Characterization of size-segregated particles turbulent flux and deposition velocity by eddy correlation method at an Arctic site. A. Donateo et al. - Responses to #RC2.

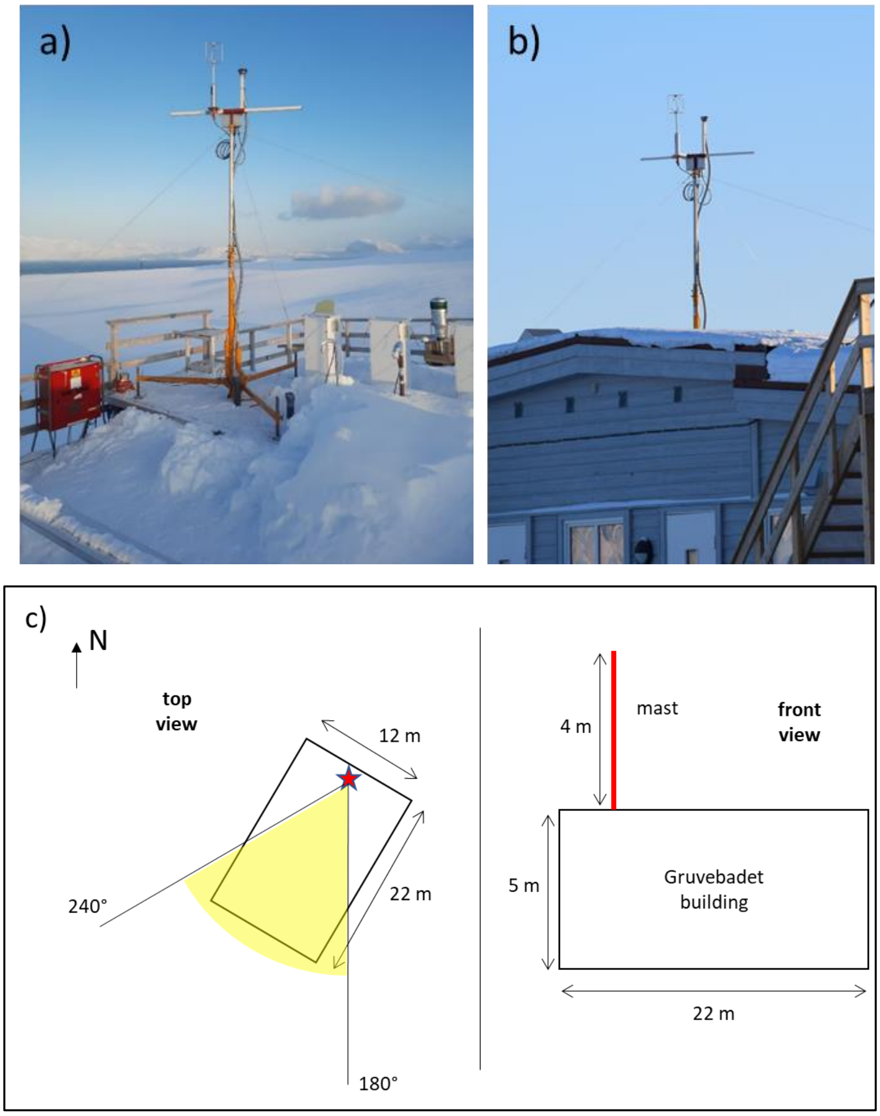
We thank the Reviewer for her/his comments and feedback and for taking the time to help improve this manuscript. Reviewer comments are presented in blue, and sections that have been added to the text are coloured green. Author's responses are reported in black. Note: Figures in the manuscript are referred to as "Fig. X", figures included in these review responses, but not in the manuscript are referred to as "R.Fig. Y".

Donateo et al. present fluxes of particles in ultrafine, accumulation and quasi-course mode over an Arctic site. They use these data to evaluate models of size-dependent particle deposition models. This is a region that is in great need of measurements, and this manuscript offers useful data to the community.

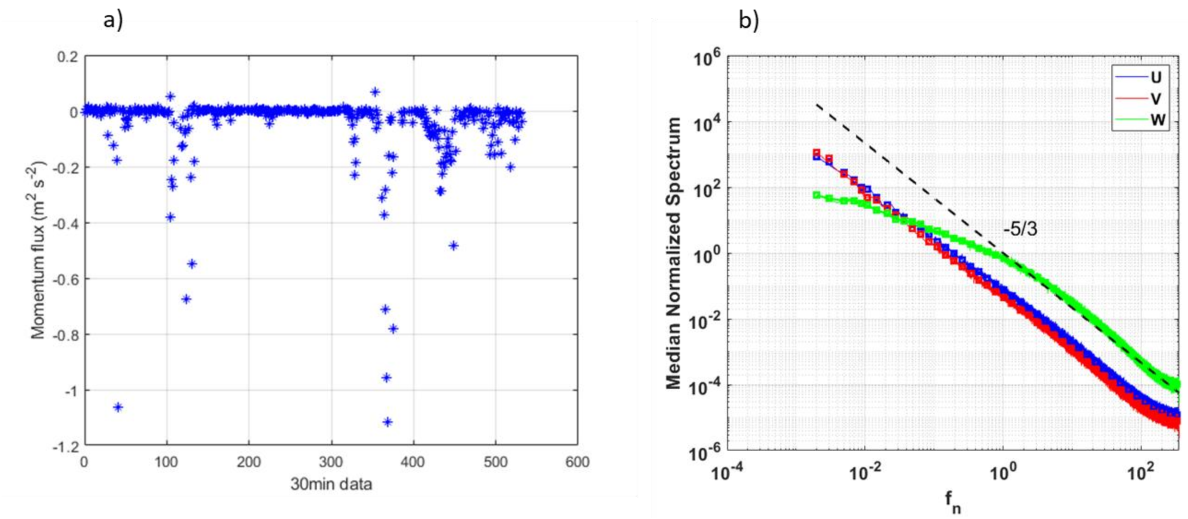
Major Comments

I do have concerns over the placement of the flux measurements. The manuscript indicates that the measurements were on a top of a building on a mast: photos would be useful, but more importantly, we need to understand the potential for the building to impact the turbulence. We need a good diagram of the inlet setup.

A comprehensive response has been provided to a similar question by the Reviewer RC1 in this ACP open discussion (RC1: 'Comment on acp-2022-768' - https://doi.org/10.5194/acp-2022-768-RC1). The mast is 4 m tall above roof level (R.Fig.1a). The building is 5 m high (above ground level) (R.Fig.1b). The anemometer sensing volume and head of the inlet is height 60 cm and 50 cm, respectively. The total height above the ground level is 9.60 m (in the manuscript is reported as "about 10 m", now "9.6 m"). The short side of the building is 12 m, and the long side is 22 m (R.Fig.1c). The mast is located very close to roof edge (northeast-facing side). Possible flow distortions could arise for the presence of the rooftop in the wind direction sector between 180° (S) and 240° (SW) (see R.Fig.1c – yellow sector). Winds coming from this sector is uncommon in this measurement campaign (about 5% of quality assured data) (Fig.3f). Furthermore, the momentum flux for the roof wind sector (see above) did not show positive anomalies (R.Fig.2). An analysis has been performed on spectra for u, v, and w wind components from the roof sector (R.Fig.2b). From this graph can be noted that both the vertical and the horizontal wind components follow very well the trend line (black dashed line) at -5/3 in the inertial subrange, without any evident anomaly. Thus, it doesn't seem to be any important flow distortions arising from the roof sector. For these reasons the Authors decided to take in consideration all the quality assured dataset, also including the wind sector corresponding to the roof area. In the new version of the paper, Authors decided to produce a Supplementary material file. Thus, R.Fig.1 was inserted in this Supplementary file as Fig.S1.

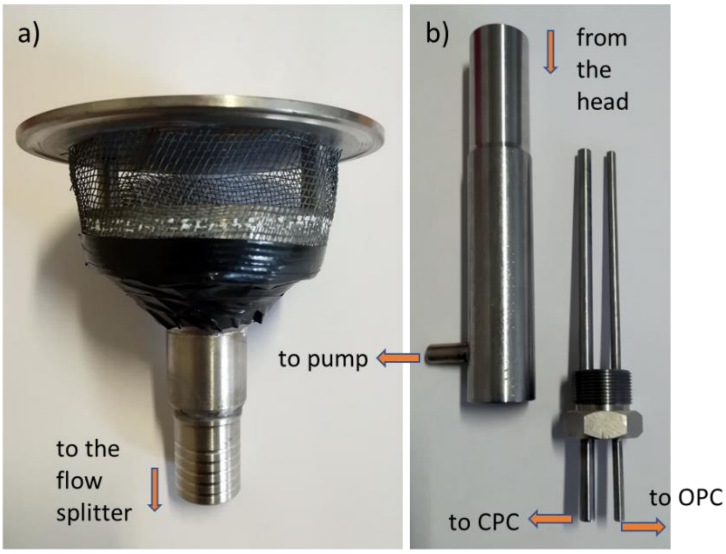


R.Fig.1. (a) Picture of the EC mast located on the roof of Gruvebadet lab and of (b) a front view of the building. (c) A sketch of the top and front view of Gruvebadet lab and EC setup.



R.Fig.2. (a) Momentum flux data points measured in the wind sector between 180° and 240° N. Please, note that the data points are not consecutive in time. (b) Normalized median spectra for u, v and w wind components as a function of the normalized frequency f_n selected from the roof sector. Black dashed line represents the slope theoretically predicted in the inertial subrange.

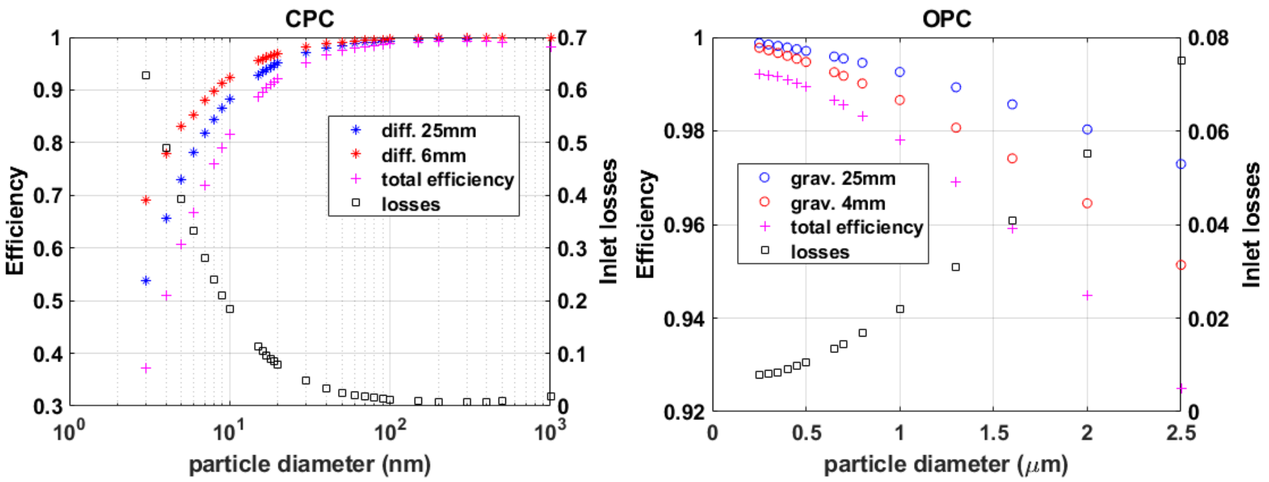
In R.Fig.1a, a picture of the inlet head is reported. It was outside on the pneumatic mast, alongside the anemometer head. The air sample was transported to the instruments, inside the lab, with a silicon tube (as described in the text). At the other end of the tube, a flow splitter (R.Fig.1b) is installed, to divide the air sample between CPC and OPC. R.Fig.1 was inserted in the Supplementary material file as Fig.S2.



R.Fig.1. A picture of the a) inlet head with a metallic hat for precipitation repair and b) the flow splitter.

The particle losses are unclear: the authors suggest that because the D50 cut off is 5 nm, they consider their system to detect particles between 5-1000nm. That is a strange assumption: the particle losses should have substantial size dependences, meaning that some particles in the size range of a given measurement will be far more effectively sampled - and as the paper shows, the size dependence of the fluxes can be substantial even within a single mode. A figure showing the size-dependent losses, and a quantitative discussion of how those losses might introduce biases to the analysis are essential.

In R.Fig.2 we reported the inlet penetration efficiency and the relative losses for the CPC (mainly diffusional) and OPC (mainly gravitational), respectively, calculated according to Hinds (1999) and Baron and Willeke (2001). The total particle losses amount to 9% (on average) for CPC and about 2.3% (on average) for OPC. Particle losses of the measurement inlet was now described clearer in the manuscript (L121-L125). Flux is affected by particle losses at the same way as particle concentration, consequently the deposition velocity should be not affected or marginally overestimated by particle losses being calculated from the ratio of flux on concentration. R.Fig.2 was inserted in the Supplementary material file as Fig.S3.



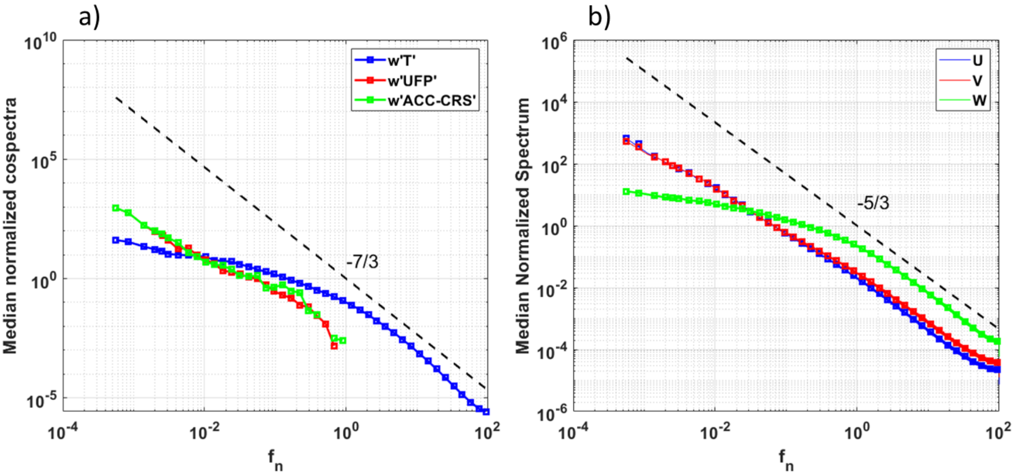
R.Fig.2. Inlet penetration efficiency. a) diffusional particle losses (both for the 25 mm and 6 mm tube) and the total penetration efficiency for the CPC. b) gravitational particle losses (both for the 25 mm and 4 mm tube) and the total penetration efficiency for the OPC. Total inlet losses (in black) of the system on the right axis for each panel.

Baron, P. A. and Willeke, K.: Aerosol Measurement: Principles, Techniques, and Applications, 2nd ed., John Wiley and Sons, New York, 2001.

Hinds, W.C.: Aerosol Technology, Properties, Behaviour, and Measurement of Airborne Particles, second ed. John Wiley and Sons, New York. 1999.

Further, an important piece of QA/QC for eddy covariance fluxes is the cospectrum of the sonic anemometer data. The data shown in Figure 2 are disconcerting, as they suggest that the sensible heat flux data do not follow the expected decay. The authors state similarity to a -7/3 slope, but the plot is on a log-log curve. What actual slope is the data (i.e. -x/3?). This suggests a substantial loss of flux or disruption of the turbulence regime. You are most definitely not in the inertial subrange. I think the authors must add a substantial paragraph acknowledging these problems and considering how they would bias the data.

The Authors agree with the Reviewer. In the original figure (Fig.2a) the kinematic heat flux has been calculated on decimated time series (w and T_s at 1 Hz) to obtain the same frequency of CPC and OPC (1 Hz). However, in this way heat flux did not clearly show its real trend. So, we decided to make again the Fig.2a, reporting the three cospectra each with its normalized frequency. At the same time the data binning has been modified to better represent the cospectral results. Now, in this new figure, the -7/3 trend (Kaimal and Finnigan, 1994) of the scalar cospectra is clearer (R.Fig.3a). CPC and OPC cospectra show their losses at high frequencies, as expected, due to their low time response. As clearly stated in the manuscript (L278-L286) the high frequency losses for CPC and OPC have been corrected using a parametric/in situ procedure proposed by Horst (1997). According to this frequency correction an average loss of about 30% and 21% has been calculated for CPC and OPC, respectively (L282-L283).



R.Fig.3. (a) Normalized median cospectra of kinematic heat flux (blue), UFP (red) and ACC-CRS particle flux (green) as a function of f_n. The binned median cospectra (about 1500) were computed from continuous period of 1h. Black dashed line represents the slope theoretically predicted in the inertial subrange (Kolmogorov, 1941); (b) median normalized ogives for kinematic heat flux as a function of natural frequency (Hz) for the three classes of atmospheric stability.

An analysis has been performed on spectra for u, v, and w wind components taking in consideration the whole dataset (R.Fig.3b). From this graph it can be noted that both the vertical and the horizontal wind components follow very well the trend line (black dashed line) at -5/3 in the inertial subrange, without any evident anomaly (Kaimal and Finnigan, 1994; Stull, 1988). In Fig.2a the line with -7/3 slope is reported. The cospectra in the figure are represented on log-log scale (Schiavon et al., 2019). Actually, they are not log-cospectra.

Horst, T.W.: A simple formula for attenuation of eddy fluxes measured with first order-response scalar sensor. Boundary-Layer Meteorol. 82, 219–233, https://doi.org/10.1023/A:100022913, 1997.

Kaimal, J. C. and Finnigan, J. J.: Atmospheric Boundary Layer flows, Oxford University Press, New York, Oxford, 2nd Edn, 1994.

Schiavon, M., Tampieri, F., Bosveld, F. C., Mazzola, M., Trini Castelli, S., Viola, A. P., Yagüe, C.: The Share of the Mean Turbulent Kinetic Energy in the Near-Neutral Surface Layer for High and Low Wind Speeds, Bound-Lay. Meteor., 172:81–106. https://doi.org/10.1007/s10546-019-00435-6, 2019.

Stull, R. B.: An introduction to boundary layer meteorology, Kluwer Academic Publishers, Dordrecht, 1988.

I generally disagree with the approach of fitting the Vdep/u* versus size range to a polynomial as this is driven purely by the data, and not at all by the underlying process. I think this needs to be made exceptionally clear that this is purely a fit, not a parameterization - and that it only applies to the accumulation / quasi-coarse mode (that should be noted in the Abstract as well).

The Authors agree with the Reviewer about the approach of fitting the deposition velocity to a polynomial because it is driven by data and not by the chemical-physical processes. However, the Authors decided to include this type of analysis to have also a comparison with other works (i.e. Deventer et al, 2015). Now, in the revised text (also in the Abstract and in the Conclusion section), it has been stated clearly that the fit was applied only to ACC and CRS particles size range, excluding the UFP size bin.

Deventer, M. J., Held, A., El-Madany, T.S., Klemm, O.: Size-resolved eddy covariance fluxes of nucleation to accumulation mode aerosol particles over a coniferous forest, Agr. Forest Meteor., 214-215, 328–340, https://doi.org/10.1016/j.agrformet.2015.08.261, 2015.

Overall, this manuscript includes unique data and is well-written. I do hold concerns over the measurement site, and request that the authors provide substantial additional details and consider the implications of not measuring fluxes in the inertial subrange. Further, the authors need to be more careful over their discussion of polynomial fits. However, the data are intriguing, and warrant publication after revision in this journal.

Minor Comments

- The Introduction is well-written, but quite difficult to read as the bulk of it is in a single paragraph. I encourage the authors to consider breaking the main paragraph into about three paragraphs, each addressing the different aspects of the motivation and background.

Corrected in the revised version of the manuscript. Thanks for the suggestions.