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New Particle Formation in the High Arctic: Observations and Model Simulations

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The central Arctic Ocean is an oceanic region far away from sources of anthropogenic sulphur gases. In summer, the region is nearly free from influences of continental or anthropogenic sources. It is generally accepted that the most likely nucleating species under tropospheric conditions are sulphuric acid (H_2SO_4), water (H_2O), and ammonia (NH_3). The formation of sulphate particles often takes place in environments where the available aerosol surface area is low and the condensational loss of nucleating vapours is not favoured. New sulphate particles may also be formed in plumes originating from point sources such as power plants or smelters. The classical homogeneous binary H_2SO_4-H_2O nucleation mechanism could be operational in plumes containing high concentrations of sulphur dioxide (SO_2). Marine particles are probably formed via a different nucleation route than the pollution-associated particles. There have been earlier model studies on the fate of sulphur-containing gas phase and particle compounds during the advection of air from the open sea over the pack ice area of the central Arctic Ocean. However, no previous attempts have been made to simulate observed new particle formation. Comprehensive observation data on gas phase compounds and on chemical composition and size distribution of particles used in this study were obtained during the Arctic Ocean Expedition (AOE-96) to the central Arctic Ocean from beginning of July until end-August of 1996. A sectional aerosol dynamics model, MAFOR, is developed in the frame of this study to predict nucleation in the marine boundary layer. MAFOR includes gas phase chemistry and aerosol dynamics and calculates number and composition distributions of particles as functions of time. Comparison with a well-documented existing aerosol box model (MONO32) and with observational data is used to evaluate the model. Several nucleation mechanisms are implemented in the model. Among the nucleation schemes are the commonly used parameterizations of the classical binary nucleation as well as more recent parameterizations of ternary, ion-induced and kinetic nucleation approaches. In the current implementation growth of newly formed particles to larger sizes can optionally be enhanced by condensation of an arbitrary organic vapour. MAFOR is used to simulate the time series of nucleation mode particle number concentrations (<10 nm diameter) measured at different locations over the pack ice area north of 80°N using different nucleation schemes. Classical homogeneous binary and ternary nucleation theories fail to predict new particle formation in the Arctic. Three nucleation mechanisms are able to predict new particle formation in the Arctic: ion-mediated, kinetic sulphuric acid and cluster activation. Condensation of organic vapour is found to be necessary for the growth of freshly generated particles to observed particle sizes. However only in rare cases do the fresh particles grow to sizes where they could become activated into cloud condensation nuclei (CCN). Thus the new model results lend support to the hypothesis that CCN size particles in the high Arctic are either directly emitted from the open water between ice floes or they are resulting breakdown products of larger particles from biological origin. Implementation of the presented aerosol box model into 3-dimensional atmospheric transport models is facilitated by the splitting of processes and by the efficient integration of gas phase/liquid phase concentrations, particle number and mass concentrations.