We would like to thank the reviewer for the comments that helped to improve the present study. Responses to each comment are provided below - see blue text.

REVIEW #2

The authors report data from a summer and a winter campaign at a monitoring site in Central Europe. The data are valuable and seem to meet the required quality standard. However, the presented analysis and interpretation is not sufficient and does not meet the standard of a publication in ACP. Many things are mentioned and connections are suggested without supporting arguments. There is no focused line that guides the reader from the measurements to the lessons that can be learned. What are the new insights and which results of the study confirm observations from previous studies? Unfortunately, I found no answers to these question, but reading this manuscript left me behind in confusion and I still wonder: What exactly can be learned from this study? Therefore I cannot recommend the publication. But I encourage the authors to improve their analysis and I hope that they find the comments below helpful.

Major comments:

The PMF analysis has been used to identify episodes of high number concentration. However it remains obscure how exactly this has been done. More explanation is needed (maybe in the appendix). Therefore, it remains unclear on which criteria the 10 summer and 13 winter episodes have been selected. In addition, these periods should be marked in Figure 2 (not in the appendix)!

PMF of PNSD is described in Section A1 (in Appendix) along with two new figures A12 and A13.

The criteria for the episode selection is described in the Section 2.2.4: … episodes of high mass concentrations were chosen based on a set of criteria: high mass size distribution of at least one main NR-PM1 species corresponding to the season (NO_3^- ≥ 0.5/0.2 μg m^{-3}, SO_4^{2-} ≥ 1/0.5 μg m^{-3}, Org ≥ 6 /2 μg m^{-3}); monomodal mass size distribution of all main NR-PM1 species; duration of the episodes min 1.5 hour. Also, the episodes were marked as well in Figure 2.

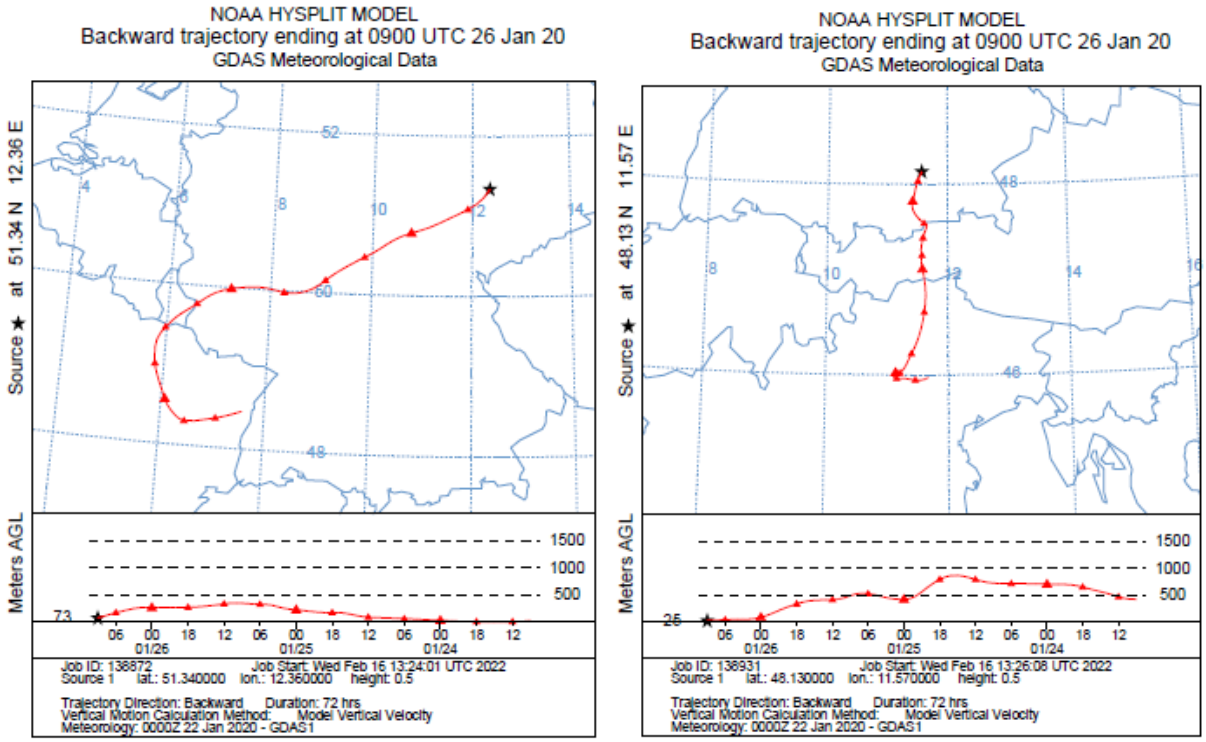
The discussion of the episodes is not convincing and too strong conclusions are drawn from the trajectory calculations without providing additional evidence. In lines 314/315 the authors link high nitrate concentrations to marine air masses. What are the arguments here? Do the authors suggest a marine source of nitrate? I do not see evidence for such a claim. In lines 328/329 maritime influence is suggested again. Given that the air crossed half a continent at low altitudes before being sampled at the site, the claim of marine influence is not convincing.

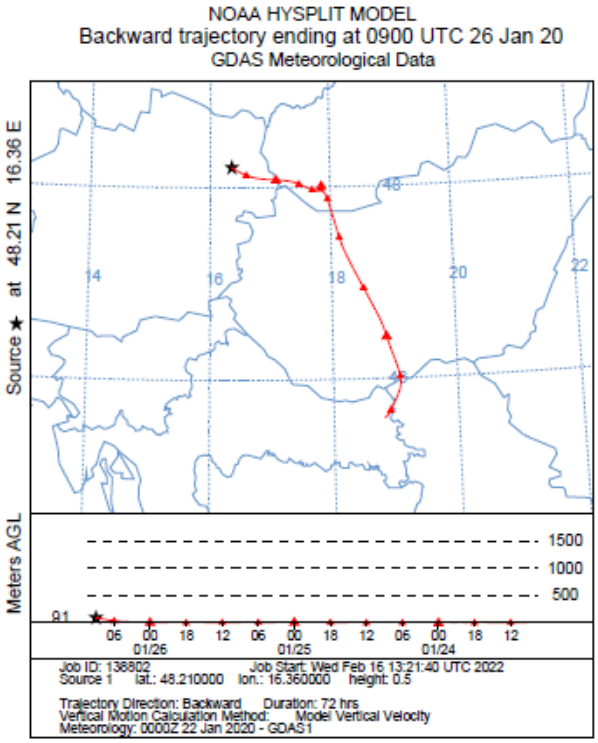
The discussion in lines 327-334 is confusing. Too many different things are mentioned nothing is followed up and or backed by arguments. For example, what is the evidence for inversion conditions in Central Europe during episode 6a? Why would this lead to higher NO3- without increasing organics as well? Moreover, the mixing layer height in Fig A3 suggests stronger inversion during W6b. What do the aauthors concude from f60 and Figure A8 that is mentioned but no interpretation is provided.

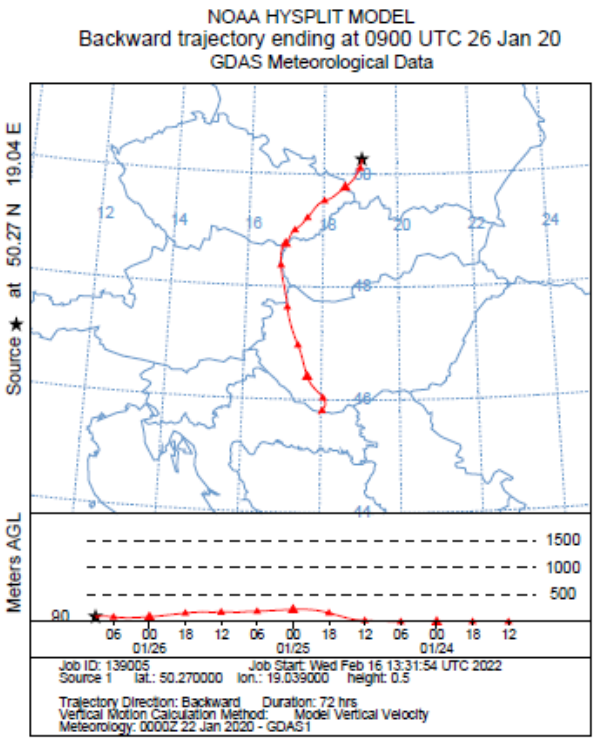
The episodes of high nitrate concentrations (lines 314/315) were discussed as follows: The episodes of high NO_3^- concentrations were mainly linked to continental air masses (from the NW-SW, Fig. A6) from northern France, Benelux, central Germany and northern Italy. These regions were traced as hotspots of high particulate nitrate concentrations related to intense agricultural activities under anticyclonic conditions in late-winter and early-spring (Waked et al., 2014; Petit et al., 2017, 2019; Favez et al., 2021).

Line 328/329 was reworded as follows: The highest concentrations of Org (15.63 μg m^{-3}) as well as low concentrations of SO_4^{2-}, NO_3^- and NH_4^+ (0.74 μg m^{-3}, 0.93 μg m^{-3} and 0.96 μg m^{-3}, respectively) measured in winter during W7 were influenced by maritime air masses crossing France and Germany before reaching the NAOK (Fig. A6).

The evidence of the inversion conditions in Central Europe during episode W6a is the mid-boundary layer height calculated using HYSPLIT for several sites (Leipzig, Munich, Vienna, Katowice and Wroclaw) see the figures below. The mid-boundary layer height ranged from 25 m AGL (Munich) to 91 m AGL (Vienna). At the NAOK the mid-boundary layer height was during for W6a 42 m AGL and W6b 155 m AGL see the figures below. Therefore, we suggested stronger inversion during W6a episode.




NOAA HYSPLIT MODEL
Backward trajectory ending at 0900 UTC 26 Jan 20
GDAS Meteorological Data
Source ★ at 48.21 N 16.36 E
Meters AGL
1500
1000
500
06 00 18 12 06 00 18 12 06 00 18 12
01/26 01/25 01/24
Job ID: 138802 Job Start: Wed Feb 16 13:21:40 UTC 2022
Source 1 lat.: 48.210000 lon.: 16.360000 height: 0.5
Trajectory Direction: Backward Duration: 72 hrs
Vertical Motion Calculation Method: Model Vertical Velocity
Meteorology: 0000Z 22 Jan 2020 - GDAS1


NOAA HYSPLIT MODEL
Backward trajectory ending at 0900 UTC 26 Jan 20
GDAS Meteorological Data
Source ★ at 50.27 N 19.04 E
Meters AGL
1500
1000
500
06 00 18 12 06 00 18 12 06 00 18 12
01/26 01/25 01/24
Job ID: 139005 Job Start: Wed Feb 16 13:31:54 UTC 2022
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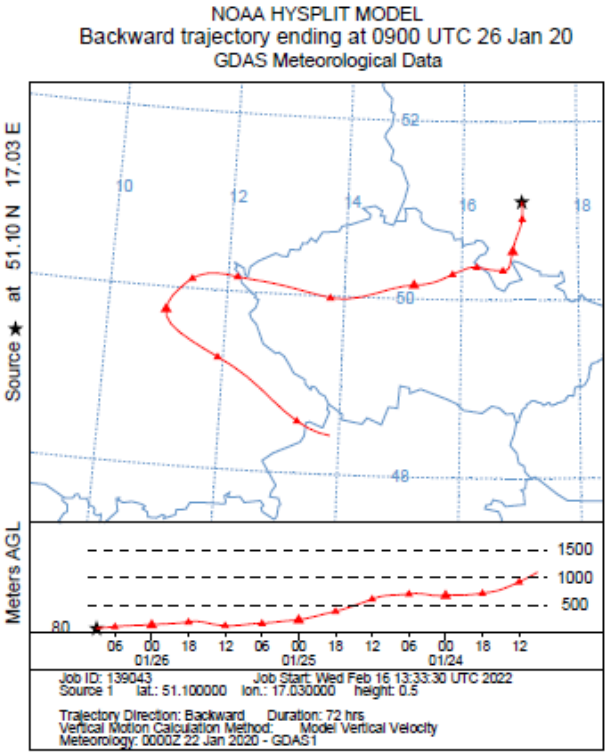
Trajectory Direction: Backward Duration: 72 hrs
Vertical Motion Calculation Method: Model Vertical Velocity
Meteorology: 0000Z 22 Jan 2020 - GDAS1


NOAA HYSPLIT MODEL
Backward trajectory ending at 0900 UTC 26 Jan 20
GDAS Meteorological Data
Source ★ at 51.10 N 17.03 E
Meters AGL
1500
1000
500
06 00 18 12 06 00 18 12 06 00 18 12
01/26 01/25 01/24
Job ID: 139043 Job Start: Wed Feb 16 13:33:30 UTC 2022
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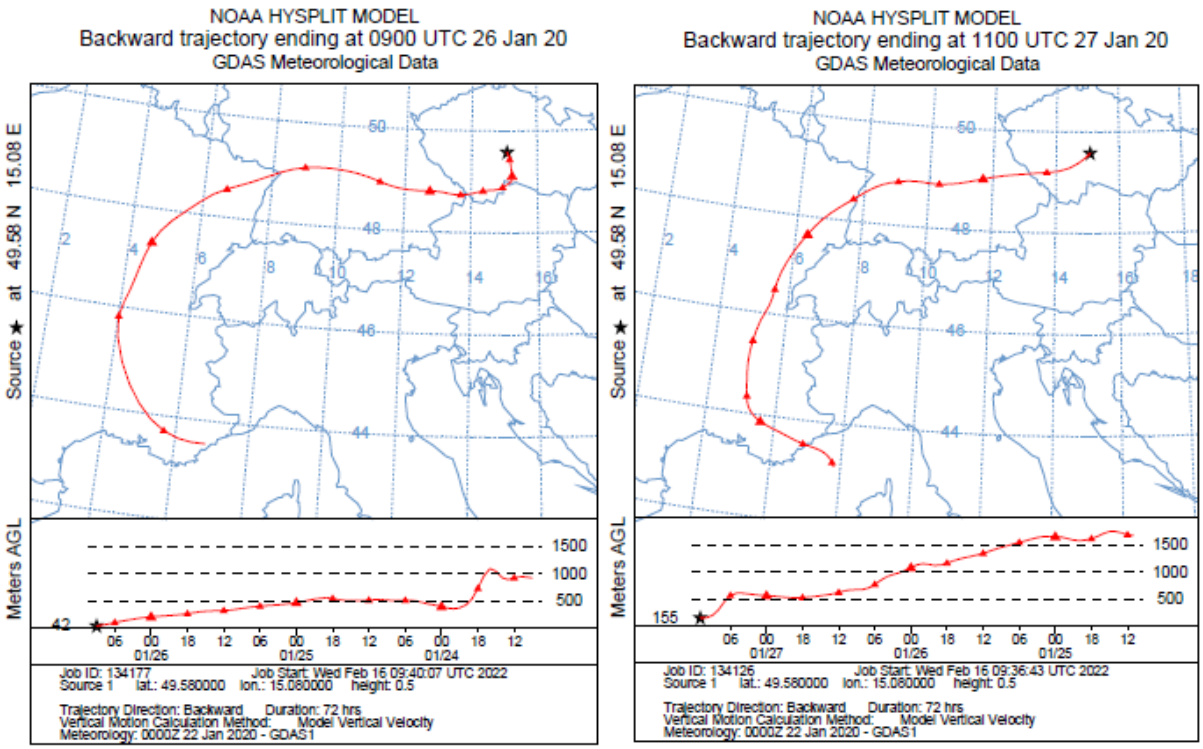
Trajectory Direction: Backward Duration: 72 hrs
Vertical Motion Calculation Method: Model Vertical Velocity
Meteorology: 0000Z 22 Jan 2020 - GDAS1



High concentrations of NO_3^- during W6a are discussed as follows: … the highest NO_3^- concentrations (10.66 μg m^{-3}) measured in the W6a episode were characterized by below-freezing temperatures, which probably arose due to inversion conditions in Central Europe. The conditions prevailing during the W6a episode, in combination with ammonia due to the agricultural activities, including the spreading of fertilizers, induce increase of particulate nitrate and ammonium concentrations similarly as reported Favez et al., 2021 for Northern France.

Fig. A7 compares organic fragments f44 and f60, which enables us to assess the presence of fresh or aged organic aerosols emitted by BB during the monitored periods.

Lines 335-337: Why does the **oxidation state** in winter point out the importance of **local sources**? Hydrocarbon aerosol can be transported as well and will remain hydrocarbon in the absence of significant photochemistry.

New figure showing origin of organic aerosol at Košetice site in winter was added and the text was rephrased as follows:

Organic aerosol ageing was examined on the f44 and f43 fragments (Fig. 3). Winter aerosols were **less oxidized** than summer aerosols. This results along with the organics diurnal trends of late evening maxima (Fig. 3) and polar plots (Fig. A5) pointing to the importance of local combustion sources during the cold part of the year. Importance of local fosile fuels combustion for residential heating as a source of fresh OA/ hydrogen-like OA in winter is presented in study by Chen et al., 2021 (under review).

Chen et al., 2021. European Aerosol Phenomenology - 8: Harmonised Source Apportionment of Organic Aerosol using 22 Yearlong ACSM/AMS Datasets. Environmental International (under review).

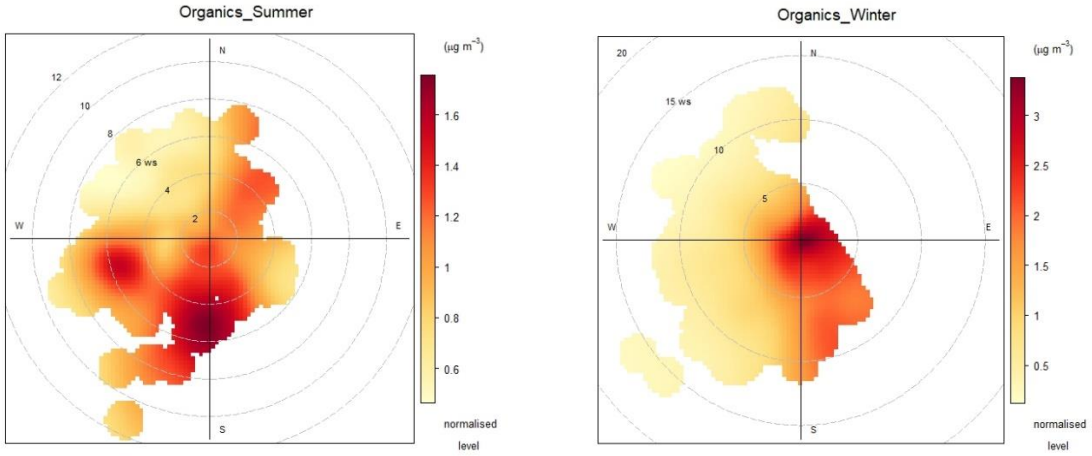


Fig. A5. Polar plots showing the origin of organics in summer (left) and winter (right).

Lines 338-339: How were MOOAs and LV-OOA retrieved? There is no information on this in the manuscript.

The sentence was left out.

Lines 347-353: This may be interesting. I understand that levoglucosan has been measured (although this is not -but should be - mentioned in the method section) about 10 times higher in the winter samples than during summer, but the f60 parameter barely indicates biomass burning influence. In my opinion this could be explored more. Apparently there is a discrepancy and something can be learned here!

In the method section 2.1 PM1 filter analysis by IC for monosaccharide anhydrides is mentioned, however levoglucosan was specifically added to the text as follows:

Additionally, 12-h PM1 filter samples were collected by a sequential Leckel LVS-3 (Sven Leckel Ingenieurbüro, Germany) for subsequent chemical analyses of cations, anions and monosaccharide anhydrides (levoglucosan, mannosan and galactosan) using ion chromatography (Dionex ICS-5000+ system, Sunnyvale, CA, USA).

lines 353-358: I do not understand this discussion and I do not agree that such conclusions can be drawn from Figure A9.

The sentence was rephrased and the number of the figure was corrected as follows: Additionally, a comparison of fragments f44 and f60 enabled us to assess the presence of fresh or aged organic aerosols emitted by BB (e.g., Milic et al., 2017) revealing that aged organic aerosols from BB influenced the site during both seasons especially in winter (Fig. A7).

The discussion in lines 359-377 is not convincing. Are the differences observed between the clusters statistically significant at all? In most cases it seems that they are not.

Yes, there were statistical differences among clusters (summer – 6 clusters, winter – 5 clusters) by all variables using Shapiro-Wilk normality test and Kruskal-Wallis rank sum test at the alpha value 0.05. The results are discussed in the text.

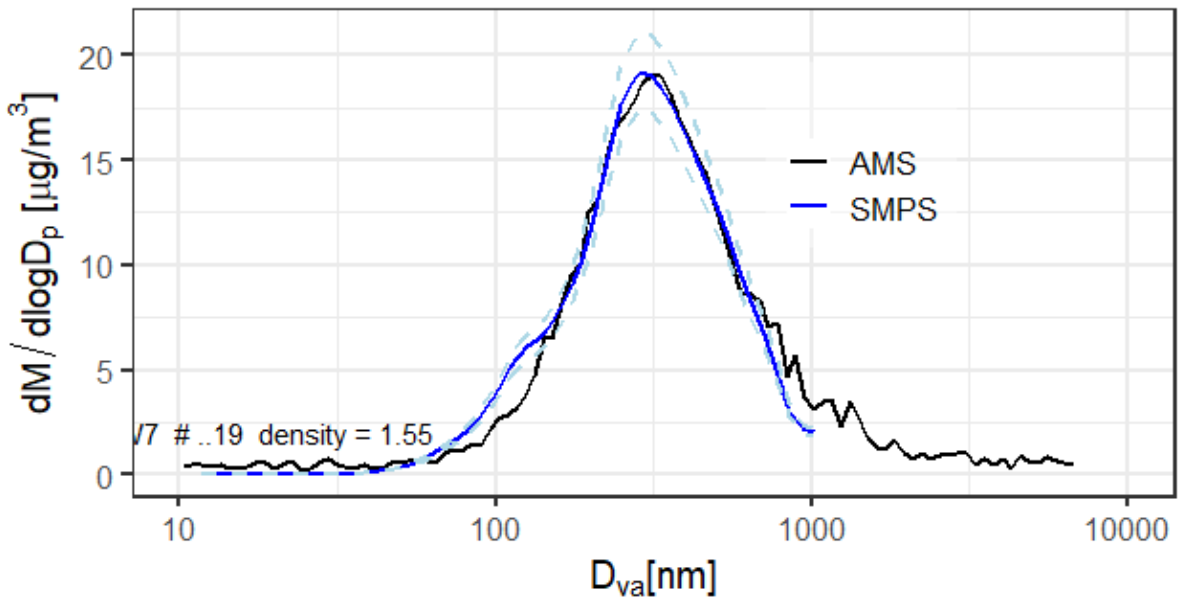
Section 3.4 (lines 388-442) could use more focus and I wonder if all the evidence from other studies for larger particles in winter is needed. More space should be dedicated instead to interpret the findings of this study. The potentially interesting Figure 5 is discussed in less than 3 lines! Certainly much more can be inferred here.

We would like to thank reviewer for his opinion. The whole paragraph was added discussing results of the figure presenting relationship between organic fragment f44 and the size of the organic fraction during episodes of high NR-PM1 species mass concentrations in both seasons.

In section 3.5 the authors focus on density retrievals during the defined episodes. The purpose of this is not clear. It is stated that a density of 1.85 g/cm3 corresponds to black carbon, but certainly nobody is claiming that these particles were predominantly black carbon. So what can be learned from this analysis? Moreover, there is no discussion of uncertainties of this analysis. is a value of 1.45 different than a value of 1.55? or is this still in the range of uncertainty?

The Section was rewritten with a focus on the episodes of high mass concentrations. In this section we discussed results of particle densities (effective and material) and shape factors (Jayne shape factor and dynamic shape factor) calculations as well as the seasonal differences between these parameter.

The density uncertainty from the first approach (density estimate from mobility and aerodynamic measurements, Eq. 1) was estimated to be +- the smallest increment used in the analysis, i.e. 0.05 g cm^{-3}. The difference of 0.05 g cm^{-3} is well visible in the results (taking the uncertainty of the sizing of SMPS – within 3%, see Wiedensohler et al. 2017 and AMS – within 8%, see Takegawa et al., 2005); for example, below the resulting density of 1.55 in winter episode #7 was plotted together with 1.50 and 1.60 (light blue dashed lines). The density uncertainty is mentioned in the Section 2.2.2 Particle density and shape factor estimation.



Takegawa, N., Miyazaki, Y., Kondo, Y., Komazaki, Y., Miyakawa, T., Jimenez, J.L., Jayne, J.T., Worsnop, D.R., Allan, J.D., Weber, R.J., 2005. Characterization of an Aerodyne Aerosol Mass Spectrometer (AMS): Intercomparison with Other Aerosol Instruments, Aerosol Science and Technology, 39:8, 760-770.

Wiedensohler, A., Wiesner, A., Weinhold, K., Birmili, W., Hermann, H., Merkel, M., Müller, T., Pfeifer, S., Schmidt, A., Tuch, T., Velarde, F., Quincey, P., Seeger, S., Nowak, A., 2017. Mobility particle size spectrometers: Calibration procedures and measurement uncertainties. Aerosol Science and Technology, 52:2, 146-164.

Minor comments:

Abstract: abbreviations 'SE' and 'SW' not explained. Better to spell this out...

The abbreviations were spelled out.

The abstract is a list of observations. But what are the lessons learned from the study? What can we conclude from the observations.

The abstract was partially rewritten.

Showing the two almost identical charts in Figure 1 is not necessary in my opinion. In theory the volume based approach should be a little bit more advanced because it takes the particle composition into account to some extent, while the other approach uses a constant density for all particles and seasons.

Yes, we agree, therefore only the result of the volume based approach is presented.

Fig A5: add a legend. The reference "common color code" is not sufficient.

The legend was added.

Fig A14: incomplete caption. ???

The figures were redone and removed to the main text as Figure 8.

Fig A10: remaiuns unclear in the current form. More explanation is needed.

The figure was redone based on the PMF results including number size distribution (dN/dlogDp) as well as volume size distribution (dV/dlog Dp) of the factor profiles (Fig. A12).