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Sulphuric acid closure and contribution to nucleation mode particle growth

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Abstract. Sulphuric acid concentrations were measured and calculated based on pseudo steady state model with corresponding measurements of CO, NO_x, O₃, SO₂, methane and non-methane hydrocarbon (NMHC) concentrations as well as solar spectral irradiance and particle number concentrations with size distributions. The measurements were performed as a part of the EU project QUEST (Quantification of Aerosol Nucleation in the European Boundary layer) during an intensive field campaign, which was conducted in Hyytiälä, Finland in March–April 2003. In this paper, the closure between measured and calculated H₂SO₄ concentrations is investigated. Besides that, also the contribution of sulphuric acid to nucleation mode particle growth rates is studied. Hydroxyl and hydroperoxy radical concentrations were determined using a pseudo steady state box model including photo stationary states. The maximum midday OH concentrations ranged between 4.1×10⁵ to 1.8×10⁶ molecules cm⁻³ and the corresponding values for HO₂ were 1.0×10⁷ to 1.5×10⁸ molecules cm⁻³. The dominant source term for hydroxyl radicals is the reaction of NO with HO₂ (56%) and the reaction of CO with OH covers around 41% of the sinks. The sulphuric acid source term is the reaction SO₂ with OH and the sink term is condensation of sulphuric acid. The closure between measured and calculated sulphuric acid concentrations is achieved with a high agreement to the measured values. In sensitivity studies, we used different values for the non-methane hydrocarbons, the peroxy radicals and nitrogen dioxide. The best fits between calculated and measured values were found by decreasing the NO₂ concentration when it exceeded values of 1.5 ppb and doubling the non-methane hydrocarbon concentrations. The ratio, standard deviation and correlation coefficient between measured and calculated sulphuric acid concentrations are 0.99, 0.412 and 0.645, respectively. The maximum midday sulphuric acid concentrations varied between 3×10⁵ to 1.9×10⁷ molecules cm⁻³ for the measurements and 3×10⁵ to 1.4×10⁷ molecules cm⁻³ for the calculations, respectively. An average participation of sulphuric acid to the nucleation mode particle growth rates is 8.8%. Classifying the days into two groups –

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