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Atmospheric sulphuric acid and aerosol formation: implications from atmospheric measurements for nucleation and early growth mechanisms

S.-L. Sihto¹, M. Kulmala¹, V.-M. Kerminen², M. Dal Maso¹, T. Petäjä¹, I. Riipinen¹, H. Korhonen², F. Arnold³, R. Janson⁴, M. Boy¹, A. Laaksonen⁵, and K. E. J. Lehtinen⁶

¹University of Helsinki, Department of Physical Sciences, P.O. Box 64, 00014 University of Helsinki, Finland

²Finnish Meteorological Institute, Climate and Global Change, Erik Palménin Aukio 1, P.O. Box 503, 00101 Helsinki, Finland

³Atmospheric Physics Division, Max-Planck Institute for Nuclear Physics (MPIK), P.O. Box 103980, 69029 Heidelberg, Germany

⁴University of Stockholm, Department of Applied Environmental Science, Air Pollution Laboratory, Frescativägen 54, 10691 Stockholm, Sweden

⁵University of Kuopio, Department of Applied Physics, P.O. Box 1627, 70211 Kuopio, Finland

⁶Finnish Meteorological Institute and University of Kuopio, Department of Applied Physics, P.O. Box 1627, 70211 Kuopio, Finland

Abstract. We have investigated the formation and early growth of atmospheric secondary aerosol particles building on atmospheric measurements. The measurements were part of the QUEST 2 campaign which took place in spring 2003 in Hyytiälä (Finland). During the campaign numerous aerosol particle formation events occurred of which 15 were accompanied by gaseous sulphuric acid measurements. Our detailed analysis of these 15 events is focussed on nucleation and early growth (to a diameter of 3 nm) of fresh particles. It revealed that new particle formation seems to be a function of the gaseous sulphuric acid concentration to the power from one to two when the time delay between the sulphuric acid and particle number concentration is taken into account. From the time delay the growth rates of freshly nucleated particles from 1 nm to 3 nm were determined. The mean growth rate was 1.2 nm/h and it was clearly correlated with the gaseous sulphuric acid concentration. We tested two nucleation mechanisms – recently proposed cluster activation and kinetic type nucleation – as possible candidates to explain the observed dependences, and determined experimental nucleation coefficients. We found that some events are dominated by the activation mechanism and some by the kinetic mechanism. Inferred coefficients for the two nucleation mechanisms are the same order of magnitude as chemical reaction coefficients in the gas phase and they correlate with the product of gaseous sulphuric acid and ammonia concentrations. This indicates that besides gaseous sulphuric acid also ammonia has a role in nucleation.

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