RESPONSE TO REVIEWER COMMENTS

Title: Aircraft-derived particle fluxes distinguish entrainment zone and decoupled layer nucleation in marine boundary layers

Journal: Atmospheric Chemistry and Physics

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Referee Comments in 12-point italicized font

Authors' Response in indented, 12-point normal font.

Changes to the manuscript in quotes, 12-point blue font.

Response to Referee's Comments

Specific Comments

#1. *Was there no measure of coarse-mode concentrations that are expected to be small in the SPE regions? This should be discussed, given that one of the main points of the paper seems to be to dispute the idea that sea-spray surface area inhibits new particle formation. As a proxy, even windspeeds (which aren't discussed except in the framework of flux measurements), might be helpful.*

> **Response:** Coarse-mode concentration data are now included in Figures 3, 6, S8, S11, S14, and S17 of the revised manuscript.

Revised manuscript text: "Figures 3–5 present data collected on January 29, 2018 with an additional example from February 10, 2018, shown in Supplementary Figs. S8–S10. Back-trajectory analysis (not shown) indicates that the sampled airmasses had been circulating around the Azores for the preceding three days and were therefore likely less polluted than North American outflow air masses. Figure 3 shows a multi-panel time series covering approximately 3.5 hours of flight operations. The aircraft initially ascended to ~2,500 m but generally remained below ~1,500 m for most of the flight (Fig. 3a). The flight trajectory (Fig. 3b) reflects predominantly east-west movement across the Azores region, spanning latitudes from approximately 38.7° to 39.4°N and longitudes from −28.4° to −27.4°W. Drizzle number concentration measured by 2DS (purple and blue lines in Fig. 3b) was absent or low during the selected SPE periods. Elevated drizzle number concentration, together with high liquid water content regions (orange in Fig. 3d) mark frequent cloud encounters. Following our quality control procedures, all N_{3-10} concentration data with LWC ≥ $3x10^{-3}$ g m^{-3} were excluded from analysis to avoid contamination from cloud droplet shattering artifacts. Pink-shaded periods mark the intervals chosen for detailed analysis, which exhibited simultaneous increases in both N_3 and N_{10} concentrations exceeding 10^4 cm^{-3} (indicating an SPE). Supermicron particle concentration (blue in Fig. 3c) as well as total particle surface area (green in Fig. 3c) were also low during the selected SPE periods, indicating the absence of particles such as sea spray aerosols."

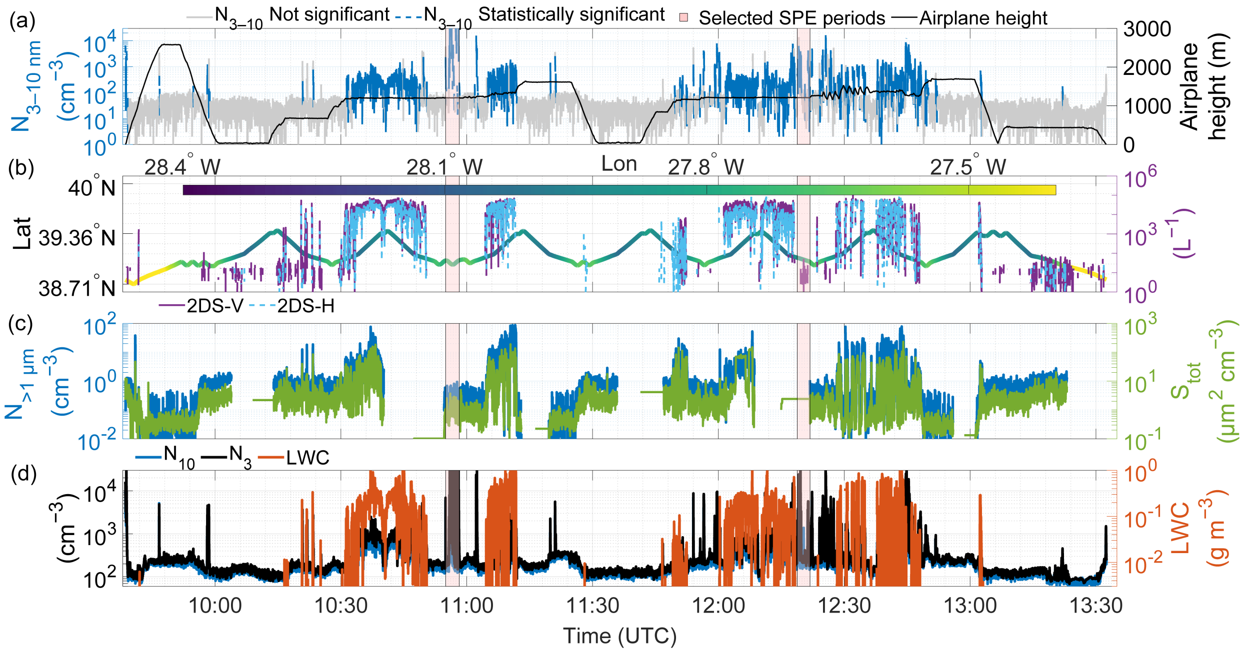

Figure 3. Multi-parameter time series from the January 29, 2018 flight. (a) $N_{3\text{-}10}$ particle concentrations and aircraft altitude; (b) aircraft position (latitude and longitude) and drizzle number concentration; (c) supermicron particle concentration and total particle surface area (S_{tot}) (d) particle number concentrations (N_{10} and N_3) and liquid water content. Gaps in the time series indicate the missing data.

"Figures 6–8 present data from June 21, 2017, with additional examples from July 7, 2017, February 18, 2018 and February 12, 2018, shown in Supplementary Figs. S11–S13, S14–16, and S17–S19. Back-trajectory analysis (not shown) indicates that the sampled airmasses originated from the Arctic and were therefore expected to be relatively clean. Figure 6 covers approximately 4 hours of flight operations, during which the aircraft initially flew at very low altitudes (~30 and 50 m) near 12:00 and 13:30 UTC before gradually ascending to ~1,000 m. Drizzle number concentration by 2DS (purple and blue lines in Fig. 6b) was absent or below the threshold during the selected SPE periods. Multiple events with $N_{3\text{–}10}$ concentrations from 10^2 to 10^4 cm^{-3} were observed throughout the second half of the flight. The flight trajectory (Fig. 6b) reflects predominantly east-west movement, spanning latitudes from approximately 38.6° to 39.3°N and longitudes from −28.4° to −27.4°W. Pink-highlighted intervals show periods with concurrent increases in N_3 and N_{10} concentrations exceeding 10^3 cm^{-3}, indicative occurrences of SPEs. As in Case 1, supermicron particle concentrations (blue in Fig. 6c) and total particle surface area (green in Fig. 6c) remained low during the selected SPE periods, indicating the absence of coarse-mode particles such as sea spray."

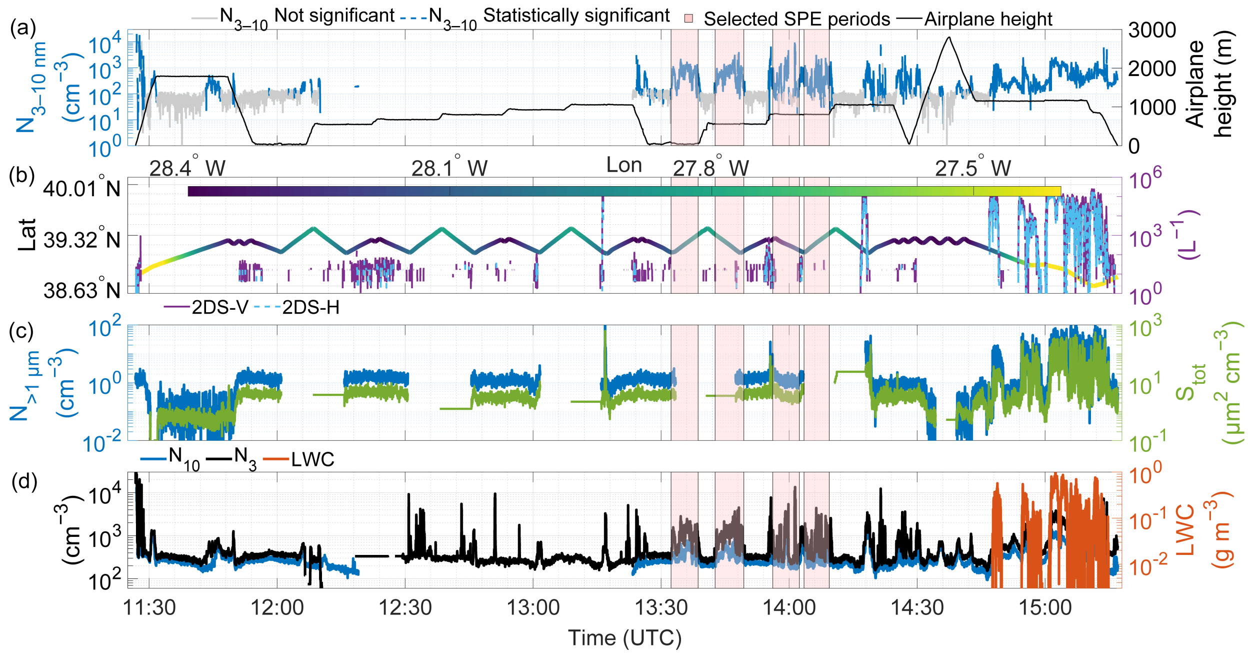

Figure 6. Same as Figure 3 but for June 21, 2017. Gaps in the time series indicate the missing data.

#2. *Also, it would be useful to show time series of aerosol surface area, at least in the sub-micron sizes, as that looks to be a potentially important change that occurs in the SPE regions. Fig 4 indicates almost no particles in the 10-1000 nm range in the second SPE period—is that correct, or is that just missing data? (Stot is shown and briefly discussed in the Fig 4 b-d vertical profiles, but these are not from the actual SPE periods.)*

> **Response:** Total aerosol surface area has been added to Figures 3, 6, S8, S11, S14, and S17. The absence of particle data in the 10–1000 nm range during the second SPE period in Figure 4 reflects a gap in data coverage.

#3. *Lines 428-436: Were these cases when clouds were present above? If so, did you check to rule out drizzle, which would breakup in the inlet and cause artifacts? Below-cloud drizzle would be independent of LWC, which is derived from the CDP (smaller sizes). Also, since droplet number concentrations are never given, it's unknown whether or not these were clean clouds, which are more likely to precipitate. The sometimes high (up to 1 g m^{-3}) LWCs would suggest drizzle as a possibility. Call me skeptical, but I've enough experience with in-situ cloud measurements from aircraft to know that artifacts are common. This point, as well as some discussion of which of the four source regimes (lines 75-78) that the presented measurements represent would be useful.*

> **Response:** Drizzle number concentration derived from the 2DS dataset has been added to the revised Figures 3, 6, S8, S11, S14, and S17. The low supermicron particle concentrations confirm that coarse-mode aerosol loading was minimal during SPE events, while drizzle concentration data confirm that drizzle was not detected outside of high-LWC cloud regions. A discussion of the source regimes represented by the presented measurements has also been added to the revised manuscript. Please also see our reply to Comment #1.

#4. *Fig 3a and discussion on lines 412-413—are these occasional spikes in 3-10 nm particles above the cloud layer--e.g., 11:20-11:25--real? Should be discussed to assure that some of the signals noted below are not from entrainment of particles from above. Unlikely, since the concentrations below are sometimes higher, and these are short spikes, so perhaps they are artifacts of some sort?*

> **Response:** The brief spikes in N_{3-10} concentrations above the cloud layer (e.g., 11:20–11:25 UTC) cannot be conclusively identified as artifacts; however, they do not contribute to any of the analysis presented. A clarifying statement has been added to the revised manuscript

Revised manuscript text: "This interpretation is supported by the near-absence of N_{3-10} at ~1,600 m during 11:14–11:25 and 12:51–13:01 UTC (Fig. 3), with the exception of brief concentration spikes of uncertain origin retained in the record due to insufficient evidence for their removal. The small particle size (3–10 nm) and limited horizontal extent of less than 10 km further argue against a free tropospheric nucleation source, as particles originating in the free troposphere would be expected to have grown substantially and the plume to have diluted during descent to measurement altitude."

#5. *Lines 43-45: "This expectation is based on the relatively high surface area of sea spray aerosols, which act as condensation and coagulation sinks for nucleating vapors and newly formed particles". There are plenty of accumulation-mode sulfate/organic particles in most MBLs that also may act as condensation and coagulation sinks. As do clouds themselves. This should be mentioned.*

Revised manuscript text: "This expectation is based on the high condensation and coagulation sink capacity of the remote MBL, which includes not only sea spray aerosols (Bates et al., 1998; Pirjola et al., 2000) but also accumulation-mode sulfate and organic particles entrained from the free troposphere (Yoon et al., 2001). Clouds further suppress NPF by scavenging Aitken-mode particles (Zheng et al., 2018), accelerating sulfate production on existing droplets through aqueous-phase SO_2 oxidation (Sanchez et al., 2021), and sequestering DMS oxidation products such as HPMTF that would otherwise contribute to sulfuric acid formation (Novak et al., 2021)."

#6. *Perhaps move some of the lengthy data and analysis criteria in Section 2 to the supplement, where some of the associated graphs already are?*

> **Response:** Section 2.3.2 is moved to the supplementary information

#7. *Lines 340-342: "We examine two flight days as case studies of SPEs observed at varying altitudes above the ocean. Additional supporting flights are presented in the Supplementary Information for each case." After this you go immediately into Table 1 that shows six different flight days, which I found confusing. Perhaps discuss Table first and then go into the case studies.*

Revised manuscript text: "We examine two flight days as case studies of SPEs observed at varying altitudes above the ocean. Additional supporting flights are presented in the

Supplementary Information. Table 1 summarizes the $N_{3\text{–}10}$ vertical turbulent flux estimates derived from all six flight days analyzed in this study, grouped by the inferred nucleation regime. Flights 1 and 2 (January 29 and February 10, 2018) are classified as entrainment zone nucleation events, where SPEs were detected near the top of the MBL at heights exceeding 1,200 m. Flights 3–6 (June 21 and July 7, 2017; February 18 and 12, 2018) are classified as decoupled layer nucleation events, with SPEs observed across a broader range of altitudes (30–837 m). For all events, the ratio of measured flux to the spectrally-complete flux $\left(\frac{F_m}{F}\right)$ exceeds 0.76, indicating minimal flux loss due to sensor response limitations. The normalized vertical velocity variance ($\sigma_w^2\ w_*^{-2}$) is generally low, consistent with relatively quiescent turbulent conditions during the measurement periods. Negative flux values indicate downward transport of freshly nucleated particles from the entrainment zone toward the surface, while positive values suggest upward transport from a source within the decoupled sub-cloud layer. Two of these flight days, January 29, 2018 (Case 1) and June 21, 2017 (Case 2), are examined in detail as case studies in the following sections, with the remaining four flights presented as supporting examples in the Supplementary Information."

#8. *Figure 4a caption: "The main panel shows size-resolved particle number concentrations (10-600 nm) from FIMS as a function of time and altitude, while N3-10 concentrations in the lower strip." I think "are shown" is missing before "in the lower strip".*

Response: Changed as suggested.

Revised manuscript text: "Figure 4. (a) Size-resolved particle number concentrations (10–600 nm) from FIMS as a function of time and altitude, with $N_{3\text{–}10}$ concentrations shown in the lower strip. Pink shading indicates selected SPE periods. (b-d) Vertical profiles of potential temperature (θ), normalized vertical velocity variance ($\sigma_w^2\ w_*^{-2}$), total particle surface area (S_{tot}), and water vapor mixing ratio (MR_{H2O}) for three time periods nearest to the selected SPE periods: (b) 09:51–10:01 UTC, (c) 11:25–11:31 UTC, and (d) 13:01–13:07 UTC. Gaps in the time series indicate the missing data."

#9) *I would suggest making 4a and 4b-d (and 7a and 7b-d) into separate plots, as these are really different from each other and this is too much information to easily assimilate in a single plot.*

Response: We respectfully disagree with this suggestion. Panel (a) provides essential context (the time evolution of size-resolved concentrations and the altitude of the aircraft) that is needed to interpret the vertical profiles shown in panels (b–d). Separating these into distinct figures would obscure the connection between the timing of SPE periods and the concurrent thermodynamic and turbulence structure of the boundary layer. Furthermore, panels (b–d) are intended to be viewed comparatively, as they document the evolution of potential temperature, normalized vertical velocity variance, total aerosol surface area, and water vapor mixing ratio across three successive time windows. We believe the current layout presents this information in the most coherent and space-efficient manner, and have added a brief note to the figure caption to guide the reader through the panel structure.

#10. *Fig 7 has a lot of missing data, so I'd suggest explicitly adding the sentence from Fig 4 caption to Fig 7 caption as well. This confused me until I figured it out. ("Gaps in the time series indicate the missing data.")*

Response: Changed as suggested.

Revised manuscript text: "Figure 7. Same as Figure 4, but for June 21,2017. Gaps in the time series indicate the missing data."