

Answers to Referee #1

We copied the referee comments and set it in italic and our answers are in normal font.

Rev.: The paper represents interesting and relevant data set on the relationship of yield and organic aerosol mass loading and further on the anthropogenic contribution to the mixed SOA. The study also shows that the increasing fraction of ASOA in the mixed particles decreased the volatility of the particles. This seems to be related to the increasing O/C ratio of the particles which resulted from the elevated OH exposure that was needed to produce ASOA. Authors also report, not so surprisingly, the overall clear correlation with increasing O/C and decreasing volatility.

All in all, the experimental methods used in the study are scientifically sound, as well as the data processing. The paper is well written and structured, and represent impressive amount of data, which also makes the paper a bit difficult to follow at some places. I have only a few minor comments that authors should take into account.

Ans.: We thank the reviewer for the kind remarks and we tried to improve the manuscript along the proposed lines.

Rev.: Table 1: it would help the reader if the corresponding values for BSOAs would be added to the table.

Ans.: First of all we would like to mention that the experiments including BSOA were not optimized for yield determinations. Nevertheless we can derive yield for exps 10/6, 11/6, 14/6, 18/6, 22/6 with substantial error bars, though. The yields are listed now in Table 1 and set in perspective to the aromatic yields at the end of section 4.1. Accordingly section 4.1 was renamed to SOA yields. The yields will be also used for testing for non-linear effects in ABSOA in a new section 4.5.

We added a new Figure 7 which made anthropogenic enhancement on VFR now clearer. We changed Conclusions accordingly.

Rev.: Experimental procedure seems to be such that in each case where AVOCs were first injected into the chamber the sunlight exposure took place right in the beginning of the experiment. If the experiments started with BVOC injection, the situation was different: the sunlight exposure took place after the beginning of the experiment. Is there some reason for this "pattern"? If there is, authors should tell it to the readers.

Ans.: In the experiments dealing with pure ASOA we expected low particulate mass concentrations. Therefore the roof of the chamber was opened before AVOC addition, since we wanted to learn about the background reactivity and chamber induced aerosol formation. The background reactivity of the chamber produced particulate matter in the range of 0.004-0.015 $\mu\text{g m}^{-3}$. This value was subtracted from the AVOC induced ASOA formation. If BVOC was involved we added O_3 to the BVOC mix in the dark just before roof opening. In these cases background reactivity is unimportant.

We added the following sentence in the experimental section:

“In the ASOA studies we opened the roof of the chamber and exposed it to sun light before AVOC addition in order to learn about the chamber induced particle formation. The background reactivity in the chamber produced particulate mass $< 0.015 \mu\text{g m}^{-3}$ and typically $0.005 \mu\text{g m}^{-3}$ - a negligible contribution in most cases. The ASOA yields only consider AVOC induced ASOA mass and the background particulate mass was treated as an offset.”

Rev.: *I'd like the authors to comment the possible artifacts related to the filter sampling. Was the sampling time short enough to prevent the possible evaporation of high vapor pressure compounds?*

Ans.: The filter sampling lasted for one hour per filter. The sampling time was necessary in order to sample sufficient mass on the filters. We used XAD-4 resin coated annular denuders to remove gases and vapors before the filter sample. High vapor pressure compounds will not survive. We are biased towards the low volatile fraction. The resin type used in the denuder is now specified in the text.

Rev.: *What are the uncertainties of the AMS measurements and O/C ratios? All in all, error bars to the figures 3, 5, and 6 should be added.*

Ans.: The reproducibility of the O/C ratio can be seen from the variation of the magenta curve in Figure 3. In addition the determination of the O/C ratio has a systematic error which was estimated to about 30% (Aiken et al., 2007). We only applied standard procedures in the AMS evaluation and we had a relative bad performance of the instrument in the W-TOF mode. Because of that we used f44 from the V-mode to characterize the ageing state, and this was stated in the paper. For f44 in Figure 6c the statistical error bars are given, as is the error for the O/C ratio at the end of the respective experiment phase in Table 2. Figure 3 demonstrates the overview over the time dependent observations in order to give an impression how direct observation and derived quantities evolve in time and relative to each other. No quantitative conclusions are drawn from the Figure 3. We therefore prefer not to add errors bars in this figure. We will state the error estimates more clearly in the tables and in the experimental section. Where missing errors estimates were added in the tables or in the text of the manuscript.

Rev.: *Authors only analyse the O/C ratio of the particle by HR-TOF-AMS and omit othermore detailed methods. From AMS data it is possible to learn a great deal about the products that form during the oxidation. It is a bit disappointing to see so little effort given here in this manuscript.*

Ans.: In our opinion we extracted from the AMS measurements what was needed for the interpretation of the VTDMA data. Moreover, we provided filter measurements with explicit speciation. We would also like to mention that our approach using f44 to characterize anthropogenic fraction corroborates the results from the simple model approach. This paper is already quite complex and we prefer not to open a new discussion thread on AMS results.

Rev.: *The results based on the model calculations are multiplied by the correction factor of 1.4 (page 9). The correction factor is defined based on two different experiments. Authors should estimate how reliable is the derived correction factor and give some reasons for this discrepancy.*

Ans.: The model calculations are conceptually very simple. It should mainly serve to classify the observations. The one product model assumes that all condensable material Psum is formed with turnover rates of the primary attack of OH to the aromatics. Psum is then partitioned into the particulate phase according our yield curve and the observed volume of the aerosol in the chamber. We described one major source of error: In this concept the model may produce too much particulate matter too fast. Since particle loss in the chamber (and the model) is faster than loss of gaseous components the fraction of Psum in the particles could be removed too fast. At long times that would lead to an underestimate of the anthropogenic fraction.

However compared to the two cases where it can be directly tested the model overestimates the anthropogenic fraction, so finally it shuffles too much Psum into the particles. Neglecting wall loss of Psum could be a cause, but the yields were also calculated neglecting wall loss of vapors, so that should be inherently taken care of. The assumed particle lifetimes in the chamber could be too short, but these are the same as derived in the yield determination. Our yields could be too large, but compared to Hildebrandt data and literature data we are at the low end side of the yields.

We added a few more information about typical times to the text.

Instead of refining the model which does fine within a factor of 1.4, we extended the description of the analysis of AMS based f44, which was also used to estimate the anthropogenic fraction and *cum granis salis* agrees with the simple model calculation. This moved from section 4.4 to the method section 3.3. The comparison is now more explicitly stated.

Rev.: *There is far too much data presented in one plot in figures 3 a and b. It would be much easier to follow the story if the data was presented in a clearer manner.*

We splitted the figure in 4 panels.