Atmospheric
Chemistry
and Physics
Discussions

# Interactive comment on "Deconvolution of FIGAERO-CIMS thermal desorption profiles using positive matrix factorisation to identify chemical and physical processes during particle evaporation" *by* Angela Buchholz et al.

**Anonymous Referee #1**

Received and published: 14 January 2020

Secondary organic aerosol (SOA) is an important fraction of aerosol particles in the atmosphere all over the world. However, its formation via different chemical and/or physical processes remains largely unquantified, mostly due to incapacity of separating these pathways. In addition to the traditional measurement of SOA by aerosol mass spectrometry, in which the molecular fingerprint of SOA is seldomly preserved, the recent development of FIGAERO inlet does allow to explore the molecular information. A primary challenge for FIGAERO data analysis is that there are also several processes going on in parallel that is difficult to deconvolute. As also pointed out by

the authors, vapor condensation and aerosol-phase (or aqueous-phase chemistry) are occurring simultaneously; the latter alone also contains many different pathways, such as oligomerization, hydrolysis, formation of organic salts, and etc. To address this major challenge in FIGAERO data analysis, the authors applied PMF analysis on such data sets for the first time, which convincibly shows that PMF is able to separate, to a big extent, different parallel processes. Thus, in general, I found this work containing enough new insights and recommend its acceptance in ACP.

However, besides these insights, I have several comments to be addressed, as list below:

1. P2 L23-24, "...,thus reducing the mass transport limitation which hinders evaporation." This sentence reads ambiguous. It can mean that reducing the mass transport limitation hinders evaporation, which I believe the opposite of what the authors meant. Please rephrase.

2. P6 L24-26, Eq. 4-6. The $Ratio_{exp}$ seems to represent the explained fraction of the measured data by PMF. How does this differ from the traditional way of calculating the explained fraction $(sum|R_{ij} – X_{ij}|)/(sum|X_{ij}|)$? What is the advantage of using this one?

3. P8 L18, Eq.8. Based on the observation of model residue, you decide to use CNerror instead of PLerror for uncertainty matrix. As shown in Figure S1, CNerror is about 1-2 orders of magnitude smaller than PLerror. However, with the CNerror, Q/Qexp got very close to 1, as shown in Figure S2, which seems to indicate that CNerror is the true error or close to that. Do you have any idea about the reason?

4. P11, L1-4. It seems that the criterion of justify the type V factors is its little change in Tmax with aerosol age or water content. As the authors stated "But this method can be applied to e.g. ambient FIGAERO-CIMS measurements as well" In the abstract (P2 L5), it is important to suggest how to determine the type V factors when applying PMF on ambient data sets, where aerosol oxidation degree (O:C), aerosol age and water content are often correlated.

5. P11 L10-14. I have difficulty in understanding why V factors in different experiments with similar Tmax may have significantly different compounds. For example, LV5, MV4, and HV4 seem to have similar Tmax but very different average elemental composition. Can you give more explanations?

6. P13 L20-23. Likewise, same molecules (e.g., $C_8H_{10}O_5$) can be separated into different V factors. Together with Point 5, do these observations suggest that the degree of thermos-decomposition may play an important (even a major) role in the final detected FIGAERO spectra?

7. P14 L3-4. Do you have any hint to explain why highOC SOA seems to be more influenced by aqueous-phase chemistry?