Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2021-722

Anonymous Referee #2

Referee comment on "Single-particle characterization of polycyclic aromatic hydrocarbons in background air in northern Europe" by Johannes Passig et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-722-RC2, 2021

Review of the manuscript acp-2021-722 with title: "Single-particle characterization of polycyclic aromatic hydrocarbons in background air in Northern Europe" by Passig et al.

General comments:

This manuscript describes the first field application of a recently developed mass spectrometric method (Schade et al., 2019) to analyse single aerosol particles for characteristic components including polycyclic aromatic hydrocarbons (PAH). This is a substantial contribution demonstrating how this new analytical tool goes beyond conventional single particle mass spectrometry to investigate atmospheric aerosol particles on a single particle basis and hence their internal mixing, aging, and potential sources. The combination of mass spectra from laser desorption ionization (LDI) and resonance enhanced multi photon ionisation allows for a more specific assignment especially of combustion related aerosol particle sources. Most of the methods are clearly outlined and the paper is well structured and written. However, some of the results could have been discussed in somewhat more detail. Overall, this manuscript should be accepted for publication after improvements focussing on the specific comments below.

Specific comments

Page 1 line 26: The appearance of Calcium in aerosol particles is not generally associated with traffic emissions. Therefore, you should reformulate this to avoid misunderstandings.

Page 3 line 76: Please give sufficient credit to previous work e.g. by Morrical et al., 1998 who showed one of the first applications of the two-step approach including PAH.

Page 3 line 90: Please use add the type and mass resolution of the mass spectrometer.

Page 3 line 100: Give the size dependent detection efficiencies and discuss their relevance for your results. E.g. to what extend would you miss PAH in smaller particles?

Page 4 line 122-123: Give the concentration factors for the whole size range of particles measured. Indicate if and how this has impact on the interpretation of your results.

Page 5 line 129: Mention that you compare to an optical particle counter.

Page 5 line 141-144: Explain the manual clustering criteria already in section 2.2 and justify why no automatic procedure was used.

Page 6 Figure 2: Enlarge the mass spectra to the full-page width. Otherwise, they are not readable.

Page 6 line 161: Also smaller particles can contain substantial amounts of secondary material. Specify the sampling and detection bias for different particle sizes and classes in the method section.

Page 6 line 163-166: Give the fraction of aged sea salt. Compare e.g. to Geng et al., 2010.

Page 7 line 173: Explain why no mineral dust was observed and compare e.g. with Marsden et al., 2018.

Page 7 line 183: Compare the particle classes with those identified in previous studies at remote locations in Europe and justify your assignment e.g. Lacher et al., 2021, Schmidt et al., 2017, Geng et al., 2010.

Page 7 line 187: Name the instrument optical particle counter.

Page 7 line 198-199: Reformulate this sentence to avoid misunderstanding. Please discuss if this could also be influence e.g. by a lower detection efficiency for sulphate rich particles?

Page 7 line 201: Please clarify what you mean with sulphur containing and sulphur rich particles.

Page 8 line 214-215: How did you identify night-time new particle formation?

Page 8 line 216: Can you really give relative contributions of different particles classes? Do you account for different detection efficiencies for different particle classes? Please discuss this addressing e.g. Shen et al., 2019a.

Page 8 line 222: Which evidence do you have for this?

Page 8 line 225: There is no data shown for October 14th.

Page 9 line 233: …peak area….

Page 11 line Figure 4: Please enlarge the mass spectra as to make them readable.

Page 12 line 315: Please reformulate this sentence, as iron is not increasing during transport.

Page 12 line 322-323: Please reformulate as you did average the mass spectra but you did not mix them.

Page 12 line 324: 53% of the PAH containing particles were not classified….

Page 12 line 325-326: Please reformulate. E.g. Their mean PAH spectrum originates from different particle types… .

Page 13 line 345: Explain the different number of PAH containing particles compared to Figure 4a.

Page 14 Table 2: Correct "Local green" in row 'alkylated LMW'.

Page 15 line 379: Explain the meaning of 'Ox'.

Page 15 Figure 5c: The grey dots are not good visible.

Page 15 Figure 5d: Choose fillings or colours that allow better to distinguish between the particle classes.

Page 15 line 388: Explain the criteria for manual classification of the subgroups and compare them with the classification in section 3.1.

Page 15 line 399-400: …among most particle subgroups….

Page 17 line 435-448: Please give an estimate of the transport times from potential source regions and typical PAH degradation time scales for typical atmospheric conditions. Demonstrate that your interpretation is reasonable.

Page 18 line 452: Do you mean: 'REMPI spectra of several PAH classes are…'.

Page 19 line 505: Please discuss the LDI spectra.

Page 19 line 515: Please use approximately instead of approx. in the text.

Page 19 line 516: …was previously observed in the analysis of….

Page 20 line 538: This statement is only correct if you could quantify the individual particle classes.

Page 20 line 548: Comparison with additional measurements, e.g. those you have already done, would probably help to do a systematic analysis for a more reliable source apportionment. However, also a comparison with dedicated transport model calculations could help to substantiate you interpretations.

Page 20 line 549: If you do this kind of measurements for the first time it can be expected that you would have taken care for suitable reference measurements either yourself or by inviting suitable other groups.

Page 21 line 555-558: If you have already a larger database for a more systematic and statistically relevant analysis, wouldn't it be possible to make use of it to achieve a better interpretation of the data collected during the measurements described in this manuscript?

Page 21 line 560: Consider giving all relevant data for your measurement campaign including the mass spectra of specific particle classes to an open data repository instead of adding 69 pages to the supplement (e.g. https://www.pangaea.de/).

References:

Geng et al., Single-Particle Characterization of Summertime Arctic Aerosols Collected at Ny-Ålesund, Svalbard, Environ. Sci. Technol., 44, 7, 2348–2353, 2010. https://doi.org/10.1021/es903268j.

Lacher, L., Clemen, H.-C., Shen, X., Mertes, S., Gysel-Beer, M., Moallemi, A., Steinbacher, M., Henne, S., Saathoff, H., Möhler, O., Höhler, K., Schiebel, T., Weber, D., Schrod, J., Schneider, J., and Kanji, Z. A.: Sources and nature of ice-nucleating particles in the free troposphere at Jungfraujoch in winter 2017, Atmos. Chem. Phys. Discuss. [preprint], https://doi.org/10.5194/acp-2021-415, in review, 2021.

Marsden, N. A., Flynn, M. J., Allan, J. D., and Coe, H.: Online differentiation of mineral phase in aerosol particles by ion formation mechanism using a LAAP-TOF single-particle mass spectrometer, Atmos. Meas. Tech., 11, 195–213, 2018. https://doi.org/10.5194/amt-11-195-2018.

Morrical et al., Coupling two-step laser desorption/ionization with aerosol time-of-flight mass spectrometry for the analysis of individual organic particles, J. Am. Soc. Mass Spectrom., 9, 1068–1073, 1998. https://pubs.acs.org/doi/10.1016/S1044-0305%2898%2900074-9.

Schmidt, S., Schneider, J., Klimach, T., Mertes, S., Schenk, L. P., Kupiszewski, P., Curtius, J., and Borrmann, S.: Online single particle analysis of ice particle residuals from mountain-top mixed-phase clouds using laboratory derived particle type assignment, Atmos. Chem. Phys., 17, 575–594, 2017. https://doi.org/10.5194/acp-17-575-2017.