



Contribution of particle formation to global cloud condensation nuclei concentrations

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[1] We use a global aerosol microphysics model to predict the contribution of boundary layer (BL) particle formation to regional and global distributions of cloud condensation nuclei (CCN). Including an observationally derived particle formation scheme, where the formation rate of molecular clusters is proportional to gas-phase sulfuric acid to the power one, improves modeled particle size distribution and total particle number concentration at three continental sites in Europe. Particle formation increases springtime BL global mean CCN (0.2% supersaturation) concentrations by 3–20% and CCN (1%) by 5–50%. Uncertainties in particle formation and growth rates must be reduced before the accuracy of these predictions can be improved. These results demonstrate the potential importance of BL particle formation as a global source of CCN. **Citation:** Spracklen, D. V., et al. (2008), Contribution of particle formation to global cloud condensation nuclei concentrations, *Geophys. Res. Lett.*, 35, L06808, doi:10.1029/2007GL033038.

1. Introduction

[2] Aerosols absorb and scatter radiation and influence the properties of clouds through a subset of the aerosol population, which act as cloud condensation nuclei (CCN). Both primary and secondary (nucleated) aerosol particles are potential sources of CCN number, but the relative contribution of each to regional and global CCN is unknown.

[3] Boundary Layer (BL) particle formation events have been observed at many locations around the world ranging from the sub-Arctic through boreal forests to polluted industrial and coastal regions [Kulmala et al., 2004b]. Particles nucleated at nanometer sizes must undergo considerable growth before they can act as CCN, and the loss by various scavenging processes limits the number that grow that large [Pierce and Adams, 2007]. Nevertheless, formation events have been shown to make an important contribution to local CCN concentrations through examination of observed aerosol size distributions before and after a nucleation event [Lihavainen et al., 2003; Kerminen et al., 2005; Laaksonen et al., 2005]. Regional models have shown that particle formation (from a ternary NH₃-H₂SO₄-H₂O nucleation mechanism) can increase CCN concentrations

(at 1% supersaturation) locally over parts of Europe by 40–100% and regionally by 1–10% over a 3 day period [Sotiropoulou et al., 2006]. In Spracklen et al. [2006] we used a global aerosol model to show that nucleation events enhance total particle number concentrations in the remote continental BL by a factor of 2–8 greater than concentrations from primary sources and upper tropospheric (UT) nucleation alone. Here, we extend this analysis to explore the contribution of BL particle formation to regional and global CCN concentrations.

2. Model Description

[4] We use the GLOMAP aerosol microphysics model [Spracklen et al., 2005a, 2005b] which is an extension to the TOMCAT 3-D chemical transport model [Chipperfield, 2006]. GLOMAP has a horizontal resolution of ~2.8° by ~2.8°, 31 vertical levels between the surface and 10 hPa and is forced by ECMWF analyses. GLOMAP includes sulfate (SU), sea-salt (SS), elemental carbon (EC) and organic carbon (OC). We treat two externally mixed distributions, each described by a two-moment sectional scheme with 20 sections spanning 3 nm to 25 μm dry diameter. One distribution, representing freshly emitted primary carbonaceous aerosol, contains OC and EC, is treated as hydrophobic and is not wet scavenged. The other distribution contains SU, SS, EC and OC, is hydrophilic and is wet scavenged. We assume that the first-stage oxidation products of monoterpenes [Guenther et al., 1995] form hydrophilic secondary organic aerosol (SOA) with a yield of 13% [Spracklen et al., 2006]. This yield is important for particle growth to CCN sizes and is increased in a sensitivity study. Hydrophobic particles age to become hydrophilic through condensation of soluble gas-phase species and coagulation with hydrophilic particles.

[5] Implementation of the nucleation scheme in GLOMAP is described by Spracklen et al. [2006]. The formation rate of 1 nm molecular clusters is given by

$$j_1 = A[H_2SO_4] \quad (1)$$

where [H₂SO₄] is the gas-phase sulfuric acid concentration and A is an empirical activation coefficient [Kulmala et al., 2006]. The formation rate of 3 nm particles is calculated using the expression of Kerminen and Kulmala [2002], which takes into account loss of 1 nm clusters onto the existing particles during growth. Particles at 3 nm are added to the smallest size bin of the model. As in the work by Spracklen et al. [2006], we restrict this mechanism to the BL and at higher levels use the binary homogeneous

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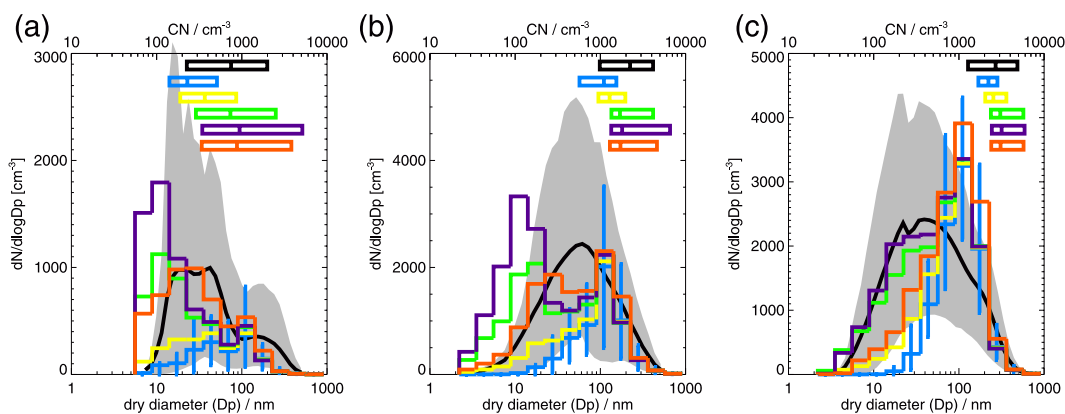


Figure 1. Monthly-mean number-size distributions and total particle number concentrations at (a) Pallas ($67^{\circ}57'N$, $24^{\circ}17'E$); (b) Hyytiälä ($61^{\circ}51'N$, $24^{\circ}7'E$) and (c) Hohenpeissenberg ($47^{\circ}48'N$, $11^{\circ}E$) in April 2000. Observations from CREATE (black), model without (blue) and with BL particle formation (yellow: activation coefficient $A = 2 \times 10^{-8} \text{ s}^{-1}$; green: $A = 2 \times 10^{-7} \text{ s}^{-1}$; purple: $A = 2 \times 10^{-6} \text{ s}^{-1}$, red: $A = 2 \times 10^{-6} \text{ s}^{-1}$ and SOA yield from monoterpenes increased from 13% to 65%). The 10th to 90th percentile variability is shown for the observations (shading) and model (vertical bars). Horizontal boxes show 5th, 50th and 95th percentiles of total particle number concentration, plotted against upper x-axis.

nucleation rate of *Kulmala et al.* [1998]. Evaluation of observed nucleation events suggests that A varies spatially and temporally by over an order of magnitude. In Hyytiälä, Finland (during March–April), calculated values of A varied from $3.3 \times 10^{-8} \text{ s}^{-1}$ to $6.0 \times 10^{-6} \text{ s}^{-1}$ whereas in Heidelberg, Germany (February–April), A varied from $3.2 \times 10^{-6} \text{ s}^{-1}$ to $3.5 \times 10^{-4} \text{ s}^{-1}$ [Sihto et al., 2006; Riipinen et al., 2007]. The reasons for this variability are not understood. Here, we investigate the sensitivity of CCN formation to this uncertainty by varying A between $2 \times 10^{-9} \text{ s}^{-1}$ and $2 \times 10^{-4} \text{ s}^{-1}$. We restrict our study to Northern Hemisphere springtime (March–May), which is the time of year for which extensive evaluation of formation events has occurred.

3. Results

[6] Figure 1 shows number size distributions and total particle number concentrations at 3 surface sites in Europe (Pallas, Finland [Tunved et al., 2003]; Hyytiälä [Tunved et al., 2003]; and Hohenpeissenberg, Germany [Birmili et al., 2003]). Observations are from the CREATE database (<http://www.nilu.no/projects/ccc/create/database.htm>) recorded using Differential Mobility Particle Sizers (DMPS). At Pallas the observed size range is 7–530 nm, Hyytiälä 3–550 nm and Hohenpeissenberg 3–730 nm. Model values are plotted for the same size range as the observations.

[7] The model without BL particle formation, where aerosol is from primary sources and UT nucleation, underpredicts total particle number concentration at all the sites due to an underprediction at diameters less than 100 nm. With BL particle formation the model better reproduces both the median and variability in total particle number concentrations. At Hohenpeissenberg, overprediction of the accumulation mode may be due to problems with primary particle emissions. At Pallas and Hyytiälä the model underpredicts the growth of small particles, probably due to underprediction of SOA which can contribute significantly

to particle growth [Tunved et al., 2006]. The modeled size distribution at these boreal forest sites is improved (Figure 1, red line) by increasing the SOA yield from terpenes by a factor of 5 (from 13% to 65%). The impact of uncertainty in SOA is explored below. The value of activation coefficient A that results in the best match with observations is between $2 \times 10^{-7} \text{ s}^{-1}$ and $2 \times 10^{-6} \text{ s}^{-1}$, consistent with values calculated from individual particle formation events [Riipinen et al., 2007].

[8] Simulated CCN concentrations are calculated using the modeled particle dry diameter and composition and a hygroscopicity parameter, κ [Petters and Kreidenweis, 2007]. We assume that the hydrophobic distribution does not contribute to CCN formation. For the hydrophilic distribution we take values of κ from Petters and Kreidenweis [2007]: SU $\kappa = 0.61$ (assuming ammonium sulfate), SS $\kappa = 1.28$, EC $\kappa = 0.0$, OC $\kappa = 0.1$ (assuming SOA from α -pinene). Simulated surface CCN concentrations without BL particle formation at 0.2% (1.0%) supersaturations are typically $50\text{--}200 \text{ cm}^{-3}$ ($100\text{--}500 \text{ cm}^{-3}$) over oceans and between $100\text{--}1000 \text{ cm}^{-3}$ ($300\text{--}2000 \text{ cm}^{-3}$) over land (Figure 2a and b) with a global surface mean value of 250 cm^{-3} (380 cm^{-3}) respectively. Modeled CCN concentrations are typically within a factor of 2–3 of observations (Figures 2a and 2b).

[9] Two model runs, with and without BL particle formation, are used to quantify the effect of particle formation on CCN (Figures 2c and 2d). All model runs include particle growth due to SOA. With an activation coefficient $A = 2 \times 10^{-6} \text{ s}^{-1}$, particle formation increases the global BL springtime (March–May) CCN (0.2%) concentration by 9% and CCN (1%) by 20% over concentrations predicted from primary sources and UT nucleation. CCN enhancements of 10–50% over Scandinavia and large regions of boreal Asia and larger enhancements over parts of the continental US, Australia and southern Africa are simulated. It is worth noting that the regional changes can be negative where particle formation rates are high and growth of these particles alters the condensation sink, thereby reducing the condensational growth of exist-

