Geophysical Research Abstracts
Vol. 12, EGU2010-7239-2, 2010
EGU General Assembly 2010
© Author(s) 2010

Aqueous aerosol may build up large upper tropospheric ice supersaturation

Anatoli Bogdan (1,3) and Mario J. Molina (2)

(1) Institute of Physical Chemistry, University of Innsbruck, Innsbruck, Austria (anatoli.bogdan@uibk.ac.at), (2) Department of Chemistry and Biochemistry, University of California, San Diego, La Jolla, (3) Division of Atmospheric Sciences, Dept. of Physical Sciences, University of Helsinki, Finland

Keywords: ice supersaturation, upper tropospheric cirrus clouds, freezing of aqueous aerosol.

Observations often reveal enhanced and persistent upper tropospheric (UT) ice supersaturation, Si up to 100%, independently of whether cirrus ice clouds are present or not (Krämer et al., 2009; Lawson et al., 2008). However, a water activity criterion (WAC) (Koop et al., 2000) does not allow the formation of Si $>\sim$67% by the homogeneous freezing of aqueous droplets even at the lowest atmospheric temperature of $\sim$185 K. For aqueous aerosol the WAC predicts the existence of a so called homogeneous ice nucleation threshold which, being expressed as Si, is between $\sim$52 and 67% in the temperature range of $\sim$220 – 185 K. The nature of the formation of large Si remains unclear. Since water vapor is the dominant greenhouse gas it is important to know the nature of the accumulation and persistence of water vapor in the UT.

We studied the freezing behavior of micrometer-scaled 3-, 4-, and 5-component droplets, which contain different weight fractions of H2O, H_2SO_4, HNO_3, $(NH_4)_2SO_4$, $(NH_4)HSO_4$, NH_4NO_3, and $(NH_4)_3H(SO_4)_2$. The study was performed between 133 and 278 K at cooling rates of 3, 0.1, and 0.05 K/min using differential scanning calorimetry (DSC) (Bogdan and Molina, 2010). The cooling rates of 0.1 and 0.05 K/min (6 and 3 K/h) are similar to the smallest reported synoptic temperature change of $\sim$2 K/h (Carslaw et al., 1998). Using the measured freezing temperature of ice, T_i, and the thermodynamic E-AIM model of the system of $H^+ - NH_4^+ - SO_4^{2-} - NO_3^- - H_2O$ (Clegg et al., 1998), we calculated the corresponding clear-sky S_i which would be built up immediately prior to the formation of ice cirrus clouds by the homogeneous freezing of aqueous aerosol of similar composition. We found that our calculated values of S_i are both larger and smaller than the homogeneous ice nucleation threshold. For example, for the droplets of compositions of 15/10 and 20/10 wt % $(NH_4)_3H(SO_4)_2/H_2SO_4$, which freeze at 194 and 186 K, respectively, the calculated clear-sky Si can exceed 80%.

Although our Si values are smaller than the largest observed value of Si $\approx$ 100%, they are nevertheless larger than the Si $\approx$ 67% predicted by the WAC at 185 K. Our results can give an impetus for the study of whether multi-component aqueous aerosol, which besides inorganic components also contains organics, may produce the observed Si $\approx$ 100%.

1. Krämer, M., Schiller, C., Afchine, A., Bauer, R., Gensch, I., Mangold, A.., Schlicht, S., Spelten, N., Sitnikov, N., Borrmann, S., de Reus, M., Spichtinger, P. (2009), Atmos. Chem. Phys. 9, 3505.

2. Lawson, R. P., Pilson, B., Baker, B., Mo, Q., Jensen, E., Pfister, L., Bui, P. (2008), Atmos. Chem. Phys. 8, 1609.

3. Koop, T., Luo, B., Tsias, A., Peter, T. (2000), Nature, 406, 611.

4. Bogdan, A. and Moilna, M. J. (2010), J. Phys. Chem. A (Published online: 5 February).

5. Carslaw, K. S., Wirth, M., Tsias, A., Luo, B. P., Dörnbrack, A., Leutbecher, M., Volkert, H., Renger, W., Bacmeister, J. T., Peter, T. (1998), J. Geophys. Res. 103, 5785.

6. Clegg, S. L., Brimblecombe, P., Wexler, A. S. (1998), J. Phys. Chem. A 102, 2137.