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Laboratory studies of H_2SO_4/H_2O binary homogeneous nucleation from the SO_2+OH reaction: evaluation of the experimental setup and preliminary results

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Abstract. Binary homogeneous nucleation (BHN) of sulphuric acid and water (H_2SO_4/H_2O) is one of the most important atmospheric nucleation processes, but laboratory observations of this nucleation process are very limited and there are also large discrepancies between different laboratory studies. The difficulties associated with these experiments include wall loss of H_2SO_4 and uncertainties in estimation of H_2SO_4 concentration ($[H_2SO_4]$) involved in nucleation. We have developed a new laboratory nucleation setup to study H_2SO_4/H_2O BHN kinetics and provide relatively constrained $[H_2SO_4]$ needed for nucleation. H_2SO_4 is produced from the $SO_2+OH \rightarrow HSO_3$ reaction and OH radicals are produced from water vapor UV absorption. The residual $[H_2SO_4]$ were measured at the end of the nucleation reactor with a chemical ionization mass spectrometer (CIMS). Wall loss factors (WLFs) of H_2SO_4 were estimated by assuming that wall loss is diffusion limited and these calculated WLFs were in good agreement with simultaneous measurements of the initial and residual $[H_2SO_4]$ with two CIMSs. The nucleation zone was estimated from numerical simulations based on the measured aerosol sizes (particle diameter, D_p) and $[H_2SO_4]$. The measured BHN rates (J) ranged from 0.01–220 cm^{-3} s^{-1} at the initial and residual $[H_2SO_4]$ from 10^8–10^{10} cm^{-3}, a temperature of 288 K and relative humidity (RH) from 11–23%; J increased with increasing $[H_2SO_4]$ and RH. J also showed a power dependence on $[H_2SO_4]$ with the exponential power of 3–8. These power dependences are consistent with other laboratory studies under similar $[H_2SO_4]$ and RH, but different from atmospheric field observations which showed that particle number concentrations are often linearly dependent on $[H_2SO_4]$. These results, together with a higher $[H_2SO_4]$ threshold (10^8–10^9 cm^{-3}) needed to produce the unit J measured from the laboratory studies compared to the atmospheric conditions (10^6–10^7 cm^{-3}), imply that H_2SO_4/H_2O BHN alone is insufficient to explain atmospheric aerosol formation and growth. Particle growth rates estimated from the measured aerosol size distributions, residence times (t_r), and $[H_2SO_4]$ were 100–500 nm h^{-1}, much higher than those seen from atmospheric field observations, because of the higher $[H_2SO_4]$ used in our study.

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