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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Heterogeneous freezing of single sulphuric acid solution droplets: laboratory experiments utilising an acoustic levitator" *by* M. Ettner et al.

Anonymous Referee #1

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This manuscript investigates the heterogeneous freezing of sulphuric acid solutions using an acoustic levitator. Several researchers have investigated heterogeneous freezing of pure water and dilute aqueous solutions, but there have only been a few studies on heterogeneous freezing of concentrated solutions such as sulphuric acid solutions, which are more relevant for ice cloud formation in the upper troposphere. Hence, this manuscript focuses on an important area of research. The paper is well written; however, I do have several comments that should be addressed.

1. In Figure 6 the authors present their homogeneous freezing results. The authors should compare these results with studies from other groups. Also related, Koop et al. [Koop et al., 2000] developed a parameterization for the homogeneous nucleation rate

coefficient of aqueous solutions. From this one can predict when 1.1 mm sulfuric acid particles should freeze homogeneously. I suggest that the authors do this calculation and include it in Figure 6 for comparison.

2. There appears to be a large scatter in the homogeneous freezing temperatures. Since the homogeneous nucleation rate for these solutions, based on previous measurements, is a strong function of temperature, one would not expect this scatter. This suggests that heterogeneous nucleation by dust or other contamination may be influencing the homogeneous measurements. Have the authors repeated the measurements on the same droplets. For example, did the 6 wt % pure H2SO4/H2O particle that froze at -16 C always freeze at this high temperature? This would suggest heterogeneous nucleation.

3. In the experiments presented by the authors the particles are constantly evaporating. This may lead to a cooling of the droplets. Is cooling by this mechanism significant in these experiments? A few lines to address this point would be useful. Could this cause a temperature gradient in the particles?

4. The authors indicate that the ultrasonic fields heat the droplet by approximately 2C. How did they measure this?

5. The authors used graphite as a proxy for soot. Is graphite a good proxy for soot? Some authors suggest that soot contains a graphite core with layers of organic material. Hence, the surface exposed to the aqueous solution may not resemble graphite? The authors should address this in the manuscript.

6. In the conclusions the authors state that soot particles that are directly emitted from aircraft engines into the UT/LS can also influence the freezing of cloud droplets. I do not think the authors can make this conclusion from their results. First, graphite may not be a good proxy for soot from aircraft engines. Second, the authors do not know the number of graphite particles present in each droplet. The number of graphite particles present in each droplet may have been orders of magnitude higher than the number

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present in liquid droplets in the atmosphere. If the authors used a surface area of graphite per droplet similar to the surface area of soot per droplet in the atmosphere, they may not have observed a significant difference in the freezing temperatures. In order to make these conclusions the authors need to consider surface area available for nucleation in their experiments.

Koop, T., B.P. Luo, A. Tsias, and T. Peter, Water activity as the determinant for homogeneous ice nucleation in aqueous solutions, Nature, 406 (6796), 611-614, 2000.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 1887, 2004.