Atmospheric Measurement Techniques Discussions

Atmos. Meas. Tech. Discuss., referee comment RC2
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Comment on amt-2021-147

Anonymous Referee #2

Referee comment on "Characterisation of the Manchester Aerosol Chamber facility" by Yunqi Shao et al., Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2021-147-RC2, 2021

This paper describes an atmospheric chamber that has been in use since at least 2011, providing details that were promised in a paper published at that time. Numerous papers have been published based upon experiments performed using this chamber though from the few I have examined, details of the chamber design, and key operating characteristics such as the mixing and wall losses have not been reported, but reports from the Manchester group show much deeper and more rigorous understanding and modeling of those factors than is evident in this manuscript. This chamber is similar to other Teflon chambers; one key difference is the connection between this chamber and a cloud chamber. The chamber can also be interfaced to emission sources such as engines, as has been done in a number of chambers around the world. The manuscript mentions, but does not discuss these features, so this work needs to be evaluated in the context of reports on other atmospheric chambers, and in the context of the prior work reported from this chamber. While the design of atmospheric chambers is certainly within the purview of AMT, and the analysis of mixing and wall effects of a facility that has been used in numerous studies of fundamental atmospheric processes would be of value in understanding the results from those and future experiments, this paper offers little in the way of new insights. Careful selection of key experiments from the many that have been performed with this system, and more critical analysis and discussion of those results in light of current understanding of nonidealities of Teflon chambers could well change that assessment. Instrumentation has changed dramatically since earlier reports on chamber characteristics, e.g., Carter et al. 2005, and Cocker et al., 2001, so with the array of instruments listed in Table 1 it may be possible to provide data on wall effects beyond what has been previously reported using existing data from this chamber. Unfortunately, this paper repeats observations that have been reported many times, and that are commonly addressed in analysis of data from chamber experiments, albeit with a different chamber.

One may ask what aspects of this chamber can be considered unique. It is a Teflon chamber operated in batch mode like many others. The air cleaning system is similar to

other facilities. The chamber suspension system is a bit different from most Teflon reactors. One difference is the illumination system, which appears to more closely approximate the ambient actinic spectrum than many other facilities. Another is the automated cleaning system, which possibly could reduce run-to-run variability. Experimental impacts of the latter items are not discussed in this paper. The mixing time in the chamber is fast, a couple of minutes. This is the primary feature of the MAC chamber. No demonstration, or even discussion is provided regarding novel experiments that might be enabled by this fast mixing time. Wall agitation is suggested to "result in sufficient mixing of its components during experiments," though no evidence is provided. Wall agitation by the air conditioning will also enhance losses, which is demonstrated. The control system that automates filling and cleaning processes could, as suggested in the discussion and conclusions, enhance comparability across experiments. Again, no evidence is provided. As I read the paper this struck me as a potentially significant benefit; data to support the hypothesis would strengthen the paper.

The experiments that are discussed in the paper focus on wall effects, wall losses of particles and vapours, and wall reactions. A key conclusion is that wall losses make the chamber sensitive to effects of deposits during prior experiments, which is well known, and has been extensively explored by Huang et al. (Env. Sci. Tech. 2018). The authors speculate that increased NO2 and O3 losses with chamber age, and speculate that the losses may reduce jNO2 (not explored by Huang), again without experimental evidence.

They further conclude that differences in the way wall losses are estimated and incorporated into data analysis makes a difference in estimates of SOA yield; unfortunately the paper merely shows different results from different data analysis methods without providing any details about how they have applied those methods. Given the work from the same group on the PyCham box model for chambers (O'Meara, …, Shao, McFiggans, Geosci Model Dev., 2021) which seems to incorporate all of the effects that are suggested in this paper, I would have expected a more insightful discussion of the treatment of wall effects that might provide some guidance regarding the best ways to account for these effects, or at least how data from this chamber has been analyzed.

The paper also concludes that alpha-pinene SOA yields from the MAC chamber are similar to those determined over the two decades from different chambers, but that differences may be attributed to differences in data analysis. Understanding of the wall effects has advanced significantly in that time, with models similar to PyCHAM playing a key role as reflected in a number of cited papers. The author thus suggest that regular chamber characterization experiments are needed, and that methods for validating data analysis approaches are needed. This has long been recognized, but bears repeating.

In summary, I find that this paper describing a chamber that has been in use for a decade does not approach the standard of prior presentations and characterizations of new chambers. Nor does the data presented lead to significant new insights. Given this system's extensive use, sufficient data almost certainly exists to provide a more rigorous and critical evaluation of the MAC chamber that would allow it to be quantitatively compared to other chambers. The instruments available to the authors could provide data that was not possible at the time that earlier chambers were characterized, so unique data

may exist, or certainly could be generated to address the key focus of wall effects. The writing also needs serious work, as does the data presentation.

In light of my conclusions and to support the authors in their efforts to , I provide additional detailed comments, questions, and points of confusion that came to mind as I read this paper.

Like numerous other chambers, the MAC is fabricated from FEP Teflon. It's volume, 18 m3, is smaller than many but still reasonably large to enable aerosol studies. A feature of the MAC is its interface to an ice cloud chamber, or to sources of gases or particles, including combustion sources.

The supporting framework differs from other designs though the description (three pairs of rectangular extruded aluminum frames, with the central rigid pair fixed and the top and bottom pairs free to move vertically. This detail may, or may not be useful to others considering building chambers but the description is uninformative, as is the schematic in Fig. 1. Why is a pair of frames needed at each level? The description in section 2. just mentions three frames. The discussion of the Teflon reactor indicates that it is maintained under slight positive pressure to minimize contamination from lab air; how much pressure is maintained in the chamber?

The chamber illumination differs from many other chambers in being designed to approximately reproduce the atmospheric actinic spectrum in Manchester. The discussion of the illumination system is confusing. It is noted in section 2.3 that the absorption of IR radiation due to water vapour in the atmosphere is simulated by inserting a water-filled container with quartz windows between the light sources and the chamber. What is the purpose of that simulation? It is also suggested that the water is there to minimize heating of the chamber, which is what I would expect is the actual purpose. This should be made clear.

The air supplied to the chamber is, according to section 2.4, dried lab air that is passed through a series of packed bed scrubbers, two to remove gases, and a HEPA filter. Figure 1 shows the air being drawn from the chamber, suggesting that chamber air is recirculated. Which is it? How pure is the processed air? This could be documented in terms of measured contaminant levels. A custom-built humidifier is included, but its description provides no hint as to how the water vapor is introduced into the air, nor is any indication as to the purity of the water used indicated. Contaminants in the water could introduce particles, depending on the humidification method. Seed aerosols are generated using an atomizer, but there is no mention of neutralizing the excess charge on the droplets. If as-sprayed particles are introduced to the chamber without neutralizing them first, charge induced wall losses could be substantial. Charan et al. (Cited) discuss the effects of particle charge on losses, though not the danger of not neutralizing the aerosol.

The MAC facility includes a control system to automate the filling and cleaning of the chamber. This could enhance reproducibility from experiment to experiment. As other systems do not have such capability, this feature of the system being reported might warrant further discussion, including data that shows the extent to which experimental conditions can be duplicated.

The discussion of the instrumentation used on the chamber (section 2.8) describes the "core instruments." Some of the additional instruments listed in Table 1 are mentioned. Since data presented in the paper include transient aerosol wall loss measurements, it is interesting that the DMA in the core set is used as a DMPS, in which the voltage is stepped. Such measurements tend to be slow due to the time required to attain steady state after each voltage step. The information provided is insufficient to assess whether this is an issue. One of the unmentioned additional instruments is an SMPS that might provide better time response. This point will come up again in the discussion of the experimental results. A filter on the outlet flow from the chamber at the end of the run. Figure 1 suggests that the filter processes both inlet and outlet flows. It should be made clear which flow is sampled. Moreover, I presume that there is a bypass flow, and valves to select whether the filter is used. That should also be made clear.

Section 3 of the paper presents chamber characterization data. Figure 2 shows that the temperature is affected by the lights, and that the temperature at the center of the chamber is higher than that of the wall. Interestingly, the relative humidity is also higher at the center than at the wall. In a well mixed chamber, one would expect the RH to decrease as T increases. For the RH data, two different sensors are used. Is the trend an artifact of the sensors, perhaps due to the sensors being radiatively heated? Some discussion of these results is needed.

Mixing is explored with both gas and particle tracers. Both approach a reasonably steady level after a few minutes. The discussion of the particle experiments indicates that neutral seed particles were introduced into the chamber. Were the particles actually neutralized?

The paper states that the light intensity as 3.5 times less intense than, but closely approximates that of the atmospheric actinic spectrum, but the lamps do not uniformly illuminate the chamber. Two different types of lamps were used to reproduce the spectrum, but they do not appear (from Fig 1) to illuminate the same portion of the chamber volume. How was the spectrum measured to produce the results shown in Fig. 4? The authors report three different photolysis rates; their respective deviations from atmospheric rates differ substantially. Since considerable effort was invested in matching the actinic spectrum, one might expect some discussion of the reasons for the differences.

The discussion of aerosol wall losses is particularly disjoint. Wall losses were shown in Crump et al. (1983 — not 2007) to depend strongly on particle size. The model from Crump, as extended to include charge effects by McMurry, is widely used to correlate observed chamber wall losses with turbulence levels within the chamber in order to properly account for losses in inferring aerosol dynamics from chamber data. Fig. 5 presents size dependent wall loss rates at two different relative humidities, and two

different turbulence levels, i.e., that when the air conditioner is disturbing the chamber walls and that with the air conditioner turned off. The data, apparently from a number of different experiments, show substantial scatter. Since no mention was made of charge conditioning (neutralizing) the seed aerosol, I must ask whether the scatter in the data could be due to charge effects. This is suggested without any elaboration in section 3.5. Discussion of these effects is needed. It would be good to compare the observed size dependent losses to the Crump model, and to assess whether the wall loss parameters change between experiments in each class (RH and AC).

The paper also discusses particle number and mass loss rates; since these are averaged over the measured size distributions, they are expected to differ from one experiment to another, due to differences in the size distribution, and within any experiment as the size distribution changes with time. Those values, and the discussion of them convey little useful information and distract from and confuse the paper. The data presented in Table 3 provides a qualitative estimation that losses in the MAC are comparable in order of magnitude to those in other chambers. The comparison with the CMU chambers in terms of the size-resolved decay rate are much more fundamental and significant.

Mixing and boundary layer effects also affect losses of gaseous species. Looking at individual species eliminates the confounding effect that size distribution introduces for particle losses, but they are confounded by differences in deposits on the walls into which the vapors may partition.

Section 3.5.1 presents a very confusing discussion of the effect of different wall-loss correction methods on estimates of SOA formation, using the mass loss rate in addition to the size-dependent loss rate. The mass loss rate is irrelevant as noted above. The Verheggen model treats the eddy diffusion and turbulent kinetic energy as empirical parameters based upon the Crump model, and uses the general dynamic equation to estimate the value of the relevant parameters. The large difference between the size resolved data inversion and the Verheggen method therefore requires some discussion. Experiments performed with seed aerosol alone provide a basis for comparison between the two models. I can only speculate as to why the Crump model gives consistently higher values than the Verheggen model which clearly includes coagulation and other aerosol dynamics processes. Since there is no discussion as to how the data were inverted it is unclear what processes were included in the data analysis.

The discussion of SOA formation analysis is further confused by using both data from the "nearest" experiment and the average of all experiments. I would be much more comfortable with an analysis of a clearly defined experiment. Teflon chambers age with use. A systematic look at the individual seed aerosol loss experiments should provide direct evidence for that.

Section 3.6 focuses on wall reactivity, which is a very real factor in data analysis (Huang et al., Env. Sci. Tech., 2018). The section should be labeled something like Chamber Wall Reactivity since calling this "Auxilliary Mechanism" conveys no useful mechanism. Huang et al. develop a model for the wall reactions that could provide a basis for considering the

processes that are very briefly discussed in this section.

The discussion of effects of wall contamination addresses problems that are encountered in every chamber that is used form multiple experiments between baking or other rigorous cleaning (which are not possible with teflon chambers. The data provided give some indications of the changes in performance of the chamber with age.

Specific points:

The abstract and introduction should make it clear that the chamber described in this paper has been used for over a decade, since papers of this sort usually report on relatively new experimental systems.

L 21: Provide a meaningful name rather than "auxilliary mechanism."

L. 65: It would also be useful to include the CLOUD chamber at CERN in the list of rigid chambers that can simulate the free troposphere.

L. 68: The discussion confuses the Caltech dual chambers (both ~28 m^3) of Cocker et al. (2001) and the University of California at Riverside dual chambers (90 m^3) (Carter et al., 2005).

L. 73: that can act as a sink

L 74: and aerosol particles (), and as a surface

L. 79: While it is obvious to those who work with chambers that chambers introduce background and memory effects, some explanation is needed for those with less experience or who may be reading this to understand data from experiments on this chamber.

L. 81: The sentence beginning "This necessitates …" is ambiguous. This what?

L. 83: MAC is used for the first time in the body of the paper. Spell it out.

L. 152: Is the specific blower type needed. There's a lot in this sentence, and inclusion of the blower details makes it difficult to follow.

L. 158: using ECD grade — and always spell out abbreviations when they are introduced for the first time.

L. 176: Edgetech and. Sensirion sensors (plural)

L. 219: The DMA uses filtered chamber air

L. 233: The company name is Cambustion - with an A

L. 235: Droplet Measurement Technologies. Spell out the name properly/

L. 239: additional instruments …

L. 250: the chamber temperature is controlled by the air conditioning system; that of the chamber itself is measured…

L 323: and O3 at concentrations ranging from 230 ppb to 350 ppb

L. 332: GIG-CAS, while the

L. 335: Measured first-order wall loss rates of selected

L. 344: Crump et al. 1983 — also correct the citation in the bibliography.

L. 381: Investigation of various particle wall-loss

L. 429: Eq. 2 should be inserted after this line

L. 433: Eq. 3 should be inserted after this line

L. 475: You really don't mean that the photolysis rates differ by 3 orders of magnitude. State the numbers independently and correctly.

L. 477: A substantial increase in the wall …and O3 was observed

L. 505: Note that the air circulation around the chamber also enhances wall losses.

L. 515: Mixing also affects losses of gases

L. 519: Differences between particle mass and number losses depend on the size distribution and are, therefore, not meaningful.

L. 535: as shown in Fig. 7

L. 538: e.g., gases … — comma is needed