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Stochastic effects in H_2SO_4-H_2O cluster growth

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Based on a recently developed particle Monte Carlo (MC) Code [C. Köhn, M. Enghoff and H. Svensmark, 2018. A 3D particle Monte Carlo approach to studying nucleation. J. Comp. Phys., vol. 363, pp. 30–38], we here analyse how the growth of sulfuric acid-water clusters is influenced by stochastic fluctuations. We present the temporal evolution of the nucleation rate and of the size distribution as well as growth rates and the onset time of the nucleation above a given cluster size with and without constant production of new monomers. We consider samples of H_2SO_4-H_2O clusters at T=200 K, with particle concentrations between 10^5 cm^{-3} and 10^7 cm^{-3} in volumes between 10^{-6} cm^3 and 10^{-2} cm^3. Simulations are performed with a MC particle code following individual clusters. After every time step, we update the position of each cluster as a function of size-dependent diffusion coefficients and check for cluster collisions enabling cluster growth. Inversely, we check after every time step whether a polymer evaporates based on evaporation coefficients from literature [Yu, 2005. J. Chem. Phys., vol. 122, 074501; Yu, 2006. Atmos. Chem. Phys., vol. 6, 5193–5211]. Conclusively, we find clear evidence of fluctuations which are not apparent in deterministic continuum models. Recent research [Olenius et al, Nature Scientific Reports 8:14160, 2018] have shown that such stochastic processes can influence the early stages of growth which are critical for the survival rate of aerosol particles. We here find that fluctuations in the MC code result in a faster growth rate of the smallest clusters compared to models that do not contain the stochastic effects.