Atmospheric
Measurement
Techniques

Discussions

Interactive comment on "Estimation of the volatility distribution of organic aerosol combining thermodenuder and isothermal dilution measurements" *by* Evangelos E. Louvaris et al.

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(1) *This paper provided a new technique to measure/estimate the volatility distribution of organic aerosols by combining a thermodenuder (TD) and a dilution system. Cooking OA was used as an example to show the performance of this technique. In general, it could be a better technique than a separated TD system, which would be suffered by the possible processes occurred during the heating, e.g. thermal decomposition. The manuscript is overall well written and the topic fits the scope of AMT. I therefore recommend this manuscript can be published after some revision.*

We appreciate the positive assessment of our work by the referee. Detail responses to

the comments can be found below.

(2) *More discussions/statements are needed to explain how these two systems are combined. What is the role of dilution system in estimating the volatility distribution of targeted aerosol?*

The dilution data, at least in these experiments, help constrain the contributions of the more volatile organic aerosol components. These are the components with C* higher or equal to 10 μg m^{-3} in this case (see Fig. 7). However, the dilution measurements cannot help constrain better the contributions of the LVOCs and ELVOCs to the organic aerosol composition. Even if the dilution results do not depend on the enthalpy of evaporation, their combination with the thermodenuder data does reduce the uncertainty of the corresponding estimate. These are now discussed in a new paragraph in the Conclusions section.

(3) *How to understand the differences between the modelled and experimented data during isothermal dilution (Fig. 4b)? It looks that the evaporation only occurred during the first few minutes, but not a continuous evaporation. Will this influence the understanding and estimation of the volatility distribution?*

This behaviour is quite useful in the estimation of both the volatility distribution of the cooking OA and the existence of any potential resistances to mass transfer. The prompt evaporation is interpreted by our model as evidence that any resistances to mass transfer in this system were modest. The resulting accommodation coefficients (evaporation coefficients in this case) were 0.06 and 0.07. The fact that only 20 percent of the COA evaporated during these first few minutes, suggests that the contribution of the more volatile OA components (IVOCs and part of the SVOCs) as also modest. After this 20 percent evaporated the system reached equilibrium and evaporation stopped. These useful insights from the behaviour of the dilution curve have been added to the paper.

(4) *It is highly possible that COA contain some ELVOC, e.g. HULIS (Nie et al., 2017@ACP). I suggest the author to provide a more bin of ELVOCs?*

This is a good point that deserves a little more attention. Almost all thermodenuder measurements stop at a temperature at which the MFR is not zero. In our case at 250 C, the MFR was approximately 5 percent. At least this fraction could in principle be ELVOCs. Our model adds this fraction to the lowest volatility bin used ($C^*=10^{-3}$ μg m^{-3}). In general, the contribution that appears in the lowest volatility bin refers to material that has volatility less or equal to the corresponding value. This point is now made in the paper together with a reference to the work of Nie et al. (2017) discussing the contributions of HULIS to ELVOCs.

(5) *How to change the dilution time, by changing flow?*

There are a number of ways to increase the time available for dilution measurements. One is to take less frequent samples from the dilution chamber. Use of lower flow rates can also work. One could also use larger dilution chambers, which also help achieve higher dilution ratios, but are a lot more difficult to operate in the field. Please note however that in these experiments the COA reached equilibrium after 30 min, so the remaining 2 hours of dilution data helped only in confirming the equilibrium state of the system.

(6) *Why the mass spectra look so similar before and after heating or dilution (25 C vs 200 C)? The volatility of organics should be correlated to their oxidation state (e.g. O/C), molecular weight et al. (E)LOVC tends to have a higher oxidation state and large molecular weight. This indicate there should be some differences for the mass fragmentation between evaporated mass and remained mass. PMF could be used for the AMS measurements in case of several heating steps, to make the change of mass spectra clearer, e.g. Hong et al., 2017@ACP.*

The modest difference in the AMS spectra of all the COA and its less volatile compo-

nents is indeed interesting. This is probably due to the fact that the AMS measures mainly the fragments of the corresponding organic molecules which probably had a lot of similarities in this case. If the differences in volatility of compounds coming from the same source were mainly related to differences in the size of the organic molecules and not so much to their chemical nature (acids, hydrocarbons, etc.) one would expect such behaviour. Please note that there are differences (a theta angle of 11 degrees does support this) but that at least from the point of view of the AMS these differences are modest. PMF does not help in this case, because for two factors it results in more or less the ambient and the 200 C spectra that have already been analysed. A brief discussion of this point has been added to the paper.

(7) *Any ideas about the phase state of produced COA, which could influence the volatility measurement.*

These experiments do not provide much information about the phase state of the produced COA. The relatively fast evaporation during dilution does suggest relatively small delays to evaporation consistent with an evaporation coefficient of 0.06-0.07. These have already been considered in the analysis. In the sensitivity analysis section (Section 3.2.1) we discuss the sensitivity of our results to these uncertain mass transfer resistances considering the case of evaporation coefficients of 0.01 and 0.1. These tests suggested that the enthalpy of vaporization is rather insensitive to the evaporation coefficient in this range, and that the SVOCs and LVOCs changed by less than 15 percent. These are discussed in the Sensitivity Analysis section.

(8) *The residence time is effective residence time or total residence time? There would be a temperature profile in the thermodenuder? An effective residence time should be carefully considered.*

The residence time of 14 s mentioned in the paper is the centerline residence time at 298 K. The temperature profile (both in the longitudinal and radial directions) in our TD

has been analysed by Lee et al. (2010) and Gkatzelis et al. (2016). The change in volumetric flowrate due to the change in temperature along the TD is taken into account by the Riipinen et al. (2010) TD model used in this work. This information has been added to the manuscript.

(9) *The given vaporization enthalpy of COA in this study is a universal value, or can only be used in this work?*

The vaporization enthalpy of COA estimated here is applicable to pork meat char broiling. We do expect that it is a useful estimate for char broiling of other meat, but clearly this needs to be supported by other experiments. Its applicability to other types of cooking (e.g., frying) is unknown. We have added a qualifying statement about the use of the corresponding value.

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