

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.

RC2: 'Comment on amt-2023-34', Anonymous Referee #1, 03 May 2023

The authors show their results about a newly constructed smog chamber for multiphase chemistry studies. The facility and characterization results have been demonstrated. This work is well organized and presented. It is publishable after the following questions have been well addressed.

Specific comments:

1) The novelty of this work should be demonstrated based on a good review of previous studies.

Response:

We have checked the Introduction Section and found that the presentation and discussion of the application of chamber in atmospheric multiphase chemistry research is a little weak, so we have made some modifications to the relevant paragraph after investigating more previous studies.

Besides, we found that the last part of the current introduction only briefly summarizes the main work of this study without emphasizing the advantages of the AIR chamber system. We have now included this in our revised manuscript.

Modification in the main text:

Line 95 – 96: added 'In addition, small chambers may have the potential for controlling RH change and simulating co-condensation phenomena';

Line 98 – 108: rewrite as 'The influence of aerosol phase state on kinetics of gas-particle interactions has received increasing attention (Virtanen et al., 2010; Berkemeier et al., 2016; Wang et al., 2015a; Reid et al., 2018), and this requires the phase state of seed particles can be controlled in chamber simulations. A laboratory study using pre-deliquescence way to control particle phase state has been reported (Faust et al., 2017), providing a feasible way for phase state control. In regard of particle morphology, some chamber-based experimental studies in recent years have preliminarily shown that organic coatings have important effects on the kinetics of aerosol multiphase transformation (Zhou et al., 2019; Zhang et al., 2018; Zhang et al., 2019), which deserves more researches. As these studies showed evidence that the morphology and phase state of aerosol particles play important roles in the atmospheric multiphase chemistry processes, focused chamber studies on multiphase chemistry require additional steps to control the morphology and phase state of seed particles in chamber design';

Line 119 – 122 (last part of Introduction): added 'Our results indicate that the AIR chamber system has more precise temperature and RH control capabilities compared to other chambers. Phase state and morphology of seed particles can also be accurately manipulated in advance, which is rare in existing smog chamber systems'.

2) Line 105, the uncertainty of RH measurement is $\pm 75\%$. What is the uncertainty for the RH

sensor?

Response:

$\pm 0.75\%$, refers to the standard deviation of measured RH data sets, which deviate from the mean value while controlling the chamber RH at a set value, that within 0.75 of an RH unit (RH is described in %). Your statement ‘the uncertainty for the RH sensor’ should refer to the extent to which measured RH values deviate from the actual RH. We calibrated the RH sensor using a hygrometer, and the deviation is less than 1%, as explained in section 3.3 of the manuscript (Line 349).

3) Lines 223-225, is the coating system stable enough for generating organic coated particles? In my experience, the coating efficiency might decrease with time at high temperatures.

Response:

Yes, we did observe such phenomenon when testing the operation conditions of the coating system. The coating efficiency would drop after running continuously for approximately four hours. We noticed that if the generated inorganic core seeds were not thoroughly dried, condensed water would appear on the bottom of the three-necked flask after several hours, submerging the organics and resulting in a reduction of coating efficiency. Increasing the drying efficiency has alleviated the issue of reduced coating efficiency, allowing the stability of the coating system to last beyond four hours. In fact, for general chamber experiments, the step of injecting seeds will not last four hours, so the stability of this coating device is sufficient.

Modification in the main text:

Line 240 – 241: added ‘The coating efficiency can keep stable within four hours, which is sufficient to meet the duration of injecting seeds for general experiments’.

4) How long does it take for introducing around 5000 cm^{-3} OA-coated seeds into the chamber?

Response:

This depends on factors such as the size distribution of generated core seeds from the aerosol generator, the selected particle size, and the aerosol flow rate. For the generator we used, TSI 3076, the generated aerosol size distribution is not wide, and the peak particle size is between 50~100 nm. We tested that when the aerosol generation efficiency was set to its maximum, the concentration of monodisperse nanoparticles in the reaction chamber reached 5000 cm^{-3} within a few seconds for 100 nm particles, about 5 minutes for 150 nm particles, and 40~50 minutes for 200 nm particles. It should be noted that the operating curve of the coating device is variable, depending on the operation goal such as injection concentration, and needs to be calibrated before formal experiments.

Modification in the main text:

Line 211 – 216: added ‘For polydisperse seeds experiments, the seed generation system can inject seeds into the reactor to the desired amount within a time scale of seconds to several minutes. For monodisperse seeds experiments, if large-sized seeds that have a lower number fraction in the generated aerosol population are selected, the time scale will expand to 40-50 minutes. The appropriate time scale for seed injection can be adjusted by changing the solution concentration and aerosol flow rate’.

5) Figure 4, why is the SOA yield higher than those reported values at high OA mass loading?

Response:

There are two reasons for this. One is that the experimental temperature we used is 293K, slightly lower than the temperatures used in the literatures. The second reason is that, as mentioned in Section 3.4, the Teflon film of our reaction chamber has been passivated by O₃, so the wall losses of intermediate products from the gas-phase reactions may be lower due to less reaction loss.

Modification in the main text:

Line 452 – 453: added ‘and the lower experimental temperature’.

6) When calculating SOA yields, the wall loss of LVOCs should be accounted for. The low SOA yield might be related to the high wall loss rate of LOVCs.

Response:

Quantifying wall losses of gaseous organic products is still a challenge in atmospheric chemistry research. 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. To our knowledge, there is no conclusive evidence to support higher wall losses of LVOCs under higher RH, which are even significant enough to cause a notable reduction in SOA yield. In addition, according to gas phase diffusion theory, if higher RH can enhance the diffusion of LVOCs towards the wall, then the diffusion of LVOCs towards the particle phase should also increase.

We still suggest attributing the lower SOA yield to higher liquid water content of the aerosol, and similar phenomena have also been observed in several groups of experiments in our another research after several months. We are currently attempting to obtain an explanation through theoretical calculations and have preliminarily found that this is related to the kinetic limitations of the particle phase dynamics, but which is different from the previously recognized viscosity and diffusion coefficient influences.

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’.

7) I am wondering why the authors do not evaluate the photochemical performance here.

Response:

The photochemical reaction is an important issue of atmospheric gas phase chemistry, which includes the study of the effects of VOCs, gas-phase oxidants, radiation, etc. However, the design of this chamber system mainly focuses on the physical and chemical properties of seed particles, so a relatively simple gas phase chemistry reaction system was chosen. For example, we adjust the phase state and liquid water content of seed particles by changing RH, but for a photochemical reaction system, the oxidant $\cdot\text{OH}$ would be strongly affected by RH, leading to the roles of particle physical and chemical properties in SOA formation becoming ambiguous. Therefore, for the photochemical behavior of this chamber system, it is mainly characterized by spectral features and photolysis rate constants in Section 3.2, and no specific oxidizing system for photochemical reactions is conducted.