that hands-on activities were generally successful at communicating complicated topics, as student understanding improved more for questions regarding the relationship between cloud droplet size and number concentration, which was directly targeted in the activity. Conversely, student understanding of cloud albedo and precipitation susceptibility, which were not included in the activity, did not increase by as large a magnitude.

Another important implication of this work is the need for outreach activities and collaborative efforts between scientists and educators to truly be collaborative. The discussions I had with the classroom teacher in the months leading up to this project were instrumental in developing the activities and lecture slides. However, in light of the IRB approval process timeline, I did not allot as much time to seeking feedback on and refining the reflection questions. Because of this, there was some phrasing that was difficult for students to understand. Discussions with the teacher during the visit of how words such as “property” are often not well-understood by students helped in interpreting some of the patterns in reflection question responses, and it would be beneficial in future iterations to preemptively check wording like this with the classroom

teacher as well.

This chapter can be viewed as one small, specific example of how atmospheric and climate science-related research can be translated into hands-on activities and demonstrations applicable at a high school level. A three-day classroom visit will not solve climate change education in K-12 schools, but it will expose several students to topics and career paths in research that they may not have known about otherwise. Additionally, if repeated and expanded by other students and researchers, more events like this could have a much wider reach. Beyond the implications for student learning and providing supplementary science content and resources for teachers, outreach activities such as this one can also greatly benefit researchers. Often we may get lost in specific, technical, and minute research details, and translating the key concepts of our work into lessons and activities applicable at the K-12 level is an excellent reminder of the motivation behind what we do and why we do it.

Chapter 5

Discussion

The primary implications of the work done in Chapters 2-4 are discussed within their respective chapters and will be summarized together in Chapter 6. This chapter outlines several recent related projects and collaborations to discuss the place of our N_{CCN} and ACI work in a broader scientific context.

5.1 ML-prediction of N_{CCN} from lidar observables

While the analyses presented in Chapter 2 were ongoing, initial findings suggested that N_{CCN} —AOP relationships are characterized by multiple non-linear dependencies. As previously mentioned, the focus of Chapter 2 was to understand and explain the governing factors of observed N_{CCN} —AOP relationships rather than being an effort to further refine the linear approximation method or develop a multivariate regression method. However, one remaining question at the time was the following: *How can we best derive N_{CCN} from lidar observables given the non-linear nature of their relationship?* The inability to relate N_{CCN} and HSRL-2 observables in a straightforward manner was a significant motivating factor for the ML work done by Redemann and Gao (2024). This study used deep learning with fully connected neural networks to predict aerosol absorption and N_{CCN} from HSRL-2 observables and RH and temperature from reanalysis (Fig. 5.1). This method predicts absorption and N_{CCN} with mean relative errors of 21% and 13%, respectively (Redemann and Gao, 2024).

Considered together, these studies each fill gaps that the other cannot. The theoretical calculations study presented in Chapter 2 did not result in a computationally feasible and accurate method for predicting N_{CCN} from remote sensing observations, but used κ -Köhler and Mie theories to investigate the physical driving mechanisms behind N_{CCN} —AOP non-linearity. ML approaches can better handle and model nonlinearities through complex parameterizations, but consequently the underlying physical relationships are generally unclear. The ML approach for predicting N_{CCN} from lidar observables will have significant impacts for the predictive capabilities of observations from the Atmospheric Lidar (ATLID) on the EarthCARE satellite, enabling the creation of a global, vertically resolved N_{CCN} data set for ACI analyses. Undoubtedly, the complementary nature of these physics-based and ML approaches emphasizes the added benefit of using both methods together and not in isolation.

5.2 N_{CCN} closure from ACTIVATE observations

One important component of the work in Chapter 2 was the theoretical calculations of N_{CCN} using in situ observed aerosol size distributions and chemical composition from the ACTIVATE campaign as input for κ -Köhler theory. At this time, we also had the opportunity to contribute to work done in Soloff et al. (2025), looking in more detail at CCN behavior and closure in the same data set. Compared to our N_{CCN} calculations, this work focuses more on characterizing CCN properties and the regional and chemical dependencies of N_{CCN} closure. For example, N_{CCN} closure was found to be most accurate for spring observations and least accurate for observations over Bermuda (Fig. 5.2). Slight over-predictions in N_{CCN} were attributed to the assumption of internally mixed particles and kappa values for organics and ammonium sulfate for inorganics. Additionally, differences between observed and calculated kappa increased

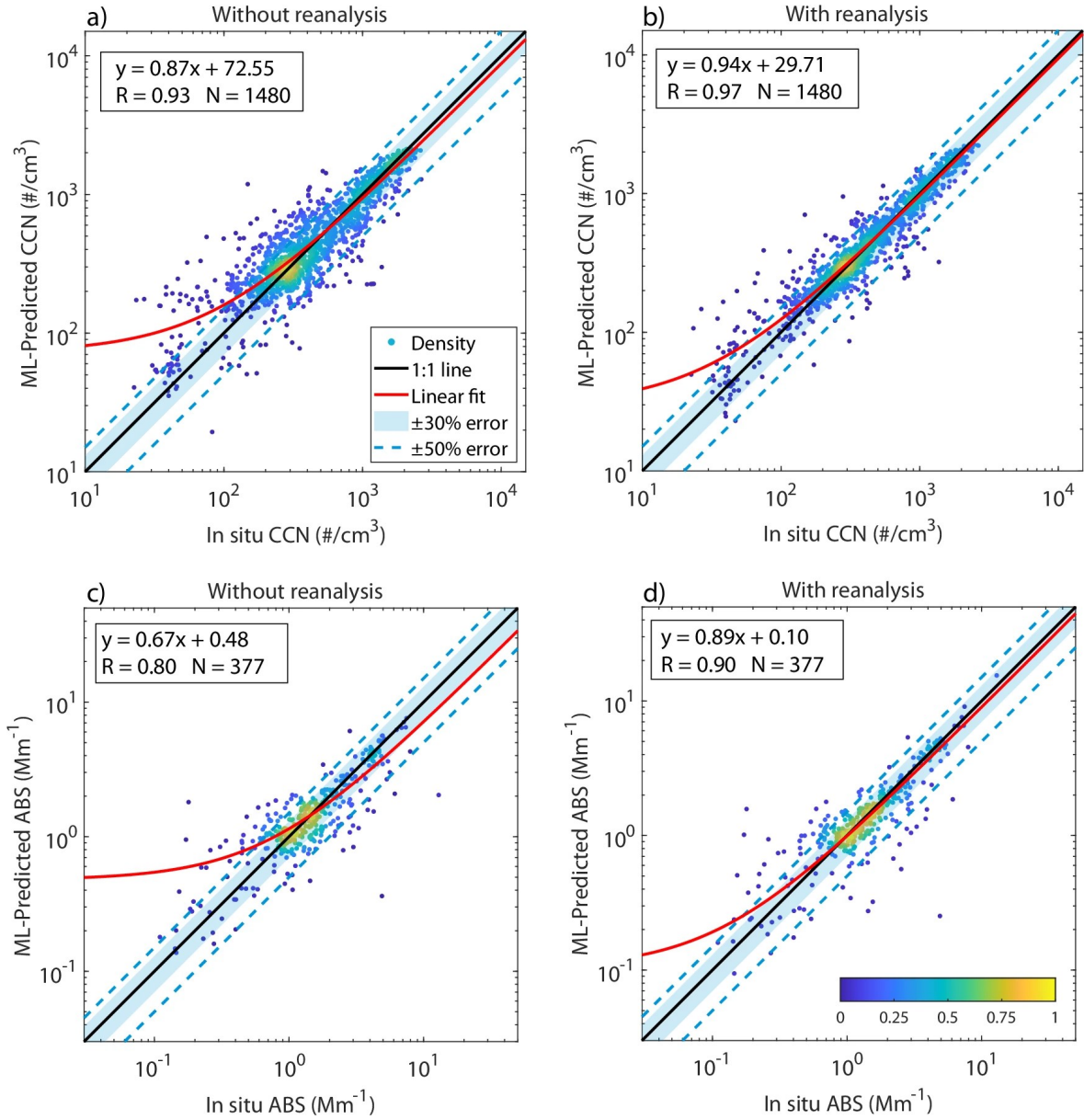


Figure 5.1: Figure 2 from Redemann and Gao (2024) showing a comparison between in situ and ML-predicted (a, b) CCN and (c, d) absorption for models with (b, d) and without (a, c) the inclusion of reanalysis data.

near Bermuda, as particles were more hygroscopic than indicated by observations.

Our results in Chapter 2 indicated that aerosol R_{eff} is the most important governing factor of the N_{CCN} —BSC relationship, and that once observations are separated by Aerosol ID the calculated kappa value is the least important governing factor. It em-

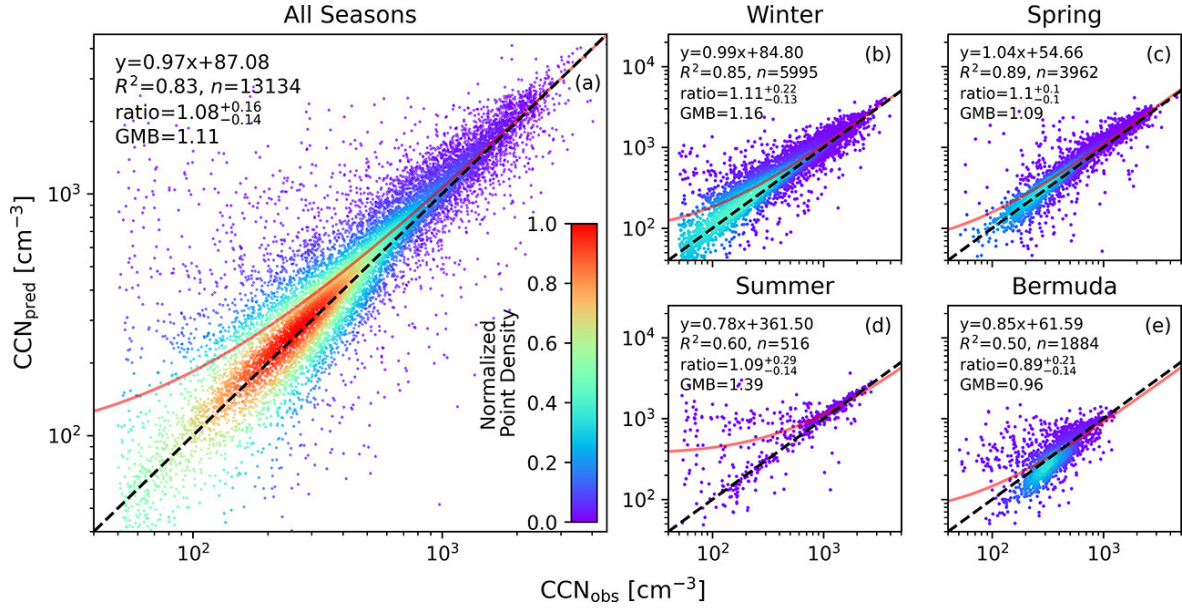


Figure 5.2: Figure 4 from Soloff et al. (2025) shows predicted vs. observed CCN concentration at all supersaturations and across (a) all ACTIVATE flights, and as separated by (b-d) season and (e) for Bermuda observations.

phasized the sensitivity of AOPs to large aerosols and showed that within N_{CCN} —AOP relationships, variability in AOPs is most influential. While that specific application did not find significant influence of aerosol chemical composition, Soloff et al. (2025) depicts in detail the importance of characterizing aerosol chemical composition for accurate CCN closure. Further, it provided important insights into CCN characteristics and activity in the northwest Atlantic, a region with both polluted and more remote oceanic regimes.

5.3 Small-Scale Variability of Solar Radiation (S2VSR) campaign

The Small-Scale Variability of Solar Radiation (S2VSR) campaign took place during summer 2023 at the Atmospheric Radiation Measurements (ARM) Southern Great

Plains (SGP) site in Billings, Oklahoma. Using a network of 60 autonomous pyranometers, this project explored the impact of shallow cumulus clouds on solar irradiance variation close to cloud shadow edges. Using five case studies of shallow cumulus clouds, one key result found that a radiation enhancement of 20% is seen at cloud edges compared to clear sky values (Fig. 5.3; Witthuhn et al., 2025). This effect depends on the size of cloud gaps, cloud base height, and geometry between the sun and clouds. Additionally, the distance over which the radiation enhancement occurs is dependent on cloud droplet R_{eff} , optical depth, and the solar zenith angle (Witthuhn et al., 2025).

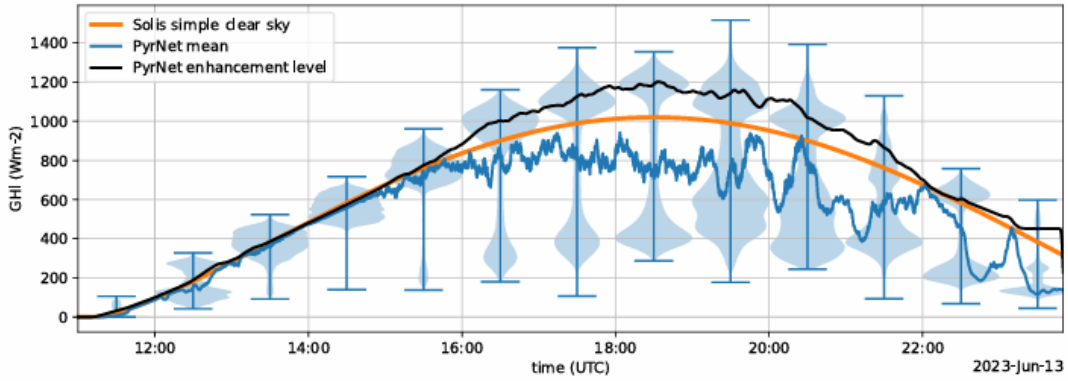


Figure 5.3: Figure 3 from Witthuhn et al. (2025) depicts observed hourly global horizontal irradiance distributions from 13 June 2023. Violin plots indicate extreme values in each bin and are bimodal in the presence of clouds. The blue line indicates the GHI mean, while the black line is the calculated enhancement level in cloud shadow gaps. The orange line is a clear sky reference modeled by the Solis simple model.

Just as low-level cloud feedbacks and the impacts of ACI need to be better represented in climate models, the 3D radiative effect at cloud edges causes uncertainty in estimating global horizontal irradiance (GHI), or the combination of diffuse and direct normal irradiance on a horizontal surface (Stein et al., 2012). GHI impacts Earth’s energy balance and water cycle, plant photosynthesis, and feedbacks that impact cloud formation and organization (Witthuhn et al., 2025). Therefore, it is important to understand and better represent its variability in climate models. One of the primary

implications of this work is that cloud shadow transition signatures are impacted by both geometric and microphysical cloud properties at cloud edges, and therefore both need to be considered in parameterizations. Similarly, in Chapter 3 we found important differences in cloud properties at cloud center compared to cloud edge, where cloud edge observations had much lower LWP and COT values than those located further from the cloud edge. While the relationships between above-cloud N_{CCN} and cloud top microphysical properties were similar for both regimes, this may not be the case for below-cloud N_{CCN} . However, our methodology was limited to assessing the impacts of below-cloud N_{CCN} only within 5 km of cloud edge, and therefore this hypothesis would need to be tested using other methods. Regardless, both studies emphasize the importance of considering both macro- and microphysical cloud properties while also acknowledging the dependence of observations relative to cloud edges.

5.4 Remote sensing-based ACI analysis in ACTIVATE region

The ORACLES ACI work done in Chapter 3 used ML-predicted N_{CCN} curtains from the Redemann and Gao (2024) method discussed in Section 5.1, which has also been applied in an ACI analysis using ACTIVATE observations. Using a very similar remote sensing-based methodology as we use in Chapter 3, Gao et al. (2026) found evidence of ACI between below-cloud N_{CCN} and cloud top microphysical properties after extrapolating cloud edge N_{CCN} below adjacent cloud bases (Fig. 5.4), thus further supporting our confidence in using this methodology to investigate ACI. While Gao et al. (2026) generally found stronger relationships between cloud properties and below-cloud N_{CCN} than we found in the ORACLES campaign, both findings speak to the importance of considering individual cloud regimes when investigating ACI. That is, the relationships

and dynamics between the SEA stratocumulus cloud deck with a thick layer of overlying smoke behave differently than a cumulus cloud field atop a more polluted MBL that is present in the ACTIVATE study region.

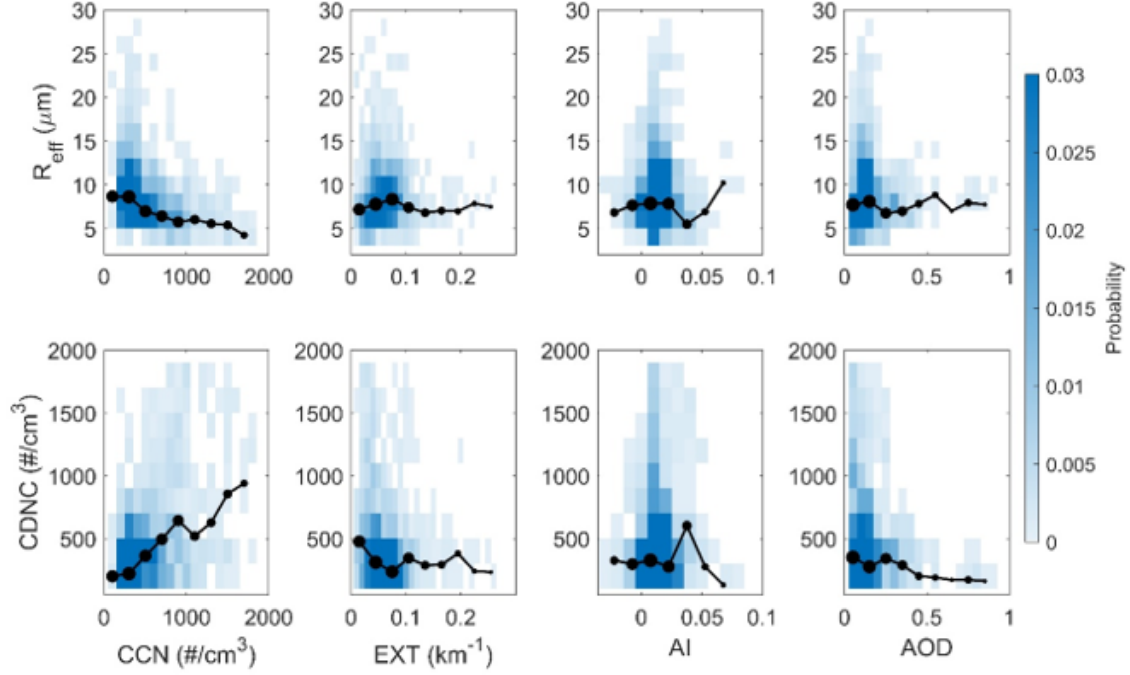


Figure 5.4: Figure 2 from Gao et al. (2026) showing joint histograms between aerosol properties and cloud top microphysical properties. Each histogram is normalized, with blue shading indicating the likelihood of observations falling within each bin. Black points represent binned mean cloud-top values.

Furthermore, this work directly supports an argument recognized by previous literature and the other results shown in this work. Many previous studies have argued that products such as AOD are not good proxies for N_{CCN} , as they cannot fully capture N_{CCN} variance. We have shown that vertically resolved EXT and BSC coefficients cannot reliably be used as simple, linear proxies for N_{CCN} . The results of Gao et al. (2026) go a step further in showing that not only do AOPs struggle to reliably predict N_{CCN} , but their use as proxies when investigating ACI relationships also tends to be unreliable. These findings corroborate the work done in Chapter 2 through their con-

firmation that the N_{CCN} —AOP relationship is non-linear and dependent on several factors.

5.5 Direct aerosol radiative effect in the SEA

Lastly, while this work has primarily focused on cloud-relevant properties of BBA in the SEA, this highly absorbing seasonal smoke plume also contributes significantly to ERF_{ari}. Work by Chang et al. (2025) targeted the direct aerosol radiative effect (DARE) of these aerosols, as its estimated magnitude varies widely in climate models. Specifically, it focused on the diurnal cycle of the stratocumulus cloud deck and its impact on quantifying DARE, since the calculation depends on the brightness of the underlying scene. When accounting for the cloud diurnal cycle using high resolution SEVIRI observations, the results show a two-fold increase in aerosol radiative warming compared to assuming a constant early-afternoon cloud field (Fig. 5.5). Higher cloud fractions and cloud optical depths in the morning result in higher DARE values, and when this is unaccounted for, it contributes to an underestimation of radiative warming over the SEA.

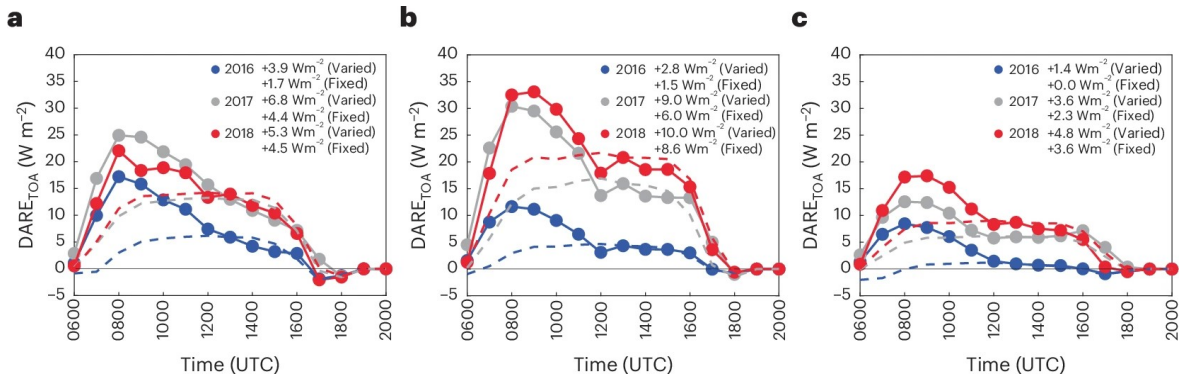


Figure 5.5: Figure 3 from Chang et al. (2025) showing monthly mean diurnal cycles of DARE_{TOA} for hourly varying cloud properties (solid lines) and diurnally fixed cloud properties from 1300 UTC (dashed line) for 2016 (blue), 2017 (gray), and 2018 (red) in August (a), September (b), and October (c).

The work discussed in Chapter 3 and in Gao et al. (2026) has focused on the impact of the spatial resolution of observations used in ACI studies. Specifically, that the vertical distribution of N_{CCN} is necessary for better understanding ACI rather than using column-integrated AOPs as a N_{CCN} proxy. Results from Chang et al. (2025) communicate the importance of considering the temporal resolution of observations when estimating ERF_{air}. We approached the ORACLES ACI analysis as an instantaneous and simultaneous look at the impacts of above- and below-cloud N_{CCN} on cloud microphysical properties. However, one way this study could be expanded in the future is by considering both the cloud diurnal cycle and the timescale of smoke entrainment from the free troposphere into the MBL. For example, Diamond et al. (2018) argues, using in situ observations, that considering the history of smoke entrainment can explain variations in cloud properties for similar instantaneous above-cloud smoke environments. Additionally, Chang et al. (2025) shows variations in cloud optical depth and cloud fraction as a function of the diurnal cycle, while we have focused on their variability as a function of proximity to cloud edge. Therefore, accounting for the diurnal cycle using the remote sensing data set in Chapter 3 may further elucidate the complexities of ACI in the SEA.

Chapter 6

Conclusion

The total aerosol impact on climate forcing is estimated to be negative, representing an overall cooling effect, but the uncertainty associated with aerosol-cloud interactions is high, creating ambiguity in the potential magnitude of this cooling effect. To reduce this uncertainty and better represent ACI in climate models, observational studies must continue to assess how aerosols impact clouds in different cloud and environmental regimes. One of the biggest pieces of missing information in observational ACI studies has been the vertical distribution of N_{CCN} . Recent studies have begun to take advantage of the vertical resolution of lidar-observed aerosol optical properties to retrieve vertical curtains of N_{CCN} and apply them to ACI studies. In this work, we have examined the factors that govern how N_{CCN} relates to lidar observables, how lidar-derived N_{CCN} profiles can be used to assess ACI in marine stratocumulus clouds, and how broad ACI- and climate-related topics can be taught at a high school level.

Using an in situ observation-informed approach to κ -Köhler and Mie theory calculations, we first assessed the theoretical relationship between N_{CCN} and BSC at 532 nm, using a random forest approach to determine the relative importance of the aerosol size distribution, chemical composition, and RH in governing this relationship (Ch. 2). Separating this analysis by aerosol types observed during the NASA ACTIVATE campaign (smoke + fresh smoke, marine + polluted marine, urban) revealed that the aerosol effective radius is the most important determining factor of the N_{CCN} :BSC ratio for all aerosol types. Ambient RH is the second most important factor, followed

by geometric mean radius and chemical composition (κ). The aerosol effective radius has a strong exponential relationship with $N_{CCN}:BSC$, such that approximately 68-79% of the variance can be explained by modeling $N_{CCN}:BSC$ using the exponential effective radius relationship alone.

The work done in Chapter 2 has several implications for remote sensing of N_{CCN} . First, many studies investigating the challenges of constraining aerosol properties from remote sensing observations have focused on errors attributable to ambient RH and hygroscopic growth of aerosols. While the impact of RH is included in our R_{eff} calculations for humidified aerosol size distributions, we still find that R_{eff} is more important in determining $N_{CCN}:BSC$ than RH alone. Therefore, future studies using remotely-sensed AOPs to constrain N_{CCN} should consider the aerosol size distribution in parameterizations in addition to hygroscopicity. Further, the use of actually observed aerosol size distributions and chemical composition measurements allowed us to represent a large, highly variable, and realistic population of aerosols that would not have been possible with a single, idealized size distribution and assumed chemistry. Though in situ observations introduce uncertainty into such analyses, they are crucial for accurately representing environments with significant aerosol variability.

The theoretical calculations work in Chapter 2 magnified the non-linearity and multivariate dependencies of N_{CCN} —AOP relationships. While this project was ongoing, Redemann and Gao (2024) developed a machine learning method to derive N_{CCN} from HSRL-2 observables that was shown to be very accurate and computationally efficient when handling non-linear and multivariate relationships. Therefore, to assess the benefit and applicability of vertically resolved N_{CCN} curtains for ACI analysis, we used the Redemann and Gao (2024) ML predicted N_{CCN} dataset (ML-CCN) in combination with cloud properties retrieved from the RSP. Because in situ analyses from ORACLES have previously found evidence of changes to cloud properties in cases of contact with

the overlying smoke plume, one of the main goals of this project was to determine whether the same relationships were evident from this larger remote sensing dataset (Ch. 3).

ML-CCN and RSP retrievals showed that N_{CCN} within 100 m of cloud top does influence cloud top microphysical properties, thus confirming the main results from the in situ based ACI study (Gupta et al., 2021). That is, as above-cloud N_{CCN} increases, cloud droplet effective radius decreases and cloud droplet number concentration increases. Additionally, we used boundary layer-reaching lidar profiles at cloud edges to assess the simultaneous impact of above- and below-cloud N_{CCN} on cloud top microphysical properties. This analysis demonstrated not only the ability for remote sensing methods to accurately observe ACI, but also the benefit of including in situ observations to provide additional, contextual information. For example, an autocorrelation analysis using in situ-observed N_{CCN} from constant altitude flight legs revealed that the N_{CCN} autocorrelation coefficient remains above 0.95 over horizontal distances of 5 km. Therefore, we applied below-cloud N_{CCN} at cloud edge to additional profiles within 5 km, which increased the CE data set threefold. Additionally, in situ-derived relationships between cloud top and cloud base heights allowed for a more accurate estimation of the below-cloud N_{CCN} on a profile-by-profile basis.

In addition to demonstrating that our remote sensing-based dataset accurately resolves above-cloud ACI, another significant finding from this work is the LTS dependence of cloud sensitivity to both above- and below-cloud N_{CCN} . Clustering the data using LTS resulted in clusters that were geographically well-defined as LTS increases from northwest to southeast across the SEA, following the climatological pattern of decreasing SST. We find that as the environment becomes more stable, cloud sensitivity to above-cloud N_{CCN} decreases, while sensitivity to below-cloud N_{CCN} increases. Therefore, we see that for this unique environment with significant above-cloud aerosol

loading, entrainment mixing of the above-cloud BBA into the cloud layer has a dominant role in modulating cloud properties in less stable conditions, while the more typical and expected role of below-cloud N_{CCN} nucleation dominates in stable conditions. Additionally, this finding highlights an important limitation of our method, in that sub-selecting lidar profiles in cloud gaps with which to assess below-cloud N_{CCN} results in a dataset of mostly low-LTS observations. Since low-LTS cases dominated the below-cloud dataset, the strong impact of below-cloud N_{CCN} on cloud properties at high LTS was not obvious when analyzing the full cloud edge dataset collectively. This corroborates many previous ACI studies that have emphasized the importance of constraining environmental factors to isolate aerosol impacts to cloud properties.

The work in Chapters 2 and 3 has implications for improving remote sensing observations of N_{CCN} and ACI, which is especially relevant for improving their representation in climate models. While it is critical to improve climate model predictions of how aerosols and clouds will impact future climate, climate change is not an issue completely isolated to the future. Rather, impacts are already occurring, and there are several actions that can and should be taken now to mitigate further warming and climate change. One suggested pathway for mitigating climate change is improving efforts in climate change education and communication, particularly in environments where climate change has become a highly politicized and divisive topic. Therefore, the work in Chapter 4 aimed to contribute to local climate science education through a three-day series of lessons and activities developed around the key aerosol and cloud concepts discussed in Chapters 1-3. While there are many suggestions in the literature for effective climate science education, this project focused on implementing two common recommendations: using local and relevant examples and engaging learners using hands-on activities and data.

This three-day series of lessons and activities was implemented in the Norman

High School meteorology elective course from April 28-30, 2026. Out of the 98 students enrolled in the course, survey and reflection question responses were analyzed for 36 students, or a little over one-third of the total enrollment. Post-survey responses showed shifts towards more correct answers for 22 of the 24 survey statements, indicating that, in general, the activities and lessons improved student understanding and confidence in skills and knowledge. When comparing results for statements whose concepts were and were not directly tied to an activity, we found that understanding generally improved more for the former. Therefore, this demonstrated the benefit of incorporating science concepts into hands-on activities. We found that some students struggled to correctly interpret questions using words such as “property” or “characterized,” and this emphasized the importance of scientists working collaboratively with teachers to better understand student background knowledge and understanding. While some jargon may be obvious, other difficult to understand words may not be as apparent. Lastly, we found that with a focus on teaching the mechanisms of aerosol and cloud interactions with solar radiation and each other, students were generally able to correctly deduce possible long-term climate impacts of these interactions.

There are also several routes for potential future work that were outside the scope of the analyses shown here. First, none of our analyses included observations of dust aerosols. While a few observations of dust are included in the training of the ML-CCN model (Redemann and Gao, 2024), we did not include any such observations in Chapter 2 since dust was not frequently observed during ACTIVATE. Because dust is typically larger in size and less hygroscopic than smoke, sea salt, and pollution aerosols, the way that N_{CCN} and BSC relate could be different for dust than for these other aerosol types. Further, this theoretical analysis was performed for observations made in a humid marine boundary layer. It is possible that this kind of environment could exacerbate the importance of large aerosols in determining how N_{CCN} relates to BSC,

so it could be informative to repeat this analysis using observations made in different environments, potentially those that are less humid or have a different mixture of typical aerosol types.

In terms of the ACI analysis, one route that should be further explored is the inclusion of additional meteorological variables for constraining the aerosol impact on stratocumulus cloud properties. While LTS was found to be a significant control on cloud sensitivity to N_{CCN} , including other parameters may result in a better understanding of the system as a whole. While we found similar above-cloud ACI metrics for observations made at cloud edge and cloud center, it may also be worth investigating the scales over which COT and LWP increase between cloud edge and cloud center and how such changes impact other cloud processes. Additionally, while we hypothesized that below-cloud ACI may be similar between cloud center and cloud edge observations, we were unable to prove this using the dataset and method shown here, meaning this question could also be investigated further in the future, potentially using a modeling approach.

Another way to expand the ACI analysis may be to combine the ML-CCN dataset with other ORACLES observations, such as in situ cloud profiles or the joint cloud water and rainwater path retrievals in Dzambo et al. (2021). Comparing remotely-sensed ACI metrics with those from in situ cloud probe data would serve to further validate the results in Chapter 3, and including the Dzambo et al. (2021) joint RSP + APR-3 (Airborne Precipitation Radar - third generation) dataset would allow for an analysis of vertical changes in cloud properties and a more direct focus on precipitation susceptibility. While the overall weak below-cloud N_{CCN} relationship at cloud edge is explained further when separating the cloud edge dataset into LTS regimes, process modeling studies could be beneficial for testing and better understanding the weaker impact of below-cloud N_{CCN} than above-cloud N_{CCN} at cloud edges in low LTS environments.

Lastly, this method should be applied to other aerosol and cloud regimes and other observational datasets. For example, Gao et al. (2026) applied a similar methodology to the cumulus cloud field observed in the ACTIVATE study area and found below-cloud N_{CCN} impacts of similar magnitude to those found for the above-cloud N_{CCN} observations here. Therefore, testing how aerosol impacts differ in various aerosol and cloud regimes would expand our overall understanding of ACI. This methodology should also be tested using ATLID observations and satellite-retrieved cloud properties to better constrain ACI on a global scale.

The work discussed in Chapter 4 should be viewed as a starting point for future outreach, teaching, and lesson development. The activities, lessons, and surveys could be improved upon through continued iterations and experimentation, and there are several possible routes for improvement. While continued collaboration with Norman High School would be beneficial for building community relationships and partnerships between the School of Meteorology and Norman Public Schools, these activities and lessons could also be tested in new schools and classrooms. More specifically, the Life Cycle of an Aerosol board game could be adapted to only focus on direct or indirect effects (not both), or to be generally more straightforward for a shorter time period. The reflection question worksheets should be examined and refined with input from the classroom teacher to avoid the inclusion of words that students are less familiar with. Determining a way to teach about cloud albedo and precipitation susceptibility through a hands-on activity or demonstration may be beneficial, and coming up with additional Oklahoma-specific data examples to demonstrate regionally-relevant impacts of aerosols and clouds on weather and climate may also strengthen this set of modules. Additionally, it could be interesting to remove the “neither agree nor disagree” option on the pre and post-surveys to see if forcing “agree” or “disagree” responses would show different results. Lastly, one limitation and area for future im-

provement is in the limited number of students whose answers were included in the results of Chapter 4. If more time had been allotted for the IRB approval process, there may have then been more time to solicit parent permission responses and to communicate with the parents more frequently before the dates of the classroom visit.

The central goal connecting each part of this work was to contribute to reducing uncertainties in aerosol-cloud interactions. Fully constraining aerosol-cloud interactions and significantly reducing the magnitude of this uncertainty on a global scale will require continued global efforts and diverse approaches. This work has taken an observational approach that has determined important considerations for remote sensing of N_{CCN} and has developed a fully remote sensing-based method for assessing ACI that confirms results seen using in situ data. The final project took a community and education-based approach to improve student understanding of how aerosols and clouds interact and impact climate on local and global scales. While perhaps not the typical view of what it means to reduce uncertainties in aerosol-cloud interactions, we found that student uncertainty of aerosol and cloud relevance to climate decreased after implementing the three-day set of lessons and activities. Finally, the three main conclusions of this work can be summarized as follows:

1. Accurately relating N_{CCN} to aerosol optical properties should involve consideration of the aerosol size distribution, specifically the coarse mode.
2. Combining the vertically resolved ML-CCN dataset with RSP-retrieved cloud microphysical properties allows for accurate remote sensing assessments of ACI, and the method can and should be translated to satellite-based instrumentation. Additionally, lower tropospheric stability is an important environmental constraint through which ACI should be interpreted and understood.
3. A three-day module focused on teaching the basic mechanisms of how aerosols

and clouds interact with solar radiation and with each other enabled students to deduce their possible long-term climate impacts. Future climate-focused outreach activities should prioritize hands-on activities, avoid combining too many topics in individual activities, and involve collaboration with classroom teachers to better understand students' background knowledge.

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