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**Explaining global
aerosol number
concentrations**

D. V. Spracklen et al.

Explaining global surface aerosol number concentrations in terms of primary emissions and particle formation

D. V. Spracklen¹, K. S. Carslaw¹, J. Merikanto¹, G. W. Mann¹, S. Pickering¹, J. A. Ogren², E. Andrews², U. Baltensperger³, E. Weingartner³, M. Boy⁴, M. Kulmala⁴, L. Laakso⁴, H. Lihavainen⁵, N. Kivekäs⁵, N. Mihalopoulos⁶, G. Kouvarakis⁶, S. G. Jennings⁷, W. Birmili⁸, A. Wiedensohler⁸, R. Weller⁹, P. Laj¹⁰, K. Sellegri¹¹, B. Bonn¹², and R. Krejci¹³

¹Institute for Climate and Atmospheric Science, School of Earth and Environment, University of Leeds, LS2 9JT, UK

²NOAA/ESRL Global Monitoring Division, 325 Broadway R/GMD1, Boulder, Co 80305, USA

³Paul Scherrer Institut, Laboratory of Atmospheric Chemistry, 5232 Villigen, Switzerland

⁴Department of Physical Sciences, University of Helsinki, Finland

⁵Climate Change, Finnish Meteorological Institute, Helsinki, Finland

⁶Department of Chemistry, University of Crete, University campus, P.O. Box 2208, 71003, Voutes, Heraklion, Crete, Greece

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⁸ Leibniz Institute for Tropospheric Research, Permoserstrasse 15, 04318 Leipzig, Germany

⁹ Alfred Wegener Institute, Am Handelshafen 12, 27570 Bremerhaven

¹⁰ Laboratoire de Glaciologie et Géophysique de l'Environnement CNRS/Université Grenoble 1, Grenoble, France

¹¹ Laboratoire de Météorologie Physique, Université Clermont-Ferrand/CNRS, Clermont-Ferrand, France

¹² J.W. Goethe University, Inst. for Atmospheric and Environmental Sciences, Frankfurt/Main, Germany

¹³ Department of Applied Environmental Science (ITM), Stockholm University, 10691 Stockholm, Sweden

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Correspondence: D. V. Spracklen (dominick@env.leeds.ac.uk)

Abstract

We use observations of total particle number concentration at 36 worldwide sites and a global aerosol model to quantify the primary and secondary sources of particle number. We show that emissions of primary particles can reasonably reproduce the spatial pattern of observed condensation nuclei (CN) ($R^2=0.51$) but fail to explain the observed seasonal cycle at many sites ($R^2=0.1$). The modeled CN concentration in the free troposphere is biased low (normalised mean bias, NMB= -88%) unless a secondary source of particles is included, for example from binary homogeneous nucleation of sulfuric acid and water (NMB= -25%). Simulated CN concentrations in the continental boundary layer (BL) are also biased low (NMB= -74%) unless the number emission of anthropogenic primary particles is increased or an empirical BL particle formation mechanism based on sulfuric acid is used. We find that the seasonal CN cycle observed at continental BL sites is better simulated by including a BL particle formation mechanism ($R^2=0.3$) than by increasing the number emission from primary anthropogenic sources ($R^2=0.18$). Using sensitivity tests we derive optimum rate coefficients for this nucleation mechanism, which agree with values derived from detailed case studies at individual sites.

1 Introduction

There are two sources of particles in the atmosphere. Primary particles are emitted directly to the atmosphere and secondary particles are formed from gas-to-particle conversion. The relative contribution of these two sources to global aerosol is poorly understood, but it is important to quantify if we are to understand long-term aerosol trends and the impact of aerosol on climate. Here we use observations of total particle number concentration at sites around the world together with a global aerosol model to understand the sources of particle number.

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Primary particles are emitted directly to the atmosphere, for example from biomass burning, combustion of fossil fuels and uplift of sea-spray and dust from the Earth's surface. Neither the emission strength nor the size distribution of primary particles is well known. A small uncertainty in the size distribution of primary particles (for a given mass) leads to large uncertainty in both the number emission and atmospheric number concentration of particles (Spracklen et al., 2005b).

Secondary particles formed from particle formation are observed at many surface locations around the world and also within the free and upper troposphere (Kulmala et al., 2004). Particle formation events are associated with a rapid increase in the number concentration of ultrafine particles followed by particle growth. Such events have been observed at sites ranging from Antarctica (Koponen et al., 2003), Arctic (Vehkamäki et al., 2004), boreal forest (Mäkelä et al., 1997; Kulmala et al., 1998b; Dal Maso et al., 2005), suburban and urban regions (Birmili and Wiedensohler, 2000; Gaydos et al., 2005) and coastal environments (O'Dowd et al., 1999). Several observations using multiple measurement stations have shown that formation events can occur more or less uniformly in air masses extending over hundreds of kilometres (Kulmala et al., 1998b, 2001; Vana et al., 2004; Komppula et al., 2006). Large concentrations of ultrafine (<20 nm) particles have also been observed in the free and upper troposphere (e.g., Clarke et al., 1998, 1999; Weingartner et al., 1999) and frequent new particle formation has now also been recorded at high altitudes (Venzac et al., 2008). The importance of secondary particle formation on a global scale is uncertain, though it may make a significant contribution to both global condensation nuclei (CN) (Spracklen et al., 2006) and cloud condensation nuclei (CCN) concentrations (Spracklen et al., 2008; Kuang et al., 2009; Makkonen et al., 2009; Pierce and Adams, 2009; Wang and Penner, 2009).

Despite such frequent occurrence in the atmosphere, and potential importance to global aerosol, the underlying mechanism of new particle formation is not known. Several mechanisms have been proposed including binary homogeneous nucleation (BHN) of $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ (Jaecker-Voirol and Mirabel, 1988; Kulmala et al., 1998a;

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Vehkamäki et al., 2002), ternary nucleation (TN) of $\text{H}_2\text{SO}_4\text{-H}_2\text{O-NH}_3$ (Napari et al., 2002) and ion-induced nucleation (Modgil et al., 2005). BHN is strongly temperature dependent and only produces new particles in the cold free and upper troposphere (Weber et al., 1999; Adams and Seinfeld, 2002; Spracklen et al., 2005a; Lucas and Akimoto, 2006). Revised parameterizations of ternary nucleation (Yu, 2006; Anttila et al., 2005; Merikanto et al., 2007) also suggest that this mechanism is not a significant source of particles in the lower troposphere (Elleman and Covert, 2009). Neither of these mechanisms is therefore likely to explain particle formation observed in the boundary layer (BL). Ion-induced nucleation potentially contributes to both free troposphere (FT) and BL particle formation (Kazil et al., 2006; Yu et al., 2008). However, some studies (Kulmala et al., 2007; Boy et al., 2008) suggest that ion-induced nucleation contributes less than 10% of particle formation in the continental BL.

While our mechanistic understanding of nucleation remains so uncertain, an alternative approach has been used to explain observed particle formation and to estimate the impact of nucleation on global aerosol properties. Observations of surface particle formation events have been analysed to develop empirical particle formation mechanisms. The formation rate of molecular clusters (1 nm to 1.5 nm in diameter) is found to be proportional to the gas-phase sulfuric acid concentrations to the power 1 to 2 (Weber et al., 1996; Kulmala et al., 2006; Sihto et al., 2006; Riipinen et al., 2007; Kuang et al., 2008). The power one dependence of the cluster formation rate on sulfuric acid has been described in terms of an activation mechanism (Kulmala et al., 2006) where sulfuric acid particles are stabilized by a secondary species such as organics. A power two dependence has been explained by proposing a kinetic nucleation mechanism (McMurry and Friedlander, 1979).

Global aerosol microphysics models can now prognose CN and CCN concentrations for the first time (e.g., Adams and Seinfeld, 2002, 2003; Easter et al., 2004; Pierce and Adams, 2006; Pierce et al., 2007; Spracklen et al., 2005a,b; Stier et al., 2005) and are being used to understand the atmospheric processes controlling global aerosol. There is therefore an urgent need to evaluate these new models against observations. The

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significant uncertainties associated both with the treatment of primary emissions and particle formation mean that model skill may be lacking. The size of primary particles emitted in global models must take into account the size distribution of the primary particles at the point of emission as well as the ageing that occurs at sub-model grid scales (Pierce et al., 2009). Because the mechanisms of atmospheric new particle formation are unknown there are also significant uncertainties in the global secondary particle formation rate (Spracklen et al., 2008).

Remote marine boundary layer (MBL) particle number size distributions can largely be explained by a combination of entrainment of particles from the FT (produced by particle formation in the upper troposphere) and primary sea-salt emissions, although there are significant regional discrepancies between model and observations (Easter et al., 2004; Spracklen et al., 2005a, 2007; Pierce and Adams, 2006; Trivitayanurak et al., 2008). Vertical profiles of CN over the remote MBL can also be explained by BHN (Spracklen et al., 2005a).

Simulated CN concentrations in the continental BL are greatly enhanced by primary sulfate (Adams and Seinfeld, 2002, 2003; Spracklen et al., 2005b; Wang et al., 2009), carbonaceous aerosol (Pierce et al., 2007) and BL particle formation (Spracklen et al., 2006, 2008; Makkonen et al., 2009; Chang et al., 2009). While Pierce and Adams (2009) show that observed CN at continental BL sites can be largely explained by primary emissions alone, other studies suggest a large contribution from particle formation (Spracklen et al., 2006, 2008; Makkonen et al., 2009).

In this study, we use observations of total particle number concentration together with a global aerosol model to better understand the contribution of particle formation and primary emissions to global aerosol number. We compile measurements of the total particle number concentration, reported for particle sizes larger than a few nanometers (typically 3–10 nm in diameter), from 36 surface sites around the world. Although such small particles are not directly relevant to climate, our dataset is the largest available for evaluating aerosol microphysics models and to understand the sources of global aerosol number. Furthermore, it is thought that subsequent growth

of these particles to larger sizes may contribute significantly to global concentrations of CCN (e.g., Spracklen et al., 2008; Kuang et al., 2009) making them indirectly important to climate.

2 Model Description

2.1 General

We use the GLOMAP aerosol microphysics model (Spracklen et al., 2005a,b), which is an extension of the TOMCAT 3-D global chemical transport model (Chipperfield, 2006) to simulate sulfate (SU), sea salt (SS), elemental carbon (EC) and organic carbon (OC) for the year 2000. Large-scale transport and meteorology is specified from 6-h European Centre for Medium-Range Weather Forecasts (ECMWF) analyses. We use a horizontal resolution of $\sim 2.8^\circ$ by $\sim 2.8^\circ$ and 31 vertical levels between the surface and 10 hPa.

GLOMAP treats the particle size distribution using either a two-moment modal (Manktelow et al., 2007) or a two-moment sectional (bin) (Spracklen et al., 2005a) scheme. Here we use the sectional scheme and treat two externally mixed distributions, each described with 20 sections spanning 3 nm to $10 \mu\text{m}$ dry diameter. One distribution, representing freshly emitted primary carbonaceous aerosol, contains OC and EC, is treated as non-hydrophilic and is not wet scavenged. The other distribution contains SU, SS, EC and OC, is hydrophilic and is wet scavenged. Non-hydrophilic particles age to become hydrophilic through condensation of soluble gas-phase species and coagulation with hydrophilic particles. Total simulated particle number is the sum of particles in these two distributions. The microphysical processes in the model include nucleation, coagulation, condensation of gas-phase species, in-cloud and below-cloud aerosol scavenging and deposition, dry deposition and cloud processing.

Concentrations of OH, O_3 and NO_3 and HO_2 are specified using 6-h monthly mean 3-D concentrations from a TOMCAT simulation with detailed tropospheric chemistry

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(Arnold et al., 2005). Concentrations of H_2O_2 are calculated as described in Spracklen et al. (2005a).

Oceanic DMS emissions are calculated using the ocean surface DMS concentration database of Kettle and Andreae (2000) and the sea-to-air transfer velocity according to Nightingale et al. (2000). Emissions of biogenic terpenes are from the GEIA inventory (Benkovitz et al., 1996) and are based on Guenther et al. (1995). Emissions of SO_2 and carbonaceous aerosol from wildfires, biofuel, fossil fuel and volcanoes are based on the AEROCOM¹ emission inventories for the year 2000 (Dentener et al., 2006).

Secondary organic aerosol (SOA) from biogenic terpenes is included assuming the reactivity of alpha-pinene including reactions with OH, O_3 and NO_3 (Spracklen et al., 2006). We assume all three reactions have a constant yield of 13% to a first-stage oxidation product that condenses with zero vapour pressure onto existing aerosol (Spracklen et al., 2006). We add SOA to the hydrophilic aerosol distribution.

2.2 Primary emissions and particle formation

We test the sensitivity of the model to the emission of primary particles and the nucleation rate through a series of experiments detailed below and in Table 1.

1. Experiment PRI includes anthropogenic and natural primary particulate emissions but no secondary particle formation. Primary sea-spray emissions are based on Gong et al. (2003). We assume that a fraction of sulfur dioxide emissions are emitted as primary sulfate either because these particles are directly emitted to the atmosphere or because they are formed through gas-to-particle conversion at spatial scales smaller than our model grid. Emissions of primary sulfate are a standard option in many aerosol models (e.g., Adams and Seinfeld, 2002; Spracklen et al., 2005a; Stier et al., 2005), however the properties of the particles are very uncertain. Primary sulfate and carbonaceous (EC/OC) particles are emitted assuming lognormal modes, which are mapped onto the model size bins. We emit primary sulfate at the rate (2.5% of SO_2

¹Available at: <http://nansen.ipsl.jussieu.fr/AEROCOM/>

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emissions) and using the size distribution suggested by AEROCOM (road transport: number median radius, $r=15$ nm, $\sigma=1.8$; shipping, industry and power-plant emissions: $r=500$ nm, $\sigma=2.0$; wildfire and biofuel: $r=40$ nm, $\sigma=1.8$; volcanic emissions: 50% at $r=15$ nm and 50% at $r=40$ nm, $\sigma=1.8$). For EC/OC emissions we use the emission size distribution suggested by Stier et al. (2005) (fossil fuel emissions: $r=30$ nm, $\sigma=1.59$, wildfire and biofuel emissions: $r=75$ nm and $\sigma=1.59$).

2. Experiment BHN includes identical primary emissions to PRI, but additionally it includes particle formation assuming binary homogeneous $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ nucleation using the parameterization of Kulmala et al. (1998a).

3. Experiments PRICAR and PRISUL test the sensitivity of the model to uncertainty in the assumed size distribution of the primary emissions. Experiment PRICAR uses the AEROCOM size distribution for emissions of EC/OC particles (Dentener et al., 2006), emitting EC/OC particles at about half the diameter of experiment PRI (fossil fuel: $r=15$ nm, $\sigma=1.8$, biofuel and wildfire: $r=40$ nm, $\sigma=1.8$) and thereby increasing the emitted EC/OC number by about a factor 8. Experiment PRISUL assumes that anthropogenic primary sulfate particles (transport, shipping, industry and power-plant emissions) are emitted at the size distribution according to Adams and Seinfeld (2003) (15% of emitted mass at $r=5$ nm, $\sigma=1.6$, 85% at $r=35$ nm, $\sigma=2.0$).

4. Experiments ACT and KIN are as experiment BHN, but additionally they include an empirical particle formation mechanism (Kulmala et al., 2006; Sihto et al., 2006) in the BL. This mechanism is based on analysis of surface particle formation events. The formation rate (J_1) of 1-nm particles is given by:

$$J_1 = A[\text{H}_2\text{SO}_4]^M \quad (1)$$

where A is the nucleation coefficient. The value of M has been found to vary between 1 and 2 and A varies spatially and temporally, for reasons which are not known. We perform model simulations for both $M=1$ (activation mechanism, ACT) and $M=2$ (kinetic mechanism, KIN) and with a range of nucleation coefficients. In each model experiment the magnitude of the nucleation coefficient is fixed globally.



The particle formation rate at 3 nm (J_3) is obtained from J_1 and the equation of Kerminen and Kulmala (2002):

$$J_3 = J_1 \exp\left(-0.153 \frac{CS'}{GR}\right), \quad (2)$$

where CS' is the reduced condensation sink (m^{-2}) and GR (nm h^{-1}) is the cluster growth rate. GR is assumed to be constant between 1 nm and 3 nm and is calculated from the gas-phase sulfuric acid concentration. Newly formed particles are added to the model at 3 nm diameter. A full description of this mechanism in the GLOMAP model is described in Spracklen et al. (2006).

The empirical particle formation mechanism is based on observation of particle formation events made at the surface and there is some evidence to suggest that formation events are less likely in the lower FT (Heintzenberg et al., 2008; O'Dowd et al., 2009). Observations of the vertical profile of CN number concentrations typically show maxima at the surface and in the UT and a minimum in the lower FT (Clarke and Kapustin, 2002; Schröder et al., 2002; Singh et al., 2002). Applying the empirical particle formation mechanism throughout the depth of the troposphere in our model does not capture this observed profile and results in overprediction of CN concentrations in the lower FT. For this reason, as in earlier work (Spracklen et al., 2006, 2008), we restrict the empirical particle formation mechanism to the BL while allowing BHN to occur above (which results in particle formation in the UT).

3 Simulation of surface CN number concentrations

Simulated surface CN (diameter >3 nm) concentrations for the different experiments described in Sect. 2.2 are shown in Table 1 and Fig. 1. With no particle formation (PRI), surface annual mean (median) CN concentrations are 105 (30) cm^{-3} over the oceans and 640 (107) cm^{-3} over the continents. Greater simulated concentrations over the continents are due to primary anthropogenic emissions. BHN increases surface mean

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(median) CN concentrations by a factor 3.3 (8.4) over the oceans and factor 1.5 (5.7) over the continents. The absolute increase in mean CN at the surface due to BHN is relatively uniform across the globe (average of 250 cm^{-3} over oceans and 310 cm^{-3} over continental regions). This is because the secondary source of particles from BHN is largely in the upper troposphere (UT) and somewhat independent of SO_2 sources at the surface (Raes et al., 2000). Increasing the number emission of primary EC/OC particles (PRICAR) further increases (over experiment BHN) mean (median) surface CN concentrations by a factor 1.4 (1.1) over the oceans and a factor 2.3 (1.4) over the continents. Increasing the number emissions of primary sulfate particles (PRISUL) increases CN concentrations by a factor 1.7 (1.1) over the oceans and a factor 2.4 (1.2) over the continents. The empirical BL particle formation scheme (with $M=1$ and $k=2 \times 10^{-6} \text{ s}^{-1}$) increases mean (median) surface CN concentrations by a factor of 2.1 (1.9) over the oceans and by a factor of 1.8 (1.4) over the continents. Surface CN concentrations are sensitive to the nucleation coefficient used: a factor 100 change in the BL nucleation coefficient (A in Eq. 1) changes J_1 by a factor 100 and increases the surface CN concentration by a factor of 2.3. In contrast, surface CN are relatively insensitive to changes in the BHN nucleation rate, changing by less than 40% for factor 100 change in the BHN rate (Spracklen et al., 2005b). This is because BHN occurs primarily in the UT and coagulation that occurs during transport and mixing of air to the surface reduces the sensitivity of surface CN to changes in BHN rate. The sensitivity of surface CN concentrations to the BL particle formation rate allows us to use observed CN concentrations and a global model to give a constraint to the global nucleation rate as we show below (Sect. 4.2).

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4 Model evaluation

4.1 The CN dataset

To evaluate the model we compiled a dataset of surface CN number concentrations recorded at 36 sites around the world. The locations of the sites are shown in Fig. 2 and Table 2. We broadly classified the sites into FT, MBL and continental BL. We did not include any sites within urban environments because the resolution of our model is not sufficient to resolve urban-scale pollution. We only included sites which have recorded CN concentrations for at least a 12 month period. At sites with more than 12 months of data we report monthly mean and median concentrations as an average over the multiannual dataset. The time period of observations used at each site is shown in Table 2. To give an indication of the interannual variability in the observations we report the standard deviation in monthly mean CN concentrations at sites where there are 4 or more years of data available.

Observations of CN were made either using condensation particle counters (CPCs), scanning mobility particle sizers (SMPS) or differential mobility particle sizers (DMPS). CPCs record total particle number concentration typically for sizes greater than 3 nm or 10 nm in diameter. SMPS and DMPS record the particle size distribution at sizes greater than either 3 nm or 10 nm in diameter. From the observed size distribution a CN concentration was calculated. We compared model experiments for each site using the same lower cutoff diameter as available from the observations. All CN concentrations are reported at ambient temperature and pressure.

All the MBL sites in this analysis are at coastal locations which are influenced by continental emissions (except Samoa which is at a very remote site in the tropical Pacific and probably experiences minimal continental influence). We did not screen the data in an attempt to remove continental influence (e.g., Reade et al., 2006) as this complicates analysis with a 3-D Eulerian model.

High altitude sites located near the top of mountains are often influenced by thermal winds resulting in diurnal cycles in aerosol (Venzac et al., 2009). Additionally, diurnal

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and seasonal variation of BL height means mountain sites can be located in either the BL or FT at different times of the day or year. For these reasons the data from high altitude sites may not represent background FT conditions without detailed screening which was not performed here. Nevertheless, the model does simulate variations in BL height though they have not been evaluated specifically at the locations analysed in this study.

We linearly interpolate the model to the horizontal location of the observations. Where sub-grid topography means that the altitude above sea-level of the observation site is not within 500 m of the grid-point elevation of the model surface level (TOMCAT uses hybrid σ -pressure coordinates), we interpolate the model in the vertical to match the altitude of the observations.

4.2 Global analysis of annual mean CN number concentrations

Figure 3 compares simulated and observed annual mean CN number concentrations. Observed annual mean CN concentrations at MBL and FT sites are less than 2000 cm^{-3} . The lowest concentrations are observed at the South Pole in Antarctica (100 cm^{-3}), Samoa in the tropical MBL (300 cm^{-3}), coastal Antarctica (350 cm^{-3}) and Point Barrow in the Arctic (450 cm^{-3}). The mid-latitude MBL sites in this analysis are at coastal locations influenced by continental aerosol sources and have CN concentrations of between 950 cm^{-3} and 1750 cm^{-3} . We did not filter out such a continental influence and so the MBL concentrations we report may not be representative for the open ocean. Observed CN concentrations at continental BL sites are up to an order of magnitude greater, spanning 800 cm^{-3} to 7000 cm^{-3} .

Figure 3 also shows the equation of the linear regression fits between model and observations. Model bias between is further quantified as a normalised mean bias ($\text{NMB} = 100 \times \sum_{i=1}^n (S_i - O_i) / \sum_{i=1}^n O_i$, where S_i is the simulated annual mean CN concentration and O_i is the observed annual mean CN concentration at site i) shown in Table 3.

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The experiment with natural and anthropogenic primary particulate emissions but no secondary particle formation (PRI) captures the spatial pattern of CN concentrations relatively well ($R^2=0.51$) but underestimates concentrations at all sites (slope of the linear regression, $m=0.29$, $NMB=-77\%$). Simulated concentrations at continental BL sites are underpredicted by a factor of 2–10, and FT and MBL sites are underpredicted by up to a factor of 10 or more.

Including binary homogeneous nucleation in the model (BHN) increases simulated BL CN concentrations by 200–400 cm^{-3} and FT CN by up to 1000 cm^{-3} but results in only a modest reduction in model bias across all sites ($m=0.32$, $NMB=-61\%$). Simulated concentrations are most improved at FT sites ($NMB=-25\%$) and annual mean particle number at FT sites are mostly predicted within a factor of 2. CN concentrations at MBL sites are still underpredicted ($NMB=-61\%$). However, annual mean CN concentrations are well simulated at Samoa which is the only remote MBL site in this analysis.

Increasing the number emission of primary anthropogenic particulate emissions (by reducing the assumed diameter of the emissions) increases simulated BL CN concentrations by as much as 10 000 cm^{-3} . The impact in the BL is regionally dependent: at sites close to anthropogenic sources (e.g., Finokalia, Hohenpeissenberg, Bondville, South Great Plains) simulated CN increases by up to a factor of 2.5, whereas at remote BL sites (e.g., South Pole, Point Barrow, Samoa, Cape Grim) they increase by less than 30%. Increasing EC/OC number emission (PRICAR) results in a smaller model bias ($m=0.93$, $NMB=-12\%$) whereas increasing primary sulfate (PRISUL) results in model overprediction ($m=1.47$, $NMB=34\%$). Such a large increase in simulated CN concentrations due to primary emissions has been seen in previous work such as Pierce and Adams (2009).

Including BL particle formation increases annual mean CN concentrations at BL sites by several thousand cm^{-3} , similar to that simulated by increasing anthropogenic primary emissions. With the activation (ACT) particle formation mechanism, model bias varies from -44% to 17% with the best match between model and observations

occurring using a nucleation coefficient of between $A=2\times 10^{-6}$ and $A=2\times 10^{-5}\text{ s}^{-1}$. With the kinetic (KIN) particle formation mechanism, model bias varies between -35% and 31% with the best match occurring when $A=2\times 10^{-12}\text{ cm}^3\text{ s}^{-1}$. These derived nucleation rate coefficients lie within the range of values calculated from observed particle formation rates: activation mechanism $A=3.3\times 10^{-8}\text{ s}^{-1} - 3.5\times 10^{-4}\text{ s}^{-1}$ (Riipinen et al., 2007); kinetic mechanism $A=2.4\times 10^{-15}\text{ cm}^3\text{ s}^{-1} - 1.3\times 10^{-10}\text{ cm}^3\text{ s}^{-1}$ (Riipinen et al., 2007; Kuang et al., 2008).

Our analysis provide support for previous work (Riipinen et al., 2007; Kuang et al., 2008) that the magnitude of the BL nucleation rate coefficient varies spatially. Observed CN concentrations at Arctic and boreal forest sites (Pallas and Hyytiälä) are well matched by the model with $A=2\times 10^{-6}\text{ s}^{-1}$ ($M=1$) or $2\times 10^{-13}\text{ cm}^3\text{ s}^{-1}$ ($M=2$). Sites in the Midwest US (Bondville and South Great Plains) would require a nucleation coefficient greater than $A=2\times 10^{-5}\text{ s}^{-1}$ ($M=1$) to match observed CN. The reason for variability in the nucleation rate coefficient is unknown, but could include varying concentrations of other atmospheric species that can stabilise sulfate clusters. It has been suggested that organic species may be a key candidate (Verheggen et al., 2007; Bonn et al., 2008). The model underpredicts CN concentrations at mid-latitude MBL sites even with the largest nucleation coefficient used in this analysis. This may be due to alternative nucleation mechanisms being important in these locations (O'Dowd et al., 2002).

The spatial pattern of annual mean CN concentrations can be best captured by either increasing the number emission of primary anthropogenic emissions, PRICAR ($R^2=0.57$) and PRISUL ($R^2=0.60$), or through including BL particle formation, ACT2 ($R^2=0.58$) and KIN2 ($R^2=0.57$). Previous studies, using a smaller dataset than we compiled here, have also shown that it is difficult to constrain particle formation and primary emissions using annual mean particle number concentrations. To provide a stronger constraint on primary emissions and particle formation we therefore examined the observed seasonal cycle of particle number.

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4.3 Accounting for the seasonal cycle in CN

Figures 4, 5 and 6 show the seasonal cycle of observed and simulated CN concentrations at FT, MBL and continental BL sites. At many locations a pronounced seasonal cycle is observed, with summer-time CN concentrations exceeding winter-time concentrations by a factor of 2–10. For example, at the South Pole summertime CN concentrations are about 250 cm^{-3} whereas wintertime concentrations are less than 20 cm^{-3} . In Pallas, Finland, summertime CN concentrations are about 1250 cm^{-3} whereas wintertime concentrations are about 250 cm^{-3} .

Table 4 shows the correlation coefficient between simulated and observed monthly mean CN concentrations for sites where monthly mean CN concentrations vary by more than a factor of 2 throughout the seasonal cycle. Figure 7 shows the average correlation coefficients for these sites calculated separately for FT, MBL and continental BL sites.

The model experiment with only primary particulate emissions (PRI) does not capture the observed seasonal cycle demonstrated by the poor correlation coefficient between simulated and observed monthly mean CN concentrations at FT ($R^2=0.09$), MBL ($R^2=0.14$) and continental BL sites ($R^2=0.11$). With BHN the model captures the observed seasonal cycle at some MBL (e.g., Neumayer) and FT sites (e.g., South Pole, Mt. Washington) resulting in a better correlation in both the FT ($R^2=0.26$) and MBL ($R^2=0.36$). While winter-time concentrations are relatively well simulated at some BL sites (e.g., Point Barrow, Hyttiälä and Pallas) underprediction during summer months results in a poorly represented seasonal cycle in the continental BL ($R^2=0.1$). Increasing primary emissions degrades model representation of the seasonal cycle in the FT (PRICAR, $R^2=0.12$, PRISUL, $R^2=0.22$) and MBL (PRICAR, $R^2=0.21$, PRISUL, $R^2=0.36$), while slightly improving simulation in the continental BL (PRICAR, $R^2=0.14$, PRISUL, $R^2=0.18$). Including BLN particle formation improves the simulated seasonal cycle at continental BL sites (ACT2, $R^2=0.25$; KIN2 $R^2=0.30$) whilst maintaining

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or improving representation of the seasonal cycle in the FT (ACT2, $R^2=0.27$; KIN2 $R^2=0.36$) and MBL (ACT2, $R^2=0.34$; KIN2 $R^2=0.43$).

4.4 Additional remote marine CN measurements

To extend the evaluation of the model to the remote MBL we use the dataset of Heintzenberg et al. (2000) who compiled observations from several field campaigns including ACE-1, ACE-2, ACE-Asia and INDOEX. Heintzenberg et al. (2000) fitted observed aerosol size distributions with two lognormal modes and gridded the observations into 15° latitude bands. The different field campaigns used a range of sampling instruments with lower cutoff diameters ranging from 3 nm to 12 nm, but this information was not captured by the analysis.

Figure 8 shows CN concentrations from the Heintzenberg et al. (2000) dataset compared with our model. We plot simulated CN (diameter >12 nm), which over the oceans is virtually identical to simulated CN (diameter >3 nm), except when BL particle formation is included in the model. For this experiment we plot both CN (diameter >3 nm) and CN (diameter >12 nm).

With primary emissions only (PRI) the model underestimates CN concentrations at all latitudes except 30° – 45° N. When binary homogeneous nucleation is included (BHN) the model still underpredicts CN concentrations in the Southern Hemisphere (between 15° S and 75° S) and Arctic (between 75° N and 90° N) oceans while either well simulating or overpredicting CN elsewhere in the Northern Hemisphere. Such underprediction in the Southern Hemisphere is a consistent feature of global aerosol models (Easter et al., 2004; Spracklen et al., 2005a,b; Pierce and Adams, 2006; Pierce et al., 2007; Trivitayanurak et al., 2008; Wang et al., 2009). Underprediction of CN above 75° N is likely due to the model not well simulating the transport of anthropogenic pollution to the Arctic (Korhonen et al., 2008). Increasing the number emission from primary anthropogenic sources results in overprediction of CN between 15° N and 60° N, while the underprediction of CN persists in the Southern Hemisphere and Arctic. Including

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BL particle formation increases simulated CN, particularly in regions of SO₂ emissions from shipping and DMS emissions from the ocean. In the Southern Hemisphere, BL particle formation scheme improves the simulation of CN, whereas in the Northern Hemisphere it results in overprediction of CN except in the Arctic where the model underprediction of CN remains. Emissions of ultrafine sea-spray (Mårtensson et al., 2003), not included in this work, may be another explanation for underprediction of CN in the Southern Ocean (Pierce and Adams, 2006).

5 Summary and conclusions

We have used the GLOMAP global aerosol microphysics model to simulate global CN number concentrations and evaluated the model against observations compiled from 36 surface sites around the globe. Our aim was to evaluate the role of primary particulate emissions and particle formation in controlling regional and global surface CN concentrations.

We conducted a range of experiments with different assumptions about primary anthropogenic emissions and particle formation. Without secondary particle formation (primary particulate emissions only) the model greatly underpredicts observed CN concentrations (NMB=-77%). While the spatial pattern of observed CN is relatively well captured ($R^2=0.51$) the model does not capture the seasonal cycle observed at FT ($R^2=0.09$), MBL ($R^2=0.14$) or continental BL sites ($R^2=0.11$).

Including binary homogeneous nucleation of H₂SO₄-H₂O (Kulmala et al., 1998a) in addition to primary emissions reduces model bias at FT sites (NMB=-25%) and improves the simulated seasonal cycle in the MBL ($R^2=0.36$) and FT ($R^2=0.26$). However, CN is still underpredicted in the continental BL resulting in little improvement in model bias (NMB=-66%) or representation of the seasonal cycle ($R^2=0.1$) at these sites.

There is considerable uncertainty in the size distribution of primary anthropogenic particulate emissions (which affects the particle number emitted for fixed mass) and

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previous studies have used a range of assumptions. We therefore evaluated the sensitivity of simulated CN to the uncertainty in the anthropogenic primary sulfate and carbonaceous aerosol size distributions. Reducing the emissions size of primary carbonaceous (PRICAR) or sulfate (PRISUL) particles (and so increasing the number emission) within the range assumed by previous studies slightly improves the spatial pattern of simulated CN (PRICAR, $R^2=0.57$; PRISUL, $R^2=0.6$) and reduces the model bias (PRICAR, NMB=-12%; PRISUL, NMB=34%) (these experiments included particle formation through binary homogeneous nucleation). However, scaling the anthropogenic primary emissions in this way does not improve the simulated seasonal cycle, slightly degrading model performance in the MBL (PRICAR, $R^2=0.21$; PRISUL, $R^2=0.36$) and FT (PRICAR, $R^2=0.12$; PRISUL, $R^2=0.22$) while only slightly improving the model in the continental BL (PRICAR, $R^2=0.14$; PRISUL, $R^2=0.18$).

We tested an empirical nucleation mechanism which assumes that the formation rate of 1-nm nuclei is proportional to sulfuric acid concentration to the power 1 (activation mechanism) or 2 (kinetic mechanism). We limited the empirical mechanism to the BL and allowed binary homogeneous nucleation to occur above. When the model included a combination of these mechanisms along with primary emissions the model captured both the spatial pattern of annual mean CN (activation, $R^2=0.58$; kinetic, $R^2=0.57$) and the seasonal cycle of CN in the FT (activation, $R^2=0.27$; kinetic, $R^2=0.36$), MBL (activation, $R^2=0.34$; kinetic, $R^2=0.43$) and continental BL (activation, $R^2=0.25$; kinetic, $R^2=0.30$).

The simulated nucleation rate depends on the nucleation rate coefficient, A , the magnitude of which is poorly known. We constrained this coefficient by running multiple experiments with different rates and finding the experiment that most reduced the model bias. The best agreement was with $A=2\times 10^{-6} \text{ s}^{-1}$ to $A=2\times 10^{-5} \text{ s}^{-1}$ for the activation scheme and with $A=2\times 10^{-12} \text{ cm}^{-3} \text{ s}^{-1}$ for the kinetic scheme, which agree with the rates derived from observed particle formation events (Riipinen et al., 2007; Kuang et al., 2008).

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5 The applicability of the BL particle formation mechanism over oceanic regions re-
quires further analysis. Observed CN concentrations at a remote tropical MBL site
(Samoa) is well matched by the model with binary homogeneous nucleation. All the
other MBL sites used in this analysis are at coastal locations influenced by continental
emissions and therefore may not well represent remote MBL conditions unless a de-
tailed screening of the observations is performed. The model underpredicts CN at
some coastal locations (e.g., Mace Head) even with the fastest BL nucleation rates
used in this study. This may be due to alternate nucleation mechanisms driven by io-
dine compounds being important in these locations (O'Dowd et al., 2002; Vuollekoski
10 et al., 2009).

Long-term datasets of the seasonal variability in CN are not available for open ocean
regions away from continental influence. In place of such observations we used a com-
pilation of data from several field campaigns that has been gridded into 15° oceanic
latitude bands (Heintzenberg et al., 2000). Without particle formation the model greatly
15 underpredicts CN throughout the global oceans. As seen in previous studies, mod-
els that include primary emissions and binary homogeneous nucleation consistently
underpredict CN concentrations in the Southern Ocean whilst either well predicting
or overpredicting CN in the Northern Hemisphere. Over the Northern Hemisphere
oceans, BL nucleation results in further overprediction of CN, whereas over the South-
20 ern Ocean BL particle formation improves simulated CN concentrations.

We have shown that particle formation is important for global aerosol concentrations.
In Merikanto et al. (2009) we use the BL particle formation rate constrained in this study
to quantify the contribution of primary particles and particle formation to global CN and
CCN.

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Table 1. Simulated annual mean (median) surface CN ($D_p > 3\text{ nm}$) concentrations (at ambient temperature and pressure) for the year 2000 for model experiments with different primary emissions and particle formation (see Sect. 2.2 for details).

Experiment Name	Size distribution of primary emissions		Nucleation mechanisms	Surface mean (median) CN	
	EC/OC	Sulfate		Ocean/cm ⁻³	Land/cm ⁻³
PRI	Stier et al. (2005)	AEROCOM	None	105 (30)	642 (107)
BHN	Stier et al. (2005)	AEROCOM	BHN	352 (252)	952 (604)
PRICAR	AEROCOM	AEROCOM	BHN	495 (265)	2230 (839)
PRISUL	Stier et al. (2005)	Adams and Seinfeld (2003)	BHN	595 (267)	2282 (721)
ACT1	Stier et al. (2005)	AEROCOM	BHN + Activation ($A=2\times 10^{-7}\text{ s}^{-1}$)	474 (323)	1262 (703)
ACT2	Stier et al. (2005)	AEROCOM	BHN + Activation ($A=2\times 10^{-6}\text{ s}^{-1}$)	729 (483)	1743 (868)
ACT3	Stier et al. (2005)	AEROCOM	BHN + Activation ($A=2\times 10^{-5}\text{ s}^{-1}$)	1157 (660)	2653 (1018)
KIN1	Stier et al. (2005)	AEROCOM	BHN + Kinetic ($A=2\times 10^{-13}\text{ cm}^3\text{ s}^{-1}$)	557 (323)	1498 (725)
KIN2	Stier et al. (2005)	AEROCOM	BHN + Kinetic ($A=2\times 10^{-12}\text{ cm}^3\text{ s}^{-1}$)	798 (426)	2003 (813)
KIN3	Stier et al. (2005)	AEROCOM	BHN + Kinetic ($A=2\times 10^{-11}\text{ cm}^3\text{ s}^{-1}$)	1181 (550)	2764 (878)

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Table 2. Observation sites used in this analysis.

Station Name	Location	Altitude (m)	Observation period	Minimum cutoff diameter (nm)	Data source ¹	Reference
Free troposphere						
Jungfrauoch	8.0° E, 46.6° N	3580	1995–1999, 2003–2007	10	E	Weingartner et al. (1999)
Puy de Dome	3.0° E, 45.8° N	1465	2005–2008	3	E	Venzac et al. (2009)
Nepal C.O.	86.8° E, 28.0° N	5079	2007–2008	10	P	Venzac et al. (2008)
Mauna Loa	155.6° W, 19.5° N	3397	1975–2000	14	W	Bodhaine (1983)
South Pole	24.8° W, 90° S	2841	1974–1999	14	W	Bodhaine (1983)
Pico Espejo	71.1° W, 8.5° N	4775	2007–2009	10	P	
Mount Washington	71.3° W, 44.3° N	1910	2002–2005	10	A	
Mount Waliguan	100.9° E, 36.3° N	3816	2005–2007	13	P	Kivekäs et al. (2009)
Zugspitze	11.0° E, 47.4° N	2650	2005–2008	12	P	
Marine boundary layer						
Mace Head	9.9° W, 53.3° N	5	2000, 2002–2007	10	E	O'Dowd et al. (1998)
Neumayer	8.3° W, 70.7° S	42	1993–2006	14	W	Weller and Lampert (2008)
Point Barrow	156.6° W, 71.3° N	11	1994–2007	14	W	Delene and Ogren (2002)
Samoa	170.6° W, 14.2° S	77	1977–2006	14	W	Bodhaine (1983)
Trinidad Head	124.2° W, 41.1° N	107	2002–2007	14	W	McComiskey et al. (2003)
Cape Grim	144.7° E, 40.6° S	94	1996–2007	3	W	Gras (1995)
Sable Island	60.0° W, 43.9° N	5	1992–2000	10	P	Delene and Ogren (2002)
Continental boundary layer						
Hyytiälä	24.3° E, 61.9° N	180.	2000–2004	3	C	Aalto et al. (2001)
Pallas	24.1° E, 68.0° N	340.	2000–2004, 2007	10	E	Komppula et al. (2003)
Finokalia	25.7° E, 35.3° N	250	1997, 2006–2007	10	E	
Hohenpeissenberg	11.0° E, 47.8° N	995	2006–2007	3	E	Birmili et al. (2003)
Melpitz	12.3° E, 51.2° N	86	1996–1997, 2003	3	C	
Bondville	88.4° W, 40.1° N	213	1994–2007	14	W	Delene and Ogren (2002)
Southern Great Plains	97.5° W, 36.6° N	320	1996–2007	10	W	Delene and Ogren (2002)
Tomsik	85.1° E, 56.5° N	170	2005–2006	3	PD	Dal Maso et al. (2008b)
Listvyanka	104.9° E, 51.9° N	750	2005–2006	3	PD	Dal Maso et al. (2008b)
Harwell	359.0° E, 51.0° N	60	2000	10	U	Charron et al. (2007)
Weybourne	1.1° E, 53.0° N	0	2005	10	B	
Botsalano	25.8° E, 25.5° S	1424	7/2006–6/2007	10	PD	Laakso et al. (2008)
India Himilaya	79.6° E, 29.4° N	2180	2005–2008	10	PD	Komppula et al. (2009)
Aspvreten	17.4° E, 58.8° N	25	2000–2006	10	PD	Dal Maso et al. (2008a)
Utö	21.4° E, 59.8° N	8	2003–2006	7	PD	Dal Maso et al. (2008a)
Värriö	29.6° E, 67.8° N	400	1998–2006	8	PD	Dal Maso et al. (2008a)
Thompson Farm	289.1° E, 43.1° N	75	2001–2009	7	A	Ziamba et al. (2007)
Castle Springs	71.3° W, 43.7° N	406	2001–2008	7	A	
Tannus Observatory	8.4° E, 50.2° N	810	2008–2009	10	P	
Po Valley	11.6° W, 44.7° N	11	2002–2006	3	P	Hamed et al. (2007)

¹ AIRMAP, <http://airmap.unh.edu/data/> (A); BADC, <http://badc.nerc.ac.uk/home/> (B); CREATE, <http://tarantula.nilu.no/projects/ccc/create/> (C); EBAS, <http://ebas.nilu.no> (E); Personal communication (P); Published data, see citation (PD); UK National Air Quality Archive, <http://www.airquality.co.uk/> (U); World Data Centre for Aerosols (WDCA), <http://wdca.jrc.ec.europa.eu/> (W).

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Table 3. Normalised mean bias between observed and simulated CN concentrations. Model experiments are described in Table 1, locations and time periods of the observations are shown in Table 2.

Stations	Model experiments									
	PRI	BHN	PRICAR	PRISUL	ACT1	ACT2	ACT3	KIN1	KIN2	KIN3
All	−76%	−62%	−12%	37%	−44%	−24%	17%	−31%	−9%	35%
FT	−88%	−25%	−6%	−18%	−9%	14%	53%	5%	28%	68%
MBL	−85%	−61%	−28%	11%	−40%	−12%	23%	−27%	−3%	21%
BL	−74%	−66%	−15%	46%	−48%	−29%	12%	−35%	−13%	31%

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Table 4. Correlation coefficient (R^2) between observed and simulated monthly mean CN concentrations at sites where monthly mean concentrations vary by more than a factor of two throughout a seasonal cycle. Model experiments are described in Table 1.

Site	PRI	BHN	PRICAR	PRISUL	ACT2	KIN2
Free troposphere						
Puy de Dome	0.01	0.01	0.05	0.07	0.19	0.06
Nepal C.O.	0.01	0.01	<0.01	0.11	0.18	0.37
South Pole	0.08	0.65	0.44	0.69	0.71	0.72
Mt. Washington	0.25	0.38	0.01	0.02	0.02	0.29
Marine boundary layer						
Neumayer	0.02	0.74	0.57	0.62	0.71	0.68
Point Barrow	0.04	0.03	0.06	0.02	0.07	0.04
Sable Island	0.37	0.31	<0.01	0.43	0.25	0.57
Continental boundary layer						
Hyytiälä	0.13	0.01	0.06	0.26	0.39	0.43
Pallas	0.12	0.19	0.07	0.25	0.55	0.59
Melpitz	0.06	0.02	0.05	0.07	0.28	0.41
Tomsk	0.14	0.05	0.26	<0.01	0.04	<0.01
Listvyanka	0.09	0.05	0.20	<0.01	0.13	0.07
Harwell	0.25	0.10	0.12	0.33	0.40	0.48
Weybourne	<0.01	0.08	0.03	0.02	0.05	0.03
India Himilaya	0.18	0.40	0.25	0.61	<0.01	0.01
Utö	0.08	0.03	0.26	0.10	0.10	0.30
Värriö	0.03	<0.01	<0.01	0.06	0.56	0.65
Tannus	0.21	0.14	0.27	0.25	0.21	0.29

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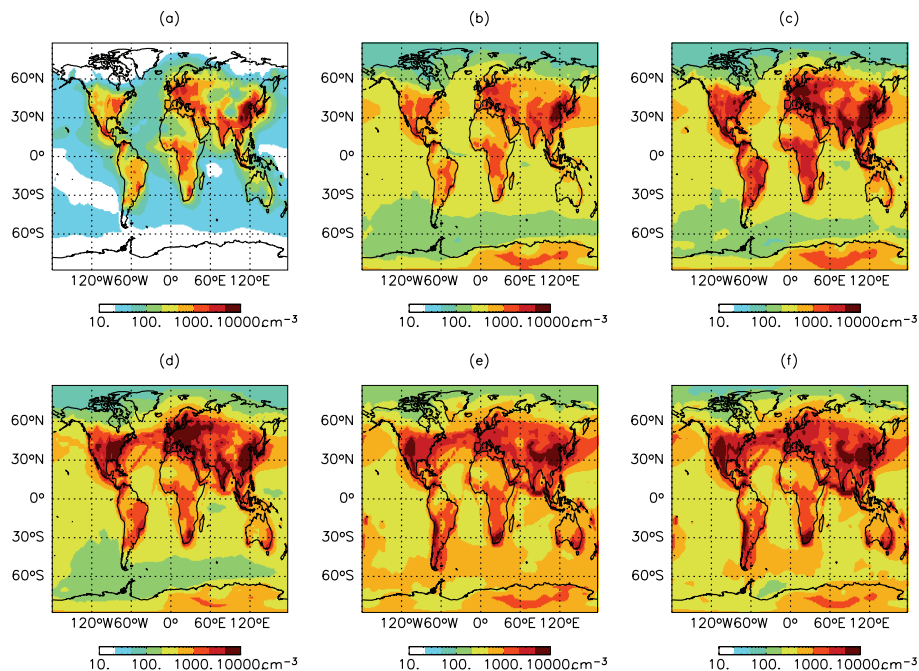


Fig. 1. Simulated surface annual mean CN ($D_p > 3$ nm) concentrations (at ambient temperature and pressure) for the different experiments described in Table 1: **(a)** Primary emissions only (PRI), **(b)** Primary emissions and binary homogeneous nucleation (BHN), **(c)** BHN and increased primary EC/OC (PRICAR), **(d)** BHN and increased primary sulfate (PRISUL), **(e)** Primary emissions, BHN and activation particle formation (ACT2), **(f)** Primary emissions, BHN and kinetic particle formation (KIN2).

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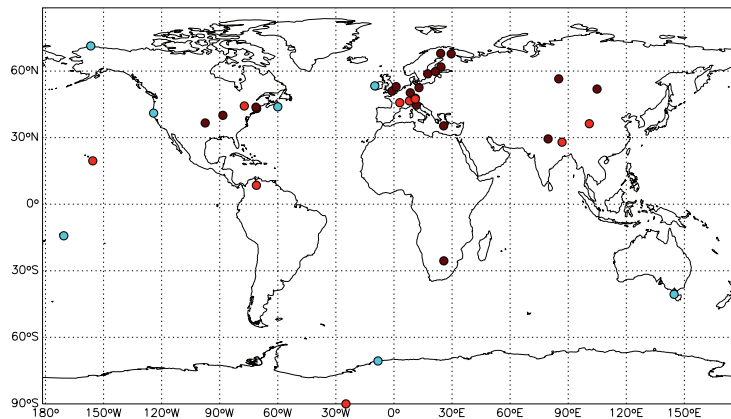


Fig. 2. Location of observation sites used in this analysis classified as in Table 2: FT (red), MBL (blue), continental BL (brown).

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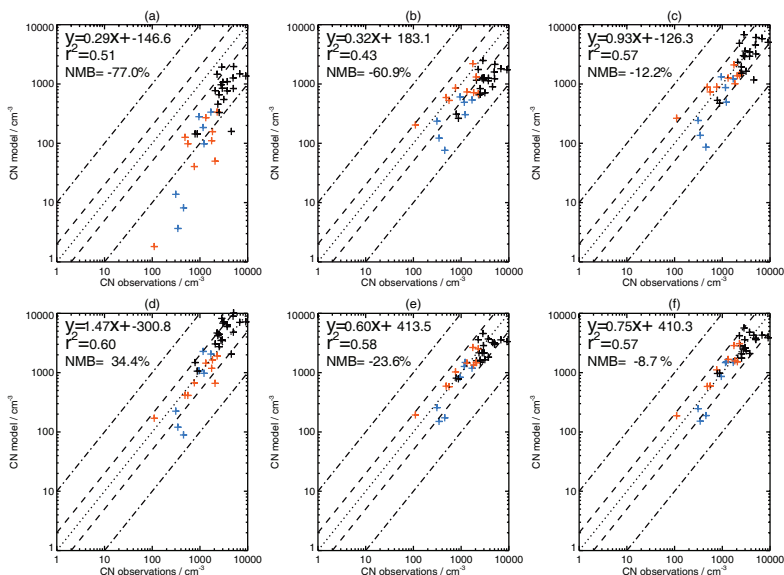


Fig. 3. Scatterplot of simulated (GLOMAP) versus observed annual mean CN concentrations (at ambient temperature and pressure) for the sites shown in Fig. 2. Sites are classified as FT (red), MBL (blue) and continental BL (black) as shown in Table 2. The different model experiments are described in Table 1: **(a)** Primary emissions only (PRI), **(b)** Primary emissions and binary homogeneous nucleation (BHN), **(c)** BHN and increased primary EC/OC (PRICAR), **(d)** BHN and increased primary sulfate (PRISUL), **(e)** Primary emissions, BHN and activation particle formation (ACT2), **(f)** Primary emissions, BHN and kinetic particle formation (KIN2). The regression equation for the reduced major axis linear regression, Pearson correlation coefficients (R^2) and normalised mean bias (NMB) are shown. The dotted line represents the 1:1 relation and the dashed lines factor of 2 and 10 deviations.

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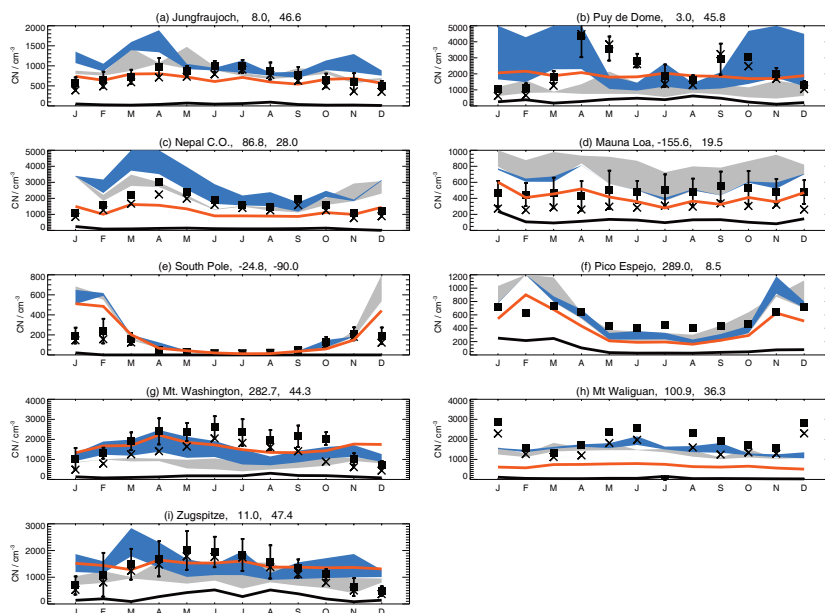


Fig. 4. Seasonal cycle of CN concentrations (at ambient temperature and pressure) at the FT sites shown in Table 2. Solid squares show observed monthly mean concentrations, crosses show observed monthly median concentrations and vertical bars show the standard deviation of the observed monthly mean (displayed only where there are 4 or more years of observations). The model experiments are described in Table 1: Black line shows the model with primary particulate emissions only (PRI), bottom of grey shading shows model with primary emissions and BHN (BHN), top of grey shading shows model with BHN and increased EC/OC number emission (PRICAR). Red line shows model with BHN and increased primary sulfate number emission (PRISUL). Blue shading shows model with primary emissions, BHN and activation BL particle formation, with the width of shading showing sensitivity to varying nucleation coefficients from $A=2\times 10^{-7}\text{ s}^{-1}$ (ACT1) to $A=2\times 10^{-5}\text{ s}^{-1}$ (ACT3).

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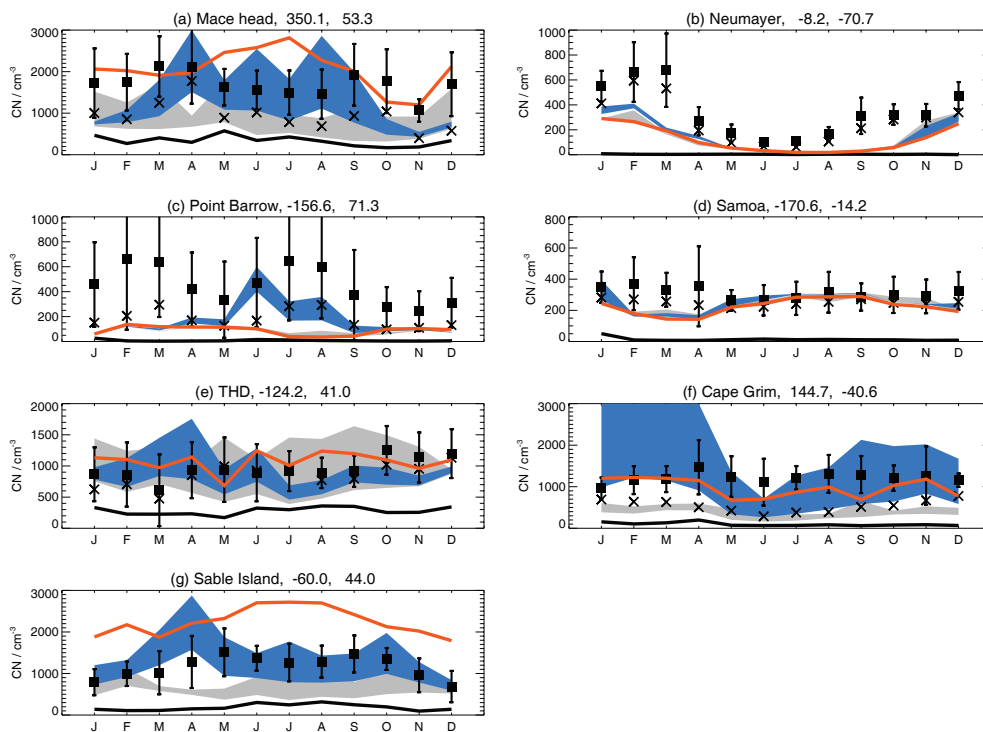


Fig. 5. As for Fig. 4 but for MBL sites.

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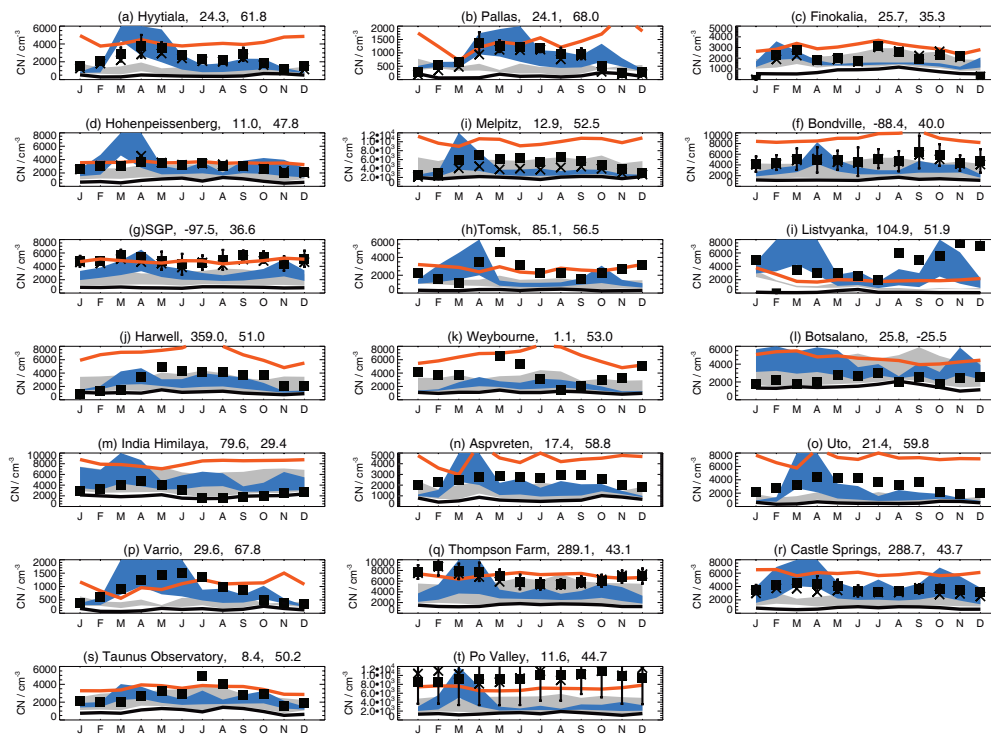


Fig. 6. As for Fig. 4 but for BL sites.

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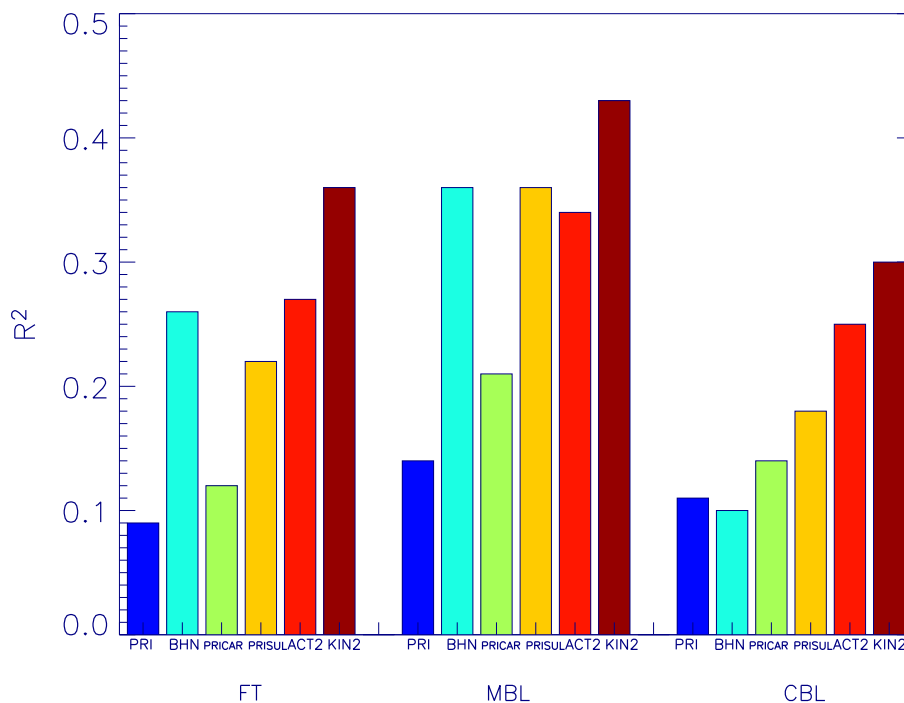


Fig. 7. Correlation coefficients (R^2) between simulated and observed monthly mean CN concentrations calculated for the FT, MBL and continental BL (CBL) sites in Table 4. Model runs are described in Table 1.

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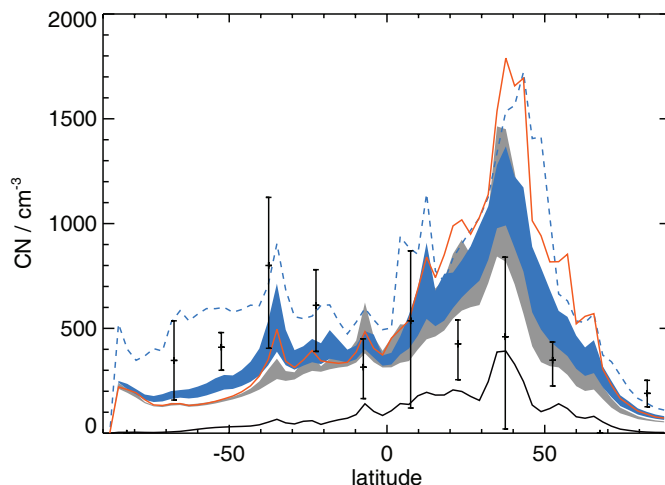


Fig. 8. Annual mean surface CN concentrations over the global oceans versus latitude. Observations (vertical bars) have been compiled from several field campaigns (Heintzenberg et al., 2000) and show mean concentrations \pm one standard deviation. The model experiments are described in Table 1: Black line shows the model with primary particulate emissions only (PRI), bottom of grey shading shows model with primary emissions and BHN (BHN), top of grey shading shows model with BHN and increased EC/OC number emission (PRICAR). Red line shows model with BHN and increased primary sulfate number emission (PRISUL). Blue shading shows model with primary emissions, BHN and activation BL particle formation, with the width of shading showing sensitivity to varying nucleation coefficients from $A=2 \times 10^{-7} \text{ s}^{-1}$ (ACT1) to $A=2 \times 10^{-5} \text{ s}^{-1}$ (ACT3). All model experiments are for CN ($>12 \text{ nm}$) except the blue dashed line which shows CN ($>3 \text{ nm}$) for the ACT2 simulation.

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