Geophysical Research Abstracts
Vol. 13, EGU2011-9149, 2011
EGU General Assembly 2011
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Temperature and size dependence of volume and surface nucleation rates for homogeneous freezing of supercooled water droplets

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The temperature and size dependences of volume and surface nucleation were investigated for the homogeneous freezing of pure water droplets. Experiments were carried out in a cryogenic laminar aerosol flow tube using supercooled water aerosols with mean radii between about 1 and 3 μm. Temperature- and size-dependent values of volume- and surface-based homogeneous nucleation rates between 234.8 and 236.2 K were derived using a microphysical model and aerosol phase compositions and size distributions determined from infrared extinction measurements in the flow tube. The results show that the contribution from nucleation at the droplet surface increases with decreasing droplet radius and dominates over nucleation in the bulk droplet volume for droplets with radii smaller than approximately 5 μm. This is interpreted in terms of a lowered free energy of ice germ formation in the surface-based process. The implications of surface nucleation for the parameterization of homogeneous ice nucleation in numerical models are considered