Atmospheric Chemistry and Physics
An Interactive Open Access Journal of the European Geosciences Union

Atmos. Chem. Phys., 6, 5631-5648, 2006
www.atmos-chem-phys.net/6/5631/2006/

The contribution of boundary layer nucleation events to total particle concentrations on regional and global scales

D. V. Spracklen[1,*], K. S. Carslaw[1], M. Kulmala[2], V.-M. Kerminen[3], G. W. Mann[1], and S.-L. Sihto[2]

[1]School of Earth and Environment, University of Leeds, UK

[2]University of Helsinki, Department of Physical Sciences, P.O. Box 64, 00014 University of Helsinki, Finland

[3]Finnish Meteorological Institute, Climate and Global Change, Erik Palmenin aukio 1, P.O. Box 503, 00101 Helsinki, Finland

[*]now at: Atmospheric Chemistry Modelling Group, Harvard University, Cambridge, MA, USA

Abstract. The contribution of boundary layer (BL) nucleation events to total particle concentrations on the global scale has been studied by including a new particle formation mechanism in a global aerosol microphysics model. The mechanism is based on an analysis of extensive observations of particle formation in the BL at a continental surface site. It assumes that molecular clusters form at a rate proportional to the gaseous sulfuric acid concentration to the power of 1. The formation rate of 3 nm diameter observable particles is controlled by the cluster formation rate and the existing particle surface area, which acts to scavenge condensable gases and clusters during growth. Modelled sulfuric acid vapour concentrations, particle formation rates, growth rates, coagulation loss rates, peak particle concentrations, and the daily timing of events in the global model agree well with observations made during a 22-day period of March 2003 at the SMEAR II station in Hyytiälä, Finland. The nucleation bursts produce total particle concentrations (>3 nm diameter) often exceeding 10^4 cm^{-3}, which are sustained for a period of several hours around local midday. The predicted global distribution of particle formation events broadly agrees with what is expected from available observations. Over relatively clean remote continental locations formation events can sustain mean total particle concentrations up to a factor of 8 greater than those resulting from anthropogenic sources of primary organic and black carbon particles. However, in polluted continental regions anthropogenic primary particles dominate particle number and formation events lead to smaller enhancements of up to a factor of 2. Our results therefore suggest that particle concentrations in remote continental regions are dominated by nucleated particles while concentrations in polluted continental regions are dominated by primary particles. The effect of BL particle formation over tropical regions and the Amazon is negligible. These first global particle formation simulations reveal some interesting sensitivities. We show, for example, that significant reductions in primary particle emissions may lead to an increase in total particle concentration because of the coupling between particle surface area and the rate of new particle formation. This result suggests that changes in emissions may have a complicated effect on global and regional aerosol properties. Overall, our results show that new particle formation is a significant component of the aerosol particle number budget.

Final Revised Paper (PDF, 8899 KB)   Discussion Paper (ACPD)

Citation: Spracklen, D. V., Carslaw, K. S., Kulmala, M., Kerminen, V.-M., Mann, G. W., and Sihto, S.-L.: The contribution of boundary layer nucleation events to total particle concentrations on regional and global scales, Atmos. Chem. Phys., 6, 5631-5648, 2006.   Bibtex   EndNote   Reference Manager