Bridging New Observational Capabilities and Process-Level Simulation: Insights into Aerosol Roles in the Earth System

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ABSTRACT: The spatial distribution of ambient aerosol particles significantly impacts aerosol–radiation–cloud interactions, which contribute to the largest uncertainty in global anthropogenic radiative forcing estimations. However, the atmospheric boundary layer and lower free troposphere have not been adequately sampled in terms of spatiotemporal resolution, hindering a comprehensive characterization of various atmospheric processes and impeding our understanding of the Earth system. To address this research data gap, we have leveraged the development of uncrewed aerial systems (UAS) and advanced measurement techniques to obtain mesoscale spatial data on aerosol microphysical and optical properties around the U.S. Southern Great Plains (SGP) atmospheric observatory. Our study also benefits from state-of-the-art laboratory facilities that include three-dimensional molecular imaging techniques enabled by secondary ion mass spectrometry and nanogram-level chemical composition analysis via micronebulization aerosol mass spectrometry. Through our study, we have developed a framework for observation–modeling integration, enabling an examination of how various assumptions about the organic–inorganic components mixing state, inferred from chemical analysis, affect clouds and radiation in observation-constrained model simulations. By integrating observational constraints (derived from offline chemical analysis of the aerosol surface using collected samples) with in situ UAS observations, we have identified a prominent role of organic-enriched nanometer layers located at the surface of aerosol particles in determining profiles of aerosol optical and hygroscopic properties over the SGP observatory. Furthermore, we have improved the agreement between predicted clouds and ground-based cloud lidar measurements. This UAS–model–laboratory integration exemplifies how these new advanced capabilities can significantly enhance our understanding of aerosol–radiation–cloud interactions.

KEYWORDS: In situ atmospheric observations; Instrumentation/ sensors; Unpiloted aerial systems; Large-eddy simulations; Aerosol hygroscopicity; Atmospheric chemistry

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SIGNIFICANCE STATEMENT: This study aims to introduce a framework harnessing the current midsize uncrewed aerial system (UAS) measurement capabilities to propel atmospheric research forward. By integrating observational constraints derived from offline chemical analysis with in situ UAS observations, we unveil the pivotal role of organic-enriched nanometer layers on aerosol optical and hygroscopic property profiles above the U.S. Southern Great Plains (SGP) observatory and showcase substantial improvements in the agreement between model results and ground-based cloud lidar measurements. This UAS–model–laboratory integration exemplifies how these advanced capabilities can revolutionize our understanding of aerosol–radiation–cloud interactions, offering unprecedented insights into the Earth system simulation.

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1. Introduction
The major role played by ambient aerosol in Earth’s energy balance has motivated significant research attention for decades (Charlson et al. 1992; Seinfeld et al. 2016; Zaveri et al. 2022). Many studies have characterized ambient aerosol in terms of concentration, size, shape, surface area, refractive index, state of matter, hygroscopicity, and chemical composition (Finlayson-Pitts and Pitts 1986; Hinds and Zhu 2022; Seinfeld and Pandis 2016). Practical sampling and monitoring of ambient aerosols under various environments started in the early 1980s (Wedding 1982) and has continued at increasing scales ever since. The U.S. Department of Energy (DOE) established the Atmospheric Radiation Measurement (ARM) program over 30 years ago in response to the growing observational needs (Mather and Voyles 2013). Initially, ARM observational data have been primarily collected from fixed-site measurement infrastructure. To meet the critical need for in situ observations to establish a comprehensive, process-level understanding of aerosols’ roles in aerosol–radiation–cloud interactions (Fan et al. 2016), the ARM climate research facility expanded to multiple instrumented platforms (ground-based and airborne) to provide comprehensive atmospheric measurements, which advanced our current knowledge of aerosol–cloud interactions and their impact on the global climate (Mather and Voyles 2013). However, measurements of the vertical distribution of ambient aerosol, especially the chemical properties, remain very sparse (Fast et al. 2019; Kulkarni et al. 2023; O’Donnell et al. 2023; Schobesberger et al. 2023). Although satellite-based and collocated surface-based remote sensing data provide additional information, their coarse spatial resolution does not provide adequate constraints on model parameterizations of aerosol optical and microphysical properties.

Aerosol chemical properties have been the subject of extensive research due to their importance for human health, climate, and air quality (Hallquist et al. 2009; Heald and Kroll 2020; Qin et al. 2022; Thompson 2018). As a result, researchers have developed various online and offline analysis techniques to describe the bulk chemical composition of ambient aerosols (DeCarlo et al. 2006; Jayne et al. 1998; Lee et al. 2003; Nizkorodov et al. 2011; Orsini et al. 2003; Xu et al. 2017; Zelenyuk and Imre 2005). Although the surface layer of aerosol particles contributes only a small fraction of the total particle mass, this
layer plays a dominant role in all essential aerosol processes and aerosol–cloud interaction, such as gas phase interaction, water vapor uptake, and cloud droplet nucleation. Therefore, researchers use surface probing tools and atmospheric simulation chambers to investigate the relationship between the aerosol properties (such as hygroscopicity and optical properties) and their chemical composition and morphology, especially for secondary organic aerosol (SOA) components (Denjean et al. 2015; Qin et al. 2022; Wingen and Finlayson-Pitts 2019). However, these data cannot quantitatively distinguish aerosol property differences between the near-cloud area and the surrounding environment (Marshak et al. 2021). In situ measurements are still vital to evaluating aerosol spatial variability in many climate-critical locations (de Boer et al. 2016; Creamean et al. 2021; Seinfeld et al. 2016).

The sparse spatial observation data also limited model performance (Brotzge et al. 2023; DeMott et al. 2022). Quantifying the uncertainties and reducing model bias associated with estimating the aerosol direct and indirect effects on radiation, especially aerosol interactions with different cloud systems, remain critical challenges for both modelers and experimentalists (Fan et al. 2016; Sanchez et al. 2017).

With the rapid development in both uncrewed aircraft systems (UAS) and miniaturized instrumentation, opportunities for these platforms to provide high-quality atmospheric measurements continue to expand (de Boer et al. 2020a,b; Frew et al. 2020; Mei et al. 2021; Sigala and Langhals 2020). The strategic plans of various agencies provide clear evidence of the momentum for employing UAS operations and the continued growth of using UAS in the international setting (e.g., the International Society for Atmospheric Research using remotely piloted aircraft) (Argrow et al. 2017; Lampert et al. 2020; Pinto et al. 2021; Schmid et al. 2020; Villa et al. 2016). Application of midsize UAS [group III, weight > 330 lb, and max airspeed > 200 kt (1 kt ≈ 0.51 m s⁻¹)] to topics across atmospheric science has also attracted increasing attention for performing remotely sensed and in situ observations (Angelopoulou et al. 2019; Pieri and Diaz 2014; Pieri et al. 2013; Telg et al. 2017). Despite the existing limitations in electrical power and payload capacity, aerosol sampling with an uncrewed system (Bates et al. 2013; de Boer et al. 2020b) can leverage valuable insights from previous field deployment experience and innovative offline chemical composition analysis techniques (Kristensen et al. 2016; Niedek et al. 2023; Zhang et al. 2011; Zhang et al. 2015). In addition, the extended flight endurance (up to 6 h) and operational altitude range (e.g., 70–6000 m) of midsize UAS enable the collection of rich datasets, filling in observational gaps and providing spatial context when deployed in combination with other ground and mobile observation stations (Bates et al. 2013; de Boer et al. 2016; Creamean et al. 2021, 2018; de Boer et al. 2020a).

A recent review paper (Kahn et al. 2023) pointed out a “three-way street” to reduce the aerosol forcing uncertainty, which combines satellite observations, in situ atmospheric measurements, and modeling and data assimilation. This three-way street is critical for any effort to understand and predict climate change. Thus, this study is motivated to provide a framework for assessing the limitations and strengths of model simulations by using new types of measurement strategies.

Addressing community needs, we utilize advanced capabilities from ARM observation platforms to integrate data collected around the Southern Great Plains (SGP) observatory with cutting-edge aerosol chemical analysis to understand the vertical structure of aerosols over the SGP. This study incorporates data from two offline chemical analyses. A micronebulization aerosol mass spectrometry (MN-AMS) chemical assay, newly developed for quantitative analysis of PM composition at the nanogram level (Niedek et al. 2023), is combined with time-of-flight secondary ion mass spectrometry (ToF-SIMS), an advanced particle surface characterization technique (Belu et al. 2003; Huang et al. 2017; Li et al. 2023), to provide comprehensive measurement data for enhanced process-level understanding and atmospheric modeling. We also...
carry out large-eddy simulations (LES), constrained by UAS sampling at the SGP observatory and routine ground-based remote sensing observations, to assess how new observational insights affect model representations of aerosol composition and their impacts on the modeled cloud properties. Additionally, an increased understanding of the sensitivity of cloud properties to the newly observed aerosol microphysical properties due to aerosol mixing state paves the way for further assessment of relevant parameterizations in Earth system models (ESMs).

2. Description of ongoing UAS capability development

a. In situ measurements. The ARM Aerial Facility (AAF) collaborated with the Mississippi State University (MSU) Raspet Flight Research Laboratory (RFRL) to operate an MSU TigerShark UAS in November 2021 and the DOE ArcticShark UAS in July 2022 (Mei et al. 2022) and in March, June, and August 2023 for engineering testing. The two midsize UAS deployments with identical scientific payloads were sampled above the SGP observatory Central Facility (Table 1). The payload provided atmospheric parameters (such as temperature, pressure, relative humidity, and 3D wind components), aerosol properties (total number concentration, aerosol size distribution, absorption, and chemical composition), trace gas concentrations (CO₂ and H₂O mixing ratios), and surface remote sensing measurements from an infrared temperature sensor and multispectral camera (Mei et al. 2022). An actively controlled community inlet system has been installed. It maintains the sampling of aerosol flow at isokinetic conditions and dries below 40% relative humidity before entering the aerosol instruments (Pekour and Newburn 2022). The payload development is an ongoing effort. The aerosol size distribution package was developed and flight-tested in 2023, and the high-time-resolution temperature and humidity measurements are still under development. One ongoing effort is exploring options for a miniaturized cloud condensation nuclei (CCN) counter capable of operating with scientific confidence at a reasonable resolution. The altitude of the UAS is plotted as the height above the ground level (AGL).

<table>
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<tr>
<th>Table 1. Baseline measurement capability of the ARM midsize UAS.</th>
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<td><strong>Instrumentation</strong></td>
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<td>Aircraft state</td>
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<td>Meteorological state</td>
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<td>Flux measurements</td>
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<td>Land surface properties</td>
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<td>Cloud properties</td>
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<td>Aerosol physicochemical properties (with isokinetic inlet)—package 1</td>
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<tr>
<td>Aerosol size distribution (with isokinetic inlet, available since 2023)—package 2</td>
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in this study. A previous publication details the payload performance and includes the archived data from the November 2021 deployment (Mei et al. 2022).

This study utilizes measurements obtained on 9 and 11 November 2021, as these dates had the necessary instrument payload and suitable sampling strategy compared to other similarly instrumented flights. Table S1 in the online supplemental material lists more information about the 2021 flights. We obtained the aerosol profiles by averaging the data into 50-m vertical intervals (Mei et al. 2022). Note that in 2021, the UAS operational altitude (AGL) was limited to 550–2000 m due to the Federal Aviation Administration (FAA) certificate of operation issued at the time, which was extended to 150–2000 m in 2023. The MN-AMS analysis was first applied with the UAS samples acquired during the November deployment. Thus, the chemical composition data were based on the combined samples from multiple flights. For the MN-AMS analysis in 2021, the aerosol particle loading from individual flights was too low to achieve a meaningful analysis. Niedek et al. (2023) compared the analysis results of collected samples with long-term ground-based measurements. They showed that the chemical composition from multiple sampling periods reflects the typical bulk chemical composition trend in the region between 9 and 16 November 2021.

b. Advanced offline chemical analysis. A primary challenge in UAS aerosol composition measurement is the lack of high temporal resolution in situ measurements with low detection limits for PM chemistry, especially for the aerosol surface chemistry, due to the high energy consumption and weight limitations of the commercially available instruments (Brady et al. 2016; Glaser et al. 2003; Hemingway et al. 2017). To overcome these challenges, Niedek et al. (2023) developed the MN-AMS technique to couple with the midsize UAS sampling, which allows high-sensitivity AMS measurement of nonrefractory aerosol species, such as sulfate, nitrate, ammonium, chloride, and organics from small samples (Canagaratna et al. 2007; DeCarlo et al. 2006). Details of the aerosol filter sample collections can be found in Niedek et al. (2023).

Specifically, this MN-AMS technique overcomes the requirement of large liquid volumes in standard aerosolization methods, reducing the solvent volume for PM extraction to as low as 10 µL. Using an isotopically labeled internal standard ($^{34}$SO$_2^-$), the MN-AMS can quantify nanogram levels of organic and inorganic substances with the detection limits determined at 0.189, 0.751, and 2.19 ng for sulfate [corresponding to (NH$_4$)$_2$SO$_4$], nitrate (corresponding to NH$_4$NO$_3$), and organics, respectively (Niedek et al. 2023). The O/C and H/C ratios were 0.363 and 1.790, respectively. Note that black carbon concentration was not measured in this study due to the current instrument payload aboard the UAS. We plan to use a soot particle aerosol mass spectrometer (SP-AMS) to measure the black carbon contribution in a future deployment.

Another unique asset, a time-of-flight secondary ion mass spectrometer (ToF-SIMS) (IONTOF GmbH, Münster, Germany), was adapted to analyze the ambient aerosol samples collected with a four-stage impactor (Sioutas Personal Cascade Impactor, SKC Inc.) (Niedek et al. 2023). ToF-SIMS has a long history of success in various scientific fields, such as materials science, geology, and biology (Huang et al. 2017; Li et al. 2023). As a surface-sensitive technique, ToF-SIMS has been developed and matured in materials science and has great potential for aerosol research (Belu et al. 2003; Benninghoven 1994; McPhail 2006). Recent developments, including argon cluster sputter ion beam and delayed extraction imaging, have made ToF-SIMS even more useful for aerosol research (Shen et al. 2015; Vanbellingen et al. 2015). Its high sensitivity and nanoscale three-dimensional imaging capabilities enable detailed analysis of molecular species and their three-dimensional distribution within a substrate-collected aerosol sample (Guo et al. 2020; Li et al. 2023; Passarelli et al. 2017). Figure 1 shows images of the representative ion species (denoted with organics,
nitrate, and sulfate to match the AMS classification) for the aerosol atomic monolayers at the particle surface (top panel) and after the outer 10 nm are removed (bottom panel). The SIMS 3D imaging also indicates that most collected aerosol particles are in a solid phase. A 10-keV Ar cluster ion beam was used for sputtering to remove 10-nm surface materials, and a 25-keV Bi\(^{3+}\) beam was used for SIMS imaging analysis before and after Ar cluster beam sputtering. After removing 10 nm of materials, the overall and sulfate signals decreased, while the nitrate signal was enhanced, as shown in the marked green circles (Belu et al. 2003; Li et al. 2023). The majority (over 85\%) of aerosol particles collected were in the solid phase. In contrast, a small number of particles are observed to shrink significantly (with greatly decreasing organic and \(\text{HSO}_4^-\) signals) after 10-nm sputtering, indicating that such particles might be liquid or semiliquid droplets, and they formed a thin film due to liquid spreading during sample collection.

Interestingly, the organic signal significantly decreased or disappeared on most solid particles (the green circles marked a few representative particles) after the 10-nm surface layer was removed, but the inorganic signals hardly changed. This observation indicates that the organic-dominated structure only exists within the thin outer surface layer. These ToF-SIMS images provide insights into the aerosol surface-layer chemical composition and mixing states of the SGP particles. Although it is well accepted that surface properties can determine aerosol particle growth, water uptake, and optical properties, direct measurements of ambient particles’ surface properties are rarely made.

By incorporating the chemical composition data from MN-AMS and ToF-SIMS, we can further examine the role of surface properties in determining particles’ chemical and physical properties. In addition, other ongoing projects from the Environmental Molecular Sciences Laboratory (EMSL) focus on elucidating various phase states of aerosol particles and molecularly identifying chemical components of the liquid and solid parts with the ToF-SIMS 3D imaging and spectra analysis. With the insight gained from the advanced techniques, we can also assess the potential bias caused by different surface property assumptions and their
impact on the cloud optical properties, microphysical properties, and associated radiative forcing uncertainty.

3. Linking UAS observations to process-level modeling

Combining UAS capabilities and advanced analytical techniques provides a unique opportunity to evaluate model treatments of aerosol processes, especially aerosol–cloud interactions (Seinfeld et al. 2016; Seinfeld and Pandis 2016). However, the main obstacle identified by previous studies is the lack of concurrent profile measurements of aerosol microphysical properties, aerosol optical properties, cloud dynamics, and cloud microphysical properties to understand the aerosol–radiation and aerosol–cloud interactions (ARI and ACI) better (Bates et al. 2013; de Boer et al. 2020b; Kahn et al. 2023).

The newly developed ARM UAS observational capability can aid the community in two ways, as illustrated in Fig. 2. First, the left portion of Fig. 2 outlines a methodology/workflow to establish observational constraints for process-level model evaluation. Second, the knowledge gained from the observations further facilitates comparison between measurements and model simulations. This comparison provides perspicacity for strengthening the simulation’s fidelity, gaining scientific insights on the process-level information for model

Fig. 2. A schematic description of the iterative process of obtaining observational constraints, based on the available airborne in situ and remote sensing measurements, for process-level model simulations. The goal is to provide an asset to evaluate the representation of aerosol–cloud processes in ESMs.
simulation and parameterization, and improving the observation capability by refining flight planning strategy and pointing out measurement limitations.

The workflow iterates in the left portion of Fig. 2 to improve the observational capability for model evaluation. In section 3a, we demonstrate the approaches of using advanced chemical analysis techniques and other UAS measurements to identify the significant influence of chemical composition and surface structure on aerosol optical and microphysical properties (more details about the ARI in the supplemental material). Through iterative refinement of aerosol assumptions and improvement of measurements, the workflow strives to produce a good agreement between the aerosol optical and microphysical closures. With better observational constraints, we then move to the right portion of the workflow and examine how the hygroscopicity parameter (based on different assumptions of the aerosol chemical compositions and their mixing state) impacts the process-level simulation results, e.g., the LES model simulated cloud properties (detailed in section 3b). Assessing the model simulations against measurements enables experimentalists to refine the measurement needs and strategy. They can also guide further advancement of the analysis technique, exploring new measurement capabilities, refining UAS flight patterns and sampling strategies, and yielding positive feedback into the observation–model interaction workflow.

While applying this workflow with the 9 November data, we identified several measurement limitations due to current research resources and technical capabilities. For example, the operational considerations limit the altitude sampling range of the UAS platform by Federal Aviation Administration Certificate of Authorization (FAA COA). Thus, we extended the lowest flight operations to 150 m above ground for the ArcticShark deployments in 2023. More potential improvements and capabilities based on the lessons learned from the flight series in 2021 have been discussed in section 4.

**a. Cloud condensation nuclei profile estimation.** As an essential element of cloud formation, ambient aerosol’s ability to serve as cloud condensation nuclei affects cloud droplet size, albedo, and lifetime. The CCN activity of aerosol particles in a saturated air parcel depends on their size and hygroscopicity parameters (Andreae and Rosenfeld 2008; Petters and Kreidenweis 2007). Meanwhile, accurately representing the mixing state of the ambient aerosols is also essential to reliably predict their climate-relevant properties in atmospheric models (Ching et al. 2016). A recent airborne closure study also points out that the measured and estimated CCN concentration often agrees better with the assumption that all particles are composed of 100% organic particles (Kulkarni et al. 2023). A critical link between aerosol thermodynamic properties and CCN activity, the hygroscopicity parameter $\kappa$ enfolds complex information about the aerosol chemical composition, surface properties, and mixing states (Petters and Kreidenweis 2007; Riemer et al. 2019). Although a miniaturized CCN counter is ideal for providing the CCN concentration, there is no reliable mini-CCN counter available to measure the ambient CCN concentration at a reasonable time resolution due to the technical difficulty. One alternative approach is to estimate the CCN concentration by combining the aerosol size distribution measurement with the chemical composition data. In this study, the aerosol size distribution is based on a size range between 140 and 3000 nm, which limits the CCN concentration estimation. A newly developed aerosol size distribution payload (package 2) can overcome this limitation but lacks the chemical composition measurement. One ongoing effort is to target this issue during the 2024 deployment.

The averaged chemical composition obtained from the filter sampler and the MN-AMS technique for the deployment period (between 9 and 16 November 2021) is summarized in Table 2, which provides critical information for CCN concentration estimation. Note that we
assume that aerosol particles were either well mixed or entirely coated with organics, and the bulk properties measured by MN-AMS represent the aerosol properties across the whole CCN size range, which does not consider other heterogeneity of mixing states and varying composition with size. First, the hygroscopicity $\kappa_{\text{org}}$ of the organic component was estimated using the relationship between hygroscopicity and O/C ratio (Cain and Pandis 2017; Cerully et al. 2015; Han et al. 2022; Mei et al. 2013; Thalman et al. 2017), giving a $\kappa_{\text{org}}$ range of 0.04–0.11. We then used the well-mixed chemical composition assumption and estimated the overall hygroscopicity $\kappa_{\text{overall}}$ of 0.23–0.33. The SIMS 3D imaging indicated that an organic-enriched surface layer usually covers the inorganic species core, forming inorganic–organic (core–shell) structures. Thus, a second assumption to test is that the organic shell properties determine the expressed aerosol optical and microphysical properties, which $\kappa_{\text{overall}}$ is the assigned values of $\kappa_{\text{org}}$. In this assignment, we also assume that surface tension depression by the organic aerosol significantly affects the formation of cloud droplets (Lowe et al. 2019).

Based on the $\kappa$-Köhler theory, vertical CCN profiles were calculated using the aerosol size distribution from the portable optical particle spectrometer (POPS, Fig. S1) and the miniaturized optical particle counter (mOPC) under a low supersaturation (SS = $\sim$0.1%), shown in Fig. 3 and Fig. S2. Due to the measurement limitation in this deployment, we assume that aerosol particles of different sizes have the same chemical composition as a proof-of-concept demonstration. Different assumptions about aerosol chemical properties and mixing states lead to significant discrepancies in CCN concentration estimation. The estimated CCN concentration under the well-mixed assumption was a factor of 3–5 higher than that under the organic-dominated shell assumption. In addition, the ground-based measurement of CCN concentration at 0.1% supersaturation was $471 \pm 44 \text{ cm}^{-3}$ during the same sampling period (1635–1745 UTC), in between most of the values generated with the two assumptions (Fig. 3). Comparison of aerosol number concentrations (in Fig. S3) showed that the ground measurement from the ARM Observation System (AOS) condensation particle counter (CPC) was always higher than the number concentration measured by the UAS CPC, which is consistent with the previous study—the measurements show a decreasing trend in aerosol number concentration above surface layer. Under the organic-dominated shell assumption, the estimated CCN concentration aligned better with the ground-based measurement data. The well-mixed organic–inorganic component assumption tends to overestimate the CCN concentration.

### b. LES modeling results and comparison to UAS and ARM measurements.

Following previous studies, we configured the Weather Research and Forecasting (WRF) Model version 4 (Skamarock et al. 2019; Gustafson et al., 2020) in idealized LES mode (Wang et al. 2009) with doubly periodic horizontal boundary conditions and prescribed large-scale advective forcings (Endo et al. 2015; Li et al. 2022). The LES domain was set to $20 \times 20 \text{ km}^2$ in horizontal dimension with a uniform grid spacing of $100 \times 100 \text{ m}^2$ per grid cell and 7 km in the vertical dimension. The model was initialized with initial conditions from ERA5 reanalysis (Copernicus 2020). The model was run for 72 hours with a time step of 120 seconds, and the results were averaged over the last 24 hours to represent the sampling period.

#### Table 2. MN-AMS analysis of chemical compositions of UAS filter samples and the estimated hygroscopicity information.

<table>
<thead>
<tr>
<th></th>
<th>(NH$_4$)$_2$SO$_4$</th>
<th>Organic$^a$ (C$<em>{1,1.79}$H$</em>{1.39}$O$<em>{0.36}$N$</em>{0.33}$)</th>
<th>NH$_4$NO$_3$</th>
<th>Well mixed</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mass concentration (µg m$^{-3}$)</td>
<td>0.59</td>
<td>1.30</td>
<td>0.22</td>
<td>2.11</td>
</tr>
<tr>
<td>Volume fraction</td>
<td>23.8%</td>
<td>66.9%</td>
<td>9.3%</td>
<td>100%</td>
</tr>
<tr>
<td>Refractive index at 532 nm</td>
<td>1.53</td>
<td>1.45–1.52</td>
<td>1.61</td>
<td>1.49–1.53</td>
</tr>
<tr>
<td>Hygroscopicity $\kappa$</td>
<td>0.61</td>
<td>0.04–0.11</td>
<td>0.67</td>
<td>0.23–0.33</td>
</tr>
<tr>
<td>Growth factor at 75% RH</td>
<td>1.04–1.10</td>
<td></td>
<td>1.19–1.23</td>
<td></td>
</tr>
</tbody>
</table>

$^a$ Average elemental composition of the organics measured by the MN-AMS.
with 153 stretched layers, having a layer thickness of around 30 m in the lower 3 km and 75 m between 3 and 7 km. We used the Morrison two-moment cloud microphysical scheme that predicts cloud droplet numbers based on the activation of aerosols in multimodal size distribution (Endo et al. 2015). Four constant lognormal size modes, fitted to the aerosol size distributions from the POPS measurements and modal hygroscopicities of 0.04 and 0.33, respectively, were used in two LES experiments. Note that the aerosol kinetics has not yet been parameterized into the LES model. Thus, the LES experiments were focused on the chemical composition effects on the bulk hygroscopicity of aerosols and its further impact on the cloud with idealized model settings for simplicity. The experiments were run from 0600 UTC 9 November to 0600 UTC 11 November. In the LES sensitivity experiments, the aerosol size distribution was based on the averaged UAS boundary layer measurements that account for the impact of transport and vertical mixing on aerosol distribution. The initial conditions, surface turbulent heat fluxes, and large-scale advective tendencies for LES were all obtained from the fifth-generation European Centre for Medium-Range Weather Forecasts (ECMWF) atmospheric reanalysis (ERA5). Domain-average horizontal wind components ($u$, $v$) were nudged to ERA5 using a 3-h time scale.

The UAS measurements of wind, atmospheric states, and aerosol properties were taken under cloudless conditions on 9 November. We first evaluated the LES cloudless turbulence against the UAS measurements. Figure S4 shows a comparison of the frequency distribution of vertical velocity. Given the limited measurement time and spatial coverage, the similarity in the frequency distribution between the WRF-LES simulations and UAS measurements is remarkable. The vertical profiles of turbulent kinetic energy (TKE) and $w^2$ ($w$, perturbation of vertical wind component) are also well captured in the WRF-LES simulation, shown in Fig. 4, except for the 100-m turbulent layer at 1100 m above the surface, near the boundary layer top.

Fig. 3. Estimated CCN concentrations (at 0.1% supersaturation) under different aerosol mixing assumptions (the black circles represent the well-mixed condition, and the blue circles represent the organic-dominated surface condition) using in situ UAS aerosol size distribution measurements from the POPS and mOPC on 9 Nov 2021. Note that the maroon symbol represents the CCN concentration from the ground site at the SGP (314 m).
This feature is likely because the inversion layer simulated in the LES (initialized and driven by ERA5 boundary forcing conditions) is less sharp (thicker and weaker) than that captured in the UAS measurements (Fig. 4), leading to a weaker mixing generated by wind shear near the inversion.

According to the WRF-LES simulation results, the assumption of aerosol mixing state and the derived bulk hygroscopicity ($\kappa = 0.04$ or $0.33$) substantially impact cloud properties and radiation. Figure 5 presents the time series of the cloud properties from 0600 UTC 10 November to 0600 UTC 11 November. The retrieved properties from the remote sensing (details included in the supplemental material) were compared with the WRF-LES simulation results. The retrieved cloud droplet number concentration $N_c$ and cloud liquid water path (LWP) are larger when using the higher $\kappa$ based on the well-mixed assumption.

Fig. 5. Time series of LWP, cloud-top height, and cloud droplet number concentration $N_c$ on 10 Nov 2021. Plus (red) and circle (blue) signs are from the WRF-LES simulations with $\kappa = 0.04$ and $\kappa = 0.33$, respectively. The model simulated LWP, cloud top, and $N_c$ are evaluated against ARM ground-based microwave radiometer and lidar retrievals averaged over cloud samples (LWP $\geq$ 25 g m$^{-2}$) every 30 min, with black squares with line bars marking the mean and one standard deviation.
However, the simulation with the lower $\kappa$, tied to the organic-dominated shell assumption, agreed better with the retrieved $N_c$. The difference between the two simulations in 12-h (from 1200 UTC) averaged LWP, $N_c$, and the effective radius of cloud droplet size distribution ($r_{\text{eff}}$) is 16.6 g m$^{-2}$, 255.5 cm$^{-3}$, and $-1.6 \mu$m, respectively. While liquid cloud water at various heights contributed to the difference in LWP, the most significant differences in $N_c$ and $r_{\text{eff}}$ were within clouds below 3 km (shown in Fig. S5).

The more abundant, smaller cloud droplets and larger LWP in the $\kappa = 0.33$ simulation induce additional short-wave cooling of $-1.2$ W m$^{-2}$ during the same 12-h period. Instantaneous differences in short-wave cooling are as large as $-10$ W m$^{-2}$ during morning hours in the $\kappa = 0.33$ simulation than in the $\kappa = 0.04$ simulation. Although the ARM measurements of $N_c$ show considerable variation, the mean values are much closer to the model results with $\kappa = 0.04$ than $\kappa = 0.33$. This comparison implies that the organic-dominated surface properties primarily determine the droplet nucleating ability of the aerosol particles, given the same aerosol size distributions. The excellent agreement of LWP and cloud-top height between the WRF-LES simulations and the ARM ground-based remote sensing measurements suggests that the model can reasonably simulate the observed clouds at the SGP site, although there might be compensating errors given the simplified experiment design for the proof-of-concept purpose.

4. Impact of the newly orchestrated ARM capabilities

This study showcases one approach to link observations with models in atmospheric studies. This approach shows great potential in constraining process-level model simulations via integrating ARM observatory, midsize UAS observational capabilities, and advanced chemical analysis. Here, we use a flight conducted on 9 November 2021, to demonstrate the importance of characterizing aerosols’ chemical composition and surface properties in simulating their microphysical and optical properties. By integrating this advanced chemical information with the vertical profiles of aerosol microphysical properties, the fidelity of LES simulations of the aerosol effects on clouds and radiation budget can be improved. Furthermore, the integration of these novel chemical analysis techniques with UAS capabilities enhances the observational capability of the U.S. DOE ARM Aerial Facility, enabling a better process-level understanding of atmospheric aerosols and their effects on climate.

One of the ideal measurement assemblies is sketched in Fig. 6, which shows a schematic representation of various observational capabilities harnessed together. The ARM observatory provides near-surface coverage and ground-based remote sensing, enhanced with airborne sampling capabilities from other collaborators. Various airborne platforms, such as UAS and piloted aircraft, cover the troposphere. Such a united measurement power provides the comprehensive inputs needed for climate modeling and constraints on modeling aerosol forcing in various perspectives (Kahn et al. 2023), such as carrying out a statistical sampling of key variables for major aerosol airmass types, characterizing an aerosol type for climate application with a suite of specific physical, chemical, and optical quantities, and capturing the aerosol evolution during transport.

Meanwhile, this study highlights the great opportunities the newly combined capabilities provide. Also, it points out several observational needs, such as extending the aerosol size distribution measurement range, enhancing the chemical composition analysis, and improving the sampling strategy.

UAS measurements with a high temporospatial resolution have a promising future in capturing critical atmospheric properties in the cloud-top entrainment zone and boundary layer turbulence structure. The approach discussed in this study has the future capacity to refine ground-based lidar retrieval algorithms and narrow the gap between observations and a hierarchy of atmospheric models. After the new measurements/parameterizations help
bridge gaps between observations and model simulations, we can improve subgrid-scale aerosol–cloud parameterizations in large-scale models and eventually the model prediction accuracy for future atmospheric or climate applications. This ARM platform may have applications for capturing aerosol chemical composition and surface properties in different altitudes of the boundary layer, tracking aerosol evolution with a Lagrangian flight pattern, characterizing aerosol concentration statistics below and above clouds, and assessing the impact of entrainment mixing on various cloud types.

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**Data availability statement.** The data mentioned in this manuscript can be freely accessed through the ARM Data Discovery website ([https://adc.arm.gov/discovery/#/](https://adc.arm.gov/discovery/#/)). The UAS data were archived through the ARM Data Center (ADC). The data collected from the ArcticShark mission can be found at [https://adc.arm.gov/discovery/#/results/s::u2](https://adc.arm.gov/discovery/#/results/s::u2). The data collected from the TigerShark mission can be found at [https://adc.arm.gov/discovery/#/results/s::u3](https://adc.arm.gov/discovery/#/results/s::u3). The source code used for the simulations of this study, the Weather Research and Forecasting (WRF) Model, is freely available on [https://code.arm.gov/lasso/lasso-wrf](https://code.arm.gov/lasso/lasso-wrf).


