

Interactive
Comment

Interactive comment on “Simulation of aromatic SOA formation using the lumping model integrated with explicit gas-phase kinetic mechanisms and aerosol-phase reactions” by Y. Im et al.

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We thank reviewer 1 for valuable comments.

1) The amount of organosulfates is calculated based on the difference of H^+ concentration derived by two methods, namely PILS measurements in combination with E-AIM thermodynamic calculations and spectrometric data (C-RUV). It is well known that in aqueous aerosol phase, organic acids can be formed which would also

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change the pH value. Thus, the estimate of organosulfates is not a comprehensive method and might be heavily biased by organic acid formation.

Response:

Organic acids are much weaker acids (high pK_a) compared to inorganic acids (e.g., H_2SO_4). For example the pK_a values of most carboxylic acids are between 2.0–5.0. Thus, their pK_a are at least 10^5 times smaller than pK_a of H_2SO_4 (–3.0). In addition, metanil yellow used in the C-RUV method is an indicator for very strong acids. Thus, the influence of organic acids on the C-RUV measurement and the estimation of organosulfates in our study is negligible.

2) Is assumed that all organics with aldehyde, epoxide and alcohol groups readily form organic sulfates? What is the basis for this assumption? It is known that such compounds undergo many more reactions in the aqueous phase than sulfate formation.

Response:

Not all organic compounds containing aldehyde, epoxide and alcohol groups immediately produce organosulfate (OS). Within each time step, only some of these compounds will produce OS. In the UNIPAR model, the OS production is operated using the model equation semi-empirically fit to the measured OS, which is estimated from the difference in $[H^+]$ between two methods (PLS-IC with E-AIM model and C-RUV). The details of OS formation are described in Supplement section S4. In the model, weighing factors were implemented for functionalities, which are directly related to reactivity of lumping groups. For example, the weighting factor of the fast reactivity group (two aldehydes, two epoxides or one aldehyde and one epoxide) is two times higher than that of the medium reactivity group (one aldehyde or one epoxide). The fast reactivity group has a weighting factor of 4, and medium reactivity group has a weighting factor of 2 in the model. For the multi-alcohol [MA] groups, the least volatile group ($i=1$) has a weighting factor of 4, and the second and third less volatile groups ($i=2$ and 3) use a weighting factor of 3.

3) It is assumed that acid-catalyzed reactions do not occur if the relative humidity (RH) is below the efflorescence RH (ERH) of ammonium sulfate since then particles are dry and do not exhibit an aqueous phase. There are several studies that have shown that ERH of mixed organic/sulfate particles is much smaller than that of ammonium sulfate alone (e.g., (Marcolli et al., 2004; Bertram et al., 2011)). How does this effect affect the results?

Response:

In the recent study by Bertram et al. (Bertram et al., 2011), the ERH of organic/ammonium sulfate particle and liquid-liquid phase separation (LLPS) under varying humidity have been parameterized as a function of elemental O:C ratio and organic:sulfate ratio. Their study reported that the ERH of inorganic salt (ammonium sulfate) is not changed due to the presence of SOA products when the O:C ratio is less than 0.7. In the recent chamber study, Nakao et al. (2011) measured O:C ratios of various aromatic SOAs. In their study, O:C ratios of toluene SOA were up to 0.65 and those of m-xylene were up to 0.45. The reported O:C ratios of aromatic SOA are generally less than 0.7 and support our assumption of phase separation (three phase: g, or, and in) in the aromatic SOA internally mixed with inorganic salt (ammonium sulfate). (Section 3.3)

In this study, the O:C ratio of SOA were not measured. In response to the reviewer's comment, we explored the estimation of O:C ratio using the product compositions predicted from the UNIPAR model. First, the O:C ratios of each lumping group was calculated based on molecular structures and then normalized with the organic mass (OM_i) of each lumping groups. Studies (Hu et al., 2011; Liu et al., 2012) have shown that organonitrates are unstable and decomposed to alcohol and NO_2 . Thus, aerosol-phase organonitrates were treated as alcohols by subtracting two oxygen atoms. Furthermore, when oligomers form through aldol condensation and esterification, one mole of water can be formed (Chen et al., 2011). The estimated O:C ratios of toluene SOA and 135-TMB SOA were around 0.62 and 0.37. Interestingly, the estimated O:C

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ratio of toluene SOA is close to the experimental O:C ratio (<0.7) reported by Nakao et al. (2011). Because the O:C ratio is smaller than 0.7, the ERH of aerosol is mainly controlled by the inorganic aerosol with minimal modification due to the presence of aromatic SOA.

4) Related to the previous comment, several studies (e.g., (Bertram et al., 2011)) have shown that the O/C ratio determines in which phase compounds will partition. What is the O/C ratio of the SOA constituents? Would a different phase partitioning impact the results?

Response:

The predicted elemental O:C ratio of SOA from the photooxidation of toluene and 135-TMB were 0.62 and 0.37. Under our O:C ratios, the liquid-liquid phase separation (LLPS) predicted using the empirical relationship derived by Bertram et al.(2011), is 65% for toluene SOA and 90% for 135-TMB SOA. In our study, the chamber experiments for both toluene SOA and 135-TMB SOA were conducted under the humidity conditions lower than 65%, except a short period at the beginning of each experiments (less than 1 hour). The amount of SOA formation for the first 1 hour was negligible due to the high zenith angle (low Sun). Hence, our assumption for phase separation can be justified for both toluene SOA and 135-TMB SOA.

5) Much progress in SOA modeling has been made since 2001. It is not clear why much more sophisticated SOA partitioning models that cover many more volatility bins (e.g. (Donahue et al., 2006; Donahue et al., 2011)) are ignored and instead the module by Schell et al. (2001) is used.

Response:

The aerosol partitioning module derived by Schell et al. (2001) is the numerical approach that calculates partitioning of semivolatile organic compounds between the gas and particle phases. In terms of partitioning calculation, there is no difference between the model by Schell et al. and the volatility basis set (VBS) model by

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Donahue (2006). The VBS model facilitates a different mathematical framework to predict partitioning OM with more volatility bins. Unlike the original Schell's model, the OM_P module used in the UNIPAR model implements 6 bins for partitioning instead of the two products to provide better lumping for volatility.

6) I cannot believe that OM/OC has not been determined more accurately in studies more recent than 1990 (p. 5847, l. 27). I also expect that this ratio is a function of time, when products become more oxidized. What is the extent to uncertainty that is introduced by using this constant value?

Response:

So far, there is no good method to predict dynamic OM/OC ratio as a function of time. It is expected that OM/OC ratio would increase as products are more oxidized in both the gas and aerosol phase reactions. The higher OM/OC ratio would lead to higher prediction of OM. In this study, we have fixed OM/OC ratio at 2.0 based on the study by Kleindienst et al. (2007) as shown in the 2nd paragraph of the section "2. Experimental Section".

7) The fact that an artificial OH source is needed and the great discrepancy between measured and predicted $NO_2 + HNO_2$ for the 1,3,5-trimethylbenzene experiments (Fig S2c and d) suggest that something fundamentally is wrong in the model. Can you speculate on missing processes in the model or inappropriate reaction parameters?

Response:

The slow production of OH radicals from oxidation of aromatics in the MCM has been reported by MCM developers and other researchers. According to previous studies (Wagner et al., 2003; Johnson et al., 2004; Bloss et al., 2005), the kinetic mechanisms of aromatic hydrocarbons in MCM overestimate ozone peaks and underestimate precursor decay due to the low production of OH radicals. Hence, an artificial OH radical was added to the aromatic HC mechanism in this study. The artificial OH

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radical production rates were determined by comparing the simulated toluene or 135-TMB decay to the measured data. Several possible explanations for the addition of artificial OH radicals were suggested in the recent study by Bloss et al. (2005): for example, the uncertainty in yields of the reactive oxygenated intermediates produced via degradation of aromatic products, unknown photolysis rates of organic compounds, and missing pathways for OH radical generation in MCM. Moreover, chemistry of a variety of peroxy radicals (RO_2) in MCM was oversimplified using surrogate coefficients instead of explicit mechanisms. Such simplified RO_2 chemistry can cause the discrepancy between the measured and the predicted concentrations of OH radicals. (See the first paragraph of Section 4.1 Gas-phase model simulation vs. Experimental data)

Minor comments

8) p. 5845, l. 9: Why is wide range of volatilities a limitation?

Response:

In the Odum's two product SOA model, SOA growth is predicted using two surrogate products associated with several parameters (two partitioning coefficients and two stoichiometric coefficients), which are semi-empirically determined by fitting to indoor chamber experiments. However, SOA growth is much more complex integrating partitioning and aerosol phase reactions of thousand chemical species. Thus, the traditional two product model is limited to represent a wide range of volatilities originating from different semivolatile products produced from multi-generation oxidation processes of hydrocarbons in the gas and the particle phases.

9) p. 5846, l. 14: In order to balance the discussion of acid-catalyzed reactions, it would be fair to add some references that state that acid-catalyzed reactions are likely not a major contributor to SOA (e.g., (Casale et al., 2007; Peltier et al., 2007; Minerath and Elrod, 2009))

Response:

The acid effect on SOA formation has been discussed in Introduction section of the revised manuscript. (2nd paragraph of Introduction section)

In general, laboratory studies have shown increases in SOA yields due to aerosol acidity. However, there have been discrepancies among field studies for the effect of aerosol acidity on SOA production (Zhang et al., 2007; Peltier et al., 2007). The effect of acidity on organic aerosol growth is complex due to compounding effects originating from various aerosol phase reactions. For example, OS is much more hydrophobic than H_2SO_4 or $(NH_4)_2SO_4$ and tends to belong to organic phase. As OS forms, the effect of acid-catalyzed reactions on SOA yields becomes less apparent due to the consumption of sulfuric acid and the reduction of the amount of water in aerosol (see Section 4.4 Effect of inorganic aerosol acidity on aromatic SOA formation). When humidity is high, the effect of acid-catalyzed reactions on SOA yields also becomes unclear due to SOA formation via the aerosol phase reactions in aqueous phase (see Section 4.4 sensitivity test).

10) p. 5846, l. 25: What is meant by ‘gas phase ageing’? Oxidation processes in the gas phase often lead to fragmentation of the carbon backbone which results in more volatile products. Is this what is meant here?

Response:

Gas phase ageing means the revolution of oxygenated products by photochemical reactions in the gas phase. Further reactions of the oxygenated products can lead to less volatile products through the addition of oxygen or nitrogen functional groups as well as more volatile products through fragmentation of the carbon backbone. To clarify this, we have changed from ‘gas-phase aging of organic compounds’ to ‘multi-generation oxidation of organic compounds in the gas phase’ in the revised manuscript.

11) p. 5852, l. 1: I think it should be either ‘self-reaction’ or ‘dimerization’. Do you assume that the acid-catalyzed reaction in the inorganic phase is also a dimerization?

Response:

“self-dimerization” is replaced with “dimerization” in the revised manuscript. The OM formation by acid-catalyzed reactions in the UNIPAR model is referring to oligomerization. The kinetic equation of acid-catalyzed oligomerization was expressed using the form of dimerization reactions.

12) p. 5853, l. 1: Do you assume that the formed OM mass is equivalent to the consumed VOC mass? How about incorporation of oxygen or sulfate into the molecules? Shouldn't that change the product mass?

Response:

OM mass formed by aerosol phase reaction (OM_{AR}) is equal to the amount of consumed oxygenated products (Not precursor VOC) that are predicted from the explicit gas kinetic model (MCM). So, mass of oxygen or nitrogen are already incorporated into oxygenated products. Sulfate containing molecules are considered by the OS formation module (Section. 4.3 Organosulfate formation).

13) p. 5853, l. 8: How are the densities calculated? Do the change over time?

Response:

In this model, the density of organic phase (ρ_{OM}) was assumed as a constant (as 1.4 g/cm^3) based on the previously reported studies (Ng et al., 2007;Sato et al., 2007;Nakao et al., 2011) for aromatic SOA. The density of inorganic phase (ρ_{in}) was dynamically calculated using the E-AIM model as a function of RH and numerical indicator for inorganic compositions related to aerosol acidity ($F_s = [SO_4]/(SO_4 + NH_4)$). For each chamber experiment, RH, SO_4^{2-} , NH_4^+ were measured as a function of time and input into the model calculation. This has been discussed at the end of the section “3. Product concentrations among the gas, organic and inorganic aerosol phases”.

14) p. 5856, l. 8: Sulfate is likely mostly formed in the aqueous phase either by S(IV) oxidation by O_3 or by H_2O_2 . Even though you did not add H_2O_2 to the reaction

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mixture it is likely that it is formed in either the gas or aqueous phase by recombination of HO_2 that originates from OH + organic reactions.

Response:

The prediction of sulfate formed from SO_2 oxidation both in the gas and the particle phase is difficult due to the lack of a model that can predict the effect of either the chamber wall on SO_2 oxidation or the oxidation on the surface of aerosols. Instead, the sulfate concentration was measured using PILS-IC and applied to the UNIPAR model.

15) p. 5856, l. 19: What are the yields in the other cited studies?

Response:

Ng et al.(2007), toluene SOA yield under no NO_X condition (H_2O_2 only) with/without H_2SO_4 are 29.5%, 28.7%, and SOA yield under high NO_X condition with/without H_2SO_4 are 19.3%, 17.4%. In the recent study by Cao and Jang (2010), toluene SOA yields under no NO_X (H_2O_2 only) condition were 20-28% with non-acidic ammonium sulfate (AS), and 25-38% with acidic sulfate seed; SOA yield under low NO_X were 13-18% with AS seed and 26-28% with acidic sulfate seed; and the SOA yield under high NO_X condition were 12-13% with both AS seed and acidic sulfate seed.

Various factors associated with temperature, RH and sunlight can influence SOA yields. SOA yields can also be different depending upon types of chambers. Overall, aromatic SOA yields of this study are relatively low compared to those from other studies (Cao and Jang, 2010;Ng et al., 2007), although similar in the trend of NO_X effects. The lower SOA yields are often observed in outdoor chambers than those using indoor chamber (Johnson et al., 2004). The temperature and sunlight for outdoor chambers have a diurnal pattern (Supplement, Fig. S8). In general, the light intensity of ambient sunlight facilitated in outdoor chambers is much stronger than indoor chambers. Johnson et al. (2004) also reported low SOA yields using the outdoor chamber. For example, toluene SOA yields under low NO_X were 4.63-5.14% and the SOA yields under high NO_X were 2.05-3.57%. (see at the end of section “4.2 Effect

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of NO_X on aromatic SOA form”)

16) p. 5856, l. 23: What is meant by ‘hypersensitivity’?

Response:

We have changed from “hypersensitivity to temperature” to “high-sensitivity to temperature”.

17) p. 5857, l. 7: Given that the prediction of organosulfate formation is an essential part of your study, more detailed discussion should be given on the predicted trends and apparent contradiction to previous results? Is the study by Wyche et al. (2008) the only one that compared SOA formation with/without SO_2 ?

Response:

There have been only few studies for the effect of NO_X and inorganic seed on the formation of 135-TMB SOA. Studies of Metzger et al. (2008) and Rickard et al. (2010) showed the similar result in NO_X trend with the study by Wyche et al. (2008). However, the investigation of the effect of SO_2 on 135-TMB SOA formation appeared only in the study of Wyche et al. (2008). This has been discussed in section 4.2.

18) p. 5858, l. 24: If the organosulfates are indeed hydrophobic, why aren't they predicted to go into the organic phase? Thus, they would not change the hygroscopicity of the aqueous phase.

Response:

In the model, the organosulfates formed in the inorganic phase are considered to go into the organic phase (OS is accounted as a part of OM_{AR}). However, OS formation still can influence the hygroscopicity of inorganic aerosol because of the consumption of sulfate ion (SO_4^{2-}). When sulfate ions are consumed due to OS formation, acidity of inorganic aerosol decreases and the hygroscopicity of aerosol also declines. As shown in section 4.4, the hygroscopicity of the NH_4^+/SO_4^{2-} /water system is lower under neutral conditions (Ammonium sulfate) than in the acidic aerosol (SA).

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19) Figure 1: More information is needed in the caption so the figure can be understood by itself. e.g., what does F, M, P, MA mean?

Response:

The caption of Fig. 1 was revised.

“Fig. 1. The conceptual structure of the UNIPAR model. $\alpha_{i,j}$ is the mass based stoichiometric coefficient. The reactivity F, M, S, P and MA indicate fast, medium, slow, non-reactivity and multi-alcohol lumping group respectively. C and K represent the concentration of lumping species and the partitioning coefficient, respectively. Subscripts “g”, “or”, and “in” denote gas, organic, and inorganic aerosol phases, respectively. OM_P , and OM_{AR} are the concentrations of organic matter produced by partitioning and aerosol phase reactions, respectively. M_{in} is the concentration of inorganic aerosol.”

Technical comments

20) p. 5846, l. 11: ‘aerosol chemistries’ should be replaced by ‘chemical reactions in the aerosol phase’

Response:

“aerosol chemistries’ is replaced by ‘chemical reactions in the aerosol phase”

21) p. 5855, l. 1: This sentence is not clear.

Response:

The sentence has been revised from

“The estimations of $C_{g,i}$ and $C_{or,i}$ by Eqs. (4)–(6) are approached those by the OM_P module and since $K_{in,i}M_{in}$ in the denominators of Eqs. (4)–(6) is relatively very small compared to $K_{or,i}OM_T$.”

to

“ $C_{in,i}$ is relatively very small compared to $C_{g,i}$ and $C_{or,i}$. The term $K_{in,i}M_{in}$ is also very

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small compared to the term $K_{or,i}OM_T$ in Eqs. (4)–(6). Hence, the estimations of $C_{g,i}$ and $C_{or,i}$ are approached by the OM_P module at the end of each time step excluding $C_{in,i}$ (Fig. 2),”

22) p. 5857, l. 21: The figures should be discussed in numerical order. Fig. 5 is mentioned before Fig. 4.

Response:

The order of Figures 4 and 5 has changed.

23) p. 5858, l. 10: Do you mean SOA from 135-TMB?

Response:

“135-TMB” is replaced with “135-TMB SOA” in the revised manuscript.

24) Figure S2: The two different shades of blue (O_3 and $NO_2 + HONO$) are hard to distinguish.

Response:

The color of $NO_2 + HONO$ has been changed to magenta

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