



Heterogeneous ice nucleation in the immersion mode- the role of water activity

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Cirrus clouds cover about 30% of the Earth's surface, affecting chemical and physical processes of the atmosphere. Their presence increases the scattering and absorption of solar radiation as well as the absorption of long wave terrestrial radiation. A change in cirrus cloud coverage may significantly alter the global radiation balance and hence the Earth's climate. So far, the exact mechanisms of cirrus cloud formation are largely unknown. Ice particles in cirrus clouds can form via homogeneous ice nucleation from liquid aerosols or by heterogeneous ice nucleation on solid ice nuclei (IN). It seems likely that solid IN may appear immersed in liquid aerosols in the upper troposphere. Therefore, heterogeneous freezing experiments with four different IN (composed of nonadecanol, silica, silver iodide and oxalic acid) immersed in aqueous solutions have been investigated in a differential scanning calorimeter and in a custom made single droplet apparatus. The results of the different IN show a consistent picture, namely the freezing temperatures are decreasing with decreasing water activity (a_w) of the solution. It was found that a constant offset ($\Delta a_{w,het}$) with respect to the ice melting curve can describe each data series very well, with $\Delta a_{w,het} = 0.101, 0.173, 0.180$ and 0.285 for IN containing nonadecanol, silica, silver iodide and oxalic acid, respectively. Heterogeneous ice nucleation rate coefficients (j_{het}) have been estimated from DSC freezing peaks for the emulsified aqueous solution samples with immersed silver iodide and oxalic acid. The obtained j_{het} values for different aqueous solutions containing the same kind of IN scatter by less than 1 order of magnitude. This implies that for a specific $\Delta a_{w,het}$ a constant j_{het} can be assumed for heterogeneous ice nucleation in the immersion mode. Therefore water activity based nucleation theory can be used to parameterize heterogeneous ice nucleation in the immersion mode analogously to homogeneous ice nucleation from supercooled liquid droplets.