Review of Tiszenkel et al, Temperature effects on sulfuric acid aerosol nucleation and growth: initial results from the TANGENT study

Scientific significance

New particle formation is a difficult and important problem, and there is currently a lack of diversity in the laboratory studies used to characterize it. Measurements from independent groups are to be encouraged.

However, the current manuscript requires revision if it is to be suitable for ACP. At first glance, it is unclear how much new information it adds to Yu et al (2017), although the new results do become clearer with a very careful reading. The last part of the introduction should be expanded to explain how the new study differs (more measurement data in the J vs temperature parameter space, and the addition of the second flow tube). The paper does not explicitly explain the point of the TANGENT apparatus, nor why it is an improvement on previous experimental setups, except via the sentence in the abstract that it allows nucleation and growth to be studied *independently*. This sentence should be revisited in the text with a better explanation for why this is an improvement on Yu et al (2017) where nucleation and growth rates are presented *separately* already. I appreciate that "separately" and "independently" are not the same, but this needs to be made more obvious in the paper text.

The lack of direct measurements of contaminant NH3 and amines during the experiments is a serious shortcoming, as the concentrations of contaminants could differ markedly between 2017 and 2018 measurement periods. This shortcoming limits the quantitative usefulness of the results, and places high demands on the quality of the data analysis and presentation if the paper is to meet the ACP publication criteria.

In addition to explaining explicitly the benefits of their new setup, the authors should consider setting their paper apart by including in their figures a more detailed, quantitative comparison with other relevant literature, for example Duplissy et al (2016) or Dunne et al (2016). The authors could try to determine from published nucleation measurements what ammonia or amine concentration would be required to reproduce the new particle formation rates they measure.

In order to make clear the usefulness of the TANGENT setup, the authors should explain explicitly and quantitatively how they can put several different concentrations of precursors in flow tube 2, or maintain them at different temperatures, and measure different growth rates, for a constant nucleation rate in flow tube 1. This is exactly what is done in Figure 6 – which is excellent. However, the figure is presented in the text as describing the situation with different temperatures in the two flow tubes. While the temperatures in the flow tubes happened to be different in the measurement presented, the figure actually describes the effect of varying ozone, and no quantitative conclusion about the effect of temperature can be extracted from it. To quantify the effect of temperature in flow tube 2, another figure is needed where the data in Figure 6 are compared to a corresponding measurement in which the two flow tubes are kept at the same temperature.

More generally, the existing plots show the parameter space of nucleation rate vs sulfuric acid, temperature and humidity is quite well explored, but this could have been achieved without the second flow tube and similar measurements were already published by Yu et al (2017). It would be useful to present more measurements where the conditions in the second flow tube are varied with those in the first tube fixed.

Scientific quality

The measurements and calculations of nucleation rate use techniques which have been published previously. The experimental apparatus is described clearly. The quality of the data is therefore reasonably well-established, apart from the lack of measurements of contamination that I already mentioned. I have only a couple of outstanding questions.

What are the temperature and RH dependences of the critical cluster size? Why is critical cluster size equal to the diameter at [H2SO4]=0 (please add reference)?

The survival of the particles in the second flow tube is clearly difficult to disentangle from the strange additional growth via sulfur dioxide and ozone. I don't have any good ideas for why this nucleation happens, beyond the obvious speculations about unmeasured contamination. Could the SO2+O3 reaction be because of a contamination by alkaline material – metallic fragments for example, or enormously high amine or ammonia concentrations – which raise the pH to something like what is seen in sea spray aerosols or cloud droplets?

Presentation quality

The written English is generally of good quality. There are a few missing articles "a" and "the" distributed through the text.

The sentence "Larger mean diameters were detected under lower temperatures for a given [h2SO4]", would imply the method used to determine the critical cluster size would give a larger critical cluster size for lower temperatures. It is clear from theory and from Figure 3 that this is not the case, so the sentence could be rephrased.

Figures 3 and 4 need error bars, if possible, or at least a careful explanation of what the uncertainties are, what is in the noise and what is a real effect.

"Our results thus show that particles were observed at the end of the room temperature nucleation tube after they were initially nucleated at lower temperatures growth tube. These results can explain the presence of newly formed particles observed in Amazon forests by (Wang et al., 2016),…..
"
It is not clear that the second sentence follows from the first. The focus on the Amazon here and in the introduction and conclusion seems odd, since this is one of the few locations on Earth where nucleation may not be dominated by the sulfuric acid clustering that is the subject of this paper. The demonstration that particles survive when the temperature increases is useful, however, and this enabled me to understand the reason for the TANGENT setup. The same mechanism that operates in the Amazon also operates in marine regions, where sulfuric acid nucleated in the upper troposphere survives to make CCN at cloud level (see recent papers by Lynn Russell's group from the North Atlantic, or much earlier work by Tony Clarke and others).