Article Discussion comments for: "Aerosol dry deposition fluxes on snow during the ALPACA

campaign in Fairbanks, Alaska", Donateo et al., Atmospheric Chemistry and Physics,

Manuscript ID: egusphere-2025-1366

We thank the Reviewer for his/her careful assessment of our manuscript and his/her valuable

suggestions. We found them very useful for improving and clarifying some of the information that

was unclear in the submitted manuscript. Below, we respond to the comments in turn, and summarise

modifications made to the manuscript. Our responses are formatted as follows:

Reviewer comments - black text (bold)

Author responses - black text

Revised manuscript text - green text

Line numbers refer to those in the original submission.

Reviewer #1: RC1: "Comment on egusphere-2025-1366' - https://doi.org/10.5194/egusphere-2025-

1366-RC1

This paper reports on calculated modal and size-resolved deposition fluxes from 2-months of

surface measurements at a site in the urban Arctic (Fairbanks, Alaska). They find an increase

in deposition flux with increasing diameter, consistent with previous work in a similar Arctic

region and a model parameterization. Using temporal trends, meteorological regime

classifications, and profiles of atmospheric thermodynamics and number concentrations, the

authors attribute the variability in deposition fluxes to regional sources (anthropogenic

pollution) with enhancements due to local meteorology and synoptic circulation.

This work provides very interesting insights on aerosol deposition in an understudied region

that is strongly impacted by intraseasonal synoptic variability and distinct aerosol sources and

transport. The discussions and presentation of results are prepared very well, and I am overall

confident in the fidelity of this work. I can recommend this paper be considered for publication

after the following minor comments, clarifications, and questions are addressed.

Comments:

1

• Lines 195-196: Can the authors please specify how the ultrafine particle mode was determined? Was this a subtraction of the OPC integrated number concentration from the total concentration measured by the CPC?

In the new version of the manuscript a sentence has been added to clarify how the ultrafine (UFP) number concentration has been calculated.

Ultrafine particle concentration (UFP) was obtained as the difference between the total number concentration (CPC measurement) and the OPC integrated concentration in the size range 0.25–1 μm.

• Lines 283-287: Particle number concentrations in the tens of thousands seem exceptionally high for ambient measurements, even during Arctic hazy periods. It makes sense that the UFP mode would be the highest particle number concentration, but the value seems dramatically high. Can the authors please provide context for these number concentrations? First, as a comparison to previous measurements of total and modal number concentrations at Fairbanks/Urban Arctic and second with respect to the meteorology.

We appreciate the reviewer's concern and agree that providing context is important. We have therefore added a short comparison with previous studies conducted in Fairbanks in Sect. 3.2.

The particle number concentrations observed in this study are consistent with previous measurements reported for Fairbanks during the winter season. For example, Robinson et al. (2023) documented a median particle number concentration above 4.5×10^4 cm⁻³ during cold stagnation events, with UFPs accounting for most particles (> 95%). Again, Robinson et al. (2023), measured the highest UFP number concentration (7.2 x 10^4 cm⁻³) in Downtown East (Fairbanks).

Robinson, E. S., Cesler-Maloney, M., Tan, X., Mao, J., Simpson, W., and DeCarlo, P. F.: Wintertime spatial patterns of particulate matter in Fairbanks, AK during ALPACA 2022, Environm. Sci. Atmospheres, 3, 568–580, https://doi.org/10.1039/D2EA00140C, 2023.

Is there a sense of how high/different these aerosol number concentrations are from "background" conditions in Fairbanks or nearby/similar regions?

We agree that it is important to contextualize our observations with respect to background conditions. We have therefore added to the revised version of the manuscript a short discussion comparing the particle number concentrations observed during our campaign with background levels reported in Fairbanks and in other Arctic remote locations.

The particle number concentrations observed in this study in the immediate outskirts of Fairbanks are comparable to, or slightly higher than, those previously reported in the surroundings of Fairbanks. For instance, Robinson et al. (2023) measured concentrations on the order of 1.5 x10⁴ cm⁻³ at sites located on the hills north of the city during strong inversion conditions. By contrast, typical particle number concentrations at pristine Arctic sites, such as Barrow in Alaska (Rose et al., 2021) or Zeppelin observatory in Ny-Ålesund (Croft et al. 2016) are two to three orders of magnitude lower (10²–10³ cm⁻³), underscoring the dominant impact of local sources and boundary-layer processes in shaping aerosol levels in Fairbanks. Our observations thus align with pollution episodes previously described for the region and highlight the strong contrast between clean background conditions and the highly elevated concentrations associated with persistent inversions and limited boundary-layer mixing.

Rose, C., Collaud Coen, M., Andrews, E., Lin, Y., Bossert, I., Lund Myhre, C., Tuch, T., Wiedensohler, A., Fiebig, M., Aalto, P., Alastuey, A., Alonso-Blanco, E., Andrade, M., Artíñano, B., Arsov, T., Baltensperger, U., Bastian, S., Bath, O., Beukes, J. P., Brem, B. T., Bukowiecki, N., Casquero-Vera, J. A., Conil, S., Eleftheriadis, K., Favez, O., Flentje, H., Gini, M. I., Gómez-Moreno, F. J., Gysel-Beer, M., Hallar, A. G., Kalapov, I., Kalivitis, N., Kasper-Giebl, A., Keywood, M., Kim, J. E., Kim, S.-W., Kristensson, A., Kulmala, M., Lihavainen, H., Lin, N.-H., Lyamani, H., Marinoni, A., Martins Dos Santos, S., Mayol-Bracero, O. L., Meinhardt, F., Merkel, M., Metzger, J.-M., Mihalopoulos, N., Ondracek, J., Pandolfi, M., Pérez, N., Petäjä, T., Petit, J.-E., Picard, D., Pichon, J.-M., Pont, V., Putaud, J.-P., Reisen, F., Sellegri, K., Sharma, S., Schauer, G., Sheridan, P., Sherman, J. P., Schwerin, A., Sohmer, R., Sorribas, M., Sun, J., Tulet, P., Vakkari, V., van Zyl, P. G., Velarde, F., Villani, P., Vratolis, S., Wagner, Z., Wang, S.-H., Weinhold, K., Weller, R., Yela, M., Zdimal, V., and Laj, P.: Seasonality of the particle number concentration and size distribution: a global analysis retrieved from the network of Global Atmosphere Watch (GAW) near-surface observatories, Atmos. Chem. Phys., 21, 17185–17223, https://doi.org/10.5194/acp-21-17185-2021, 2021.

Croft, B., Martin, R. V., Leaitch, W. R., Tunved, P., Breider, T. J., D'Andrea, S. D., and Pierce, J. R.: Processes controlling the annual cycle of Arctic aerosol number and size distributions, Atmos. Chem. Phys., 16, 3665–3682, https://doi.org/10.5194/acp-16-3665-2016, 2016

Were the aerosol concentrations (total and modal) different between meteorological regimes?

Yes, this aspect is addressed in the manuscript. In Sect. 3.6 we specifically discuss the variability of aerosol concentrations, fluxes, and deposition velocities (V_d) under different meteorological regimes. Tables 4 and 5 report the corresponding concentration values and flux characteristics for the main synoptic categories (cyclonic, anticyclonic, etc.), showing that concentrations and fluxes indeed vary

depending on the prevailing regime. A sentence has been added at the end of Sect. 3.2 to reiterate more clearly these differences.

The highest concentrations occurred during persistent anticyclonic periods with strong surface-based inversions, weak winds, and low mixing heights, which favour the accumulation of locally emitted particles. By contrast, during frontal passages and enhanced mixing, number concentrations dropped by an order of magnitude. These results therefore fall within the expected range for Fairbanks wintertime conditions and reflect the strong modulation of aerosol concentrations by meteorology.

Additionally, typically in CPCs the laser becomes saturated above 10,000 cm-3 and coincidence errors become quite large. Can the authors speak to and justify why they feel such high concentrations measured by the CPC can be trusted?

We acknowledge that coincidence errors and laser saturation can be a concern in CPC measurements. However, in our case the instrument used was a TSI 3756, whose specifications (manufacturer's datasheet - https://tsi.com/getmedia/cb4a10a6-3ae8-4cb0-bb5d-1d9dfc841ded/3756_A4_5002016_RevB_Web?ext=.pdf, last access 25/08/2025) report a maximum measurable concentration of 3.0 × 10⁵ particles cm⁻³ before coincidence errors exceed 10%. The highest concentrations observed in our dataset were on the order of 2–3 × 10⁴ particles cm⁻³, well below this upper threshold. We therefore consider the measurements to be reliable and not significantly affected by coincidence errors. Although the CPC 3756 has not been the explicit subject of dedicated publications, many of its key features (2.5 nm sensitivity, butanol capacity, and data rates up to 50 Hz) are direct implementations of the same technology used in the CPC 3776, which is well described in the literature (Takegawa et al. 2016).

Takegawa, N., Iida, K., Sakurai, H.: Modification and laboratory evaluation of a TSI ultrafine condensation particle counter (Model 3776) for airborne measurements. Aerosol Science and Technology, 51:2, 235-245, DOI:10.1080/02786826.2016.1261990

• Table 1: is the mean diameter (dp) in this table taken as the average of the range of diameters in each mode/size range? I believe this should be specified somewhere in the text and table legend as not to confuse with a geometric mean diameter from a modal fit to a size distribution.

In the revised version of the manuscript, we have replaced the previous values with the mean diameter (d_p) of each size range, and we have specified this clearly in the caption of the corresponding table to avoid any confusion with a geometric mean diameter derived from a modal fit.

 d_p represents the arithmetic mean diameter of each size range.

• Did the authors test for independence in the 3 modes considered? More specifically, did they find coverability between any of the modes and does that have any influence on the interpretation of sources and deposition?

We thank the reviewer for this question. To ensure that the three modes considered were statistically independent, we evaluated the correlation among the different size channels measured by the Optical Particle Counter (OPC). Channels showing correlations higher than 50% were merged into the same size class. This procedure minimized overlap between adjacent modes and reduced the risk of double-counting or misinterpreting correlated particle populations. As a result, the three modes presented in the manuscript can be considered sufficiently independent for the purpose of source attribution and deposition analysis. We have clarified this point in the revised Sect. 2.3.

To assess the independence of the particle size modes, we calculated Pearson correlation coefficients between number concentrations in adjacent OPC size channels, merging those with a correlation > 0.5. This approach reduces coverability between size classes and ensures that the reported modes represent distinct particle populations for interpretation of sources and deposition processes.

• I believe it is appropriate to provide standard deviations when means are reported.

In Table 3 we already report both the standard deviation and the standard error together with the mean values, to provide a comprehensive representation of the data variability and uncertainty.

• Section 3.2: Figures 3 and A1 should be cited in lines 283-287 (preferably with Table 1) before their first citation later in this section so the reader can see the trends and ranges discussed.

In the revised version of the manuscript a citation to Fig. 3 and (now) Fig. A3 was inserted early in Sect. 3.2.

• Lines 288-301: Can the authors please quantify the particle differences in this passage. Phrases like "enhanced pollution", "concentration significantly smaller", "close to levels" and reference to the figure panels are used, but it helps the reader glean differences when these values are

quantified. Did the authors also consider performing statistical tests on the differences between number concentrations in different meteorological regimes?

In the revised manuscript, we have added explicit percentage differences to quantify the particle differences previously described qualitatively with terms such as "enhanced pollution," "concentration significantly smaller," or "close to levels". In addition, we have explicitly referenced Tables 4 and 5, from which readers can extract the numerical values. These additions allow the reader to clearly assess the magnitude of particle variations under different conditions.

We have performed statistical significance tests (Kruskal–Wallis) on the differences in number concentrations between the various meteorological regimes. The results indicate that all differences are statistically significant (p < 0.05). These results have been added to Sect. 3.2.

Statistical analysis using the Kruskal-Wallis test confirms that number concentrations differ significantly among all meteorological regimes (p < 0.05), supporting the observed variations discussed above.

• Lines 300-301: There is higher NQ-CRS in periods before Feb 7-10 (gleaning from Fig. 3 and Fig. A1), but the authors state that a maximum is observed during Feb 7-10. Do the authors mean elevated number concentrations during that period? Please clarify.

Yes, the authors indeed refer to elevated number concentrations during the period of February 7–10. We have clarified the text in the revised manuscript to make this explicit, specifying that the "maximum" mentioned corresponds to higher particle number concentrations rather than absolute peak fluxes.

An increase in particle number concentration N_{Q-CRS} (up to 0.61 cm⁻³) was observed (Fig. 3) during the C period (7 - 10 February).

Additionally, the authors should clarify what is meant by "... is characteristic of this class of particles and may witness a contribution from regional transport (e.g. dust or sea salt particles)." I believe what the authors are saying here is that dust and sea salt are primarily in the coarse mode and an elevation in the concentration of these particles may suggest an increase

in their transport. This should be clearly stated and with citations supporting these claims (1 - dust/sea salt in the coarse mode, 2 – elevation in number concentration from transport).

We thank the reviewer for pointing out this ambiguity. We agree that our statement should be clarified. What we meant is that dust and sea salt particles typically occur in the coarse mode, and that an increase in coarse-mode number concentrations may therefore indicate a contribution from regional transport of these particles. We have revised the text accordingly and added supporting citations.

This behaviour is characteristic of coarse-mode particles, which are commonly associated with primary emissions such as mineral dust and sea salt (Seinfeld & Pandis, 2016). An increase in the number concentration of coarse particles may therefore be indicative of long-range or regional transport events, when enhanced advection can bring dust or sea-salt aerosols into the measurement area (Textor et al., 2006), as already observed in the Fairbanks area since the early 90s' (Shaw 1991a,b).

Seinfeld, J.H., Pandis, S.N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, Wiley, New Jersey, 2016

Shaw, G. E., Aerosol Chemical Components in Alaska Air Masses 1 Aged Pollution, Journal of Geophysical Research, 96, 22357-22368, 1991a.

Shaw, G. E., Aerosol Chemical Components in Alaska Air Masses 2 Sea Salt and Marine Products, Journal of Geophysical Research, 96, 22369-22372, 1991b.

Textor, C., Schulz, M., Guibert, S., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., Berglen, T., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Feichter, H., Fillmore, D., Ghan, S., Ginoux, P., Gong, S., Grini, A., Hendricks, J., Horowitz, L., Huang, P., Isaksen, I., Iversen, I., Kloster, S., Koch, D., Kirkevåg, A., Kristjansson, J. E., Krol, M., Lauer, A., Lamarque, J. F., Liu, X., Montanaro, V., Myhre, G., Penner, J., Pitari, G., Reddy, S., Seland, Ø., Stier, P., Takemura, T., and Tie, X.: Analysis and quantification of the diversities of aerosol life cycles within AeroCom, Atmos. Chem. Phys., 6, 1777–1813, https://doi.org/10.5194/acp-6-1777-2006, 2006.

• Section 3.3: The authors should place the calculated values in context with previous measurements. How do these flux values compare to other Arctic or continental regions? A diel cycle of modal flux is an interesting result that needs to be compared to similar and dissimilar calculations.

We agree with the reviewer that contextualizing the calculated fluxes is important. However, as we have already highlighted in the manuscript, studies reporting aerosol particle fluxes in Arctic or urban snow-covered environments are still very scarce. Flux measurements are inherently site-specific, as they strongly depend on local sources and surface characteristics. This makes direct comparison of absolute flux values across sites problematic.

For this reason, in the literature the more meaningful comparison between sites is often performed using deposition velocity (V_d) , i.e. the flux normalized by particle concentration, which reduces the influence of site-specific concentration levels and allows a better understanding of how the underlying surface and micrometeorological conditions control deposition. As also noted in previous studies (Mathes et al., 2025), for a general understanding it is more instructive to compare typical deposition velocities over different surface types rather than absolute flux values. In line with this approach, we also compare our calculated deposition velocities with observations reported in the literature, as shown in Sect. 3.4 (Fig. 7), rather than focusing solely on the absolute fluxes.

Mathes, T., Guy, H., Prytherch, J., Kojoj, J., Brooks, I., Murto, S., Zieger, P., Wehner, B., Tjernström, M., and Held, A.: Particle flux–gradient relationships in the high Arctic: emission and deposition patterns across three surface types, Atmos. Chem. Phys., 25, 8455–8474, https://doi.org/10.5194/acp-25-8455-2025, 2025.

• Lines 336-337: Please state the quantified difference between FACC during AC compared to the other periods that is referenced here.

In the revised manuscript we now quantify this difference: F_{ACC} during the anticyclonic period is on average 3.4 times higher compared to the cyclonic period, and 1.4 times higher compared to the transition period T_2 (Table 4 and Table 5).

• Figure 3 and 5: Please denote that (if I assume correctly) dots in the box plots represent the mean and line in the box is the median.

We thank the reviewer for this observation. We confirm that in all box plots, the line inside the box represents the median value, while the dots indicate the mean. We have clarified this in the revised figure captions.

The line inside each box indicates the median, while the squares represent the mean values.

• Lines 358-361: The authors suggest wind-driven resuspension as a potential mechanism contributing to the variability in FQ-CRS. Did the authors observe more variable FQ-CRS with variable or stronger wind speeds?

In response to this comment, we selected $F_{Q\text{-}CRS}$ values under conditions of high and low wind speed. The mean fluxes are $0.02~\text{cm}^{-2}~\text{s}^{-1}$ and $-3.92\times10^{-5}~\text{cm}^{-2}~\text{s}^{-1}$, respectively. This indicates that under high wind conditions (> 2.35 m s⁻¹, where the mean wind velocity was 2.35 m s⁻¹) the flux is more than two orders of magnitude larger and positive (consistent with wind-driven resuspension), whereas under low wind speeds (< 2.35 m s⁻¹) it is close to zero and slightly negative (indicating deposition). We have added a brief discussion in the revised manuscript to clarify the relationship between $F_{Q\text{-}CRS}$ variability and wind conditions.

Periods with stronger or more variable winds were associated with increased variability in F_{Q-CRS} . Selecting F_{Q-CRS} by wind speed, the mean flux was $0.02\,\text{ cm}^{-2}\,\text{s}^{-1}$ under high wind conditions (> 2.35 m s-1), while it was close to zero and slightly negative ($-3.92\times10^{-5}\,\text{ cm}^{-2}\,\text{s}^{-1}$) under low wind speeds (< 2.35 m s-1), supporting the hypothesis of wind-driven resuspension of particles from the surface contributes to the observed fluctuations in deposition fluxes.

• Lines 360-361: The authors spend little time discussing the differences in the flux diurnal cycle between synoptic regimes. I think the larger fluxes throughout the day during the anticyclonic regime is consistent with the overall finding of higher number concentrations and higher fluxes during the anticyclonic regime and should be stated.

We agree that the diurnal cycle of fluxes is an important aspect to emphasize. Following the suggestion, we have expanded the discussion by explicitly stating that the larger fluxes observed throughout the day during the anticyclonic regime are consistent with the higher number concentrations and overall higher fluxes already reported for this regime. The following revised text has been added in the new version of the manuscript.

The comparison of the diurnal cycle between the two synoptic regimes further highlights the role of large-scale circulation in controlling particle concentrations and exchange processes. During anticyclonic conditions, fluxes remain consistently higher throughout the day for the ACC particles, and on average higher between 0:00 and 6:00 LT for UFP and Q-CRS particles, during a time of the day when wind speed and TKE are enhanced during the AC period. The larger fluxes observed in the

anticyclonic period with respect to the cyclonic conditions therefore not only reflects the overall increase in particle number concentrations but also suggests more favourable micrometeorological conditions for upward and downward transport, such as enhanced turbulence and stronger coupling between the surface and the boundary layer.

• Line 384-385: Citation to previous literature should be provided here for this claim.

Citations to relevant previous studies have been provided later in the manuscript, a few lines below, where the broader context of the study is analysed and compared to existing literature. We chose to introduce these references in that section to maintain a logical flow of discussion.

• Line 397-400: Are the deposition velocities cited from previous measurements in these lines from Arctic/polar regions? Please clarify.

Yes, all deposition velocities cited in these lines refer to measurements conducted in remote Arctic or Antarctic locations. We have clarified this in the revised manuscript to make explicit that these values come from polar environments, ensuring a consistent comparison with the present study.

• Line 401-402: Are the authors able to correlate the parameterized deposition velocities using Slinn (1982) with the calculated median values shown in Fig. 5b from this study? How well do they agree? Further, are the author able to speculate on why the Slinn parameterization has such (visually) great agreement with your calculated values from these measurements?

To address the comment, we compared our calculated median deposition velocities with the values obtained using the Slinn (1982) parameterization (SL82) at the same particle diameters. The scatter plot (Fig. R1) of observed vs parameterized values shows a very strong agreement, with a Pearson correlation coefficient of 0.96. This indicates that the SL82 parameterization can reproduce our observations with remarkable accuracy.

As for the reason behind this close match, we do not have a definitive explanation. However, we note that the SL82 formulation explicitly accounts for deposition processes relevant under the conditions of our study, such as Brownian diffusion and impaction, which might dominate the observed particle size ranges. This may contribute to the good agreement found here.

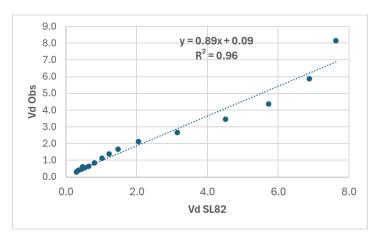


Fig. R1. Scatter plot of observed median deposition velocities vs. values parameterized with Slinn (1982, SL82) at the same particle diameters.

Slinn, W.: Predictions for particle deposition to vegetative canopies, Atmos. Environ., 16, 1785–1794, https://doi.org/10.1016/0004-6981(82)90271-2, 1982.

• Figure 8 caption: it should be clarified that Vn is the deposition velocity normalized by the friction velocity.

 V_n is indeed defined as the deposition velocity normalized by the friction velocity ($V_n = V_d / u^*$). This definition was already provided in the main text (line 425), and we believe it is unnecessary to repeat it in the figure caption to avoid redundancy.

\bullet Line 435: I might suggest referring to the 0.54-0.89 μm size range as the "large" accumulation mode here.

We wish to thank the Reviewer for his/her valuable suggestion. Now, in the revised version of the manuscript, we added large to accumulation mode.

• Line 438: I understand the suggestion that physical characteristics (like different particle densities) may affect the deposition behaviour in this mode, but can the authors clarify why hygroscopicity would affect the deposition behaviour? A citation might be helpful here.

Hygroscopicity can influence particle deposition because it affects particle size and, consequently, the mechanisms controlling deposition. Under conditions of higher relative humidity, hygroscopic particles can grow due to water uptake, increasing their effective diameter. This growth enhances gravitational settling and impaction processes and may also alter the particle's ability to follow

turbulent eddies, thereby changing deposition efficiency in the accumulation mode. For example, hygroscopic growth has been shown to significantly modify particle size distributions and deposition rates (e.g., Seinfeld and Pandis, 2016). Thus, differences in hygroscopicity between sites or particle types could contribute to the observed differences in deposition behaviour. In the revised version of the manuscript a literature citation has been added.

Seinfeld, J.H., Pandis, S.N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, Wiley, New Jersey, 2016.

• Lines 438-441: Citations are needed here to allude to these suggested effects.

We highlight that under polar conditions, electric charging processes induce surface charges on ice and snow particles. Such charging enhances electrostatic attraction of other particles, potentially altering deposition dynamics - especially for intermediate-sized particles that are most sensitive to changes in adhesion and transport mechanisms. In the revised version of the manuscript a literature citation has been added.

Tkachenko, K. and Jacobi, H.-W.: Electrical charging of snow and ice in polar regions and the potential impact on atmospheric chemistry, Environ. Sci.: Atmos., 4, 144, https://doi.org/10.1039/d3ea00084b, 2024

• Fig 9: The authors should provide units for the y-axis in panel d).

In panel d), the same y-axis displays both deposition velocity (V_d) and sensible heat flux (H), which have different units. Therefore, units were not indicated on the axis itself to avoid confusion. However, the units can be clearly inferred from the main text: V_d is expressed in mm s⁻¹, and H in W m⁻². We have added a clarifying note in the figure caption to make this explicit.

Panel d) shows both deposition velocity (V_d , mm s⁻¹) and sensible heat flux (H, W m⁻²) on the same y-axis; units for each variable are given here and can also be found in the main text.

• Fig 10 caption: It should be specified that panels (b,d) are zoomed in versions of panel (a,c).

Thank you for this suggestion. In the revised version of the manuscript a clarification has been added in the caption of Fig. 10 and Fig. 11

In (b) and (d) a zoom of the graph in (a) and (c) panels is reported.