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From molecular clusters to nanoparticles: second-generation ion-mediated nucleation model

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Abstract. Ions, which are generated in the atmosphere by galactic cosmic rays and other ionization sources, may play an important role in the formation of atmospheric aerosols. In the paper, a new second-generation ion-mediated nucleation (IMN) model is presented. The new model explicitly treats the evaporation of neutral and charged clusters and it describes the evolution of the size spectra and composition of both charged and neutral clusters/particles ranging from small clusters of few molecules to large particles of several micrometers in diameter. Schemes used to calculate the evaporation coefficients for small neutral and charged clusters are consistent with the experimental data within the uncertainty range. The present IMN model, which is size-, composition-, and type-resolved, is a powerful tool for investigating the dominant mechanisms and key parameters controlling the formation and subsequent growth of nanoparticles in the atmosphere. This model can be used to analyze simultaneous measurements of the ion-mobility spectra and particle size distributions, which became available only recently. General features of the spectra for ions smaller than the critical size, size-dependent fractions of charged nanoparticles, and asymmetrical charging of freshly nucleated particles predicted by the new IMN model are consistent with recent measurements. Results obtained using the second generation IMN model, in which the most recent thermodynamic data for neutral and charged $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ clusters were used, suggest that ion-mediated nucleation of $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ can lead to a significant production of new particles in the lower atmosphere (including the boundary layer) under favorable conditions. It has been shown that freshly nucleated particles of few nanometers in size can grow by the condensation of low volatile organic compounds to the size of cloud condensation nuclei. In such cases, the chemical composition of nucleated particles larger than ~ 10 nm is dominated by organics.

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