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Comment on acp-2021-722

Anonymous Referee #1

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-RC1, 2021

Review of “Single-particle characterization of polycyclic aromatic hydrocarbons in background air in Northern Europe”

Passig and coauthors describe a measurement campaign at a remote site in coastal Sweden using a two-step LDI/REMPI single particle mass spectrometer designed to measure inorganic, general organic and more specific PAH particle content. Because the instrument separately records more typical LDI mass spectra and more unique REMPI PAH mass spectra, two approaches were taken to bring this compositional information together. Firstly, the LDI mass spectra were clustered using the regular neural network approach, and the corresponding REMPI PAH signatures were examined to gain insight on sources/processing. Secondly, the REMPI dataset was clustered and within those resulting PAH classes the LDI mass spectra were further examined to highlight differences between sources and processes for PAH-containing particles. Although PAHs were detected in relatively few particles in the overall dataset (which can be explained by the remote location and the dominance of aged particles), some interesting connections between PAH mass spectral patterns and the refractory core composition of the single particles can nonetheless be obtained. While the article represents a ‘proof-of-concept’ for the method, the features of the dataset are useful for informing future particle classifications at other sites. A few things stand out. Firstly the connection between potassium-rich particle cores and pyrogenic PAH signatures (m/z 228/252) is emerging as a useful signature for woodburning particles, even after long atmospheric processing times. Secondly, the connection between iron (detected with high sensitivity using the excimer laser here) and petrogenic PAH signatures (m/z192/206) is useful for identifying aged engine exhaust. Thirdly, more fresh soot particles associated with engine exhaust are characterized by LDI signals for calcium and alkylated phenanthrenes. This information is useful for identifying the original primary sources of these particles, even at remote sites. The discussion of limitations is also useful, as there remain potential hurdles for this type of analysis, including the difficulty in measuring substituted or heterocyclic PAHs. Overall, however, as a first examination of how to parse the complex ambient datasets generated using this new approach, the findings here are valuable for the single particle and source apportionment communities. I only have minor comments to suggest.

Line 90 define continuous wave

Line 91: “A few”

Line 114: It would be helpful to discuss how the vigilance factor was arrived at for the REMPI dataset

Line 139: M for million

Line 163: Na_2^+

Line 166: Na_2Cl for m/z 81/83

Line 179: Was this EC/OC regrouping used to inform the final 10 classes in some way?

Line 186 subscript 2.5

Line 187 m^3

Figure 3 part of caption unclear: top row and bottom row?

Line 287 caption: “absolute number and number fraction”

Table 2: bottom row, “Local”

Line 405: Refer to the name of this PAH class- HMW?

Figure 8: Concerning the acidity aspect- it is interesting that there is no detectable signal for ammonium in these sulfate-containing particles. Although this appears to be the case for several of the PAH classes. Is detection of ammonium less efficient in this system

relative to single-step LDI instruments. This would be worth discussing.