

Interactive comment on “Formation of anthropogenic secondary organic aerosol (SOA) and its influence on biogenic SOA properties” by E. U. Emanuelsson et al.

Anonymous Referee #2

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This work describes chamber studies of secondary organic aerosol (SOA) generated from both aromatic (anthropogenic) and monoterpene (biogenic) precursors. The focus is on mixed anthropogenic-biogenic SOA (“ABSOA”), with the goal of understanding how the anthropogenic fraction influences the properties of the aerosol as a whole. The topic is certainly of interest to the ACP readership. The dataset, which includes measurements of particle loading, composition, and volatility - is extensive and quite impressive. However, the interpretation of results in terms of how aromatic SOA influences monoterpene SOA is not very clear; additionally, some of the data analysis needs to be explained in more detail. These need to be addressed prior to publication in ACP.

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Major points:

The authors provide an excellent, extensive dataset on mixed aromatic-terpene SOA, and then attempt to describe how one component affects the other. However, at no point is the mixed aerosol (“ABSOA”) ever described in terms of simple additivity – how well are the key SOA parameters (yield, O/C, VFR) described by a combination of the pure aromatic and biogenic components? For example, how much of the decrease in volatility when ASOA is added to BSOA is just a result of the mixing in of lower-volatility ASOA, and how much is from some additional synergistic effect? This is a crucial question in understanding anthropogenic-biogenic interactions. This question of additivity needs to be in the manuscript (probably starting in section 4.2) if any conclusions about aromatic influences on biogenic SOA are to be made.

Throughout the paper, “aromatic” and “anthropogenic” are taken to be synonymous, as are “monoterpene” and “biogenic”. However there are many more anthropogenic SOA precursors than just aromatic species, and many more biogenic ones than just monoterpenes. The language throughout should be altered to reflect this.

Also throughout the manuscript, volatility is described in terms of “persistency”, with low-volatility particles referred to as “persistent”. I’m not sure I understand this non-standard use of the word “persistent”. The particles are indeed longer-lasting upon dilution or heating, but in general that won’t greatly affect their lifetime in the atmosphere – the main sink of SOA is usually deposition. Thus I would recommend the VFR data be interpreted in terms of volatility rather than persistence.

Other points:

P. 20315, Line 15: “Low NO_x” (1 ppb) and “High NO_x” (10 ppb) conditions are described, but not explained. Whether conditions are considered low or high NO_x usually refers to the fate of RO₂ radicals – high NO_x refers to when RO₂ radicals react with NO. Was this definitely the case here?

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Additionally, there is little discussion of NO_x concentrations later in the paper (Results, Discussion). Did NO_x levels have any bearing on results?

Section 2: Much more information needs to be provided as to how yields are calculated. What value is used for density? How are losses particle wall loss treated? Vapor wall losses? How is dilution (which affects both dHC and Coa) accounted for?

Table 1 and Figure 2: Extremely low aerosol loadings (<0.02 ug/m³) and yields (<0.0001) are reported. At such low loadings, background aerosol formation, which can be of this level (or higher), needs to be considered. Were blank runs (irradiation of NO_x in the absence of VOC) carried out?

Tables 1 and 2: both the initial HC and the amount of HC reacted away should be given.

Fig 3: What are the sources of OH in the chamber? Are the fluctuations in [OH] real (from changing amounts of sunlight), or a function of noise in the LIF?

P 20320: the calculation of ASOA fraction needs to be described in more detail. I assume "P" refers to semivolatile products only? A major assumption here is that partitioning is not only instantaneous but also ideal: aromatic oxidation products mix in with BSOA as well as they do ASOA. A second assumption is that the extent of oxidation of a given precursor is independent of the presence of other species. The validity and implications of these assumptions should be discussed.

P 20320, Line 22: A reference is made to experimental determination of ASOA fraction, in order to validate the model, but as far as I can tell there is no description of how the experimental value is determined. This needs to be discussed. Additionally, Table 3 includes ASOA fractions determined by AMS data; how these are determined is also not explained (other than a very vague statement in the table caption). The model-measurement discrepancies in this table are very different from those discussed in the text (p 20320) – why is this?

P 20321: Comparison with other yield results other than just Hildebrandt et al would

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be useful.

P 20322, Line 13: "remains in the condensed phase" is more accurate than "is non-volatile".

Section 4.4 (and Figure 6): It seems the strongest correlation is between VFR and ASOA fraction, but this is never plotted directly. It probably deserves its own panel in Figure 6.

P 20329, L7: I don't understand this sentence. For a smaller carbon backbone, more oxidation steps are NOT necessary for a given increase in O/C. In fact, aromatic oxidation mechanisms involve relatively large increases in O/C per reaction step, since multiple O₂ molecules can be added.

P 20331, L 5-19 (an P 20333, L 15): Here the results are described in terms of previous work showing "anthropogenically influenced biogenic SOA". However, the authors have not clearly shown that the aromatic SOA is truly changing the biogenic SOA – rather, it just appears that when aromatic SOA mixes in, it alters the properties of the overall SOA, on account of its high O/C and low volatility. To show the biogenic fraction is truly being influenced by the aromatic fraction, the additivity calculations (described above) would seem to be necessary.

Typos: 20321, L21: principal; 20325, L28: disentangle; 20331, L28: (units)

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