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Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Homogeneous nucleation of sulfuric acid and water mixture: experimental setup and first results" *by* D. Brus et al.

**Anonymous Referee #1**

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Review of Brus et al. 'Homogeneous nucleation of sulfuric acid and water...' In an overall sense, the introduction of a new experimental apparatus for studying nucleation processes is a worthwhile endeavour. The difficulty of these types of experiments is 'legendary' and their accuracy is not established: this field needs experimental results to coalesce for confidence in them to grow.

This paper adds to the list of experiments/results that have been presented over the decades. It has been improved from that originally submitted but questions remain on the techniques and measurements (listed below.) Whether it should be published depends on their answers. The English also needs work (e.g. p. 23876 singular-plural



on l.5, sentence fragment on l.19; 'of' vs. 'from' on l.1 and missing article 'a' in l.16,17, next page. Missing article 'the' in l.26, 27 p. 23879.)

(i) Laminar flow tube. The name given to the overall apparatus could be a little misleading because (1) a warm gas flowing into a region with cooled walls will experience natural convection (bouyancy) and (2) the entrance of the cooled tube likely has some regions where the flow is not even steady with time. The authors should add a section or at least a few sentences discussing the likely flow profiles in the various regions of the flow tube. Typical Reynold's and Grashof numbers should be presented.

(ii) The use of filters in the experiment is an area of concern. (iia) HEPA filters can elute some vapors depending upon materials while the Teflon filter inside the apparatus, that needed an 'autoclaving' (probably not a correct usage here) cleaning procedure, is particularly troubling. How do the authors know about its final state of cleanliness? The use of polymer filter materials at elevated temperatures ($\sim$200 C?) does not seem to be a good idea. Was a glass wool or fibre filter found to be inadequate (much less likely to introduce contaminants?) Also, the temperature and location of the internal Teflon filter are unclear. (iib) Since the H2SO4 vapor concentration for the final results are ultimately derived from losses obtained from the IC measurements, it seems another source of H2SO4 could be used. The hot filter might be unnecessary if the jet is not used, and cleanliness of the system could be bolstered. (iic) There are a few lines on p.23879 that are troubling. It seems that a new filter must be used to measure again nucleation at the low H2SO4 (high RH) conditions. The reason given is that the walls of the instrument become a source of sulfuric acid vapor. But this is not a satisfying explanation. The walls of the cooled tube cannot be a significant source because the vapor must be highly supersaturated with respect to the bulk (i.e., acid solution on the walls) for nucleation to occur. If the 470 K (or mixing) region is an 'additional' source of H2SO4, then the IC measurements can pick that up. It is suggested that data from a rerun at high RH without changing filters and replacing mass balance with IC measured H2SO4. These results should be in agreement with the previous run. Hopefully, the

Teflon filter did not become ‘degraded’ by the hot and acidic conditions it experiences over the course of the experiment. Sulfuric acid at 470 K and low water vapor can lead to SO3 which can be very reactive towards plasticizers or even the plastic itself.

(iii) Plateau in particles is not well demonstrated for the 10% RH data (23881, l10.)

(iv) Three ways to count particles are discussed and they seem to not agree (the PH-CPC results did not agree with 3025A in an early version of the paper and now they are not being compared.) Details for results that are shown need to be presented: charging efficiency assumed for DMPS system; was the effluent from DMPS neutralized before the 3025A? Certainly the DMPS using a given detector should agree with the detector deployed without the DMPS.

(v) Figure 10 is a nice picture that is presumably interpolated results from the PH-CPC. Firstly, the raw data should be shown: the particle numbers vs. radius at each axial position should be plotted in a separate plot. What is the sample flow rate for the PH-CPC in these measurements? The difference in distributions on going from 8 to 12 sLpm flows is puzzling. (Firstly, what do the numbers in the legend mean? The same scale for each should be used.) Much higher numbers at the end of the reactor for the lower flow rate experiments is counterintuitive as less sulfuric will survive to this region compared to the higher flow conditions. Note also that these distributions are notably different than those presented by Ball et al. who also had natural convection concerns. Are the current temperature gradients larger than those of this earlier work?

(vi) A measured WLF that is less than the very lowest it can be (fully developed laminar flow) raises concerns about the measurements of H2SO4. The authors should present their explanation for such low WLFs.

(vii) There is additional concern that the J results for 30% RH are greater than the 50% RH results. This is not consistent with either theory or what has been published previously. Please comment.

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