Anonymous Referee #3 Received and published: 27 December 2020

GENERAL COMMENTS: The authors describe a new type of enclosure chamber which allow to observe particle growth by seeding a real mix of ambient atmospheric air exposed to outside light conditions. Long residence time of the seed particles is provided by rotating the cylindric chambers around the center axis which reduces gravitational settling and convective mixing. Gas exchange is provided by permeation through a membrane, such that the injected seed particles are not subject to flush out. The latter concept was also applied before (QUALITY camber). The presented chambers provide still a quite substantial progress compared to the previous versions. The results are overall presented well and look promising. And I think the manuscript shows indeed proof of concept. However, from the descriptions in the manuscript, I could not understand all features the new chamber. Some important information is missing. The manuscript is within the range of AMT and could be published after the authors addressed the comments below.

SPECIFIC COMMENTS: In the manuscript the authors seem not to distinguish between mixing time and exchange time (e.g. line 22, abstract).

In my opinion there is a difference. If the mixing time is short compared to the residence time, the chamber will be always well mixed, even when the boundary conditions are (slowly) changing. Isn't fast mixing a prerequisite for treatment of the chamber as a continuously stirred tank reactor? If the chamber is not mixed fast, in how far is the sampling from one (or two?) points inside the chamber representative for the processes in and state of the chamber? Please describe, how the mixing within the volume of the chamber is assured, the mixing time and mixing behavior in the CAGE chamber?

This is an excellent question and one for which we do not have an excellent answer. We feel there is some optimal amount of mixing within the chamber – too much and gas and, especially, particle wall losses will be excessive, and too little and there will be significant gradients in gas phase concentrations, especially for more reactive species. It has been our opinion that mixing in the chamber is typically on the high side of that optimum, meaning that we have sought to minimize it. Of course, the balance between the two effects depends on the nature and objectives of the experiment. We did not perform any experiments that would allow us to separate the mixing time from the exchange time. For now, we have replaced "mixing" with "exchange" for both instances in the text where it was mistakenly used (abstract and original line 266).

A bit related to the previous point. You are taking samples for measurements from the chamber by two DMAs, APS, and AMS. How large is the total sample flow? I guess it is significant compared to the residence time? How much does this sample flow affect the exchange time? Does the sample flow enhance the mixing in the chamber?

The APS was configured with separate sample and sheath flows, such that it pulled only 1 L/min from the chamber (and not the total 5 L/min including the sheath flow rate). The SMPS had a sample flow rate of about 2.1 L/min. Adding in the much smaller AMS flow rate results in an overall sample of about 3.5 L/min. However, a sampling sequence was used to minimize extraction from either chamber. Specifically, the repeated sequence used during the study described in the manuscript was Chamber A

→ Chamber B → 4 x ambient. The result is that sample was extracted from each chamber only 1/6th or 16.7% of the time, meaning that the effective withdrawal rate was only about (3.5 L/min) x 0.167 = 0.58 L/min. The particle injection flow rate was about 3 L/min. The particle injection frequency and duration varied, with an average of around 20 to 30 min per day, resulting in an average chamber air displacement rate of about 0.05 L/min. The corresponding loss time constant is 1000 L / 0.63 L/min = 1,600 min or about 26 h. Of the particle size categories considered, the maximum residence time was about 8.2 h. Thus, extraction contributes to the loss, but is less important than other contributors such as diffusion and electrostatic loss. The reported particle residence times of the three size categories of 6.0 h (small particles), 8.2 h (medium particles), and 3.9 h (large particles) would be approximately 7.8 h, 11.9 h, and 4.6 h, respectively, without any sample or injection flows.

The intermittent sampling approach was clarified in the manuscript by revising the following sentence in Section 2.5 of the original manuscript:

"The instrumentation was configured to sample from both the inside of each of the two chambers and ambient air."

To now be:

"The instrumentation was configured to sample from both the inside of each of the two chambers and ambient air, with a repeated sampling sequence of Chamber A → Chamber B → 4 x ambient, such that sample was extracted from each chamber only 1/6th of the time in order to minimize the loss rate of the captive particles."

Was the sheath for the DMA(s) taken from the chamber? If not in how far could not using sheath air from the chamber and drying of the aerosol have affected the observation that only condensation took place (that growth rate always > 0)?

The sheath flow in the SMPS DMA was configured in a recirculation loop. Thus, all of the air in the DMA came from the same source. The interior volume of the DMA is about 1.6 L. The filter and blower enclosure in the recirculation loop probably bring the total volume up to about 2 L. As noted above, the SMPS sample flow rate was about 2.1 L/min, resulting in a turnover time constant for the air in the DMA of about 1 min, which is short compared with the 5 min sampling time used. If carryover from a previous measurement (e.g., from Chamber A when sampling from Chamber B) had a significant impact, it would be reflected in differences between the up and down scans in the SMPS measurements, which was not observed. It also seems unlikely that the gas phase would result in a positive bias in measured growth rate because low volatility species would likely be depleted in the stainless sampling line or in the DMA itself.

Line 126: I have difficulties to understand how the sampling is done. Is the centers axis a tube which serves as a sampling line at the same time? Is that little perpendicular extension in Figure S2 to the very right one of the sampling tubes? Do you sample from both tubes or are you using one for sampling and the other for injection of seed particles? Do all instruments sample from the same point?

Particles are injected into one side of the chamber and are extracted from the other side. The two tubes that are perpendicular to the axle in Figure S2 are the sampling and injection ports. (The third, which is at about a 45 degree angle was intended for gas sampling, but was not used.) Those 0.95 cm OD tubes are bent 90 degrees inside the hollow axle and then continue through opposite ends of the axle where they are sealed into fixed unions using radial o-rings. Thus, the injected or sampled flow remains within a single 0.95 cm OD stainless tube from outside of the chamber to the end of one of those perpendicular tubes.

Line 323: I think you use “Dp” as size parameter for the size distributions and as mode diameter. I suggest using two different notations and call latter Dmode, or so.

We have changed instances of D_p used to describe the fitted mode diameter with D_{mode}, including those in Figures 11 and 13.

Line 423 and Figure 15. I can see that the traces look different, but also different quantities are plotted. Please, extent the description why exactly we can see from this Figure the difficulty in determining secondary aerosol production from mass measurements. I guess you have all information to calculate the mass production from your growth rates using an average particle density (from AMS measurements)? Or I don’t understand at all what you want to address.

Our objective in including that figure was simply to argue that it can be challenging to quantify the SA production rate from measurements of mass (or volume) concentration as would typically be necessary if a polydisperse seed aerosol were instead injected. For a typical batch chamber, the rate of SA mass production can be controlled to be greater (or much greater) than the rate at which SA already on particles is lost to the walls. Within the CAGE chambers, the SA production rate is comparatively low and highly variable. The result is that the noise caused by variability in the wall loss rate is often greater than the signal resulting from added SA to the particles in the chamber. This is reflected in the noisy curve showing the production rate inferred from the AMS measurements.

Line 459: Since O3 is substantially lower in the night than during day time, ozonolysis ([VOC]*[O3]) is probably not driving the night time maximum.

As shown in Figure 17, the average rate of decay of the O_3 concentration at night is comparable to that calculated for $NO_3\cdot$ (which is, of course, not surprising because O_3 reaction produces $NO_3\cdot$). With our model, the sort of production rate estimate shown in Figure 18 was not very sensitive to whether the O_3 reacted with the precursors or the O_3 produced $NO_3\cdot$, which reacted with the precursors. Though O_3 decreases rapidly in the evening, the concentrations of precursor VOCs (especially monoterpenes) increase rapidly, with the result that the product of the concentrations often reaches a maximum around the time of the peak in particle growth rate.