



Measurement Report: Tethered Balloon Observations of Vertically Resolved Aerosol Size Distributions during the U.S. DOE ARM CoURAGE and BNF Campaigns

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Abstract. The vertical distribution of aerosol particle size is important for aerosol transport, atmospheric mixing, and surface
15 air quality, yet vertically resolved aerosol size-distribution observations remain limited. Here, a miniature scanning electrical mobility spectrometer (mSEMS) was deployed aboard the U.S. Department of Energy Atmospheric Radiation Measurement (ARM) Tethered Balloon System (TBS) during the 2025 CoURAGE and Bankhead National Forest (BNF) campaigns. Aerosol particle number size distributions were measured from the surface to approximately 1200 m AGL over a diameter range of 10–300 nm. A total of 63 aerosol profiles were obtained, providing one of the most comprehensive vertically resolved aerosol
20 size-distribution datasets collected using a tethered balloon platform. Within the planetary boundary layer (PBL), observations revealed vertically homogeneous profiles associated with efficient turbulent mixing and heterogeneous profiles reflecting incomplete mixing, surface influences, or elevated particle layers. Profiles extending above the PBL further resolved distinct aerosol transitions into the lower free troposphere and nocturnal residual layer. A representative case on 27 March 2025 at BNF documented the evolution of particles associated with a regional new particle formation and growth event: particles
25 formed during the previous day were preserved overnight within the residual layer, vertically reorganized during early-morning boundary-layer development, and subsequently remixed throughout the daytime mixed layer. These observations demonstrate strong coupling between boundary-layer evolution and aerosol vertical structure and establish an observational framework linking aerosol size distributions, residual-layer storage, and atmospheric mixing. The dataset provides valuable constraints for understanding lower-tropospheric aerosol life cycles and evaluating atmospheric chemistry and transport models.



30 1. Introduction

Atmospheric aerosol properties often vary substantially with altitude due to the combined influences of emissions, atmospheric transport, boundary-layer evolution, aerosol formation and growth, and cloud processing (Engvall et al., 2008; Yu et al., 2016; O'Donnell et al., 2023; Wang et al., 2023; Chen et al., 2025). As a result, aerosol populations observed at the surface may not be representative of those present throughout the lower troposphere. Characterizing aerosol vertical profiles is therefore important for understanding aerosol evolution, atmospheric mixing, and the exchange of aerosol populations between surface layers, residual layers, and the free troposphere (Mei et al., 2024a; Rosenfeld et al., 2014; Lin et al., 2023).

Despite decades of surface-based aerosol size distribution measurements (Kulmala et al., 2012; Nieminen et al., 2018; Ling et al., 2019; Yu et al., 2020; Jokinen et al., 2022), vertically resolved observations of aerosol particle size distributions remain relatively limited (Wang et al., 2023, Kuang, 2023a). Previous studies have reported elevated aerosol layers, residual-layer particle reservoirs, and vertically separated particle growth events, suggesting that aerosol evolution frequently occurs within layers that are only weakly coupled to the surface (Chen et al., 2018; Carnerero et al., 2018, Williamson et al., 2019, Zhao et al., 2020, Lampilahti et al., 2021; Lai et al., 2022, O'Donnell et al., 2023; Wang et al., 2023, Kuang et al., 2023a). However, routine observations of vertically resolved high resolution aerosol size distributions throughout the lower troposphere remain sparse, particularly across contrasting atmospheric environments. Tethered balloon systems (TBS) provide a unique observational platform for characterizing aerosol vertical profiles because they can repeatedly measure the lower atmosphere with high vertical resolution while carrying compact aerosol instrumentation (Ran et al., 2022; Mei et al., 2025, 2026; Fan et al., 2026). Such observations can reveal aerosol layering, boundary-layer coupling, and vertical redistribution processes that are difficult to identify using surface measurements or remote-sensing observations alone. However, to date, vertically resolved observations of aerosol particle size distributions with high resolution in altitude, time, and particle size, spanning the ultrafine and accumulation modes—where the majority of aerosol number concentration resides and where particles relevant to cloud condensation nuclei (CCN) are found (Salma et al., 2011; Pöhlker et al., 2021; Dedrick et al., 2024)—remain scarce, limiting our understanding of how aerosol populations evolve and interact with boundary-layer processes under different atmospheric environments.

To address this observational gap, the DOE Atmospheric Radiation Measurement (ARM) TBS deployments (Dexheimer et al., 2026) equipped with a miniature scanning electrical mobility spectrometer (mSEMS) were conducted as part of the DOE ARM-supported Seasonal/Spatial Vertical NPF Variation (SSVNV, Zhang et al., 2026a) campaign during the ARM Mobile Facility 1 Coast-Urban-Rural Atmospheric Gradient Experiment (AMF1 CoURAGE) in the Baltimore region (Davis et al., 2024) and the ARM Mobile Facility 3 deployment at Bankhead National Forest (AMF3 BNF), Alabama (Kuang et al., 2023b and 2026. **Figure S1** for the locations of CoURAGE and BNF). The SSVNV field campaign was designed to investigate vertically resolved aerosol variability, boundary-layer structure, and new particle formation (NPF) processes across contrasting atmospheric and emission environments. The resulting observations provide a unique opportunity to characterize aerosol vertical structures and particle size distributions under substantially different atmospheric, emission, and boundary-



layer conditions.

The primary objectives of this study are to present this high-resolution, vertically resolved aerosol dataset and to (1) characterize recurring aerosol vertical profile regimes within the planetary boundary layer (PBL), (2) identify elevated layers of small particles and other vertically heterogeneous aerosol structures, and (3) examine how these vertical structures evolve in relation to PBL-top transitions and residual-layer processes. This study provides new observational insights into the complexity and temporal evolution of aerosol vertical distributions and highlights the importance of vertically resolved measurements for understanding aerosol–boundary-layer interactions across diverse atmospheric environments.

2. Method

2.1 Vertical aerosol size distribution measurements

A miniature scanning electrical mobility spectrometer (mSEMS; Brechtel Manufacturing Inc.) was deployed onboard the DOE ARM TBS to obtain vertically resolved aerosol particle number size distributions throughout the lower troposphere. The mSEMS is a compact scanning mobility particle sizer that determines aerosol size distributions based on electrical mobility classification using a differential mobility analyser coupled with an advanced mixing condensation particle counter. More detailed information can be found in the “Miniaturized Scanning Electrical Mobility Sizer (mSEMS) Instrument Handbook” (Mei et al., 2024b).

The airborne mSEMS measured aerosol particle number size distributions over a nominal mobility diameter range of approximately 10–300 nm using 48 logarithmically spaced size bins, with a time resolution of 15s for all bins. The instrument provided continuous high-time-resolution measurements suitable for resolving rapid vertical changes in ultrafine, Aitken-mode, and accumulation-mode aerosol populations within the evolving boundary layer. The observations enabled characterization of aerosol layering, elevated new-particle populations, and vertically decoupled aerosol structures under contrasting forest and urban/coastal atmospheric environments. The mSEMS was integrated into the TBS payload using a lightweight conductive aerosol inlet positioned near the instrument package to minimize particle transmission losses. Routine flow checks and quality-control procedures were conducted throughout the campaign, and data collected during unstable flow conditions or launch and landing transitions were excluded from subsequent analysis.

2.2 TBS deployments at AMF1 CoURAGE and AMF3 BNF

TBS observations were conducted at the AMF1 CoURAGE and AMF3 BNF sites during multiple intensive operating periods spanning different seasons in 2025 (Tables 1 and S1, Figure 1 as examples of TBS launches during CoURAGE and BNF). The CoURAGE deployment represented an urban–coastal environment influenced by anthropogenic emissions, marine air masses, and land–sea interactions (Davis et al., 2024), whereas the BNF deployment represented a forested southeastern U.S. environment characterized by strong biogenic emissions and frequent new particle formation (Kuang et al, 2023b, 2026). Together, these deployments encompassed a broad range of atmospheric, emission, and boundary-layer conditions sampled during the 2025 field campaigns. At BNF, the TBS was operated at the main site (Figure S1b), allowing direct comparison with collocated surface and remote-sensing measurements, including Scanning Mobility Particle Sizer (SMPS), ceilometer,

and Doppler lidar observations. In contrast, during CoURAGE, the TBS launch site was located approximately 36 km from the AMF1 core site (**Figure S1c**). Therefore, the PBLH used for the CoURAGE analysis was derived from ceilometer measurements at the AMF1 core site and is used to provide broader context for boundary-layer evolution rather than as a strictly collocated measurement.

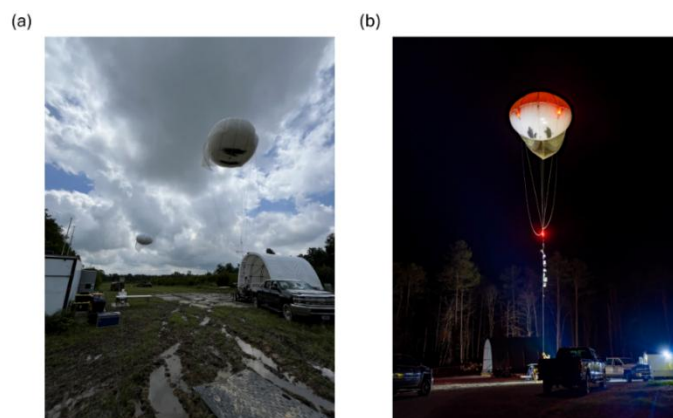


Figure 1. DOE ARM TBS deployment with mSEMS at (a) AMF1-CoURAGE and (b) AMF3-BNF in 2025

The TBS flight operations were coordinated with routine ARM observations and conducted under favourable meteorological conditions. Measurements acquired during the ascent and descent were averaged within the same altitude layers to construct the vertical aerosol size distribution profiles used in this study. Across the CoURAGE and BNF deployments, a total of 63 successful TBS flights were completed (**Table 1** summarizes the campaign statistics, while detailed flight information is provided in **Table S1**). The TBS flights were launched from the surface, with maximum flight altitudes ranging from 106 to 1201 m above ground level (AGL), depending on flight conditions and operational constraints. Most flights remained entirely within the PBL, whereas six flights extended above the PBL into either the residual layer (4 flights) or the lower free troposphere (2 flights). The resulting dataset captures a broad range of boundary-layer conditions, including well-mixed daytime PBLs, shallow nocturnal boundary layers, residual-layer environments, and periods characterized by enhanced ultrafine particle concentrations. To better represent the diurnal evolution of the PBL and associated aerosol processes, all times reported in this study are given in local time (LT), unless otherwise specified.

Table 1. Summary of TBS intensive observation periods (IOPs) and aerosol profile classifications during the CoURAGE and BNF campaigns in 2025

Campaigns	IOP	Time period	Season	Flights	Number of aerosol profile type				Cross PBL (Y/N)
					I.A	I.B	II.A	II.B	
CoURAGE	#1	02/13-02/27	Late winter	6	1	1	3	1	1
	#2	07/14-07/28	Summer	19	8	8	2	1	0
BNF	#3	03/27-04/26	Spring	15	10	0	3	2	5
	#4	05/29-06/15	Early Summer	15	8	6	0	1	0
	#5	08/09-08/23	Summer	8	3	5	0	0	0
Total				63	30	20	8	5	6



115 3. Results

The TBS-mounted mSEMS observations revealed several recurring aerosol vertical profiles associated with different stages of boundary-layer evolution during the CoURAGE and BNF deployments in 2025. Within the planetary boundary layer, aerosol profiles ranged from vertically homogeneous structures indicative of efficient turbulent mixing to vertically heterogeneous structures reflecting incomplete mixing and localized aerosol processing (**Section 3.1**). Profiles extending above the PBL further captured distinct aerosol transitions into the lower free troposphere or the nocturnal residual layer (**Section 3.2**). Case studies demonstrated that particles formed during regional growth events can remain stored within nocturnal residual layers overnight and subsequently undergo vertical redistribution as the daytime boundary layer redevelops (**Section 3.3**). Together, these observations illustrate how vertically resolved aerosol size distributions provide direct insight into aerosol transport, mixing, and atmospheric evolution that cannot be inferred from surface observations or boundary-layer height alone.

125 3.1 Aerosol profiles within the PBL

Aerosol profiles within the PBL exhibited a wide range of vertical variability during the CoURAGE and BNF deployments. To characterize aerosol variability associated with mixing processes within the PBL, measurements below the ceilometer-derived PBL height were examined for all TBS profiles, regardless of whether the flights remained entirely within the PBL or extended above the PBL top. Based on the degree of vertical variability in aerosol number size distributions, the within-PBL segments of the profiles were classified into two primary categories: **Type I**, characterized by vertically homogeneous aerosol structures, and **Type II**, characterized by vertically heterogeneous aerosol structures. Type I profiles were further divided into single accumulation-mode (“**Type I.A**”) and bimodal profiles (“**Type I.B**”), whereas Type II profiles were separated into surface-influenced profiles (“**Type II.A**”) and upper-PBL aerosol-enriched profiles (“**Type II.B**”) according to the location of the dominant aerosol enhancement. The transitions observed above the PBL top are discussed separately in **Section 3.2**.

3.1.1 Vertically homogeneous aerosol profiles (“Type I”)

Vertically homogeneous aerosol structures were the dominant profile type observed during both the CoURAGE and BNF deployments, accounting for 50 of the 63 TBS measurements (**Table 1**), with representative examples shown in Figure 2. In these profiles, aerosol size distributions remained nearly constant with altitude, indicating efficient vertical mixing and strong coupling between surface and elevated aerosol populations (Zhao et al., 2025; Canales et al., 2025). Most homogeneous profiles (30 of 50) were characterized by a single accumulation-mode peak centered at approximately 80–150 nm and were classified as “**Type I.A**” (single accumulation-mode profiles, **Figures 2a and 2b**). All Type I.A profiles were obtained during late morning, afternoon, or evening, when the PBL was well developed. Under these conditions, the maximum TBS sampling altitude remained below the ceilometer-derived PBL height, and the measurements therefore captured only the well-mixed portion of the boundary layer. The accumulation mode dominated the aerosol population throughout the sampled vertical range with little variation in modal diameter or relative contribution, consistent with efficient turbulent mixing that minimized vertical gradients in aerosol size distributions. Type I.A profiles were observed during both winter, spring and summer



deployments for both CoURAGE and BNF, indicating that this vertically homogeneous single accumulation-mode profile is not restricted to a particular season but represents a common feature of well-mixed boundary layers under a range of atmospheric conditions.

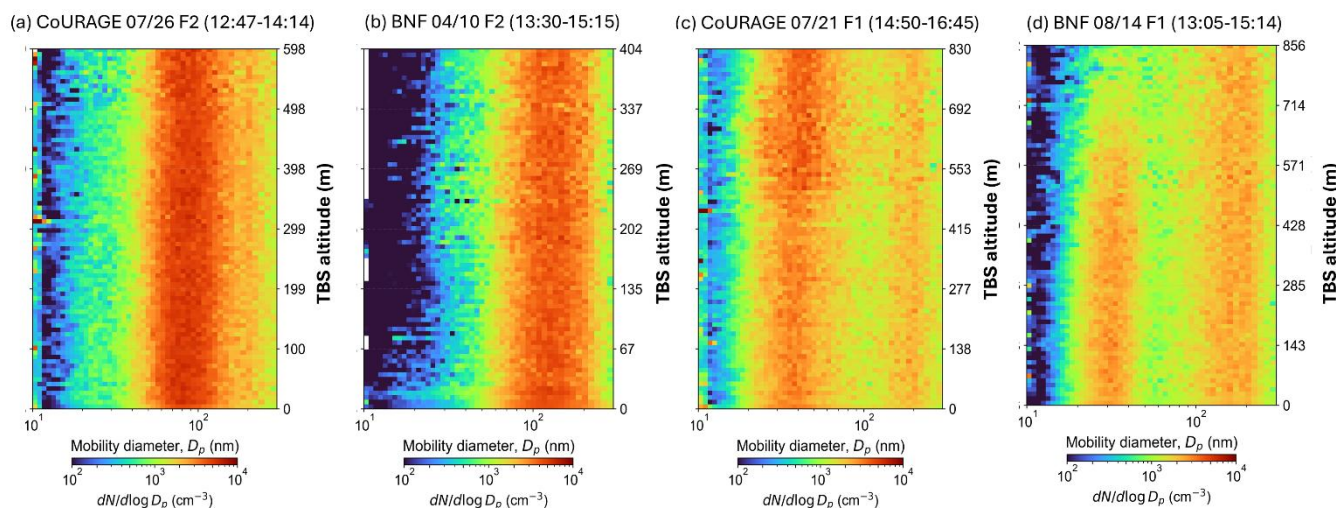


Figure 2. Aerosol vertical size distribution observed by TBS mSEMS during (a) CoURAGE 07/26/2025 Flight 1 between 10:40-12:03 LT, (b) BNF 04/10/2025 Flight 2 between 13:30-15:15 LT, (c) CoURAGE 07/21/2025 Flight 1 between 14:50-16:30 LT, and (d) BNF 08/14/2025 Flight 1 between 13:05-15:14 LT

The remaining 20 vertically homogeneous profiles exhibited persistent bimodal aerosol size distributions and were classified as “**Type I.B**”. These profiles were characterized by a distinct Aitken-mode population centered at approximately 20–50 nm together with an accumulation-mode peak near 100–200 nm (**Figures 2c and 2d**). Similar to Type I.A, both modes showed little variation with altitude throughout the sampled PBL, indicating that the two aerosol populations were well mixed within the boundary layer rather than being confined to either the surface layer or a narrow elevated layer.

A pronounced seasonal and diurnal preference was evident for Type I.B profiles. Nineteen of the twenty cases occurred during the late-spring and summer deployments, while fourteen were observed during afternoon flights and most of the remaining cases occurred during late morning (approximately 11:00–12:00 LT). Thus, bimodal profiles were observed almost exclusively from late morning through the afternoon, when photochemical activity and boundary-layer mixing were generally strongest (Lee et al., 2009; Seinfeld and Pandis, 2016; Fischer et al., 2019). The occurrence of Type I.B profiles during both the CoURAGE and BNF deployments indicates that this behavior was not unique to a single site despite the contrasting atmospheric environments of the two campaigns. The CoURAGE deployment sampled a coastal agricultural environment influenced by marine and anthropogenic air masses (Davis et al., 2024), whereas the BNF deployment represented a forested southeastern environment with strong biogenic influences (Kuang et al., 2023b, 2026). The presence of similar vertically homogeneous bimodal structures at both locations suggests that this profile type represents a recurring feature of summertime boundary-layer aerosol evolution across different environmental settings.

Concurrent surface SMPS observations at the BNF site provided valuable temporal context for interpreting the TBS

observations. The surface measurements showed that a secondary Aitken mode centered at approximately 20–50 nm repeatedly emerged from late morning through the afternoon and frequently persisted into the evening on multiple days, as illustrated by the weekly SMPS measurements covering the BNF TBS deployments on 29 May, 14 August, and 16 August (**Figure 3**). The repeated daytime appearance of this mode indicates that the bimodal aerosol structure was associated with systematic daytime atmospheric processing rather than isolated events. Together, the complementary surface and TBS observations suggest that, by the time of the balloon measurements, the Aitken-mode particles had become broadly distributed throughout the daytime mixed layer, producing vertically homogeneous bimodal aerosol profiles.

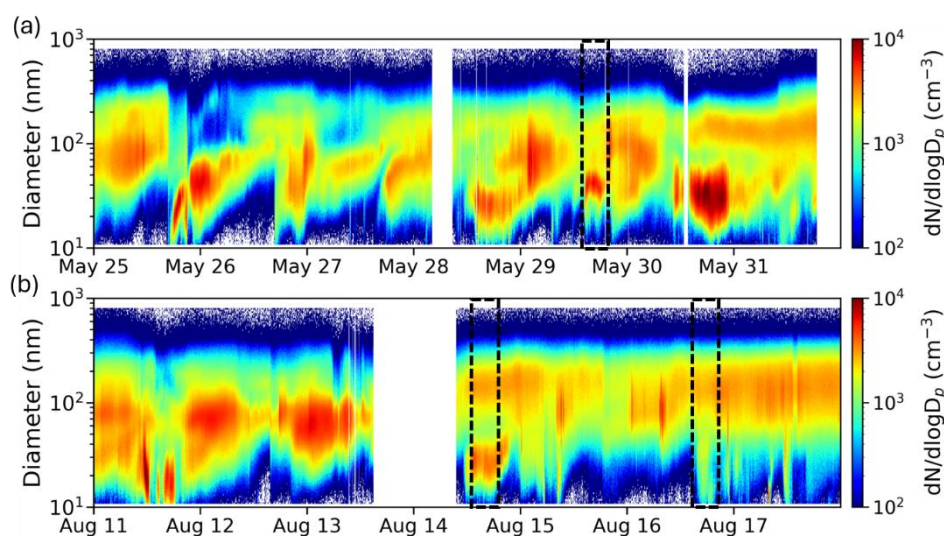


Figure 3. Time series of surface aerosol particle number size distributions measured at BNF during May 25–31 and August 11–17, 2025 (local time, UTC–5). The dashed black boxes indicate the TBS deployment periods.

Although the observed bimodal profiles shared similar vertical characteristics, the available observations suggest that the processes responsible for the secondary Aitken mode likely varied among individual cases. For many summertime cases, the repeated afternoon appearance of the 20–50 nm mode could be consistent with daytime new particle formation followed by condensational growth into the Aitken-size range (Seinfeld and Pandis, 2016; Young and Keeler, 2007; Carlton et al., 2018). Pronounced classical banana-shaped growth was not evident in many events; however, the recurring daytime emergence of the secondary mode suggests that new particle formation and growth may nevertheless have occurred under relatively high background aerosol loadings, where the earliest stages of growth were either not resolved or partially obscured by efficient particle sinks (June et al., 2026). Consequently, the observed Aitken mode likely represented a relatively mature ultrafine-particle population rather than freshly nucleated particles. Given that this study is intended primarily as an observational characterization of aerosol vertical structures, distinguishing among these potential formation pathways is beyond the scope of the present work. Future studies integrating the TBS observations with chemical transport model simulations and additional process analyses will provide a more comprehensive assessment of the mechanisms responsible for the observed Type I.B



195 profiles.

Not all Type I.B profiles, however, appeared to evolve through the same pathway. A notable example was the 16 August 2025 BNF flight, during which the evolution of the secondary Aitken mode was captured simultaneously by the TBS (**Figures 4a and 4b**), continuous surface SMPS measurements (**Figure 3b**), and Doppler lidar observations (**Figure 4c**). During the TBS ascent (**Figure 4a**), the 20–50 nm particle population was largely absent in the lowest portion of the boundary layer, whereas the same mode became evident near the surface during the subsequent descent approximately 30 minutes later (**Figure 4b**). Concurrently, the surface SMPS recorded the appearance of the secondary Aitken mode at approximately 16:00 LT (**Figure 3b**), closely matching the timing of the TBS observations. Doppler lidar measurements during the same period indicated active vertical motions within PBL, providing additional evidence for enhanced turbulent exchange between the upper boundary layer and the mixed layer. Taken together, these independent observations are consistent with rapid downward mixing or entrainment of an aerosol population from aloft, followed by efficient turbulent redistribution throughout the daytime boundary layer. The observed evolution also suggests a possible link between the upper-PBL aerosol-enriched profiles (Type II.B in following section) and the vertically homogeneous bimodal profiles (Type I.B). Under favorable mixing conditions, the elevated Aitken-mode particle population may be mixed downward and redistributed throughout the boundary layer, providing one plausible pathway for the development of Type I.B profiles. Further observations will be required to assess the prevalence of this evolutionary pathway.

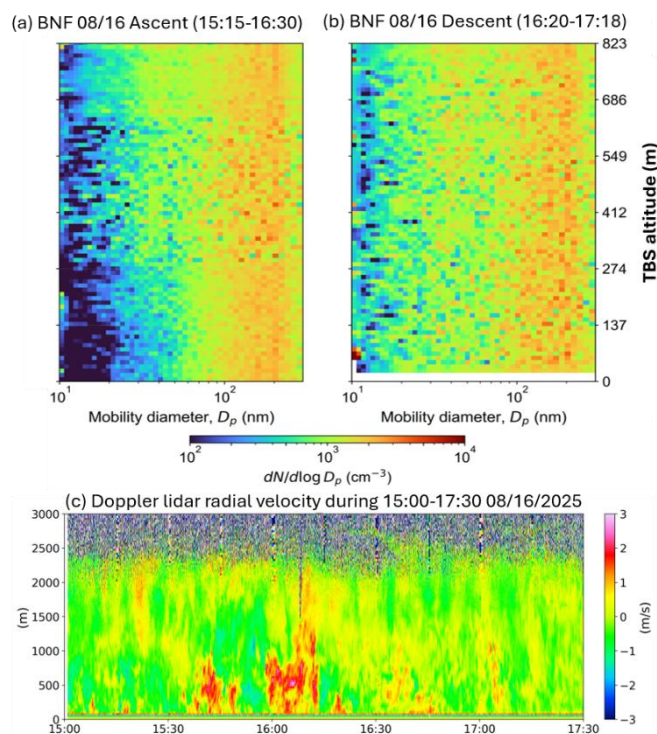


Figure 4. TBS mSEMS measurements during the BNF 16 August 2025 (a) ascent deployment (15:15–16:30 LT) and (b) descent deployment (16:30–17:18 LT), together with (c) Doppler lidar radial velocity measured during the TBS deployments.



Overall, these observations indicate that Type I.B represents a recurring summertime aerosol profile rather than a
 215 unique formation mechanism. While many cases are consistent with daytime photochemical production and subsequent
 particle growth, others appear to be more strongly influenced by boundary-layer dynamics, including entrainment and rapid
 turbulent redistribution. Regardless of the specific formation pathway, the common characteristic of all Type I.B profiles is
 the vertically homogeneous coexistence of Aitken- and accumulation-mode particles throughout the sampled boundary layer.

3.1.2 Vertically heterogeneous aerosol structures (“Type II”)

220 Although vertically homogeneous profiles were the dominant aerosol type observed during the TBS deployments, a
 substantial fraction of the measurements (13 of the 63 profiles) exhibited pronounced vertical heterogeneity in aerosol size
 distributions within the sampled PBL. In these profiles, aerosol populations varied considerably with altitude, indicating
 incomplete vertical mixing and reflecting the combined influences of boundary-layer dynamics, aerosol processing, and
 localized transport. The heterogeneous profiles comprised two distinct patterns, with (1) eight profiles exhibiting pronounced
 225 surface influence and were classified as **Type II.A** (Table S1, Figures 5a and 5b) and (2) five profiles exhibiting upper-PBL
 aerosol-enriched layers and were classified as **Type II.B** (Table S1, Figures 5c and 5d).

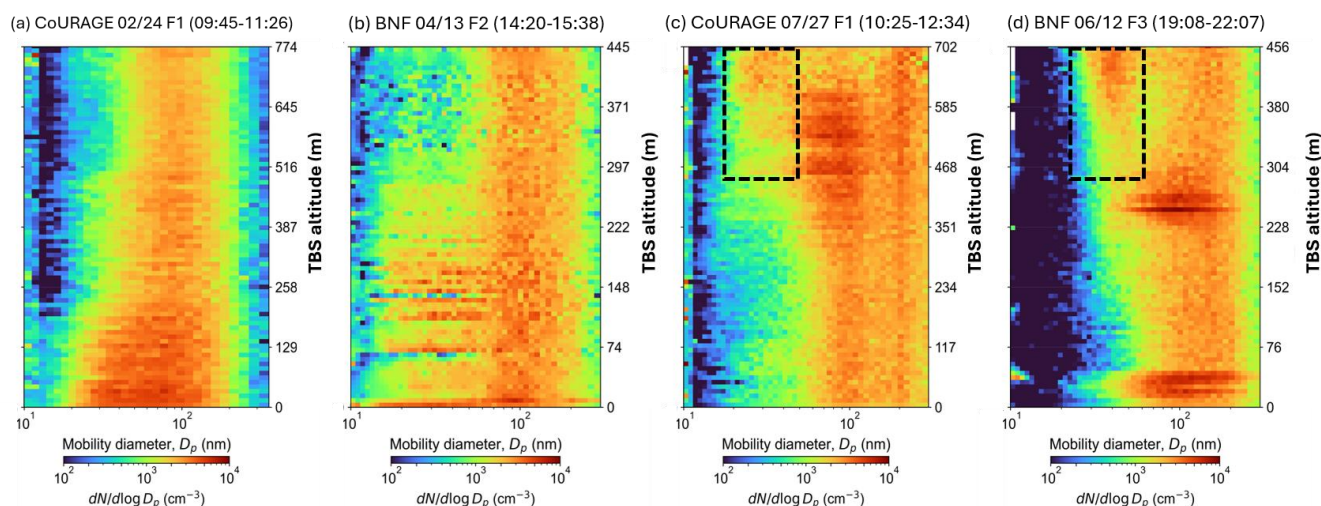


Figure 5. Aerosol vertical size distribution observed by TBS mSEMS during (a) CoURAGE 02/24/2025 Flight 1 between
 09:45-11:26 LT, (b) BNF 04/13/2025 Flight 2 between 14:20-15:38 LT, (c) CoURAGE 07/27/2025 Flight 1 between 10:25-
 12:34 LT, and (d) BNF 06/12/2025 Flight 1 between 19:08-22:07 LT
 230

The **Type II.A** profiles were characterized by enhanced concentrations of Aitken-mode particles and broader aerosol
 size distributions within the lowest several hundred meters of the boundary layer. With increasing altitude, the contribution of
 smaller particles gradually decreased, and the aerosol population transitioned toward a relatively uniform accumulation-mode
 distribution centered at approximately 80–150 nm. Unlike the upper-PBL aerosol-enriched profiles described in the following
 235 section, Type II.A profiles did not contain distinct elevated aerosol layers. Instead, aerosol properties changed gradually with
 altitude, indicating a progressive weakening of surface influences away from the ground rather than an abrupt transition



between separate aerosol populations. The enhanced abundance of Aitken-mode particles near the surface is consistent with stronger impacts of local emissions and near-surface aerosol processing, whereas aerosol populations aloft appeared more representative of the regionally mixed boundary layer. These observations suggest that local surface sources can remain evident within the lower PBL despite substantial daytime turbulent mixing. One notable example of a Type II.A profile was observed under nocturnal boundary-layer conditions, when a shallow stable PBL confined locally influenced aerosol populations within the lowest few hundred meters while the overlying residual layer remained comparatively well mixed. This case illustrates how a shallow nighttime boundary layer can preserve strong vertical gradients in aerosol size distributions despite relatively weak changes in total particle concentrations. A more detailed analysis of this event is presented in **Section 3.3** for “Case Study”.

In contrast, the **Type II.B** profiles were characterized by enhanced concentrations of Aitken-mode particles confined to the upper portion of the boundary layer, while the surrounding atmosphere remained dominated by broader accumulation-mode populations centered at 80–150 nm. Representative examples were observed during both the CoURAGE (27 July) and BNF (12 June) deployments, and aerosol layers enriched in particles between 20 and 80 nm were identified within the upper PBL, typically between 300 and 700 m above ground level. During the 27 July CoURAGE flight (**Figure 5c**), the elevated concentrations of 20–60 nm particles occurred between 450 and 700 m. During the 12 June BNF flight (**Figure 5d**), a distinct layer enriched in 30–80 nm particles was observed between approximately 300 and 450 m. In addition, a relatively concentrated accumulation-mode aerosol layer centered near 100–150 nm was present at 230–260 m, indicating that multiple vertically stratified aerosol populations coexisted within the boundary layer during this flight. In each case, the enhanced small-particle population was vertically confined and clearly distinguishable from the aerosol populations both above and below it.

Looking back at the 16 August 2025 case (Type I.B, **Section 3.1.1**), this observation further suggests a possible link between the upper-PBL aerosol-enriched profiles (Type II.B) and the vertically homogeneous bimodal profiles (Type I.B). During this event, an Aitken-mode population initially confined to the upper portion of the boundary layer was subsequently observed throughout the lower boundary layer within approximately 30 minutes, coincident with enhanced turbulent mixing inferred from Doppler lidar observations (**Figure 4c**). This evolution suggests that, under favorable mixing conditions, upper-PBL aerosol-enriched profiles may evolve into vertically homogeneous bimodal profiles through entrainment and turbulent redistribution. Although additional observations are required to determine how frequently this pathway occurs, the present observations demonstrate that such an evolution is physically plausible. The formation of these upper-PBL aerosol-enriched profiles likely involves multiple atmospheric processes. Possible mechanisms include particle formation and growth within the upper PBL, entrainment associated with boundary-layer growth, residual-layer storage, downward transport of aerosol populations from above the PBL, and subsequent turbulent redistribution within the boundary layer. The observations presented in **Section 3.2** and **Section 3.3** further demonstrate that Aitken-mode particle populations can also be present within the residual layer and lower free troposphere, suggesting these regions may serve as temporary reservoirs for upper-PBL aerosol populations. The vertically confined Aitken-mode particle populations observed in these profiles suggest that aerosol processing can occur within the upper PBL independently of near-surface conditions.



3.2 Aerosol transitions across the PBL top

Although most TBS flights remained entirely within the PBL, six profiles extended above the ceilometer-derived PBL height, providing direct observations of aerosol vertical structures across the PBL top. These measurements captured transitions from the daytime mixed layer into the lower free troposphere or from the nocturnal boundary layer into the overlying residual layer. Compared with the relatively homogeneous aerosol structures frequently observed within the PBL, the profiles crossing the PBL top exhibited more pronounced changes in aerosol size distributions, reflecting the contrasting aerosol populations on either side of the boundary-layer interface. Because the physical characteristics of the lower free troposphere and the nocturnal residual layer differ substantially, these two transition types are discussed separately below.

3.2.1 Transitions from the daytime PBL to the lower free troposphere

Two morning TBS flights extended above the planetary boundary layer (PBL) into the lower free troposphere, providing direct observations of aerosol vertical transitions across the daytime PBL top (**Figures 6a and 6b**). These transitions were captured during the morning, when the daytime PBL was still relatively shallow and the maximum TBS sampling altitude was sufficient to extend above the PBL top. In both cases, aerosol size distributions changed abruptly upon crossing the ceilometer-derived PBL height (red dashed lines in **Figure 6**; Table S1). In contrast to the relatively homogeneous aerosol profiles observed within the well-mixed boundary layer below the transition, the overlying lower free troposphere exhibited substantially different aerosol profiles, indicating a sharp transition in aerosol characteristics rather than a gradual decrease in particle concentrations with altitude.

Despite occurring during different field campaigns, the 02/22 CoURAGE and 04/13 BNF flights exhibited remarkably similar aerosol transition profiles. In both cases, aerosol size distributions remained nearly uniform throughout the daytime mixed layer before changing abruptly near the ceilometer-derived PBL height (approximately 750 m on 22 February CoURAGE flight and 850 m on 13 April BNF flight). Above the transition, accumulation-mode particles decreased substantially whereas smaller particles remained detectable, producing aerosol profiles distinctly different from those observed within the underlying mixed layer.

Although aerosol number concentrations generally decreased above the transition altitude, aerosol particles remained present within the lower free troposphere. In both cases, accumulation-mode particles centered at approximately 80–150 nm were substantially depleted above the PBL top, whereas Aitken-mode particles at around 20–30 nm remained detectable, resulting in a relative enhancement of smaller particle populations aloft. Consequently, aerosol size distributions above the PBL differed markedly from those within the underlying mixed layer, reflecting a pronounced reduction in accumulation-mode particles while retaining a detectable Aitken-mode population. Similar observations of persistent ultrafine particles in the lower free troposphere have been reported previously (Chen et al., 2018; Lampilahti et al., 2021; Wang et al., 2023; O'Donnell et al., 2023; Liu et al., 2024; Shan et al., 2025; Zhang et al., 2026). These observations demonstrate that the lower free troposphere can contain aerosol profiles distinct from those within the underlying daytime mixed layer, highlighting the importance of vertically resolved aerosol size-distribution measurements for characterizing aerosol stratification across the



PBL top.

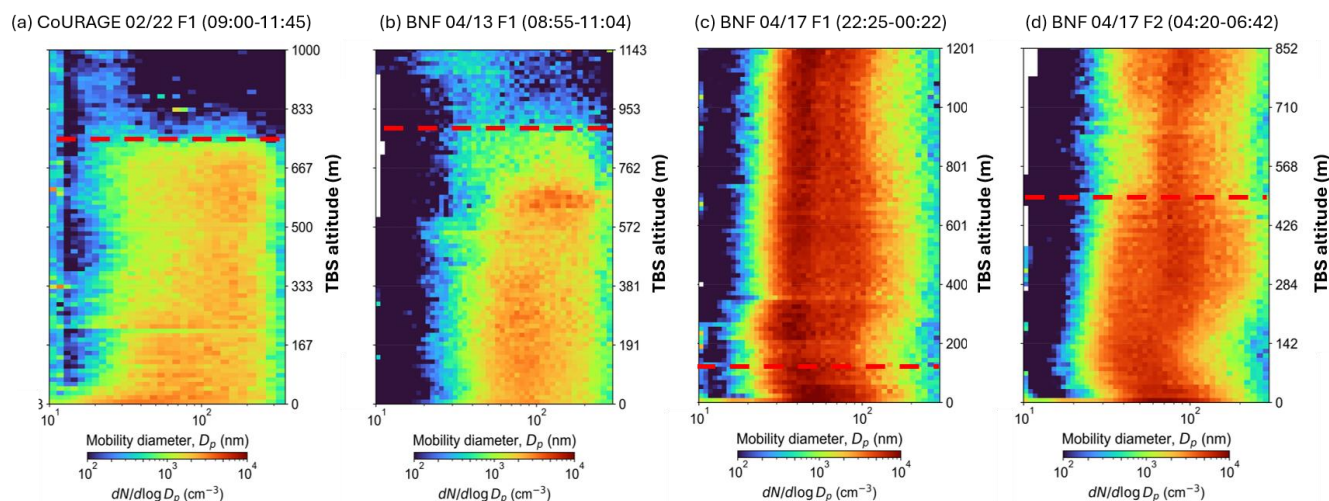


Figure 6. Aerosol vertical size distribution observed by TBS mSEMS during CoURAGE (a) 02/22/2025 Flight 1 between 09:00-11:45 LT, (b) BNF 04/13/2025 Flight 1 between 08:55-11:04 LT, (c) BNF 04/17/2025 Flight 1 between 22:25-00:22 LT, and (d) BNF 04/17/2025 Flight 2 between 04:20-06:42 LT. The red dashed lines indicated the ceilometer-derived boundary layer heights of each flight.

3.2.2 Transitions from the nocturnal PBL to the residual layer

Unlike the daytime transitions into the lower free troposphere, four nighttime TBS flights extended above the shallow nocturnal planetary boundary layer into the overlying residual layer (03/27 BNF F1, F2 and 04/17 BNF F1, F2), providing direct observations of aerosol vertical transitions across the nocturnal PBL top (Figures 6c and 6d for the representative examples from the 04/17 BNF flights, with 03/27 BNF flights discussed in Section 3.3). These transitions were captured after sunset or before sunrise, when the stable nocturnal boundary layer was substantially shallower than the daytime mixed layer, allowing the TBS to simultaneously sample the near-surface stable layer and the overlying residual layer. In contrast to the daytime PBL-top transitions, aerosol size distributions generally changed more gradually across the nocturnal PBL top, reflecting the greater similarity between aerosol populations in the residual layer and those remaining within the underlying stable boundary layer.

During the late-evening 17 April BNF flight (22:25–00:22 LT; Figure 6c), the ceilometer-derived nocturnal PBL was only about 100 m deep, whereas the TBS sampled aerosol populations throughout the overlying residual layer to an altitude of 1200 m. Aerosol size distributions remained broadly similar above the shallow nocturnal boundary layer, indicating that a substantial fraction of the aerosol population resided within the residual layer rather than being confined to the stable surface layer. During the subsequent early-morning flight (04:20–06:42 LT; Figure 6d), the nocturnal PBL had deepened to 450 m. Overall, aerosol populations aloft remained broadly consistent with those observed during the preceding late-evening flight, although noticeable changes in aerosol size distributions were evident near the residual-layer interface, suggesting the presence of vertical stratification within the residual layer. These aerosol transitions around 450m coincided with enhanced wind shear



near the nocturnal PBL top (**Figure S2**), consistent with the dynamical separation between the shallow stable boundary layer and the overlying residual layer.

330 The overall similarity between the late-evening and early-morning observations indicates that the aerosol populations within the residual layer were largely preserved overnight, with no evidence of substantial changes in the overall aerosol size distributions during the intervening period. Compared with the daytime lower free troposphere, the residual layer retained aerosol populations that were much more similar to those within the underlying daytime boundary layer, consistent with the residual layer acting as a reservoir of aerosols that had been mixed throughout the daytime PBL prior to nocturnal stabilization.

335 Representative case studies illustrating the evolution of aerosol populations within the nocturnal residual layer are presented in **Section 3.3**. In particular, the 27 March 2025 BNF flight observations document the evolution from daytime new particle formation to the subsequent preservation of the resulting aerosol population within the nocturnal residual layer, providing one of the clearest examples of the aerosol life cycle captured during the TBS deployments

3.3 Case study: aerosol evolution from daytime new particle formation to nocturnal residual-layer stratification

340 To investigate how particles formed during NPF events are redistributed vertically during boundary-layer evolution, we selected the TBS observations on 27 March 2025 at BNF as a representative case. Surface SMPS measurements (**Figure 7a**) showed repeated NPF and subsequent particle growth between 22 and 28 March 2025, with elevated concentrations frequently persisting into the following day. The observations on 27 March captured the transition from a shallow nocturnal boundary layer to a fully developed daytime mixed layer (**Figure 7b**), allowing the vertical evolution of particles associated

345 with the previous day's growth event to be examined.

During Flight 1 (03/26 21:22–23:15 LT; **Figure 7c**), aerosol size distributions were nearly identical throughout the sampled profile from the surface to 700 m. Only a subtle reduction in particles smaller than 30 nm was observed within a narrow layer near the top of the shallow nocturnal boundary layer (approximately 70–100 m), while particles larger than 50 nm exhibited little vertical variability. The close agreement in aerosol size distributions between the near-surface layer and the

350 overlying residual layer indicates that the aerosol population remained largely continuous following the collapse of the daytime mixed layer. These observations suggest that particles formed and grown during the previous afternoon's NPF event were well preserved overnight within the residual layer, with only limited modification occurring near the top of the shallow stable boundary layer. The observed reduction in ultrafine particles accompanied by a modest enhancement of accumulation-mode particles is potentially associated with weak turbulent mixing and particle coagulation within this transition layer, although the

355 available observations do not allow these mechanisms to be distinguished and further investigation are warranted.

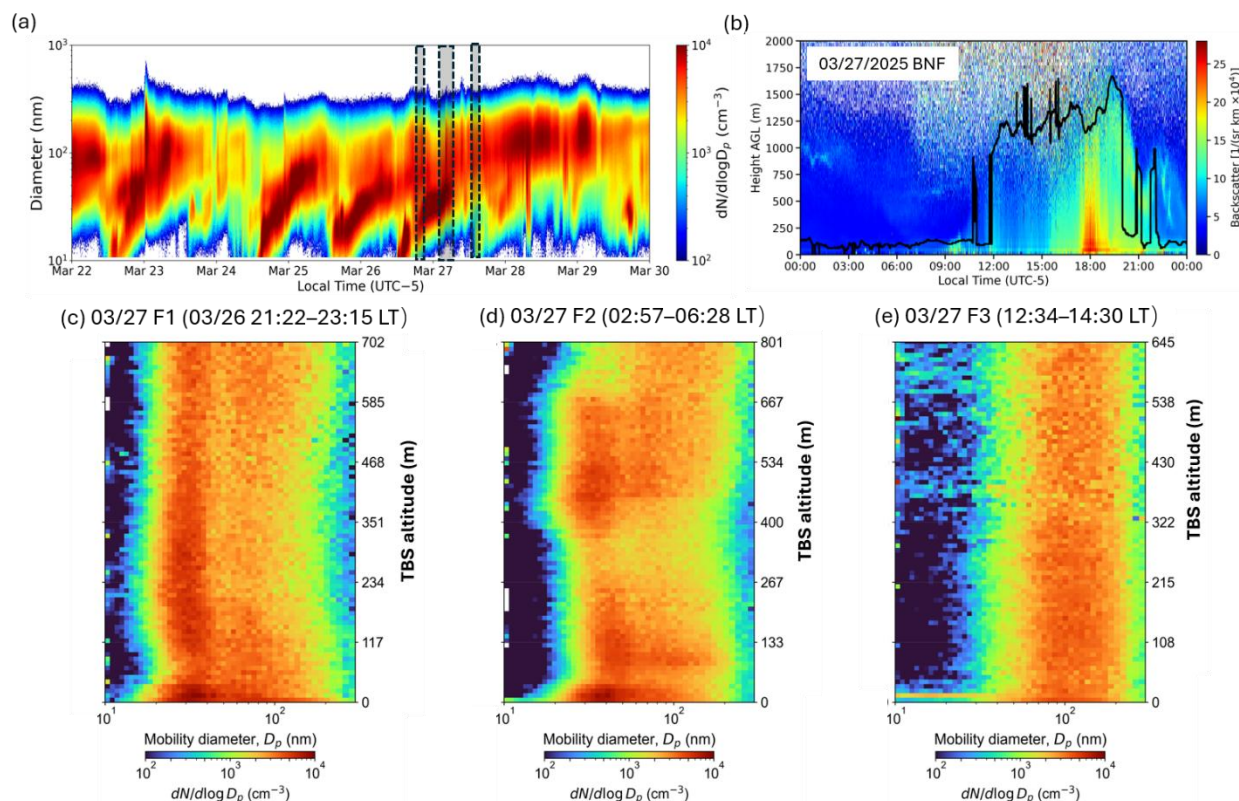


Figure 7. (a) Aerosol number size distributions measured by the surface SMPS at the BNF main site from 22 to 29 March 2025 (The dashed boxes indicate the TBS measurement periods in panels (c), (d), and (e)). (b) Ceilometer-derived boundary-layer structure and layer heights (black line) at the BNF main site on 27 March 2025. Vertical aerosol size distributions measured by the TBS-mounted mSEMS during (c) Flight 1 (03/26 21:22–23:15 LT), (d) Flight 2 (02:57–06:28 LT), and (e) Flight 3 (12:34–14:30 LT) on 27 March 2025.

By the 03/27 F2 flight during 02:57–06:28 LT (**Figure 7d**), the aerosol structure had become more heterogeneous than during F1. Within the shallow near-surface layer below 100–150 m, particle populations shifted toward slightly larger diameters, with enhanced concentrations extending into the 50–100 nm size range and a corresponding reduction in particles smaller than 30 nm. This evolution was consistent with the concurrent surface SMPS measurements (**Figure 7a**), indicating that aerosol aging within the shallow developing surface layer was not confined to the immediate surface but extended throughout the lower boundary layer. Above the shallow surface layer, particle concentrations decreased markedly between 150 and 400 m, forming a distinct transition layer. Doppler lidar wind profiles obtained during the same period indicated that this altitude range broadly coincided with a transition in the wind field separating the shallow surface layer from a dynamically distinct layer aloft (**Figure S3**), suggesting incomplete vertical coupling during the early stages of daytime boundary-layer development. Above 400 m, a relatively stable residual aerosol layer persisted up to about 700 m. Within this layer, the aerosol size distributions remained remarkably similar to those observed during F1, with the persistent 30–50 nm mode largely preserved and particle concentrations slightly enhanced. The close agreement between the two flights indicates that particles



formed and grown during the previous day's NPF event remained well preserved within the weakly mixed residual layer, while the developing surface layer had only begun to modify aerosol properties below. Above approximately 700 m, particle modal diameters became slightly larger, coinciding with changes in the wind profile and indicating increasing vertical variability toward the upper portion of the residual layer. Overall, F2 represents an intermediate stage of boundary-layer evolution, characterized by a shallow developing surface layer, a transition layer between approximately 150 and 400 m, and a relatively undisturbed residual-layer core above.

During the 03/27 F3 deployment (12:34–14:30 LT, **Figure 7e**), the layered aerosol structure had largely disappeared and aerosol size distributions became nearly homogeneous throughout the sampled atmosphere. Concurrent ceilometer observations indicated rapid daytime boundary-layer growth, with the mixed-layer height reaching approximately 650 m, encompassing nearly the entire TBS sampling range. The disappearance of the vertical aerosol gradients coincided with the development of the deep mixed layer, indicating efficient entrainment and recoupling of aerosol populations throughout the lower atmosphere. Taken together, these observations depict a complete evolution in which particles formed during the 26 March NPF event were initially preserved within a vertically homogeneous residual layer overnight, became vertically reorganized during the early-morning transition as boundary-layer growth began, and were subsequently remixed throughout the daytime mixed layer. The persistence of the 30–50 nm particle mode throughout the entire evolution further indicates that the observed vertical structures primarily reflected redistribution of pre-existing particles rather than fresh particle formation.

This case study demonstrated that particle growth events can influence aerosol vertical distributions well beyond the period of active formation and growth. Particles produced during regional growth episodes may remain stored within residual layers overnight and subsequently undergo redistribution as the boundary layer evolves on the following day. Such processes are difficult to infer from surface observations alone and highlight the value of size-resolved vertical measurements for characterizing aerosol evolution within the lower troposphere.

4. Conclusions

This study presents one of the first comprehensive datasets of vertically resolved aerosol particle size distributions obtained using a miniature scanning electrical mobility spectrometer (mSEMS) aboard the DOE ARM Tethered Balloon System across contrasting urban/coastal and forested environments. The observations demonstrate that aerosol vertical structure is closely coupled to boundary-layer evolution and can exhibit substantial variability even when surface aerosol measurements appear relatively uniform. Within the planetary boundary layer, aerosol populations ranged from vertically homogeneous distributions associated with efficient turbulent mixing to heterogeneous structures characterized by surface influence or elevated particle layers enriched in Aitken-mode aerosols. Measurements extending above the PBL further resolved distinct aerosol transitions into the lower free troposphere and the nocturnal residual layer, highlighting atmospheric structures that cannot be fully characterized using boundary-layer height or surface observations alone.

The case studies further demonstrate that boundary-layer evolution strongly regulates the vertical redistribution of aerosol populations. In particular, the 27 March 2025 BNF TBS observations show that particles produced during a regional



growth event were preserved overnight within a residual layer, became vertically reorganized during early-morning boundary-layer redevelopment, and were subsequently remixed throughout the daytime mixed layer. Together with Doppler lidar observations, these measurements provide direct observational evidence linking aerosol vertical structure to boundary-layer dynamics and identify nocturnal residual layers as important reservoirs connecting aerosol evolution across consecutive days. These results highlight the important role of boundary-layer processes in controlling the transport, storage, and redistribution of atmospheric particles within the lower troposphere.

The vertically resolved aerosol size-distribution measurements presented here provide substantially more information on aerosol transport and atmospheric processing than can be obtained from surface observations or boundary-layer height alone. The dataset therefore provides valuable observational constraints for evaluating atmospheric chemistry and transport models and demonstrates the importance of routine vertically resolved aerosol measurements for improving our understanding of aerosol life cycles and boundary-layer processes.

Implications for Future TBS Deployments: Future TBS deployments would benefit from continuous multi-day observations beginning before the onset of new particle formation and continuing through subsequent particle growth, to capture the complete evolution of aerosol populations. Additional flights extending above the evolving boundary-layer top would further improve characterization of aerosol exchange between the mixed layer, residual layer, and lower free troposphere. Coordinated observations combining tethered-balloon measurements with higher-altitude uncrewed aerial systems and ground-based remote sensing would provide a more complete picture of aerosol transport and mixing throughout the lower troposphere.

Data availability. The TBS mSEMS data in this study are available at ARM Data Discovery portal (https://adc.arm.gov/discovery/results/iopShortName::amf2025SSNPF/instrument_code::tbsmsems) (Zhang et al., 2026b). Other ARM data, including the surface SMPS measurements as well as the ceilometer measurements are publicly available through the ARM Data Discovery portal (<https://adc.arm.gov/discovery/>).

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Competing interests. At least one of the (co-)authors is a member of the editorial board of Atmospheric Chemistry and Physics.

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