

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.

RC1: 'Comment on amt-2023-34', Anonymous Referee #2, 26 Apr 2023:

The manuscript by Zong et al., titled “A new smog chamber system for atmospheric multiphase chemistry simulation: design and characterization” characterized the 2 m³ indoor smog chamber (AIR) and conducted a series of characterization experiments. The characterization results, as well as the yield experiments of α -pinene ozonolysis combined with a box model simulation, supported that the AIR chamber could be used to simulate multiphase atmospheric chemical reactions. The manuscript was well written and organized, but the reviewer thinks that detail characterizations related to new designs for multiphase chemistry may be further strengthened.

Specific comments:

1) Figure 2: the conditions of the test should be listed, such as the fan condition. Is it batch mode? Will irradiation affect the temperature? Can water molecules permeate the FEP film? If so, the RH should decrease with time due to the water permeation.

Response:

This chamber system is designed to operate in Batch Mode. The fans are only turned on during the injection period and kept off after finishing injection. The irradiation can change the temperature range of stable temperature control, but it does not affect the stability of temperature control. This is mentioned in Section 3.3: ‘The illumination of lamps raises the lowest achievable temperature by 3 °C for every 10 lights on. However, the illumination of the reactor does not affect the stability of temperature and RH inside the reactor’.

FEP film is semi-permeable, and gaseous molecules can penetrate through it, but at a slow rate. For example, the permeation rate of nitrogen is about 5L/m²/24h/atm, while that of water molecules is only, 0.007L/m²/24h/atm. For batch mode experiments, which typically last around 3 hours, this permeation amount can be ignored. In fact, the stable RH observed in Figure 2 also suggests this point. These details were clarified in the revised manuscript.

Modification in the main text:

Line 374 – 375 (the description of Figure 2): added ‘The chamber was operated in batch mode’;

Line 353 – 355, added ‘The RH fluctuation caused by the water permeation through the FEP filter can be ignored due to the slow permeation rate of water molecules (0.007L/m²/24h/atm)’.

Line 384 – 386, added the discussion about the fans ‘In Table S6, when turn on the fans, the wall loss is usually much higher, so the fans will only be turned on during the injection period and kept off after injection’.

2) What is the size distribution of the particles before and after condensing organics on the particles? Is it efficient to introduce monodisperse seeds (number concentration and size

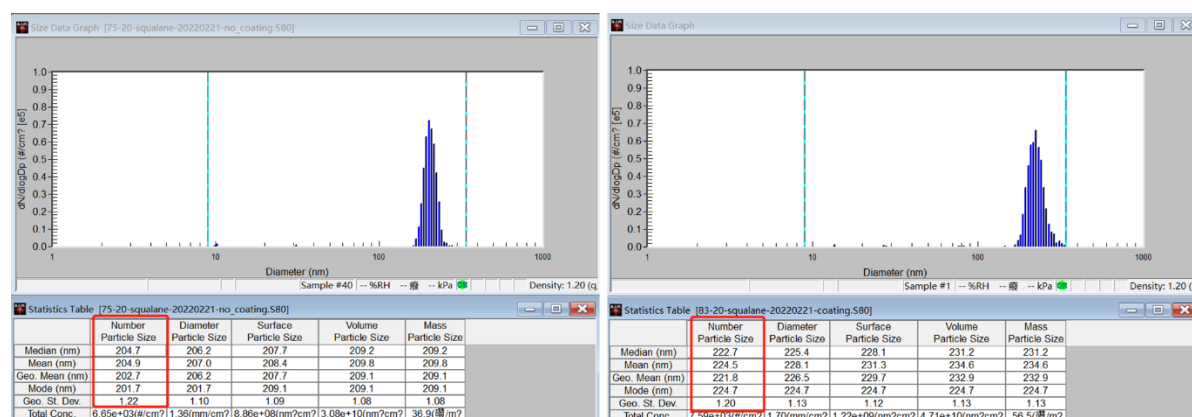
distribution)? Is there homogeneous nucleation?

Response:

In coating tests, the homogeneous nucleation of organic vapors after cooling is related to several factors such as the volatility of organic species, the number concentration/surface area concentration of the existing seed particles, and the cooling rate. As one part of the chamber system, the detailed operating conditions of the coating device need to be carefully considered and tested before each set of experiments to obtain monodisperse particles with coating morphology. For the results shown in Figure 3b, we have optimized the operating conditions and achieved monodisperse coating particles without nucleation of organic vapors.

The size distribution of aerosols was measured by SMPS, and two images are attached below for comparison, where the left graph shows the size distribution of 200nm monodisperse NaCl particles that have not passed through the coating device, and the right graph shows the size distribution of aerosols generated after passing through the coating device and forming organic coatings. This suggests that the monodisperse seeds are sufficient and no homogeneous nucleation of organic vapors.

Regarding the homogeneous nucleation of organic vapors in the coating device, we will add relevant statements in the revised manuscript for clarification.



Modification in the main text:

Line 414 – 417: added ‘The surface area concentrations of the introduced seed ($> 800 \mu\text{m}^2/\text{cm}^3$) are sufficient that no homogeneous nucleation of organic vapor occurs. Both the size distributions of the particles before and after condensing organics on the particles are monodisperse’.

3) For the pinene ozonolysis experiments, is there homogeneous nucleation with the high concentrations of pinene and ozone. I think the experimental conditions should be optimized for multiphase chemistry. For example, SOA is generated on the seed surfaces as much as possible.

Response:

During the preliminary experiments, a nucleation-growing particle population did exist due to the insufficient surface area concentration of seed particles. We did not use the data from these preliminary experiments. Later, we increased the loading of seed particles to avoid this situation. The corresponding time series of particle size distribution can be found in Figure S11 in SI, which supports this point.

Modification in the main text:

Line 467 – 468: added 'We used seeds with sufficient surface area concentration to prevent the gas phase products of VOC from homogeneous nucleation'.

4) Section 4: The vapor wall loss can have significant influence on SOA formation. How this will affect the SOA formation and SOA yield in this study, especially for different systems in the absence or presence of seed aerosols?

Response:

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. Therefore, we cannot determine the specific role of wall losses of gas-phase products in the formation and yield of SOA. The wall loss behavior of gases essentially depends on the concentration gradient between the gas phase and the wall. 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. However, the extent of this difference 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 described and analyzed with models that carefully consider wall loss behavior and physicochemical properties of particles, but to our knowledge, it seems that such models are lacking and need to be established first. We are currently working on this task.

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’.