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Modeling the possible role of iodine oxides in atmospheric new particle formation

S. Pechtl[1], E. R. Lovejoy[2], J. B. Burkholder[2], and R. von Glasow[1]
[1]Institute for Environmental Physics, University of Heidelberg, Heidelberg, Germany
[2]Aeronomy Laboratory, National Oceanic and Atmospheric Administration, Boulder, CO, USA

Abstract. We studied the possible role of iodine oxides in atmospheric new particle formation with the one-dimensional marine boundary layer model MISTRA, which includes chemistry in the gas and aerosol phase as well as aerosol microphysics. The chemical reaction set focuses on halogen (Cl-Br-I) chemistry. We included a two-step nucleation parameterization, where in the first step, the "real" nucleation process is parameterized, i.e., the formation of cluster-sized nuclei via homogeneous condensation of gases. We considered both ternary sulfuric acid-ammonia-water nucleation and homomolecular homogeneous OIO nucleation. For the latter, we derived a parameterization based on combined laboratory-model studies. The second step of the nucleation parameterization treats the "apparent" nucleation rate, i.e., the growth of clusters into the model's lowest size bin by condensable vapors such as OIO. We compared different scenarios for a clean marine versus a polluted continental background atmosphere. In every scenario, we assumed the air to move, independent of its origin, first over a coastal region (where it is exposed to surface fluxes of different reactive iodine precursors) and later over the open ocean. According to these sensitivity studies, in the clean marine background atmosphere OIO can be responsible for both homogeneous nuclei formation and the subsequent growth of the clusters to detectable sizes. In contrast to this, in the continental case with its higher levels of pollutants, gas phase OIO mixing ratios, and hence related nucleation rates, are significantly lower. Compared to ternary H_2SO_4-NH_3-H_2O nucleation, homogeneous OIO nucleation can be neglected for new particle formation in this case, but OIO can contribute to early particle growth, i.e., to apparent nucleation rates. In general, we found OIO to be more important for the growth of newly formed particles than for the formation of new nuclei. According to our studies, observations of particle "bursts" can only be explained by hot spot-like, not by homogeneously distributed emissions.

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