

We thank reviewers for reviewing our manuscript and also for useful hints and suggestions. Below, comments from the reviewer are given in blue while our answers are given in black, with passages including new text given in red.

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RC3: 'Comment on amt-2023-34', Anonymous Referee #3, 11 May 2023

This study designed and characterized a new smog chamber that can be used to study atmospheric multiphase chemistry. The smog chamber system can achieve precise control of temperature and humidity to generate seed particles at different phases states. Some key parameters for the new chamber, including leak proofness, mixing performance, the wall losses for gas and particle phases etc, were systematically characterized. The new smog chamber system shows its ability to simulate secondary aerosol formation and atmospheric multiphase processes like other chambers. Overall, the manuscript is well organized. Some questions shall be clarified before publication:

**Major comments:**

1) Line 138-140: Have the authors measured the temperature differences in different positions of the chamber? The sensor was set in the bottom now. How about the spatial distribution of the sensor.

**Response:**

We did not measure the spatial distribution of temperature. On the one hand, the internal space of the chamber is quite small, which is not suitable to arrange many temperature sensors. On the other hand, it is unnecessary. The internal design of our chamber enclosure is that, the circulating air for temperature control surrounding the reactor, and the temperature ultimately reaches equilibrium from the outside of Teflon film to the inside. The temperature control design of the chamber system reported in Wang et al (2014) is similar to ours, and their reactor is 30 m<sup>3</sup>, with a spatial distribution difference in temperature within  $\pm 0.5\text{K}$ . Our reaction chamber is only 2 m<sup>3</sup>, and the spatial difference in temperature can be ignored.

**Modification in the main text:**

Line 344 – 348: added 'The internal design of our chamber enclosure ensures that the circulating air, which controls the temperature surrounding the reactor, reaches equilibrium (taking < 2 hours as shown in Fig. 2) from the outside of the Teflon film to the inside. This design guarantees that the temperature distribution is spatially homogeneous, even for a chamber system with a 30 m<sup>3</sup> reactor (Wang et al., 2014)'.

2) Line 442-443: Are there any other studies about NaCl seed found the same conclusion? Is the dry NaCl seed not competitive with sorption of organic vapors onto the chamber wall? Or the condensing SOA compounds form a separate phase from the seed?

**Response:**

The seeds we used in the  $\alpha$ -pinene ozonolysis experiments were ammonium sulfates, which was clarified in Section 4.2. This was selected to allow comparison with previous chamber studies since ammonium sulfate is the most commonly used seed while sodium chloride is relatively less used. In terms of SOA yield in the  $\alpha$ -pinene ozonolysis, we found that in chamber studies, liquid water

tends to inhibit its formation (Cocker Iii et al., 2001; Kristensen et al., 2014), while in oxidation flow reactor studies, it tends to enhance the SOA yield (Faust et al., 2017; Zhao et al., 2021). Unfortunately, to our knowledge, these studies did not analyze the reasons behind this phenomenon in detail. In a subsequent study a few months later, we observed the same phenomenon and have preliminarily focused on the kinetic limitation of liquid water, but further theoretical calculations are still needed to explain it in detail.

Dry seeds still compete with the sorption of organic vapors onto the chamber wall, and in gas-particle partitioning, this is related to the vapor pressure of organic species in the condensed phase, and for particulate phase, curvature effect should be considered. Dry seeds have relatively higher vapor pressure of organic materials due to their lower Raoult effect, so they are not conducive to SOA formation in the initial stage of the reaction, as shown in the subplot of Figure 5. However, as SOA continues to form, the difference in SOA yield reverses.

The homogenous nucleation of organic vapor was avoided by increasing the seed number concentration/surface area concentrations. The particle size distribution in Figure S11 demonstrates this.

**Modification in the main text:**

Line 484 – 501: added the paragraph ‘It is worth noting that, the organic vapor wall loss can have significant influence on SOA formation. However, quantifying wall losses of gaseous organic products is still a challenge in chamber experiments. Gaseous intermediates are difficult to be quantitatively measured, and the theoretical calculations of wall losses also have large uncertainties due to the lack of data on some parameters, such as the effective wall mass concentration and eddy diffusion coefficient inside the reactor. The wall loss behavior of gases essentially depends on the concentration gradient between the gas phase and the wall. To our knowledge, there is no conclusive evidence to support higher wall losses of gaseous intermediates under higher RH, which are even significant enough to cause a notable reduction in SOA yield. In addition, if higher RH can enhance the diffusion of gaseous intermediates towards the wall, then the diffusion of gaseous intermediates towards the particle phase should also increase. Compared with experiments without seed particles, when seed particles exist, gases condense on the particles while condensing on the walls, causing the gas-phase concentration to decay more rapidly, resulting in less wall loss of gases and higher SOA yields during the initial period of the experiment, as shown in the subplot of Fig. 5. However, the final difference in SOA yields is still unclear because, under the condition without seed particles, particles generated through nucleation continue to grow and can provide a considerable amount of condensation sink after the reaction proceeds for a period of time. This process needs to be numerically described and analyzed that carefully consider wall loss behavior and physicochemical properties of particles in future studies’.

Line 466 – 467: added ‘We used seeds with sufficient surface area concentration to prevent the gas phase products of VOC from homogeneous nucleation’.

3) Line 372-373: How to determine the segmentation of the shift and whether the results of the shift accurately represent the actual wall loss of the substance in different experiments?

**Response:**

The determination of the shift points can be roughly determined by human judgment. We had the curve of the wall loss rate constant  $\beta$  as a function of particle size  $D_p$  for each measured particle diameter, which allows us to visually identify two shift points. Then, we observed the corresponding particle size bin in the raw data where  $\beta$  changes. For instance, as  $D_p$  increases,  $\beta$  first increases and reaches a maximum value at 45.3 nm. Starting from the next particle size bin at 52.3 nm,  $\beta$  decreases with increasing  $D_p$ . Therefore, the first shift point is determined to be between 45.3 nm and 52.3 nm, and the value can be taken as 50 nm. So was the identification of another shift point

As for the second question, there may be some differences in  $\beta$  for identical  $D_p$  in each experiment, but the two shift points remain the same. Therefore, in the formal experiments using monodisperse seeds, a 20-30 minute period is reserved before each reaction to determine  $\beta$  for this monodisperse particle size in that particular experiment. Then, the  $\beta$  for other particle sizes can be determined by an up-and-down shift of the  $\log_{10}(k)$ - $\log_{10}(D_p)$  function curve (as mentioned in Section 3.4). For experiments with polydisperse seeds, the total volume wall loss rate constant (mentioned in the last paragraph of Section 3.4) is typically used, and this parameter also needs to be determined 20-30 minutes before each experiment.

**Modification in the main text:**

Line 394 – 396: changed ‘In this study, two inflection points are identified at 50 nm and 150 nm (Fig. S8)’ as ‘In this study, two inflection points are selected at 50 nm and 150 nm according to the identified inflection particle size bin of 45.3 – 53.2 nm and 143.3 – 165.5 nm, respectively (Fig. S8)’.

**Technical comments:**

4) Line 239-240: Please note the use of subscripts ( $\text{SO}_2$  and  $\text{O}_3$ ).

**Response:**

Thank you, we have corrected it now.

**Modification in the main text:**

Line 255 – 256: changed the subscripts ( $\text{SO}_2$  and  $\text{O}_3$ ).

5) The description of figures should be more accurate:

Line 259-261: “more than 25 hours” could not be seen in Fig. S2a.

Line 284-285: Most of the mixing time to uniformity for gas showed in the figure is longer than 1 minute with the fans on.

**Response:**

Thank you for your correction. We have changed it to ‘within 24 hours’ for more precise description.

We apologize for any potential confusion caused by the left panel of Fig. S5, but most mixing times are indeed within one minute. We have added a sentence in the main text to clarify: ‘the duration between the two plateaus in Fig. S5’.

**Modification in the main text:**

Line 276: changed 'more than 58 hours' as 'within 24 hours'.

Line 300 – 301: added '(the duration between the two plateaus in Fig. S5)'.

6) Line 321-323: The number of lights of scheme 'left and right' is 20 but not 40, and is 10 lights for 'only left' and 'only right'.

**Response:**

Thank you for your correction, we have changed.

**Modification in the main text:**

Line 338 – 339: changed the correct numbers of lights: 'the scheme 'left and right' (20 lights) was the sum of that of 'only left' (10 lights) and 'only right' (10 lights)'.

7) Line 378-380: Please note the use of operational symbol (\* and Í).

Line 198: The format of unit shall be checked through, e.g., should be no 'space' before %. The same unit with different format such as cm<sup>-3</sup> and µg/m<sup>3</sup> were used.

**Response:**

Thank you for your correction, we have changed the symbol '\*' as '×' (Line 386) and '·' (Line 361).

**Modification in the main text:**

Line 404: changed the symbol '\*' as '×';

Line 379: changed the symbol '\*' as '·';

Line 446: changed the symbol '\*' as '·'.

8) Please unify the format of the graphs and optimize the graphs. For example, the font size is different in Fig. 3a and 3b. There should be a 'space' between 20 and °C for Fig. 3b. And we could even see some grey lines outside the graphs, it seems that the graphs were simply pasted from other software and combined together. Please redraw these figures and unify the format.

**Response:**

Thank you for your suggestion, we have unified the format of the graphs and optimized the graphs.

**Modification in the main text:**

We have optimized Figure 3, and unified the format of Figure 2 to Figure 5.

9) Please doublecheck the citation of references. For example,

Line 85, the citation of "Ravishankara, 97" should be 1997 but not 97.

Line 762-764, the year was missing for this citation.

**Response:**

Thank you for your reminding, we have double checked and corrected the improperly formatted citations.

**Modification in the main text:**

Line 85: corrected as '1997';

Line 540, 554 – 555, 557, 559, 573 - 574, 578, 587, 655, 692, 694, 704, 752, 784, 812: completed the year of these citations.

**Reference:**

Cocker Iii, D. R., Clegg, S. L., Flagan, R. C., and Seinfeld, J. H.: The effect of water on gas–particle partitioning of secondary organic aerosol. Part I:  $\alpha$ -pinene/ozone system, *Atmospheric Environment*, 35, 6049-6072, [https://doi.org/10.1016/S1352-2310\(01\)00404-6](https://doi.org/10.1016/S1352-2310(01)00404-6), 2001.

Faust, J. A., Wong, J. P. S., Lee, A. K. Y., and Abbatt, J. P. D.: Role of Aerosol Liquid Water in Secondary Organic Aerosol Formation from Volatile Organic Compounds, *Environmental Science & Technology*, 51, 1405-1413, [10.1021/acs.est.6b04700](https://doi.org/10.1021/acs.est.6b04700), 2017.

Kristensen, K., Cui, T., Zhang, H., Gold, A., Glasius, M., and Surratt, J. D.: Dimers in alpha-pinene secondary organic aerosol: effect of hydroxyl radical, ozone, relative humidity and aerosol acidity, *Atmospheric Chemistry And Physics*, 14, 4201-4218, 2014.

Wang, X., Liu, T., Bernard, F., Ding, X., Wen, S., Zhang, Y., Zhang, Z., He, Q., Lü, S., Chen, J., Saunders, S., and Yu, J.: Design and characterization of a smog chamber for studying gas-phase chemical mechanisms and aerosol formation, *Atmospheric Measurement Techniques*, 7, 301-313, [10.5194/amt-7-301-2014](https://doi.org/10.5194/amt-7-301-2014), 2014.

Zhao, R. R., Zhang, Q. X., Xu, X. Z., Zhao, W. X., Yu, H., Wang, W. J., Zhang, Y. M., and Zhang, W. J.: Effect of experimental conditions on secondary organic aerosol formation in an oxidation flow reactor, *Atmos Pollut Res*, 12, 392-400, [10.1016/j.apr.2021.01.011](https://doi.org/10.1016/j.apr.2021.01.011), 2021.