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Comment on acp-2022-842

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Referee comment on "A 1-year aerosol chemical speciation monitor (ACSM) source analysis of organic aerosol particle contributions from anthropogenic sources after long-range transport at the TROPOS research station Melpitz" by Samira Atabakhsh et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-842-RC1, 2023

Atabakhsa et al. have reported a nice study of one-year ACSM/MASP measurement. Using rolling PMF and a multilinear regression model, they have conducted detailed source appointments for non-refractory aerosol components and eBC. In addition, to better identify the origin of aerosol sources, clustering analysis was also applied using the back-trajectory cluster method. Based on these approaches, the variations of chemical composition, aerosol mass concentrations, and diurnal cycles among the meteorological seasons were carefully examined and discussed, especially the comparison was performed between the cold and warm seasons. I have a couple of comments on the determination of the emission sources of sulfate, nitrate and org. The comments are not serious but need to be clarified and justified. Overall, the manuscript is well written and easy to read. I would recommend its publication after my comments are addressed.

General comments

The interpretation of sources and diurnal cycles of aerosol components relies heavily on the wind rose patterns and NMR analysis in Sections 3.1-3.3. If I understand correctly, both techniques can only determine local emission sources. However, you have claimed the significance of long-range transported aerosol sources for different measured aerosol species in this study. The statement appears bit abrupt and needs more clarification. Since you have conducted nice back-trajectory and cluster analysis later, I would suggest you already cite those results to support your conclusions in Sections 3.1-3.3.

Specific comments.

L16, How about nucleation? Should it be a source of secondary aerosol?

L48-49, PM1 also has an effect on air quality, right?

Sec. 2.2 How did the temperature vary at the measurement station where ACSM was located, since the ambient temperature may affect the instrument sensitivity from season to season?

Sec.3.1 Since ACSM is associated with a couple of measurement uncertainties, such as the

determinations of collection efficiency and response factor, it would make a lot of sense to compare ACSM derived concentration to other instruments in the parallel measurements.

L228-231, As you have mentioned in the nitrate results in the next paragraph, nitrate concentration dropped in the afternoon due to the vertical mixing. Similar phenomenon is expected for the sulfate species too. However, a flat diurnal cycle was observed in winter when photochemistry was weak. Did it indicate that, except for long-range transport, there were additional sources for sulfate around noon in wintertime? Can you comment on it? In addition, you observed a high peak at noon in other three seasons, and you interpreted it to be the photochemistry of SO2. I just wonder if enough OH are available for SO2 photochemistry to explain such a diurnal peak given the effect of boundary layer mixing? A rough estimation on this point would not go amiss.

L231-232, You should make it clear that it is valid only for the same season that a high sulfate concentration was shown at low wind speed, but does not hold when you compare it among different seasons.

L248-250, You had very good discussion on the local contributions to eBC and org in section 3.1.2, did the local source contribute to nitrate during winter season? As far as I understand, NWR only determines local sources, nor the long-range transported ones (fig.S2). If you argue the dominant sources of nitrate from long-range transport, you might need to justify it. Furthermore, you cluster analysis shows a high local nitrate mass concentration (CS-ST), should this suggest a local source of nitrate?

L254-256, Based on my rough estimation of chemical composition during winter (Fig. 2), nitrate and sulfate were fully neutralized by ammonium as ammonium nitrate and sulfate. It sounds like the contribution of organic nitrate was negligible in this study.

L281-283, Traffic is a good source of nitrate, how does it affect your observed nitrate in this study?

L304-306, I agree with you that boundary lay mixing played a significant role to determine the measured org diurnal, but I don't think you can neglect the fact of evaporation of semi-volatile organic compounds in the middle day, especially in spring and summer seasons.

L331-332, Since you have observed very similar nighttime HOA concentrations in all seasons (Fig. 6), could the slightly different HOA concentration between summer and winter be explained by evaporation of HOA or photochemical conversion to LO/MO-OOA in summer?

L335-337, HOA concentrations observed in summer and winter were very similar, (Fig. 6), sounding more like the characteristics of local emission sources. This is also in line with the usual view of HOA as a local emission factor. The city of Leipzig is located in the sector of HOA wind rose (Fig. 7). Considering that Leipzig is 50km away from to the measurement station, we assume that the average wind speed is 4m/s, and it took only 3 hours to transport Leipzig city aerosol particles to the measurement station. As you have already stated that HOA is mainly emitted by household heating and a minor source of traffic, is it possible that the city of Leipzig, as a local/or regional source, is a HOA contributor? Moreover, here you have attributed HOA to be a long-range transport one in winter. You have shown two peaks in the morning and evening in the diurnal cycle, which sounds they were more likely related to traffic rush hours, do you think if the long-range transported HOA can explain this typical diurnal cycle?

L441-443, Though I agree to that night chemistry may play a role, how about the effect of boundary layer mixing on the diurnal cycle during the daytime?

L445-447, Sulfate is also considered as a regional or long-range transported chemical species (e.g. Zhang et al., 2005). Does a better correlation between MO-OOA and sulfate indicate the nature of long-range transported MO-OOA in this study?

L454-456, are you saying nitrate was formed locally or via long-range transport?

L458-459, As you have stated in lines 248-251that nitrate is a long-range transported species, does a good correlation between LO-OOA and nitrate indicate that LO-OOA is also mainly from long-range transport?

Technical corrections:

L23, The sentence "Melpitz represents due to its location the Central European aerosol" reads odd. Please reword it.

L123, Please give a full name of the acronym when it appears for the first time.

L248, Fig. S3□Fig. 3?

L301, …in night time, …

Fig.S2, The pixel resolution in Fig. S2 is rather poor. What does the radius axis in each rose plot represent?

L372, Fig. 11?

L375, Fig. 4b□Fig.4c

L510, Fig. S3 and S4□Fig. S4 and S5?

L579, Fig. 11?

Reference:

Zhang, Q., et al., Hydrocarbon-like and oxygenated organic aerosols in Pittsburgh: insights into sources and processes of organic aerosols, Atmos. Chem. Phys., 5, 3289-3311, 2005.