

Interactive comment on “Secondary organic aerosol formation from acetylene (C₂H₂): seed effect on SOA yields due to organic photochemistry in the aerosol aqueous phase” by R. Volkamer et al.

Anonymous Referee #2

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This paper presents new and interesting results on the formation of secondary organic aerosol based on laboratory experiments. The topic is of great interest and the data presented are important. The paper is a valuable contribution to understanding SOA formation. The manuscript is, however, somewhat challenging to read and many details are not presented; it comes across as hurriedly written. Some issues are easily addressed through editing, and hopefully the suggestions below will be of some help. In many ways the problems seem to be the result of the authors attempting to cover a large number of topics, with actually more focus on a general discussion versus de-

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tailed description of the experiments. Thus, the experimental results section is a small portion of the manuscript, with the bulk of the paper being a comparison, critique, and interpretation of other studies, and a rather long-winded discussion of possible implications. The result is a panacea on SOA formation at the expense of a careful presentation and analysis of the experiments. The paper would be improved if more attention were paid to a detailed discussion of the experimental results, possibly by a reorganization of the results section. This could include more background information, why certain experiments were performed (eg, the hypotheses to test) and more detailed descriptions of the results. Although the Discussion is very long, it does contain insightful and valuable comments, but overall could likely be reduced in length.

Specific Comments

Acronyms, especially those for various aerosol seed composition, are often used without prior definition (or the definitions are hard to find). For example, Table 1 is very difficult to understand until all the meanings of all acronyms are tracked down. The paper would be greatly improved if a table of acronyms were included.

Experimental section

Pg 14844, lines to 15, What is the expected OH concentration in the chamber. This is a critical component in the chamber influencing both the gas and possibly the heterogeneous chemistry and was likely predicted by the chemical model. Comparison of expected chamber OH to typical ambient levels could also be discussed.

Pg 14846 lines 20–23; and pg 14847 line 1 It is stated that aerosol is produced in the chamber via vaporizing dilute salt or salt/organic solutions. Can this be right? Is nebulizing what is meant (that is what is shown in Fig 1).

pg 14847 line 15, what is the $\frac{dN}{dt}$;

pg 14847 line 19, does this refer to measurement of CHOCHO in the chamber.

Fig 1. The figure needs clarification. Provide labels for vacuum pumps (at least I believe

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that is what it is). Also the arrows representing DMA flow directions don't seem to make sense.

Since aerosol mass (LWC + solute) is determined via the DMA, and this is a crucial quantity in the calculated yields, some information should be given on the SMPS operation, including: 1. Are losses in the DMA and related sample lines considered 2. Was the RH monitored in the DMA to make sure particle sizes in the column did not change relative to the chamber due to heating or cooling (ie, water loss/uptake). 3. How were the DMA results inverted (eg, was multiple charging considered) 4. What is the uncertainty from these factors

One might argue that Fig 2 would be better if $dV/d\ln D_p$ were plotted instead of number concentration. Or the volumes associated with these distributions could be given on the number distribution plot.

Table 1. Since the composition of the aerosol formed in the chamber was not measured how sure are the authors that only CHOCHO contributed. in one form or another, to increase in aerosol mass (ie, SOA). Could other C₂H₂ oxidation products be involved in some manner?

pg 14850 line 11 How were yields determined? It is stated that SOA mass is determined from the SPMS, but how was the mass of Delta(ROG) determined (the mass of C₂H₂ reacted). Also, in Fig 3, how were the various masses of organics in the particles determined (Mo). How was uncertainty in yield determined?

pg 14850 line 11, give r^2 or some other proof of an excellent correlation;. Also, it is stated that the Y vs LWC is linear but no proof (eg regression result) is supplied.

Pg 14852, lines 5 through 10. A brief discussion providing more details on the light vs dark experiments would be helpful. It would be worthwhile including a statement that, according to my understanding, the major difference is the light experiments produce

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gas phase oxidants that may then participate in liquid phase reactions, whereas there are no oxidants produced in the dark expts. so no oxidation chemistry is expected in the liquid phase under dark experimental conditions (I may have this wrong, but more details of this type would make the paper clearer).

Pg 14852, line 20, how is [Gly]t determined?

Figure 4. The explanation for panel b is not clear. There is no dashed line, as stated.

Pg 14853 line 22. The line that ends in CHOCHO is a building block for the observed SOA formation. I assume is based on the assumption or knowledge (which is it) that only CHOCHO , water vapor and seed particles were in the chamber during dark experiments.

Generally, I have a hard time understanding Fig 4 and Section 3.3. From Fig 4 it looks like when the lights are turned on, V_{norm} jumps to some higher value (type A in Fig4b) then follows a slower growth rate (type B). But why is this, if the lights remained on why does the very high slope of type A (looks more like a step function) suddenly switch to type B. Or are the lights only on for a very short duration, as appears to be the case in Fig 5. If so, why not leave the lights on for a much longer period of time. Is the problem here that the precursor vapours are rapidly consumed? This section should be clarified by providing more details on the experiments.

Page 14858 line7 and 8, Strictly, I don't see any direct proof for the statement that CHOCHO uptake is actually to the seed (AS or SucA), thus forming some compound containing both the seed and some form of the CHOCHO ; could these seeds play some other indirect role (apart from liquid water formation as noted) other than direct chemical reaction with CHOCHO or its various aqueous forms.

Page 14868, line 21, more explanation should be provided setting up the issue of double counting, the intent of this section is not clear from the first few lines.

Finally, there is a question on the use of a Henry's law constant to describe

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the experimental results. Maybe it should be called something else. Strictly speaking Henry's law describes the concentration of a specific chemical species in the aqueous phase relative to the gas phase, and the effective Henry's constant includes various aqueous forms of the original species. In this case Henry's law constants are given for CHOCHO based on experimental results, where, I presume, the aqueous phase concentration comes from the non-chemically specific SMPS mass measurement. Isn't it possible that other C₂H₂ oxidation products, other than CHOCHO, also contributed to the observed mass. Thus, the Henry's law constants given may depend on, for example, how the C₂H₂ was oxidized, hence on the specific experiments performed.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 14841, 2008.

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