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Laboratory studies of H₂SO₄/H₂O binary homogeneous nucleation from the SO₂+OH reaction: evaluation of the experimental setup and preliminary results

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Abstract. Binary homogeneous nucleation (BHN) of sulphuric acid and water (H₂SO₄/H₂O) 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 H₂SO₄ and uncertainties in estimation of H₂SO₄ concentration ([H₂SO₄]) involved in nucleation. We have developed a new laboratory nucleation setup to study H₂SO₄/H₂O BHN kinetics and provide relatively constrained [H₂SO₄] needed for nucleation. H₂SO₄ is produced from the SO₂+OH→HSO₃ reaction and OH radicals are produced from water vapor UV absorption. The residual [H₂SO₄] were measured at the end of the nucleation reactor with a chemical ionization mass spectrometer (CIMS). Wall loss factors (WLFs) of H₂SO₄ 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 [H₂SO₄] with two CIMSs. The nucleation zone was estimated from numerical simulations based on the measured aerosol sizes (particle diameter, D_p) and [H₂SO₄]. The measured BHN rates (J) ranged from 0.01–220 cm⁻³ s⁻¹ at the initial and residual [H₂SO₄] from 10⁸–10¹⁰ cm⁻³, a temperature of 288 K and relative humidity (RH) from 11–23%; J increased with increasing [H₂SO₄] and RH. J also showed a power dependence on [H₂SO₄] with the exponential power of 3–8. These power dependences are consistent with other laboratory studies under similar [H₂SO₄] and RH, but different from atmospheric field observations which showed that particle number concentrations are often linearly dependent on [H₂SO₄]. These results, together with a higher [H₂SO₄] threshold (10⁸–10⁹ cm⁻³) needed to produce the unit J measured from the laboratory studies compared to the atmospheric conditions (10⁶–10⁷ cm⁻³), imply that H₂SO₄/H₂O BHN alone is insufficient to explain atmospheric aerosol formation and growth. Particle growth rates estimated from the measured aerosol size distributions, residence times (t_p), and [H₂SO₄] were 100–500 nm h⁻¹, much higher than those seen from atmospheric field observations, because of the higher [H₂SO₄] used in our study.

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