

Interactive comment on “Oxalic acid as a heterogeneous ice nucleus in the upper troposphere and its indirect aerosol effect” by B. Zobrist et al.

B. Zobrist et al.

Received and published: 13 July 2006

The authors would like to thank anonymous referee #2 for his/her constructive comments. We have addressed the referee’s concerns point-by-point below.

Conditioning temperature:

We have performed experiments similar to those of Zuberi et al. 2001 (Zuberi et al., J. Phys. Chem. A, 105, 6458, 2001.) by varying the conditioning temperature for two differently concentrated oxalic acid solutions. However, we did not observe any effect on the heterogeneous ice nucleation temperature, in contrast to the ammonium sulfate case reported by Zuberi et al. We did not include the results of these experiments in

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the original paper, but we will add the following paragraph on page 3577 after line 28 to the revised manuscript: "In a similar study with solid $(\text{NH}_4)_2\text{SO}_4$ as a heterogeneous ice nucleus, Zuberi et al. have found that heterogeneous freezing temperature of ice on ammonium sulfate crystals occurred at warmer temperatures if the conditioning temperature was closer to the $(\text{NH}_4)_2\text{SO}_4 - \text{H}_2\text{O}$ eutectic temperature. The authors concluded that the surface area and surface microstructure of the $(\text{NH}_4)_2\text{SO}_4$ crystals changed with different conditioning temperatures, leading to a change in the heterogeneous freezing temperature. We have performed similar heterogeneous freezing experiments with a 4.95 wt% and a 9.76 wt% oxalic acid solution by varying the conditioning temperature over a range of 277–283 K and 279–293 K, respectively. When the conditioning temperature for the 4.95 wt% and the 9.86 wt% oxalic acid solution was above $T \geq 281$ K and $T \geq 291$ K, respectively, heterogeneous ice nucleation was no longer observed, i.e., a complete melting of the oxalic acid dihydrate crystals had occurred (equivalent to the case of a 2.5 wt% oxalic acid solution in the first row in Table 1). For all other conditioning temperatures we did not observe any change in the heterogeneous ice freezing temperature, indicating that the crystal microstructure is probably unimportant for heterogeneous ice nucleation on oxalic acid dihydrate."

Oxalic acid dihydrate formation:

We agree with the referee that oxalic acid dihydrate most likely crystallizes heterogeneously on the homogeneously nucleating ice crystals. We note, however, that once ice forms the concentration of oxalic acid and, hence, also the supersaturation of oxalic acid dihydrate increases dramatically at temperatures far below the eutectic. This occurs both, in our supermicron laboratory dropets as well as in submicron atmospheric aerosol droplets. We believe this strong increase in supersaturation is so large that size differences become unimportant. For example, reducing the droplet radius (and the corresponding ice particle radius) by a factor of 10, i.e., from $4\mu\text{m}$ in the laboratory to 400 nm in the atmosphere, will reduce the ice surface area by only a factor of 100 and, consequently, also the heterogeneous nucleation rate by the same

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factor. However, the strong increase in concentration when ice forms will most likely increase the nucleation rate coefficient by many orders of magnitude making it very likely that oxalic acid dihydrate will also form in submicron droplets.

The sentence suggested by the referee will be adopted in Table 1.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 3571, 2006.

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