Dear ACP Editor:

As requested by the editor Prof. Dara Salcedo, here we corrected our manuscript for a possible publication in ACP. We greatly appreciate the time and effort that editor and reviewers spent in reviewing our manuscript. Their comments are really thoughtful and very helpful for us to improve the quality of our paper. After reading the comments from the reviewers, we have carefully revised our manuscript. All the changes in the manuscript are marked in blue color. Our responses to the comments are itemized below.

Anything for our paper, please feel free to contact me via ghwang@geo.ecnu.edu.cn.

All the best

Gehui Wang

Nov.4, 2024

Reviewer(s)' Comments to Author:

Reviewer: 1

Comments:

The authors have done an excellent job responding to reviewer comments thoroughly, and their revisions have certainly improved the manuscript. I have a few minor concerns detailed below, primarily about the wording and level of detail used in newly added sections, which aren't always sufficiently comprehensive. Further, the added analysis has revealed one particularly major concern which needs to be addresses regarding the importance of photolysis.

Response: We thank the comments and carefully revised our manuscript. Our responses to the comments are itemized as follows.

Comments:

1. The authors should clearly state according to the OBM-MCM model what fraction

of acetone is photolyzed vs. reacted with OH in these experiments. The fact that modeled OH is "two orders of magnitude lower than the values calculated by the loss rate of acetone" would suggest to me that photolysis is two orders of magnitude faster than OH reaction in here -- so does that mean these chamber experiments consisted of ~99% acetone photolysis and ~1% acetone + OH? If so, that needs to be very clearly stated throughout, because the reactive pathways following acetone photolysis (a minor though not entirely negligible pathway in the atmosphere) are not at all the same as those following reaction with OH. That would mean that this study effectively isolates the SOA formation potential from acetone photolysis in low-NOx environments -- which is scientifically interesting and worthy of publication, but is far from being an accurate representation of how most acetone reacts in the atmosphere. More time should be spent in the introduction and discussion sections on previous experimental efforts related to acetone photolysis (both SOA formation and gas-phase mechanism), what's included in models currently for acetone photolysis, and how the results here compare with both of those.

Response: Suggestion taken. (1) According to the OBM-MCM results, the reaction rates of photolysis and OH oxidation are 3.66×10^6 and 1.32×10^7 molecules cm^{-3} s^{-1} respectively in this work. This suggests that OH oxidation plays a more significant role than the photolysis of acetone. Figure R1 displays the time series of three main RO_2 concentrations during the experiments. Initially, CH_3O_2 and CH_3CO_3 are predominantly formed from the photolysis of acetone, and $CH_3COCH_2O_2$ is generated from oxidation by OH radicals (Ge et al., 2017). Meantime, both CH_3O_2 and CH_3CO_2 can be produced within the $CH_3COCH_2O_2$ chemistry, resulting in their higher concentrations compared to $CH_3COCH_2O_2$ and a consistent increase in concentrations throughout the experiments (Figure R1).

(2) During the photolysis of acetone, products such as organic hydroperoxides and formaldehyde can promote the formation of OH and HO_2 radicals effectively. This indicates that certain gaseous products can be generated through both photolysis and OH reaction pathways of acetone. Therefore, it is difficult to effectively isolate the SOA

formation potential from acetone photolysis. In this study, we use photochemical reactions of acetone to cover photolysis and OH reaction of acetone. Moreover, our primary focus is on the heterogeneous formation mechanism of SOA in the aqueous phase and the impact of seed physicochemical properties on SOA formation. As a result, gas-phase reactions are presented in a simplified manner.

(3) Photolysis and OH oxidation are main sinks of acetone in atmosphere, with photolysis contributing 45% of the sink, OH oxidation 30%, ocean uptake and dry deposition to land 25% (Jacob et al., 2002). The photolysis of acetone leads to the production of hydrogen oxide radicals. Hence, in current models, the photolysis of acetone is used in the global chemical transport model (GEOS-Chem) and the community atmosphere model with chemistry (CAM-chem) to estimate the hydrogen oxide radical production in the upper troposphere and lower stratosphere, respectively (Wang et al., 2020; Arnold et al., 2004). However, the role of acetone in SOA heterogeneous formation remains unclear.

We have added the information related to photolysis into the introduction and discussion sections of the revised manuscript. This can be found on page 4 lines 84-90 and page 14 lines 382-391.
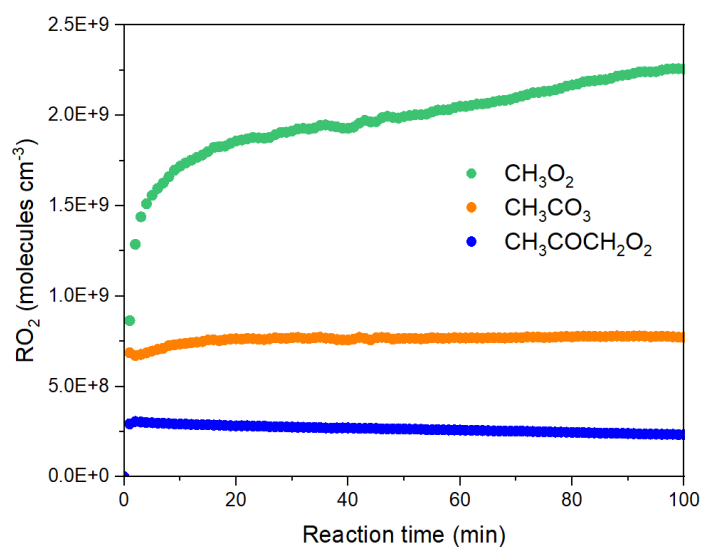
Figure R1 The times series of three main RO_2 concentrations during Phase I.

Comments:

2. 90-91: "MGly is an important product of acetone photochemical reactions with 14% molar yield" where does the 14% come from? Surely that's different under different conditions (RO_2 fate, photolysis vs. OH reaction)? Or is it (actually or expected to be) 14% in these specific experimental conditions?

Response: The 14% molar yield represents the global annual mean molar yield of MGly produced formed from acetone, as calculated by GEOS-Chem (Fu et al., 2008). This yield includes all formation pathways of MGly formed from acetone, including photolysis and OH oxidation. The information has been added on page 4 lines 97-98 in the revised manuscript.

Comments:

3. 94-96 "Currently, estimation of acetone-derived SOA by chemical transport models only consider its product MGly as the SOA precursor (Fu et al., 2008)." Are there any more recent examples? Models have been thoroughly updated since 2008, and surely some include more comprehensive SOA formation pathways, so perhaps there are counter-examples where people do include some other route? I'm thinking in particular of Dovrou et al.'s analysis of the potential contribution from formaldehyde (DOI: 10.1073/pnas.2113265119), but perhaps there are others.

Response: Suggestion taken. We have searched for the most recent articles related to model studies. In chemical transport models, the SOA module typically comprises two components: firstly, the gas-particle equilibrium partitioning of all nonradical organic compounds followed by oligomerization in the organic phase; and secondly, the formation of aqueous aerosols from dicarbonyls and isoprene epoxides (Huang et al., 2024b; He et al., 2024; Huang et al., 2024a). Zheng et al. (2023) simulated the organic aerosol concentrations from 2005-2019 with Community Multiscale Air Quality (CMAQ, version 5.3.2) coupled with a Two-Dimensional Volatility Basis Set (2D-VBS) module. The 2D-VBS organic aerosol framework lumps all organic compounds into O/C and volatility bins, taking into account the multigenerational aging of these organic

compounds across various volatility bins. The VBS module mainly focuses on the homogeneous formation of SOA. The SOA from heterogeneous reaction primarily includes the uptake of isoprene epoxydiol, glyoxal, methylglyoxal, hydroxymethyl-methyl-a-lactone, 2-methylglyceric acid and 2-methyltetrols. Huang et al. (2024a) updated the Comprehensive Air Quality Model by considering primary organic aerosol evaporation and secondary organic aerosol formation from VOCs, IVOCs, and SVOCs. However, their model only considered the heterogeneous uptake of glyoxal and methylglyoxal on aerosol surface and the production of non-volatile product from the hydrolysis of organic nitrates in the aerosol phase. Therefore, it is clear that the SOA module in current chemical transport models primarily encompasses the homogeneous reactions of various volatility VOCs and heterogeneous reactions of specific compounds on the aerosol surface. Notably, these models do not account for SOA formation from the heterogeneous photochemical reactions of acetone on aerosols. We have added this information into the revised manuscript, page 5, lines 101-107.

Comments:

4. 405-406 "especially in polluted urban regions, which are often characteristic of high loadings of acetone and aerosols" -- these regions would have predominantly $RO_2 +$ NO chemistry, which was not studied here. This mismatch should at least be acknowledged here, and it would also be worth mentioning in the abstract what general conditions were probed in these experiments (e.g.: "low-NO, photolysis dominated oxidation of acetone")

Response: We agree with your comments totally and have taken your suggestion. Indeed, "the polluted urban regions" is mismatch with the experimental conditions in this work due to the different NOx levels. We have revised the sentence "suggesting that only considering the role of MGly will inevitably underestimate the contribution of acetone to SOA production in the atmosphere especially in polluted urban regions, which are often characteristic of high loadings of acetone and aerosols." to "suggesting that only considering the role of MGly will inevitably underestimate the contribution of acetone to SOA production in continental atmosphere, which is often characteristic

of high loadings of acetone and aerosols." This modification can be found on page 16, lines 438-440 of the revised manuscript. And we have mentioned that this study was carried out under low NOx condition in the abstract of the revised manuscript (page 2, line 24).

Comments:

5. Fig 1: do the authors gain any insight from these experiments as to why the SOA plateaus so early, even though acetone continues to oxidize at a fairly constant rate? Does this tell us anything about either the self-limiting nature of acetone SOA formation, or the rates of oxidative loss of the acetone-derived aerosol, or both?

Response: We hypothesize that the observed plateaus in SOA may correlate with an increase in acidity within the aerosol phase during the reaction. We calculated the pH values of particles at the beginning and end of the Phase I (Figure 1) using the E-AIM model IV. At the beginning of the Phase I, the pH of particles is 4.89. When the UV lights are turn on, abundant gas-phase intermediate products containing hydrophilic functional groups such as alcohol, ketone, and organic acids are formed from acetone photochemical reactions. These products can dissolve into the aqueous phase and undergo further oxidation, esterification reaction and radical-radical reactions to form SOA on particles (Poulain et al., 2010; Ge et al., 2017). These reactions contribute to the rapid formation of SOA on particles within a reaction time of less than 30 minutes during Phase I.

The uptake of organic acid products and the reaction of carbonyl compounds with the $(NH_4)_2SO_4$ seeds can significantly lower the pH of particles due to the formation of NH_4HSO_4. This conclusion is supported by the decrease in the molar ratio of $[NH_4^+]/[SO_4^{2-}]$ and the increase of fraction of CO_2^+ species in aerosols. The pH value of particles reduced rapidly from the initial 4.89 to 1.77 after a 30 min reaction. This increased acidity can hinder the gas-particle partitioning of gas-phase reaction products such as formic, acetic acids, and methylglyoxal (Lv et al., 2022; Zhao et al., 2006), as most weak acids are unable to dissociate at pH<2 (Tilgner et al., 2021). Therefore, SOA formation by the partitioning process sharply decreased after the 30 min reaction time,

resulting a slow increase of the SOA concentration. Subsequently, the SOA is primarily formed through aqueous reactions on aerosols, leading to a persistent increase in the O/C, N/C ratios and oxidation state of SOA. The above information has been added into the revised manuscript, page 9, lines 218-228.

Comments:

6. Details of the OBM-MCM need to be clearly stated somewhere (probably the SI) -- are the observational constraints imposed only at the beginning or throughout? Was the full MCM mechanism used, or a subset, and what was used for the light intensity? At what time during the experiment were the OH radical concentrations calculated to be $5.89e^6$ molec/cm^3 -- right when the lights were turned on? Did they change much over the course of the experiment? If the model calculated HO2 and RO2 concentrations throughout the experiment, some discussion of these would also be useful.

Response: Thanks for your suggestion. We have enhanced the description of the OBM-MCM in the manuscript. The observation levels of acetone, acetaldehyde, formic acid and acetic acid throughout the photochemical reactions, along with meteorological parameters (temperature and relative humidity) and the initial H_2O_2 concentration, were incorporated into the OBM-MCM model as constraints. The comprehensive MCM mechanisms related to the photochemical reactions of acetone and other VOCs observed in this work were also incorporated into the OBM-MCM. The photolysis rate for H_2O_2 is 9.1×10^{-6} s^{-1}. The average concentration of OH radicals during the reactions is 5.89×10^6 molecules cm^{-3}. The time series of OH and HO$_2$ radical concentrations are shown in Figure R2. In the experiments, OH radicals are formed from the photolysis of H_2O_2 and organic hydroperoxides. HO$_2$ radicals are formed from the reaction between H_2O_2 and OH, as well as the photochemical reactions of aldehydes (photolysis and OH oxidation). Aldehydes are formed from the oxidation of organic hydroperoxides by OH radicals. Concurrently, HO$_2$ can interact with another to yield H_2O_2 (Seinfeld and Pandis, 2006). In this work, in addition to the circulation of H_2O_2-OH-HO$_2$, organic hydroperoxides and aldehydes, which are formed from the photolysis of acetone, also contribute to the formation of OH and HO$_2$. Therefore, the concentrations of OH and

HO$_2$ consistently increase throughout the reaction. The details of the OBM-MCM have been added to the revised manuscript, page 8, lines 189-197.

Figure R1 displays the time series of three main RO$_2$ concentrations during the experiments. Initially, CH$_3$O$_2$ and CH$_3$CO$_3$ are primarily formed from the photolysis of acetone, and CH$_3$COCH$_2$O$_2$ is generated from oxidation by OH radicals (Ge et al., 2017). Meantime, both CH$_3$O$_2$ and CH$_3$CO$_2$ can be produced within the CH$_3$COCH$_2$O$_2$ chemistry, resulting in their higher concentrations compared to CH$_3$COCH$_2$O$_2$ and an increase in concentrations throughout the experiments. The information can be found on page 14-15 lines 385-391.
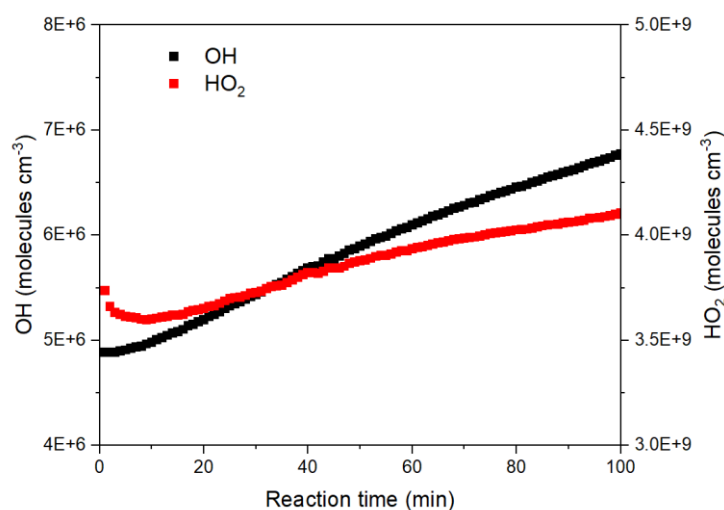
Figure R2 The times series of OH and HO$_2$ radical concentrations during Phase I.

Reviewer: 2

Comments:

Acetone is abundant in the urban atmosphere. However, less attention has been paid to its contribution to secondary organic aerosol and brown carbon formation in the presence of ammonia. Zhang et al. investigated the SOA and BrC formation for the oxidation of acetone in the presence of different seeds and ammonia. They found that the uptake of other products besides methylglyoxal is important for SOA formation and aerosol acidity is vital for BrC formation via a formation of NOC. Overall, the results are interesting and the manuscript is well organized. It is publishable after a minor revision.

Response: We thank the above comments and have carefully revised our manuscript. Following is our response to the comments.

Comments:

1. The reaction conditions including concentrations of pollutants should be shown in a table.

Response: The reaction conditions including concentrations of pollutants are shown in Table R1 (refer to the revised supporting information, Table S1). And the associated description in the manuscript can be found on page 6, line 149.

Table R1. The reaction conditions of the experiments

Exp. No.	Acetone (ppb)	OH^a (molecules cm^{-3})	NH_3^b (ppb)	SO_2 (ppb)	Seeds	RH (%)
1	750	5.89×10^6	73	/	$(NH_4)_2SO_4$	85±1
2	750	5.89×10^6	73	/	$(NH_4)HSO_4$	85±1
3	750	5.89×10^6	73	/	Na_2SO_4	85±1
4	750	5.89×10^6	73	500	Na_2SO_4	85±1
5	750	5.89×10^6	73	/	/	85±1

[a] The concentrations of OH radicals in all experiments are average concentrations during Phase I. These concentrations are determined using an OBM-MCM.

[b] The concentrations of NH_3 are wall-loss corrected average concentrations during Phase II, without considering the reaction process.

Comments:

2. The concentration of NH_3 is according to the wall loss rate and assumed initial concentration. This may lead to great uncertainty because NH_3 is a sticky compound. Thus, a long time is usually required to establish a steady state.

Response: The concentrations of NH_3 in Table S1 represents the average concentration during Phase II (Figure 1), without considering the reaction process. These values are calculated using the first-order wall-loss corrected method reported by Li et al. (2021a), which has been widely adopted in chamber studies (Li et al., 2021a; Li et al., 2021b; Zhang et al., 2024). To provide clarity, we have included a supplement note in Table S1: "[b] The concentrations of NH_3 are wall-loss corrected average concentrations during Phase II, without considering the reaction process."

NH_3 is a sticky compound and requires a long time to establish a steady state after introduced into chamber. In this work, NH_3 was introduced into the chamber along with

a N_2 flow using a glass syringe. This approach effectively reduces the time required for uniform diffusion of NH_3 in the chamber. The reaction lasted for 45 min to ensure complete reactions in chamber (as shown in Figure 1). Therefore, the potential impact of NH_3 diffusion on the results can be considered negligible.

Comments:

3. In the chamber, several tens ppb NH_3 might exist in the first experimental phase even though you do not add any NH_3. The influence of trace NH_3 in Phase I on SOA and BrC formation should be discussed.

Response: Thanks for your suggestions. It is possible that trace amounts of NH_3 may exist in Phase I, even though no NH_3 is intentionally introduced. These trace amounts of NH_3 could potentially facilitate the gas-particle partitioning of gaseous organic acids, thereby promoting the formation of SOA and BrC on aerosols. The influence of trance amounts NH_3 on SOA and BrC formation appears to be more significant on acidic seeds than on neutral seeds. However, the normalized concentration of SOA on Na_2SO_4 seeds is two times larger than that on $(NH_4)_2SO_4$ and NH_4HSO_4 seeds, respectively, indicating that the impact of trace NH_3 on SOA formation is significantly weaker than that of the inherent properties of the seed. We have analyzed the influence of trace NH_3 on SOA formation on different seeds. In experiments with $(NH_4)_2SO_4$, the decrease in the molar ratio of NH_4^+/SO_4^{2-} suggests that the uptake of trace NH_3 on aerosols is negligible, and its effect on SOA and BrC formation can also be considered negligible. On neutral Na_2SO_4 seeds, based on the gas-to-particle phase partition coefficients of NH_3 ($\epsilon(NH_4^+)$) (Figure 4b), it is difficult for NH_3 to uptake on Na_2SO_4 seeds. The influence of trace NH_3 may be more obvious in NH_4HSO_4 seeds. The SOA formation with NH_4HSO_4 seeds is similar to that with $(NH_4)_2SO_4$ seeds, which is possibly caused by the promotion of residual trace NH_3 in the chamber on the uptake of acidic organics (Figure 3a). This information has been added on page 11, lines 288-290 of the revised manuscript.

Comments:

4. In Figure 4c, BrC at 270 nm of wavelength is still observable in the presence of

Na$_2$SO$_4$ seed instead of invisible as the author described. You can see it is weaker than that over (NH$_4$)$_2$SO$_4$ seed.

Response: Thanks for your suggestion. We have revised the sentence "However, there was no absorption peak at ~270 nm for the products of the Na$_2$SO$_4$ particles in the absence of SO$_2$ (Figure 4c)" to "However, the absorption peak at ~270 nm for the products of the Na$_2$SO$_4$ particles in the absence of SO$_2$ was weaker than that with (NH$_4$)$_2$SO$_4$ seeds significantly (Figure 4c)". This information can be found on page 14, lines 366-368 of the revised manuscript.

Comments:

5. I do not agree with the authors for classifying Na$_2$SO$_4$ as a mineral dust. It should be a neutral inorganic salt.

Response: Na$_2$SO$_4$ is the main component of mirabilite and has been identified as a saline mineral particle during dust storm events (Wu et al., 2019; Wu et al., 2020). As reported by Wu et al., during the dust storms in Shanghai in 2019, Ca^{2+} and Na^{+} showed a linear correlation with SO$_4^{2-}$, particularly under the strong dusty weather conditions (R^2 > 0.80, p < 0.05). This suggests that the dust plumes carried effluent gypsum and mirabilite into Shanghai (Wu et al., 2020). Thus, we classify Na$_2$SO$_4$ as a saline mineral particle to distinguish it from (NH$_4$)$_2$SO$_4$. We have updated the term "mineral dust" to "saline mineral particle" throughout the revised manuscript.

Comments:

6. UV lights with 254 nm wavelength were used in the chamber experiments. It is weird to choose such a short wavelength of UV light which might have an important influence on the photolysis of organics.

Response: Photolysis and OH reaction are the primary atmospheric sinks for acetone. In this work, we selected H$_2$O$_2$ as the OH radical precursor to simulate the oxidation reaction of acetone by OH. The UV absorption cross section of H$_2$O$_2$ is notably higher at shorter wavelengths compared to longer UV wavelengths (Atkinson et al., 2004). The 254 nm wavelength has been widely used in numerous studies to photolyze H$_2$O$_2$

and generate OH radicals (Tan et al., 2009; Raff et al., 2005; Song et al., 2019). Therefore, we used UV lights with 254 nm wavelength to produce OH radicals in our chamber experiments.

In addition, in our previous study, we employed 365 nm UV to photolyze H_2O_2. However, no significant decrease in acetone concentration or SOA formation was observed, suggesting that significant reactions did not occur under the 365 nm UV irradiation. In contrast, a marked reduction in acetone levels was observed under 254 nm UV irradiation. Consequently, this study utilized 254 nm UV lights to explore the photochemical reactions of acetone and the subsequent SOA formation.

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