Supplemental Material

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

I. κ-Köhler theory

In κ-Köhler theory, the water vapor saturation ratio ($S$) over an aqueous solution droplet is given by:

$$S = \frac{D^3 - D_p^3}{D^3 - D_p^3} \exp\left(\frac{4\sigma_w M_w}{RT \rho_w D}\right)$$  \quad (1)

where $D$ is the droplet diameter, $D_p$ is the dry diameter of the aerosol particle, $M_w$ is the molecular weight of water, $\sigma_w$ is the surface tension of pure water, $\rho_w$ is the density of water, $R$ is the gas constant, and $T$ is the absolute temperature. When $\kappa$ is greater than 0.1, it can be conveniently derived as:

$$\kappa = \frac{4A^3}{27D^3S_c^2}$$  \quad (2)

where $A = \frac{4\sigma_w M_w}{RT \rho_w D}$, and $S_c$ is the particle critical supersaturation that was derived using the approach detailed in references (Cerully et al. 2015; Mei et al. 2013; Petters and Kreidenweis 2007, 2013; Thalman et al. 2017).

II. The procedure of CCN estimation

We used the following steps to estimate cloud condensation nuclei (CCN):

1. **Supplementary Materials for**

2. **Bridging observational capabilities and process-level simulation: new insight into aerosol roles in the Earth system**


*Corresponding authors: Email: fan.mei@pnnl.gov (Fan Mei) , hailong.wang@pnnl.gov (Hailong Wang)
1) Obtain the chemical composition of the particle of interest, including the bulk chemical composition from the MN-AMS and the surface properties from the SIMS.

2) Estimate the overall hygroscopicity ($\kappa$) weighted by the volume fraction of each species when assuming the well-mixed case.

3) Determine the activation size under a given critical supersaturation (0.1% in this study) using $\kappa$-Köhler theory (equation 2).

4) Integrate over the POPS size distribution, assuming the chemical composition of the aerosol population remains the same, to obtain an estimate of the total CCN concentration.

Note that we can improve the CCN estimation with size distribution from other instruments if available.

III. The procedure of estimating aerosol optical properties

Estimating the optical properties of aerosols (such as extinction coefficient and single-scattering albedo) based on their size distribution, relative humidity (RH), and refractive index involves the following steps:

1) Get the aerosol input data, such as the size distribution of aerosols and refractive index: This step can use any size distribution measurement and has been demonstrated using POPS measurement in Fig. S6. Note that the limited size range of the POPS (140 - 3000 nm) introduces uncertainty in extinction estimation. The uncertainty range depends on various factors, including the accuracy of the size distribution measurement, the range of sizes included in the calculation, and the size distribution model used. For the refractive index of the aerosol material, we used the previously published values based on the laboratory measurements, listed in Table 3. The effective refractive index of an aerosol mixture is the weighted average of refractive indices of its constituent components, where the weighting factor is the component mass fraction. This study used the bulk mass fractions of each chemical species from the MN-AMS analysis as the weighting factor.

2) Estimate the extinction coefficient: The extinction coefficient is the sum of the scattering and absorption coefficients and has been calculated from the effective refractive index and size distribution of the aerosols based on the Mie theory (the code from Bohren and Huffman version), and we assumed that all aerosol particles are spherical.

3) Compute the single-scattering albedo: The single-scattering albedo is the ratio of the scattering coefficient measured by the STAP to the estimated extinction. It's important to
note that the error propagation in the estimation of the optical properties significantly impacts the accuracy of other calculations that rely on the extinction coefficient, such as the single-scattering albedo.

4) Consider the effects of relative humidity: The optical properties of aerosols depend on relative humidity, and the size distribution and complex refractive index of the aerosols should be adjusted accordingly. In this study, we neglect the RH effect on the refractive index, and only consider the effect on the size distribution. Firstly, the growth factor (GF) was calculated as a function of relative humidity and hygroscopicity (equation 11 in Petters and Kreidenweis’s paper, 2008), which was based on the chemical analysis from the MN-AMS and SIMS. Secondly, we assumed that the same GF would weigh the whole size distribution. Thirdly, we used the weighted size distribution to estimate the extinction under the ambient RH condition.

5) Determine the Aerosol Optical Depth (AOD): AOD can be calculated based on the extinction at each altitude by integrating the extinction coefficient along the vertical column of the atmosphere. $\text{AOD} = \int \beta dx = \Sigma (\beta * \Delta x)$

IV. Observational input for the LES model

The LES model takes measurements of atmospheric state (e.g., profiles of temperature, humidity, wind), surface turbulent heat and moisture fluxes, and aerosol properties to constrain and/or initialize simulations. Notably, in this study, aerosol size distributions from the POPS measurements are averaged to account for the impact of transport and vertical mixing on aerosol distribution and fitted into four log-normal modes. In addition, bulk hygroscopicity for each size mode, which affects the calculation of aerosol activation and cloud droplet number concentration, is obtained from the offline calculation based on the core-shell particle structure revealed by ToF-SIMS chemical analysis.

V. Ground-based remote sensing retrievals

Operated by various ARM deployments, ground-based remote sensing instruments provide continuous observations of clouds, aerosols and meteorological properties across different regions. The co-located deployment of UAS with the ground measurements at the SGP provided an excellent opportunity to take advantage of ample ground-based remote sensing data. This study
compared various retrieved cloud properties with the model simulation results (discussed in section 3.b).

The ARM Raman lidar measures profiles of water vapor and aerosol optical properties with data provided via several VAPs (Newsom et al. 2021; Turner et al. 2016). The Raman Lidar Profiles—Feature Detection and Extinction (RLPROF-FEX) VAP (https://www.arm.gov/capabilities/vaps/rlprof-fex) provides height- and time-resolved estimates of cloud and aerosol optical properties, including aerosol backscatter coefficient, extinction, scattering ratio, lidar ratio, and linear depolarization ratio (Chand et al. 2019).

Cloud liquid water path (LWP), with a temporal resolution of ~28 s, is derived from microwave radiometer (MWR) measurements and is available from the ARM MWR retrievals (MWRRET VAP, https://www.arm.gov/capabilities/vaps/mwrret). In addition, cloud-top height is obtained from the ARM Active Remote Sensing of Clouds product using the Ka-band ARM zenith radar VAP (KAZRARSCL, https://www.arm.gov/capabilities/vaps/kazrarscl). KAZRARSCL uses merged measurements from radar and lidars to provide cloud boundary detection for up to 10 cloud layers with a vertical resolution of 30 m and a temporal resolution of 4 s. Cloud droplet number concentration (Nc), LWP, and the effective radius of cloud droplet size distribution ($r_{eff}$) are retrieved using the Raman lidar particulate extinction profile from the Raman Lidar Vertical Profiles Feature Detection and Extinction VAP, based on the methodology developed by (Snider et al. 2017). Zhang et al. (2023) evaluated Nc retrievals with in situ probe measurements during the ARM Aerosol and Cloud Experiments in the Eastern North Atlantic (ACE-ENA) field campaign (Zhang et al. 2023). They showed that lidar-based Nc retrievals have relative differences between ~10% to ~30% with respect to Fast-Cloud Droplet Probe (FCDP) measurements, with smaller differences for overcast clouds and larger differences for broken clouds.

VI. Boundary layer profiling

Using the UAS in-situ measurements, we captured the partial mixing layer and entrainment zone of the planetary boundary layer between 16:35-17:45 on November 9, 2021, as shown in Fig. S7. The airborne aerosol data were averaged every 50 m in height in Fig. S6 and generally showed consistency with the ground observations in Table S2. The ambient temperature profile in Fig. S6 (a) shows a weak inversion layer between 750 and 850 m. Then the temperature slightly decreased with altitude until a strong inversion at 1200 m. Between 850 and 1200 m, the constant relative
humidity and accumulation-mode particle concentration, indicated the well-mixed boundary layer in the sampling altitude range. According to the temperature profile, the entrainment zone near the boundary-layer top was relatively shallow (less than 100 m). However, as shown in Fig. S7 (d), the wind profiles demonstrated a relatively strong wind shear with wind speed increasing from 1 m/s to 4 m/s between 1100 and 1300 m. Meanwhile, the vertical velocity switched from positive to negative values over the same height range. Both profiling figures illustrated that the downdraft of free tropospheric air helped cap the inversion and limit the uplift of boundary-layer moisture and aerosol particles.

Supplementary Figures and Tables

Fig. S1. The averaged POPS size distribution for flight legs above 1500 m and below 1500 m between 16:40-17:40 on November 9, 2021
Fig. S2. Different profiling trends were observed on the same day, November 11, 2021. The later period (16:00-17:10 UTC) has an opposite vertical trend compared to the earlier period (14:50-15:40 UTC).
Fig. S3. Comparison between aerosol number concentrations from the ArcticShark flight and ground measurements from the ARM Observation System (AOS) on November 9, 2021.

Figure S4: Comparison of the frequency distribution of cloudless vertical velocity (w, m s\(^{-1}\)) between UAS measurements and WRF-LES simulations on November 9, 2021. WRF-LES output is sampled within the timeframe and altitude range of UAS measurements but with more data points. Thus, the frequencies within the bins are normalized by the respective total samples. The
5 Hz UAS samples are taken at a 10-point moving average to be comparable with the model grid samples at 100 m spacing.

Figure S5: Vertical profiles of liquid water content (LWC), cloud droplet number concentration ($N_c$), and effective droplet radius ($r_{eff}$) averaged between 12 UTC on November 10 and 00 UTC on November 11, 2021, for the two WRF-LES simulations with $\kappa=0.04$ and $\kappa=0.33$, respectively.

Figure S6. UAS in situ measured and estimated optical properties under different aerosol surface property conditions (well-mixed condition and organic-dominated surface condition). The left panel is extinction coefficients at 525 nm estimated using the dry POPS size distribution, ambient humidity, and aerosol refractive index listed in Table 3. The middle panel shows the absorption
coefficient from the STAP (525 nm, filter correction applied), and the right panel is the estimated single scattering albedo under two assumptions.

Figure S7. Profiles of (a) ambient temperature, (b) ambient relative humidity (RH), (c) potential temperature, (d) the vertical wind speed, and (e) the horizontal wind speed from the flight between 16:35-17:45 UTC on November 9, 2021.

Table S1: UAS November flight information

<table>
<thead>
<tr>
<th>Date</th>
<th>Takeoff</th>
<th>Landing</th>
<th>Flight info</th>
</tr>
</thead>
<tbody>
<tr>
<td>11/8/2021</td>
<td>14:42</td>
<td>15:12</td>
<td>AIMMS-30 calibration flight (no sample collection)</td>
</tr>
<tr>
<td>11/9/2021</td>
<td>15:57</td>
<td>17:54</td>
<td>Profiling above SGP central facility (collecting aerosol samples with the sampler)</td>
</tr>
<tr>
<td>11/11/2021</td>
<td>14:34</td>
<td>17:58</td>
<td>Profiling above SGP central facility (collecting aerosol samples with the sampler)</td>
</tr>
<tr>
<td>11/13/2021</td>
<td>15:15</td>
<td>18:19</td>
<td>Lawn-mowing pattern (collecting aerosol samples with the sampler)</td>
</tr>
<tr>
<td>11/14/2021</td>
<td>18:18</td>
<td>19:04</td>
<td>Troubleshooting a connection issue (no sample collection)</td>
</tr>
<tr>
<td>11/15/2021</td>
<td>14:26</td>
<td>17:46</td>
<td>Profiling above SGP central facility (collecting aerosol samples with the sampler, issue with AIMMS recording)</td>
</tr>
<tr>
<td>11/16/2021</td>
<td>14:38</td>
<td>16:00</td>
<td>Attempt to fly another lawn-mowing pattern (no sample collection)</td>
</tr>
</tbody>
</table>
Table S2: Ground measurements from the Aerosol Observation System (AOS) at the SGP central facility

<table>
<thead>
<tr>
<th>Periods</th>
<th>Ambient Temperature, C</th>
<th>N_{CCN}, cm(^{-3})</th>
<th>N_{CCN}, SS=0.1%, cm(^{-3})</th>
<th>N_{CCN}, SS=0.2%, cm(^{-3})</th>
<th>σ_{SC}, Mm(^{-1}) (at 532 nm)</th>
<th>σ_{abs}, Mm(^{-1}) (at 532 nm)</th>
<th>SSA (at 532 nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>11/09/2021, 16:40 -17:40</td>
<td>15.42±0.76</td>
<td>3348±58</td>
<td>471±44</td>
<td>1256±197</td>
<td>37.86±4.35</td>
<td>4.21±0.03</td>
<td>0.90±0.01</td>
</tr>
<tr>
<td>11/11/2021, 14:50 – 15:40</td>
<td>7.50±0.50</td>
<td>1556±48</td>
<td>34±11</td>
<td>202±20</td>
<td>3.06±0.85</td>
<td>0.36±0.04</td>
<td>0.89±0.02</td>
</tr>
<tr>
<td>11/11/2021, 16:00 – 17:10</td>
<td>10.66±1.00</td>
<td>1479±51</td>
<td>32±18</td>
<td>189±42</td>
<td>3.48±0.68</td>
<td>0.36±0.05</td>
<td>0.90±0.02</td>
</tr>
</tbody>
</table>

Reference


