

# Effects of aerosol organics on cloud condensation nucleus (CCN) concentration and first indirect aerosol effect

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## Abstract

Aerosol microphysics, chemical composition, and CCN properties were measured on the Department of Energy Gulfstream-1 aircraft during the Marine Stratus/Stratocumulus Experiment (MASE) conducted over the coastal waters between Point Reyes National Seashore and Monterey Bay, California, in July 2005. Aerosols measured during MASE included free tropospheric aerosols, marine boundary layer aerosols, and aerosols with high organic concentration within a thin layer above the cloud. Closure analysis was carried out for all three types of aerosols by comparing the measured CCN concentrations at 0.22% supersaturation to those predicted based on size distribution and chemical composition using Köhler theory. The effect of aerosol organic species on predicted CCN concentration was examined using a single hygroscopicity parameterization. For aerosols with organics volume fraction up to 70%, such as the marine boundary layer and free troposphere aerosols, CCN concentration and the corresponding first indirect aerosol effect are insensitive to the properties of organics, and can be accurately predicted with a constant hygroscopicity for all organic species. This simplification can facilitate the prediction of indirect aerosol effects using physically-based parameterizations in large scale models. However, for the aerosols within the thin layers above clouds, organics contributed up to 90% of the total aerosol volume, and a detailed knowledge of organic hygroscopicity is required to accurately predict CCN concentrations. Derivations of organic properties in future closure studies, when aerosols are dominated by organic species, would help constrain the descriptions of organics and aerosol-cloud parameterizations in large scale models.

## 1 Introduction

Atmospheric aerosols affect the global energy budget by scattering and absorbing sunlight (direct effects) and by changing the microphysical structure, lifetime, and coverage of clouds (indirect effects). An increase in aerosol concentration would lead to smaller

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cloud droplet size and higher cloud albedo, i.e. brighter clouds (Twomey, 1977). This effect, which is known as the first indirect aerosol effect, tends to cool the global climate. The smaller cloud droplet size resulting from increased aerosol concentration also inhibits precipitation, leading to an increase in cloud lifetime and coverage, (second indirect aerosol effect, Albrecht, 1989). Although it is widely accepted that the indirect effects can strongly influence the global climate, and potentially mask the warming effect due to anthropogenic CO<sub>2</sub>, the magnitudes of indirect aerosol effects are poorly understood. The Intergovernmental Panel on Climate Change (IPCC, 2007) considers the indirect effects of aerosols the most uncertain components in forcing of climate change over the industrial period.

A key challenge in quantifying the aerosol indirect effects is to determine the spectrum of cloud condensation nuclei (CCN) and its spatial and temporal variations. CCN's are particles that can grow into cloud droplets at atmospherically relevant supersaturations. At a given supersaturation, CCN concentration ( $N_{\text{CCN}}$ ) is determined by aerosol size distribution and chemical composition. For a particle consisting of inorganic compounds, CCN activity can be effectively predicted using Köhler theory (Köhler, 1936) based on physicochemical properties of the solute, such as its mass, molecular weight, density, and activity coefficient. However, as ambient particles are often comprised of substantial amount of organic species (Kanakidou et al., 2005), Köhler theory has been extended to include the influence of organic species on surface tension (Facchini et al., 1999) and their contributions as solute (Shulman et al., 1996). While laboratory studies have shown that CCN activities of both single- and some multi-component aerosols can be successfully described using extended Köhler theory (Raymond and Pandis, 2002, 2003; Bilde and Svenningsson, 2004; Huff-Hartz et al., 2006; Svenningsson et al., 2006), the prediction of  $N_{\text{CCN}}$  for ambient aerosols remains a challenge. Ambient aerosols often consist of hundreds of organic species. Despite recent improvements in measurement techniques, a complete speciation of the organic components in ambient aerosols remains very difficult if not impossible. For the prediction of  $N_{\text{CCN}}$  in large scale models, the hundreds of organic species cannot be simulated individually due to

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computational constraints as well as the lack of knowledge of the precursors and formation mechanisms. Furthermore, the properties of many organic species in ambient aerosols (such as the particle density, molecular weight, solubility, and water activity coefficient) are often not available to initiate calculations using extended Köhler theory.

5 As a result, the treatment of aerosol indirect effects needs to be greatly simplified in large scale models including Global Circulation Models (GCM). Some GCM's avoid the calculation of  $N_{\text{CCN}}$  by using empirical relationships between aerosol mass and cloud droplet number concentration (Menon et al., 2002). These relationships are based on observations and are easy to implement in global forcing calculations, but they do not  
10 reflect the physical and chemical processes that occur during cloud nucleation, which depends on the size and chemical composition of the aerosol, as well as the updraft velocity. In addition, these relationships are based on limited measurements, and may not be sufficiently representative for use in global calculation or to predict future scenarios. More rigorous approaches, which are also more computationally intensive, are  
15 parameterizations that are based on particle size distribution, chemical composition, and Köhler theory (e.g. Abdul-Razzak and Ghan, 2000). However, due to the inability to resolve individual species in global models, the description of organic species is often greatly simplified, such as by assuming a constant hygroscopicity for all organics (Takemura et al., 2005).

20 The relationships among particle size distribution, chemical composition, and the ability of particles to serve as "CCN" have also been examined in a number of closure studies, which compared  $N_{\text{CCN}}$  predicted using measured ambient aerosol size distribution and composition to that directly measured. An overview of the results from previous closure studies can be found in Broekhuizen et al. (2006) and Medina et al. (2007).  
25 In these closure studies, a variety of simplified representations of particle composition and organic properties were necessary due to a limited knowledge of the identity and properties of these organic species. Therefore, these closure studies also provided tests for predicting  $N_{\text{CCN}}$  using extended Köhler theory and simplified description of aerosol organics, which is necessary in large scale models. In many closure studies,

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organics are assumed to be insoluble (Liu et al., 1996; Chuang et al., 2000; Cantrell et al., 2001; Roberts et al., 2002; Snider et al., 2003; Broekhuizen et al., 2006; Rissman et al., 2006; Medina et al., 2007). Other studies used different simplifications of aerosol chemical composition. For example, Van Reken et al. (2003) assumed particles observed during CRYSTAL-FACE study were pure ammonium sulfate, which was generally supported by Aerosol Mass Spectrometer (AMS) measurements. Roberts et al. (2002) also assumed about half of the organics were water soluble (e.g. dicarboxylic acids). Stroud et al. (2007) represented the organics as a mixture of levoglucosan, pinic acid, and fulvic acid. However it was found that assuming insoluble organics resulted in better agreement between the predicted and measured  $N_{CCN}$ . While closure was achieved for all or most of the data sets in some of the studies (Liu et al., 1996; Cantrell et al., 2001; Roberts et al., 2002; Van Reken et al., 2003; Broekhuizen et al., 2006; Medina et al., 2007), others showed substantial discrepancies (Chuang et al., 2000; Rissman et al., 2006; Stroud et al., 2007). In general, better closure agreements were achieved when the aerosols were not strongly influenced by anthropogenic emissions and contained low organic content. The difficulty in achieving closure in some of the studies could be due to a number of reasons. First, the CCN measurements could have substantial uncertainties and/or biases. Second, aerosol chemical composition measurements in some of the earlier closure studies were often crude and incomplete. In addition, for aircraft-based measurements, slow measurement techniques could have also contributed to the failure to achieve closure due to the spatial variability.

In this work, CCN closure studies were conducted using data collected during the Marine Stratus/Stratocumulus Experiment (MASE). Due to their extensive spatial coverage and high susceptibility, marine stratus/stratocumulus is considered a major contributor to first indirect aerosol effect. Based on sampling location, the aerosols observed during MASE were classified into three different types, which showed very different chemical characteristics. CCN concentrations at 0.22% supersaturation were predicted using measured aerosol size distribution and chemical composition, and compared to simultaneously measured  $N_{CCN}$  for the three aerosol types observed.

The closure study was performed assuming a wide range of organic properties, which is represented using a single hygroscopicity parameterization. The sensitivity of the predicted  $N_{\text{CCN}}$  to organic properties is examined. Theoretical analysis was also carried out to explore the feasibility of predicting  $N_{\text{CCN}}$  and the first indirect aerosol effect using a simplified representation of organic species. The accuracy of predicted  $N_{\text{CCN}}$  and the first indirect aerosol effect when using the simplified representation is examined.

## 2 Measurements

### 2.1 Location and instruments

The Marine Stratus/Stratocumulus Experiment (MASE) field campaign was carried out in July 2005 off the coast of Northern California between Monterey and Pt. Reyes, to study the aerosol-cloud interactions in the climatically important regime of the eastern Pacific marine stratocumulus (Daum et al., 2008<sup>1</sup>). The Department of Energy (DOE) Gulfstream-1 (G-1) aircraft flew a total of 11 scientific flights. All G-1 sampling flights originated from Sacramento, CA where the aircraft was based. The flight strategy was to climb to an altitude of  $\sim 1.5$  km or more after takeoff from Sacramento and head west towards Pt. Reyes, where the mobile facility of the DOE Atmospheric Radiation Program (ARM) was deployed. Upon completion of sampling at Pt. Reyes, the aircraft headed offshore, and flew repetitive multi-altitude patterns (about 10–15 min/leg) with legs above cloud, at multiple altitudes in-cloud, and as possible, below cloud.

During MASE, the G-1 carried a suite of instrumentation to measure the microphysics and chemical composition of aerosol, cloud microphysics, state parameters, winds, and radiation fields. Only the measurements related to this study are discussed

<sup>1</sup>Daum, P. H., Liu, Y. G., McGraw, R. L., Lee, Y. N., Wang, J., Senum, G. I., Miller, M., and Hudson, J. G.: Microphysical properties of stratus/stratocumulus Clouds during the 2005 Marine Stratus/Stratocumulus Experiments (MASE), in preparation, 2008.

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below. Aerosol size distribution was measured by a Scanning Mobility Particle Sizer (SMPS) (Wang et al., 2003). The major components of the SMPS are a cylindrical Differential Mobility Analyzer (Model 3081, TSI Inc., Minneapolis, Minnesota) and a Condensation Particle Counter (Model 3010, TSI Inc., Minneapolis, Minnesota). Prior to measurements, the relative humidity (RH) of aerosol sample was reduced to below 25% inside a Nafion drier. Aerosol size distribution ranging from 15 nm to ~450 nm was measured every 60 s. The SMPS was calibrated using polystyrene latex standards. Data from the SMPS were reduced using the data inversion procedure described by Collins et al. (2002).

CCN concentration at 0.22% supersaturation was measured by a CCN counter (Droplet Measurement Technology, Boulder). The 0.22% supersaturation is within the typical supersaturation range of stratocumulus clouds. The CCN counter consists of a cylindrical continuous-flow gradient diffusion chamber. A constant stream-wise temperature gradient is applied such that the difference between water vapor mass diffusivity and air thermal diffusivity leads to a quasi-uniform centerline supersaturation. CCN's, which are confined near the centerline, grow into super-micrometer droplets and are detected by an Optical Particle Counter downstream (Roberts and Nenes, 2005). The CCN data during the descent and ascent of G-1 were excluded from this analysis, as the pressure change inside the CCN counter led to instability of sample flow and supersaturation.

The size-resolved chemical composition of ambient aerosol was determined using an Aerodyne Quadruple aerosol mass spectrometer (AMS, Jayne et al., 2000). The details of the AMS operation onboard the G-1 has been given in detail in Kleinman et al. (2007), and will be only briefly described here. The AMS operation alternates between pTOF (particle time of flight) and MS (mass spectrum) modes. In the pTOF mode, the particle beam is chopped and the particles are sized according to their flight time between the chopper and a particle vaporizer. In the MS mode, the chopper moves in and out of the aerosol sample beam to provide total and background signals, which allow the total mass concentrations to be deduced. For most flights, the sam-

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pling average time was 30 s, which was split between the pTOF and MS modes. The AMS analysis software (Allan et al., 2004) determines concentration from MS mode and partitions that concentration into size bins according to the pTOF measurements. Measurement of aerosol species depends on the aerosol impinging on the particle vaporizer, after which it is vaporized, ionized and detected by a quadruple mass filter. During MASE, the temperature of the vaporizer was 600–650°C, adequate to vaporize mixed  $\text{SO}_4^{2-}$ – $\text{NO}_3^-$ – $\text{NH}_4^+$  salts and organic compounds which together are expected to constitute most of the mass for the particles with diameters less than 500 nm. Refractory materials such as NaCl, mineral dust, or blackcarbon are not detected by the AMS. For 9 of the total 11 research flights during MASE, valid data were available for all three measurements described above.

### 3 Data overview: typical aerosols observed during MASE

Aerosols observed during MASE were grouped into three major types based on the sampling altitude and the characteristics of aerosol microphysics and composition. The first was “Free Troposphere (FT)” aerosol observed during the ferries between Sacramento and Pt Reyes (Fig. 1). The ferries were mostly at an altitude of 1500 m or higher and were above the boundary layer except on 18 July 2005 and 19 July 2005. Figure 2 shows the size distributions of total number,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , and organics measured during the ferries on 27 July 2005 and 18 July 2005. The size distributions of the FT aerosols were often unimodal (Fig. 2a), and the aerosol mass loading was typically very low (e.g. less than  $1.4 \mu\text{g}/\text{m}^3$  on 27 July 2007). Due to the low FT aerosol mass loading and the short average time associated with aircraft measurements, the mass size distributions of chemical species measured by the AMS were quite noisy (Fig. 2b). However, aerosols observed during the ferry flights on 18 July 2005 and 19 July 2005 showed much higher number and mass concentrations, which were likely due to the upward mixing of boundary layer pollution. During MASE, the composition of FT aerosols was dominated by sulfate and organics, and the contribution from nitrate was small.

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The size distributions of sulfate and organics exhibited a similar shape based on the measurements on 18 July 2005 and 19 July 2005, which had higher counting statistics due to elevated aerosol mass concentrations.

“Boundary layer (BL)” aerosols were sampled over the Pacific at multiple altitudes within the boundary layer. Two examples of typical BL aerosols observed are shown in Fig. 3. Aerosol size distributions were often bimodal, with the modal diameters of the Aitken and accumulation modes near 60 and 160 nm, respectively. Similar to the FT aerosols, sulfate and organics dominated the composition of the BL aerosols, and the shapes of the sulfate and organic size distributions were similar.

During MASE, a thin layer of aerosol with high mass concentration was frequently observed just above the cloud. Aerosol within this thin layer, which was often ~60 m thick, is referred to as the “Above Cloud (AC)” type. Figure 4 shows aerosol composition within the thin layer was dominated by the organics, especial for particles with diameters less than 200 nm. Unlike the FT and BL aerosols, the size distribution of organics was quite different from that of sulfate. The sulfate mass concentration often reached its maximum at vacuum aerodynamic diameter of ~600 nm, which was substantially larger than the peak size of the organics.

## 4 CCN closure study

### 4.1 $\kappa$ -Köhler theory

In our closure studies, the critical dry diameter that corresponds to activation at 0.22% supersaturation was calculated using the “ $\kappa$  Köhler theory” (Petters and Kreidenweis, 2007), which uses a single parameter  $\kappa$  to describe the Raoult effect on CCN activation. In  $\kappa$  Köhler theory, the water vapor saturation ratio over the aqueous solution droplet  $S$

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is given by:

$$S = \frac{D^3 - D_p^3}{D^3 - D_p^3(1 - \kappa)} \exp\left(\frac{4\sigma_w M_w}{RT \rho_w D}\right) \quad (1)$$

where  $D$  is the droplet diameter,  $D_p$  the dry diameter of the particle,  $M_w$  the molecular weight of water,  $\sigma_w$  the surface tension of aqueous solution,  $\rho_w$  the density of water,  $R$  the gas constant,  $T$  the absolute temperature. For particles comprised of multiple components, the value of  $\kappa$  is given by a simple mixing rule (Petters and Kreidenweis, 2007):

$$\kappa = \sum_i x_i \kappa_i \quad (2)$$

where  $x$  is the volume fraction and subscript  $i$  denotes species  $i$ . For soluble inorganic species, such as  $\text{NH}_4\text{HSO}_4$ ,  $\text{NH}_4\text{NO}_3$ ,  $\kappa_i$  can be derived as:

$$\kappa_i = v_i \frac{\rho_i M_w}{\rho_w M_i} \quad (3)$$

where  $\rho$  is the density,  $M$  the molecular weight, and  $v$  the van't Hoff factor.

For organic species, the derivation of  $\kappa$  from experimental data is based on the assumption that  $\sigma_w = 0.072 \text{ J m}^{-2}$  (pure water, Petters and Kreidenweis, 2007). The derived  $\kappa$  is positively biased for surface active organic species due to this assumption. However, in calculation of  $\kappa$  for a mixture containing the same surface active species, the larger  $\kappa$  of the organic species partially compensates for the lower surface tension of the mixture. Although this approach is not thermodynamically rigorous, Petters and Kreidenweis (2007) showed that it predicts the hygroscopicity within measurement uncertainty for mixtures that contain both surface active species and surface inactive compounds. In addition, at the critical droplet diameter that corresponds to the maximum supersaturation at the droplet surface, the concentration of surface active

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organics is often very dilute due to the large water mass. As a result, the effect of surface active organic species is usually small except for organics with extreme properties (Ervens et al., 2005).

#### 4.2 Derivation of aerosol chemical composition from AMS measurements

5 While AMS represents a significant improvement over traditional filter-based techniques, its characterization of aerosol composition is still incomplete. To carry out CCN closure study, we constructed aerosol chemical composition from the AMS data using following approach. First, the aerosols were considered consisting mainly of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , and Organics. This assumption should be valid for the aerosols observed  
10 in the free troposphere and in the thin layers above clouds. The BL aerosols might contain substantial amount of sea salt. However, it is expected that sea salt is present mainly in large particles (Lewis and Schwartz, 2005), which contribute to only a small fraction of CCN due to their low number concentrations. Therefore, although sea salt could contribute to the total aerosol mass concentration, neglecting the sea salt should not lead to an appreciable error in predicted  $N_{\text{CCN}}$ . Observed  $\text{NO}_3^-$  was assumed to  
15 exist as  $\text{NH}_4\text{NO}_3$ , and  $\text{SO}_4^{2-}$  is assumed as  $\text{NH}_4\text{HSO}_4$ . This picture is consistent with the average concentration of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$  observed during MASE. Given the similar CCN activity of  $(\text{NH}_4)_2\text{SO}_4$  and  $\text{NH}_4\text{HSO}_4$ , the uncertainty associated with these assumptions is expected to be small. The sensitivity of the predicted  $N_{\text{CCN}}$  to this  
20 assumption is discussed further below.

For the FT and BL aerosols, the size spectra of sulfate and organics showed a similar shape and the aerosol composition was dominated by sulfate and organics. Therefore, aerosol chemical composition was treated as independent of particle size with all species internally mixed. This assumption allowed derivation of volume fraction of  
25 each species using the AMS MS-mode measurements, which have much higher signal to noise ratio than the pTOF-mode measurements. During MASE, the average volume fraction for ammonium bisulfate, nitrate, and organics were 43%, 7% and 50% for the

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FT aerosols, and 51%, 3%, and 46% for the BL aerosols.

For the AC aerosols, the shapes of sulfate and organic size distributions were quite different. To describe this size dependence of chemical composition, we assumed the AC aerosols to be external mixtures of lognormal modes, and that within each mode, the chemical composition was constant, and independent of particle size. The mode diameter, concentration, geometric standard deviation, and the mass fraction of each species were derived for each mode using the approach described as follows. First, the total mass size distribution was fitted by lognormal modes (two modes for the example given in Fig. 5a). Second, the size spectra for the three species ( $\text{NH}_4\text{HSO}_4$ ,  $\text{NH}_4\text{NO}_3$  and organics) were each expressed as a linear combination of the lognormal modes determined from the total aerosol mass. The linear coefficients, which are the mass fractions of individual species within the lognormal modes, were determined using a least square fitting procedure. Figure 5 shows that the fitted size distributions of total mass,  $\text{NH}_4\text{HSO}_4$ ,  $\text{NH}_4\text{NO}_3$  and organics agree closely with the measurements, supporting the external-mixture nature of the aerosols. For each mode, the volume fractions were calculated from fitted mass fraction using the densities of participating species. For organics, a density of  $1.2\text{ g/cm}^3$  is used in this study. For the AC aerosols observed on 27 July 2005, the fitted lognormal mode parameters and the volume fraction of each species are listed in Table 1. The vacuum aerodynamic diameters ( $D_{va}$ ) of the two modes were 190 nm and 410 nm, respectively. The smaller mode was dominated by organics, which contributed over 90% of the total volume. For the larger mode, the sulfate volume fraction was about 20%, a substantial increase from the fraction in the smaller mode. The fitting procedure described above was carried out for the AC aerosols observed on all flights.

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### 4.3 CCN closure study and the sensitivity of predicted $N_{\text{CCN}}$ to the properties of organic species

#### 4.3.1 CCN Closure study with assumption of insoluble organics

Closure studies were carried out for  $N_{\text{CCN}}$  at 0.22% supersaturation using the following approach. For the FT and BL aerosols, which were treated as internal mixtures of sulfate, nitrate, and organics, the fraction of each species was derived from the AMS MS-mode measurements averaged over the SMPS measurement intervals. We point out that due to the lower time resolution of SMPS, size distributions during which substantial variation in  $N_{\text{CCN}}$  was observed were excluded. Based on the derived composition, the critical dry particle activation diameter ( $D_{pc}$ ) was calculated using the  $\kappa$  Köhler theory for each SMPS measurement interval, and  $N_{\text{CCN}}$  was then derived by integrating the size distribution from  $D_{pc}$  and above. The AC aerosols were considered as external mixture of lognormal modes. As described in Sect. 4.2, the lognormal mode parameters and the composition of each mode were derived by fitting the size distributions of total mass and each species averaged over the layer. For each mode, the  $N_{\text{CCN}}$  was predicted using the same method described above, and the total  $N_{\text{CCN}}$  is the sum of the contribution from all modes. As a starting point, the organic species were assumed to be water insoluble (i.e.  $\kappa=0$ ), as in many previous closure studies. Figure 6 shows the comparison of predicted  $N_{\text{CCN}}$  to the measured at 0.22% supersaturation for all research flights. When the organic species are assumed to be insoluble, the predicted  $N_{\text{CCN}}$  for the FT and BL aerosols agrees well with the measured  $N_{\text{CCN}}$ . The ratio of predicted to measured  $N_{\text{CCN}}$ , (i.e. the slope of the best fit) is 0.95 and 0.94 for the FT and BL aerosols, respectively. For the AC aerosols, the ratio is only 0.46, suggesting that assuming insoluble organics substantially underestimates the  $N_{\text{CCN}}$ .

As described earlier, sulfate was assumed to exist as  $\text{NH}_4\text{HSO}_4$ . To examine the uncertainty associated this assumption, we also carried out closure study treating all sulfate as completely neutralized  $(\text{NH}_4)_2\text{SO}_4$ . The results are very similar; the ratios are 0.93, 0.91, and 0.46 for the FT, BL, and AC aerosols, respectively. The sensitivity

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of predicted  $N_{\text{CCN}}$  to particle composition was also investigated by varying the mass concentration of sulfate. When the sulfate mass concentration is increased by 20%, the ratio only increases slightly to 0.99 and 0.97 for the FT and BL aerosols, respectively. Similarly, both ratios decrease slightly to 0.90 for the FT and BL aerosols when the mass loading of sulfate is decreased by 20%. As the assumption that the chemical composition was independent of particle size may not be appropriate for all FT and BL aerosols, the good agreement may be partially due to the low sensitivity of predicted  $N_{\text{CCN}}$  to the chemical composition for these two types of aerosols.

#### 4.3.2 Sensitivity of predicted $N_{\text{CCN}}$ to the $\kappa$ value of organic species

Many organics species observed in the atmosphere are water soluble and therefore have  $\kappa$  values greater than zero. Petters and Kreidenweis (2007) showed that  $\kappa$  values for organic species commonly observed in the atmosphere range mostly from 0 to 0.25. The sensitivity of predicted  $N_{\text{CCN}}$  to the  $\kappa$  value of organic species ( $\kappa_{\text{org}}$ ) is investigated in this section. For each of the three aerosol types defined above,  $N_{\text{CCN}}$  was predicted using a wide range of  $\kappa_{\text{org}}$ ; the ratio of predicted to measured  $N_{\text{CCN}}$  is shown in Fig. 7 as a function of  $\kappa_{\text{org}}$ . For the FT and BL aerosols, the predicted  $N_{\text{CCN}}$  is insensitive to  $\kappa_{\text{org}}$ . When  $\kappa_{\text{org}}$  increases from 0 to 0.25, the ratio of predicted to measured  $N_{\text{CCN}}$  increases only slightly from 0.95 to 1.15 for the FT aerosols, and from 0.94 to 1.10 for the BL aerosols. In contrast, the ratio increases from 0.46 to 1.50 for the AC aerosols. The difference in the sensitivity to  $\kappa_{\text{org}}$  is due to the following reason. Whereas organics contributed up to 50% of the aerosol volume for the FT and BL aerosols, the ion concentrations were dominated by the inorganics due to the high solubility of sulfate and nitrate (i.e. high  $\kappa$  values). The additional contribution from soluble organics, which has relatively low  $\kappa_{\text{org}}$ , was only secondary. As a result, for a wide range of  $\kappa_{\text{org}}$ , the variations in the predicted  $N_{\text{CCN}}$  and the ratio of predicted to measured  $N_{\text{CCN}}$  are small for the FT and BL aerosols. For the AC aerosols, organics dominated the composition (i.e. up to 90% of the total aerosol volume concentration), and the ratio is highly sensitive to  $\kappa_{\text{org}}$ . The  $\kappa_{\text{org}}$  for the AC aerosols was derived as

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0.1 by matching the predicted  $N_{\text{CCN}}$  to the measured. For the FT and BL aerosols, the low sensitivity of predicted  $N_{\text{CCN}}$  to  $\kappa_{\text{org}}$  is an important reason for the good agreement between predicted and measured  $N_{\text{CCN}}$  even when the organics are assumed to be completely insoluble, which may not be true for many organic species observed away from the source regions.

### 4.4 Uncertainty in first indirect aerosol effect

#### 4.4.1 Uncertainty in first indirect aerosol effect due to the uncertainty in $\kappa_{\text{org}}$

As discussed above, ambient aerosols often consist of hundreds of organic species, which cannot be speciated with current techniques or simulated in global models. The lack of knowledge of the organic properties will also lead to an uncertainty in predicted  $N_{\text{CCN}}$ , as indicated in Fig. 7. The uncertainty in  $N_{\text{CCN}}$  translates into uncertainties in cloud droplet number concentration and albedo, and ultimately the uncertainty in indirect aerosol effects. The uncertainty in average first indirect aerosol effect,  $\Delta F_c$ , can be estimated by (Schwartz and Slingo, 1996):

$$\Delta F_c \approx -\frac{F_T}{4} A_{\text{mst}} T_r^2 \Delta R_c \quad (4)$$

where  $F_T$  is the solar constant,  $A_{\text{mst}}$  the cloud fractional coverage, and  $T_r$  the fractional transmission of shortwave radiation above the cloud layer, and  $\Delta R_c$  the uncertainty in cloud albedo. For global mean calculations, the cloud fraction is taken with the assumption (Charlson et al., 1987) that non-overlapped marine stratus and stratocumulus clouds having a fractional area of 30%, and  $T_r$  is 76% (Schwartz and Slingo, 1996). For the range of cloud albedo  $0.28 \leq R_c \leq 0.72$ , which is characteristic of the prevalent and climatically important marine stratus clouds,  $\Delta R_c$  can be estimated using the following equation with error less than 10% (Schwartz and Slingo, 1996):

$$\Delta R_c = \frac{1}{3} [R_c (1 - R_c)] \cdot \Delta \ln N_d \approx 0.075 \cdot \Delta \ln N_d \quad (5)$$

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where  $N_d$  is the cloud droplet number concentration. Sotiropoulou et al. (2006) studied the uncertainty in  $N_d$  associated with the uncertainty in  $N_{CCN}$  using field data and found that for majority of the cases, the relative uncertainty in  $N_d$  is less than 50% of the relative uncertainty in  $N_{CCN}$ . Therefore, the upper limit of the relative uncertainty of

$N_d$  can be estimated as:

$$\Delta \ln N_d \approx 0.5 \Delta \ln N_{CCN} \quad (6)$$

The lower relative uncertainty in  $N_d$  is due to the feedback of the maximum supersaturation inside clouds. For example, at a constant updraft velocity, an increase in  $N_{CCN}$  leads to reduced maximum supersaturation inside the cloud due to the competition of water vapor among more particles. As a result, the fractional increase in  $N_d$  will be less than that of  $N_{CCN}$ . Combining Eqs. (4), (5), and (6), we can estimate the upper limit of  $\Delta F_c$  from the uncertainty in  $N_{CCN}$  as:

$$\Delta F_c \approx -\frac{F_T}{4} A_{mst} T_r^2 (0.038 \Delta \ln N_{CCN}) \approx -2.2 \cdot \Delta \ln N_{CCN} \left( W/m^2 \right) \quad (7)$$

It is important to point out that Eq. (7) represents the upper limit of  $\Delta F_c$ , the true uncertainty in the first indirect aerosol effect could be substantially less. A recent study based on GCM model (Sotiropoulou et al., 2007) found that a 9% global average uncertainty in  $N_{CCN}$  leads to an uncertainty of  $0.1 W/m^2$  in global average first indirect aerosol effect, only 50% of  $0.2 W/m^2$  predicted using Eq. (7).

The uncertainty in first indirect aerosol effect was estimated using Eq. (7) for the three types of aerosols observed during MASE. For the FT and BL aerosols, the predicted  $N_{CCN}$  is insensitive to  $\kappa_{org}$  (Fig. 7). For the typical range of  $\kappa_{org}$  from 0 to 0.25, the uncertainties in predicted  $\ln N_{CCN}$  (i.e.  $\Delta \ln N_{CCN}$ ) are 0.18 and 0.16, which correspond to  $\Delta F_c$  of 0.40 and  $0.35 W/m^2$  for the FT and BL aerosols, respectively. For the AC aerosols, the high sensitivity of  $N_{CCN}$  to  $\kappa_{org}$  leads to a  $\Delta F_c$  of  $2.6 W/m^2$  for  $\kappa_{org}$  ranging from 0 to 0.25. It is expected that ambient aerosols have a wide range of sensitivities of  $\ln N_{CCN}$  to  $\kappa_{org}$ . The global average  $\Delta F_c$  is based on the average  $\Delta \ln N_{CCN}$ , and is likely somewhere between the cases examined here.



#### 4.4.2 Uncertainties of predicted $N_{\text{CCN}}$ and first indirect aerosol effect when assuming a constant $\kappa_{\text{org}}$

Given the complexity of organic species in ambient aerosol, a simplified description of organic species is necessary to predict aerosol indirect effects using physically-based parameterizations in global models. In this section, we estimate the uncertainties in predicted  $N_{\text{CCN}}$  and first indirect aerosol effect for a simplified representation, in which an average  $\kappa_{\text{org}}=0.12$  is used to represent all organic species. To simplify our analysis, we start with internally mixed aerosols and represent all inorganic species using  $\text{NH}_4\text{HSO}_4$ . As the  $\kappa$  values of other common inorganic species (e.g.  $(\text{NH}_4)_2\text{SO}_4$  and  $\text{NH}_4\text{NO}_3$ ) are very close to the  $\kappa$  of  $\text{NH}_4\text{HSO}_4$ , we do not expect this assumption will affect the conclusion of our analysis. For internally mixed aerosols such as the FT and BL aerosols observed during MASE,  $N_{\text{CCN}}$  can be calculated as:

$$N_{\text{CCN}} = \int_{\ln D_{pc}}^{+\infty} n(\ln D_p) d \ln D_p, \quad (8)$$

where  $n(\ln D_p)$  is particle size distribution,  $D_{pc}$  the critical dry activation diameter. The sensitivity of predicted  $N_{\text{CCN}}$  to the overall  $\kappa$  of particle is given by:

$$\frac{d \ln N_{\text{CCN}}}{d \ln \kappa} = \frac{d \ln N_{\text{CCN}}}{d \ln D_{pc}} \cdot \frac{d \ln D_{pc}}{d \ln \kappa}. \quad (9)$$

Based on Eq. (8), the sensitivity of  $N_{\text{CCN}}$  to  $D_{pc}$  is given by:

$$\frac{d \ln N_{\text{CCN}}}{d \ln D_{pc}} = \frac{1}{N_{\text{CCN}}} \frac{d N_{\text{CCN}}}{d \ln D_{pc}} = - \frac{n(\ln D_{pc})}{N_{\text{CCN}}} \quad (10)$$

For  $\kappa > 0.2$ , which is typical for atmospheric particles, we have (Petters and Kreidenweis, 2007):

$$\kappa = \frac{4 \left( \frac{4\sigma_w M_w}{RT\rho_w} \right)^3}{27D_{pc}^3 \ln^2 S_c}, \quad (11)$$

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where  $S_c$  is the critical supersaturation. From Eq. (11), the following equation can be derived:

$$\frac{d \ln D_{pc}}{d \ln \kappa} = -\frac{1}{3} \quad (12)$$

Combining Eqs. (9), (10), and (12), we can derive the sensitivity of  $N_{CCN}$  to overall  $\kappa$  as:

$$\frac{d \ln N_{CCN}}{d \ln \kappa} = \frac{1}{3} \frac{n (\ln D_{pc})}{N_{CCN}} \quad (13)$$

When all organic species are represented using  $\overline{\kappa_{org}}$ , the fraction error in calculate  $N_{CCN}$  can be estimated using Eq. (13):

$$\frac{\Delta N_{CCN}}{N_{CCN}} \approx \Delta \ln N_{CCN} = \ln \left( \frac{N'_{CCN}}{N_{CCN}} \right) = \frac{1}{3} \frac{n (\ln D_{pc})}{N_{CCN}} \ln \left( \frac{(1 - x_{org}) \kappa_{ABS} + x_{org} \overline{\kappa_{org}}}{(1 - x_{org}) \kappa_{ABS} + x_{org} \kappa_{org}} \right) \quad (14)$$

where  $N'_{CCN}$  is the CCN concentration predicted using the  $\overline{\kappa_{org}}$  (0.12),  $\kappa_{ABS}$  the  $\kappa$  of ammonium bisulfate (0.7), and  $x_{org}$  the volume fraction of organic species. For aerosols with composition (i.e.  $x_{org}$ ) varying with particle size,  $x_{org}$  represents the volume fraction of organics at  $D_{pc}$ .

For externally mixed aerosols, the total  $N_{CCN}$  is the sum of  $N_{CCN}$  of each participating population:

$$N_{CCN} = \sum_j N_{CCN,j} \quad (15)$$

where  $N_{CCN,j}$  represents the CCN concentration of each aerosol population. The fractional error of the total  $N_{CCN}$  is then given by:

$$\frac{\Delta N_{CCN}}{N_{CCN}} = \sum_j \frac{\Delta N_{CCN,j}}{N_{CCN}} = \sum_j \left( \frac{N_{CCN,j}}{N_{CCN}} \right) \frac{\Delta N_{CCN,j}}{N_{CCN,j}} \quad (16)$$

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Equation (16) shows that for externally mixed aerosols, the fractional error in overall  $N_{\text{CCN}}$  will be a weighted average of the fractional error for each aerosol population. In the rest of this section, we examine internally mixed aerosols. According to Eq. (14), for  $\kappa_{\text{org}}$  ranging from 0 to 0.25, the maximum fractional error in  $N_{\text{CCN}}$  for using  $\overline{\kappa_{\text{org}}}$  occurs when  $\kappa_{\text{org}}$  is either 0 or 0.25. The corresponding error on the aerosol first indirect effect,  $\Delta F_c$ , can be derived by combining Eqs. (7) and (14):

$$\Delta F_c \approx -2.2 \cdot \Delta \ln N_{\text{CCN}} = -0.7 \frac{n(\ln D_{pc})}{N_{\text{CCN}}} \ln \left( \frac{0.70(1 - x_{\text{org}}) + 0.12x_{\text{org}}}{0.70(1 - x_{\text{org}}) + \kappa_{\text{org}}x_{\text{org}}} \right) \quad (17)$$

The value of  $\frac{n(\ln D_{pc})}{N_{\text{CCN}}}$  depends on the shape of particle size distribution and  $D_{pc}$ . The average  $\frac{n(\ln D_{pc})}{N_{\text{CCN}}}$  is 1.60 and 1.35 for the FT and BL aerosols, respectively. For typical marine, rural, urban, and remote continental aerosol (Seinfeld and Pandis, 2006)  $\frac{n(\ln D_{pc})}{N_{\text{CCN}}}$  ranges from 0.2 to 2.3 for  $D_{pc}$  between 50 and 150 nm, which is the typical range of  $D_{pc}$  under climatically relevant supersaturations. For an average  $\frac{n(\ln D_{pc})}{N_{\text{CCN}}}$  of 1.3,  $\Delta F_c$  is calculated as a function of  $x_{\text{org}}$  and  $\kappa_{\text{org}}$  using Eq. (17) and the results are shown in Fig. 8. As expected,  $\Delta F_c$  increases with increasing  $x_{\text{org}}$  (Fig. 8a). When organic volume fraction was lower than  $\sim 60\%$ , using  $\overline{\kappa_{\text{org}}}$  for all organic species leads to  $\Delta F_c$  of less than  $0.2 \text{ W/m}^2$  for typical  $\kappa_{\text{org}}$  ranging from 0 to 0.25. However, since Eq. (17) is based on Eq. (6), which assumes the upper limit of the uncertainty in  $N_d$ ,  $\Delta F_c$  can be as low as 50% (i.e.  $0.1 \text{ W/m}^2$ ) of the estimate using Eq. (17) (Sotiropoulou et al., 2007). Furthermore, it is expected that ambient organics have a wide range of  $\kappa_{\text{org}}$  distributed about the central value of 0.12. As a result, when averaged over a large area and/or extended time period,  $\Delta F_c$  may be substantially less than the  $0.2 \text{ W/m}^2$  limit shown in Fig. 8a. However, if organics dominate the aerosol volume,  $\Delta F_c$  becomes significant: when organics contribute to  $\sim 90\%$  of the total aerosol volume,  $\Delta F_c$  is  $-1.0$  and  $0.6 \text{ W/m}^2$  at  $\kappa_{\text{org}}$  of 0 and 0.25, respectively. The  $\Delta F_c$  is less than  $0.2 \text{ W/m}^2$  only for  $\kappa_{\text{org}}$

within a narrow range from 0.09 and 0.16. This suggests that when organics dominates the aerosol volume, detailed knowledge of the organic properties is required to accurately predict  $N_{CCN}$  and first indirect aerosol effect.

Based on previous field measurements, the  $\frac{n(\ln D_{pc})}{N_{CCN}}$  of marine aerosols ranges from 0.2 to 1.5 for  $D_{pc}$  between 50 and 150 nm (Quinn et al., 1995; Van Dingenen et al., 1995; Jensen et al., 1996; Seinfeld and Pandis, 2006). Unlike urban and continental aerosols whose number concentrations are often dominated by small nucleation mode and/or Aitken mode particles, marine aerosols often consist of a larger fraction of accumulation mode particles (Seinfeld and Pandis, 2006). As accumulation mode particles are likely to be CCN due to their large size, marine aerosols often have larger percentage of total particles that are CCN, which preferentially increases the denominator of Eq. (10). As a result, marine aerosols tend to have relatively lower values of  $\frac{n(\ln D_{pc})}{N_{CCN}}$ . Figure 8b shows the  $\Delta F_c$  calculated using an average  $\frac{n(\ln D_{pc})}{N_{CCN}}$  of 0.9 for marine aerosols. For  $\kappa_{org}$  ranging from 0 to 0.25, using  $\overline{\kappa_{org}}=0.12$  for all organic species results in a  $\Delta F_c$  less than  $0.2 W/m^2$  when organic species contribute to less than 70% of the aerosol volume, which is likely true for most marine aerosols. Zhang et al. (2007) presented aerosol chemical composition measured by AMS over the Northern Hemisphere mid-latitudes, where the aerosols are influenced by anthropogenic emissions. The volume fraction of organics estimated from the data shows that among the 37 datasets presented, the volume fraction of organics is less than 60% for 26 datasets and less than 70% for 32 datasets. The less than  $0.2 W/m^2$   $\Delta F_c$  corresponds to less than 10% uncertainty in predicted  $N_{CCN}$ . The low uncertainty in the predicted  $N_{CCN}$  when organic species make up less than 60–70% of aerosol volume is consistent with the results from the closure study using MASE data described in earlier sections. This can also potentially explain the results of previous closure studies, which showed that better closure agreements were generally archived when the aerosol contained low organic content.

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## 5 Summary

For each of the three aerosol types observed during MASE, closure analysis was carried out by comparing the  $N_{\text{CCN}}$  predicted from the measured size distribution and chemical composition using  $\kappa$  Köhler theory to simultaneously measured  $N_{\text{CCN}}$ . For FT and BL aerosols, the predicted  $N_{\text{CCN}}$  is insensitive to the properties of organics. The predicted  $N_{\text{CCN}}$  shows little variation and agrees with the measured  $N_{\text{CCN}}$  to within 15% when  $\kappa_{\text{org}}$  is assumed to be between 0 to 0.25, which represents the typical range of  $\kappa_{\text{org}}$  for ambient organic species. In contrast,  $N_{\text{CCN}}$  is a strong function of  $\kappa_{\text{org}}$  for the AC aerosols. The predicted  $N_{\text{CCN}}$  is only 46% of the measured  $N_{\text{CCN}}$  if the organic species are assumed to be insoluble (i.e.,  $\kappa_{\text{org}}=0$ ), and 150% when  $\kappa_{\text{org}}$  is assumed to be 0.25. The  $\kappa_{\text{org}}$  of the AC aerosols is consistent with a value of 0.1 derived by matching the predicted  $N_{\text{CCN}}$  to the measured. For the FT and BL aerosols, the low sensitivity of predicted  $N_{\text{CCN}}$  to  $\kappa_{\text{org}}$  arises because inorganic species, such as sulfate and nitrate, are more water soluble (i.e. higher  $\kappa$  value) compared to typical organic species observed in the atmosphere. Whereas organic species contributed up to about half of the total aerosol volume for the FT and BL aerosols, the total soluble ions, which dictate particle CCN activity, were dominated by inorganic species, and the additional contribution from the organic species was only secondary. For the AC aerosols, organics contributed up to 90% of the total aerosol volume at  $D_{\text{pc}}$ . As a result, the predicted  $N_{\text{CCN}}$  is highly sensitive to  $\kappa_{\text{org}}$ .

Atmospheric aerosols often consist of hundreds of organic species, which cannot be simulated individually in global models due to computational constraints. The inability to simulate individual species, coupled with the lack of thermodynamic data of many organic compounds, necessitate simplified representations of organic species when using physically based parameterizations in large scale models to evaluate indirect aerosol effects. A simplified representation in which an average  $\overline{\kappa_{\text{org}}}$  of 0.12 is used for all organics is explored. The analysis shows that for typical aerosol size distribution, the predicted  $N_{\text{CCN}}$  and the corresponding first indirect aerosol effect are insensitive

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to the properties of organic species when the volume fraction of aerosol organics is less than 60%. As a result, the estimated uncertainty in the first indirect aerosol effect ( $\Delta F_c$ ) for using an average  $\overline{\kappa_{\text{org}}}$  (0.12) is low, between  $-0.2$  and  $0.2 \text{ W/m}^2$  for  $\kappa_{\text{org}}$  between 0 and 0.25, the typical range of  $\kappa_{\text{org}}$  for ambient aerosols. It is worth noting that  $0.2 \text{ W/m}^2$  represents the upper limit of the uncertainty. Since the ambient aerosol organic species are expected to have a wide range of  $\kappa_{\text{org}}$  surrounding 0.12,  $\Delta F_c$  may be substantially less than  $0.2 \text{ W/m}^2$  when averaged over large area and/or extended time period. As expected,  $\Delta F_c$  due to the simplified treatment increases with increasing organics volume fraction. For example, when organics contribute to 90% of the total aerosol volume, the  $\Delta F_c$  can reach  $0.6$  and  $-1.0 \text{ W/m}^2$ . Analyses show that  $\Delta F_c$  associated with the simplification depends on not only the volume fraction of organic species but also the shape of the aerosol size distribution, which controls the sensitivity of the predicted  $N_{\text{CCN}}$  to  $D_{pc}$ . For marine aerosols that often consist of larger fractions of accumulation particles, the sensitivity of predicted  $N_{\text{CCN}}$  to  $D_{pc}$  is low, which leads to lower sensitivity of predicted  $N_{\text{CCN}}$  to  $\kappa_{\text{org}}$ . Based on the marine aerosols measured during previous field studies, using  $\overline{\kappa_{\text{org}}}$  for all organic species lead to  $\Delta F_c$  less than  $0.2 \text{ W/m}^2$  provided that the organic volume fraction is less than 70%, which is likely to be true for most marine aerosols. Due to the extensive spatial coverage and high susceptibility of marine stratus/stratocumulus, the low uncertainties in predicted  $N_{\text{CCN}}$  and the first indirect aerosol effect for marine aerosols will contribute to low uncertainties in global averages. The low sensitivity of predicted  $N_{\text{CCN}}$  to  $\kappa_{\text{org}}$  for aerosols with an organic volume fraction less than 60–70% can potentially explain the results of previous closure studies, which showed better agreements were generally archived when the aerosol contained low organic content.

It is important to note that the analyses presented in this work are based on a number of assumptions. Nevertheless, they suggest that for many situations, first indirect aerosol effect can be predicted accurately using an average  $\overline{\kappa_{\text{org}}}$  of 0.12 for all organics present in the aerosol. This can significantly simplify the calculation of  $N_{\text{CCN}}$ , and

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facilitate the prediction of indirect aerosol effects using physically-based parameterizations in global models. More field data and modeling studies will be needed to assess the distribution of organics in the atmosphere, and the spatial and temporal coverage of aerosols dominated by organics, as detailed knowledge of the organic properties is required to accurately predict  $N_{\text{CCN}}$  for these aerosols. Derivations of  $\kappa_{\text{org}}$  in future closure studies, when aerosols are dominated by organic species, would help constrain the descriptions of organics and aerosol-cloud parameterizations in large scale models. We note that the uncertainties in first indirect aerosol effect estimated here represent upper limits, which could be a factor of 2 greater than the true values, as suggested by the results from Sotiropoulou et al. (2007). Nevertheless, the analyses here show that using an average  $\overline{\kappa_{\text{org}}}$  for all organic species may be sufficient for many aerosol types. The accuracy of globally averaged aerosol indirect effects for using  $\overline{\kappa_{\text{org}}}$  should be evaluated using global models, which will be a subject of our future studies. An efficient and accurate representation of organics may require a combination of the average  $\overline{\kappa_{\text{org}}}$  for most aerosol and  $\kappa_{\text{org}}$  constrained by field studies for major aerosol types that are dominated by organics.

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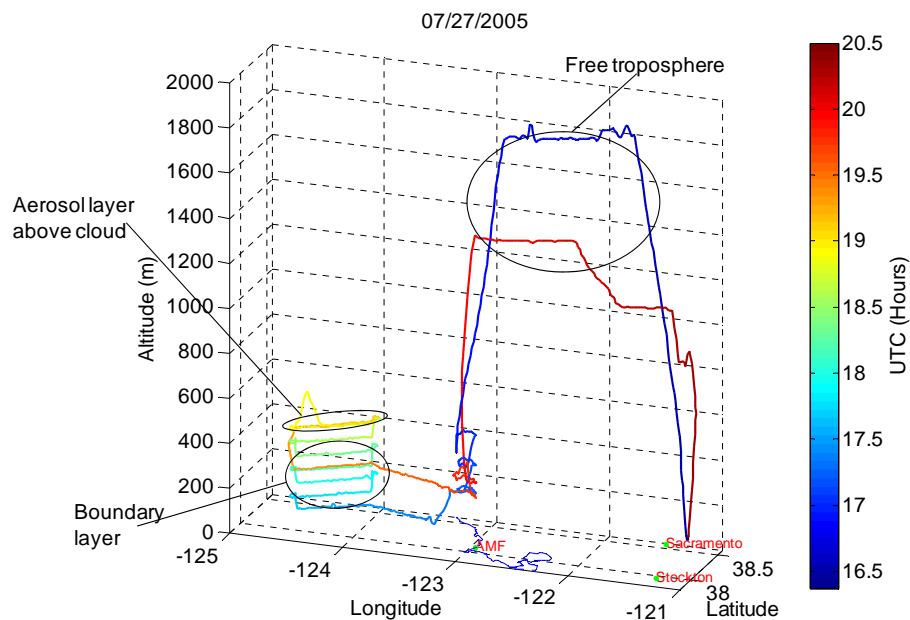
**Table 1.** The fitted lognormal mode parameters and volume fraction of  $\text{NH}_4\text{HSO}_4$ , organics, and  $\text{NH}_4\text{NO}_3$  for the aerosol observed in a thin layer above cloud on 27 July 2005.

Mode	$D_{va}$ (nm)	M ( $\mu\text{g}/\text{m}^3$ )	$\sigma$	Vol. fraction ( $\text{NH}_4\text{HSO}_4$ )	Vol. fraction ( $\text{NH}_4\text{NO}_3$ )	Vol. fraction (Organics)
1	190	5.0	1.38	3.0%	3.0%	94%
2	410	4.7	1.45	20%	2.0%	78%

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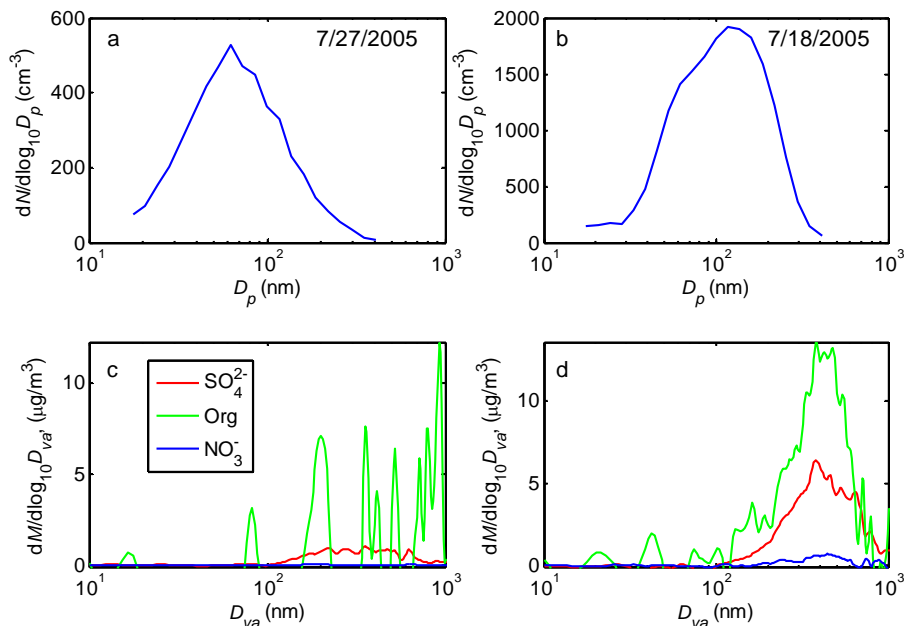


**Fig. 1.** The flight track of G-1 on 27 July 2005 and the classifications of aerosol types.

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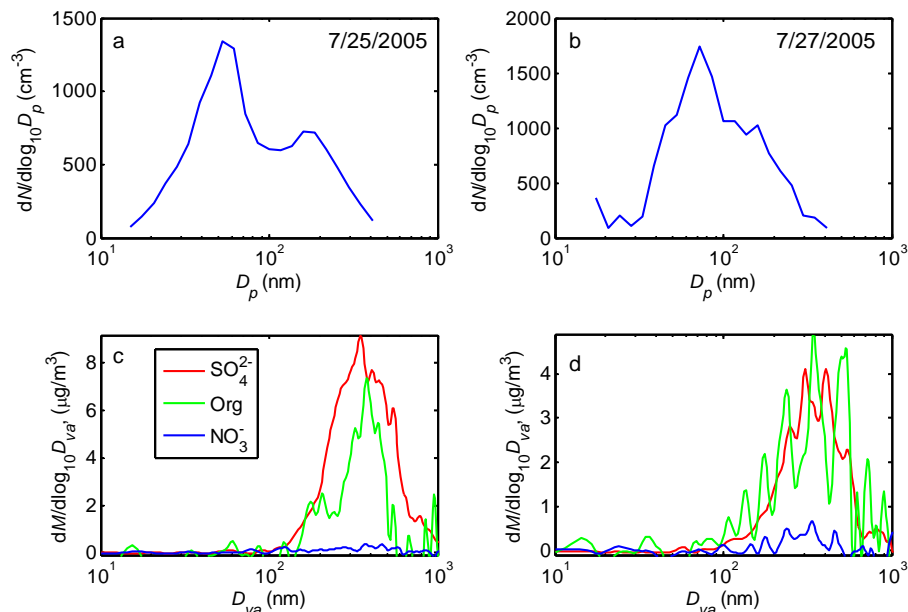


**Fig. 2.** Aerosol number size distribution (a and b) and mass size distributions of  $\text{SO}_4^{2-}$ , organics, and  $\text{NO}_3^-$  (c and d) for aerosols observed during ferry flights on 27 July 2005 (a and c) and 18 July 2005 (b and d).

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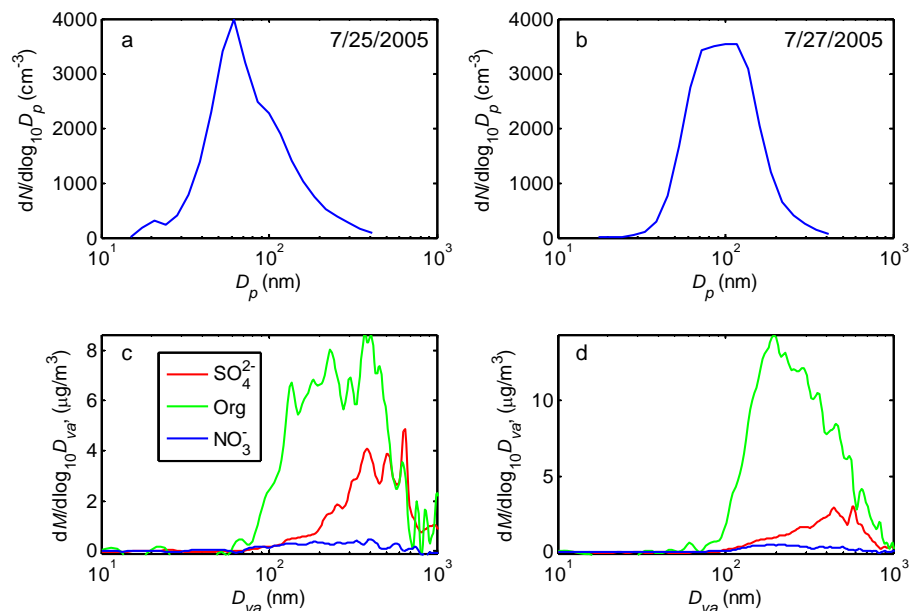


**Fig. 3.** Aerosol number size distribution (**a** and **b**) and mass size distributions of  $\text{SO}_4^{2-}$ , organics, and  $\text{NO}_3^-$  (**c** and **d**) for aerosols observed in boundary layer over ocean on 25 July 2005 (a and c) and 27 July 2005 (b and d).

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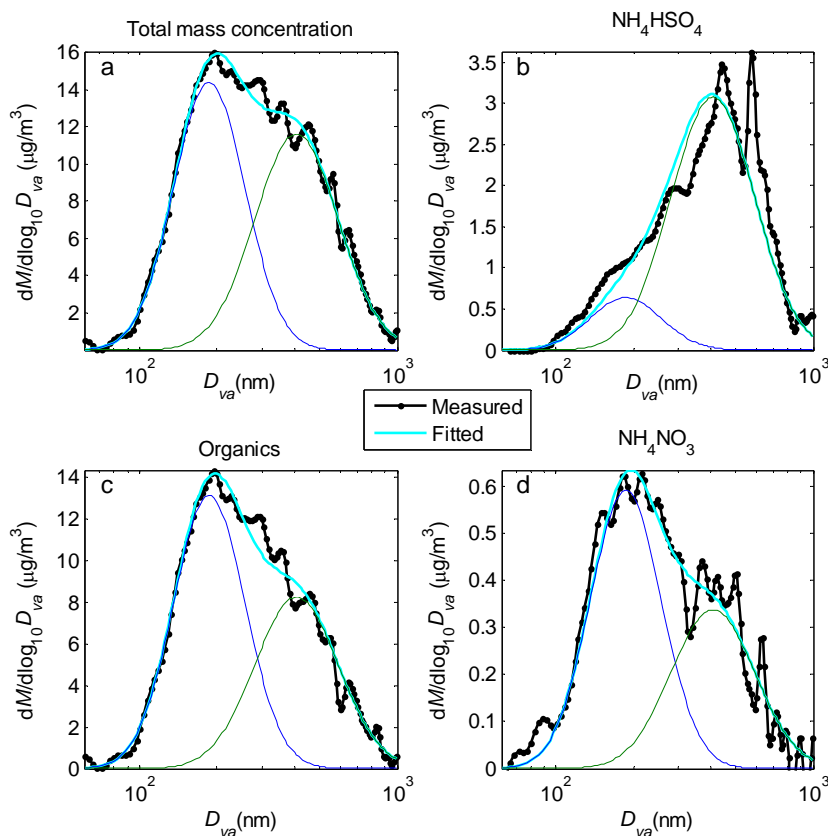
**Fig. 4.** Aerosol number size distribution (**a** and **b**) and mass size distributions of  $\text{SO}_4^{2-}$ , organics, and  $\text{NO}_3^-$  (**c** and **d**) for aerosols observed in thin layers above clouds on 25 July 2005 (a and c) and 27 July 2005 (b and d).

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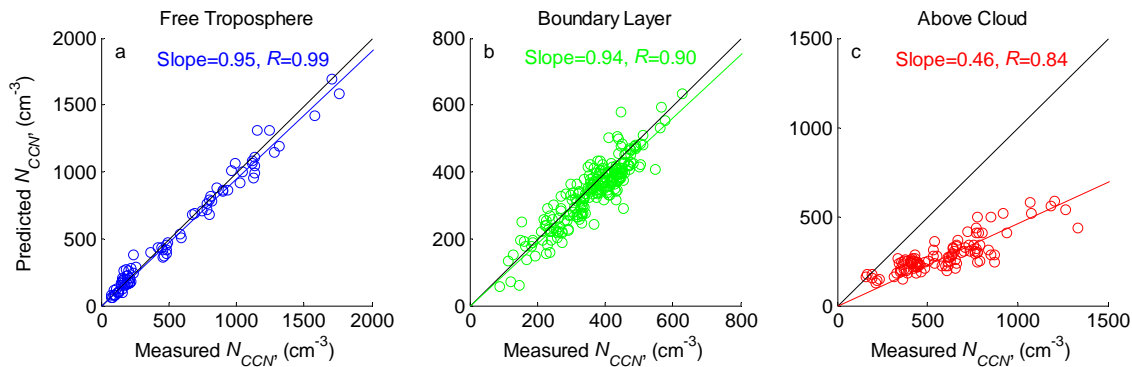


**Fig. 5.** Measured and fitted mass size distributions of total mass,  $\text{NH}_4\text{HSO}_4$ , organics, and  $\text{NH}_4\text{NO}_3$  for the aerosol observed in a thin layer above cloud on 27 July 2005. The thick black and cyan lines represent the measured and fitted mass size distributions. The thin blue and green curves are fitted mass size distributions for each lognormal mode.

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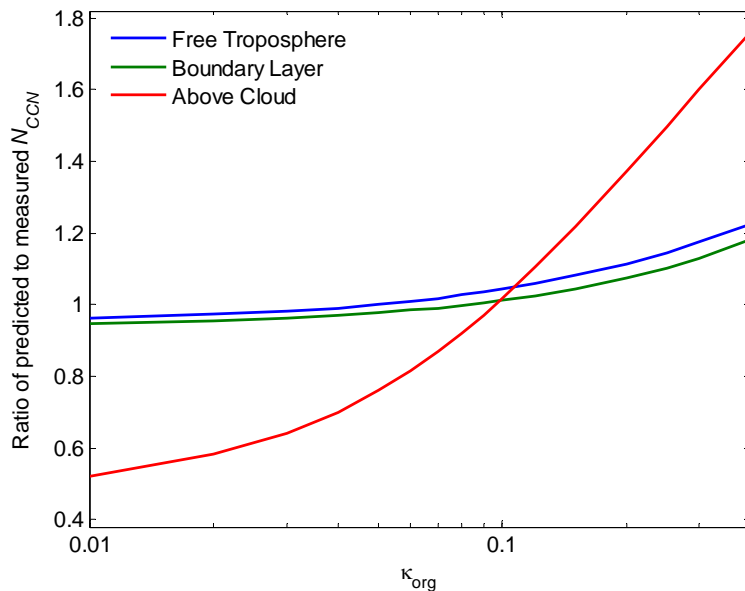


**Fig. 6.** Comparison of predicted and measured CCN at 0.22% for **(a)** Free Troposphere, **(b)** Boundary layer, and **(c)** Above Cloud aerosols.

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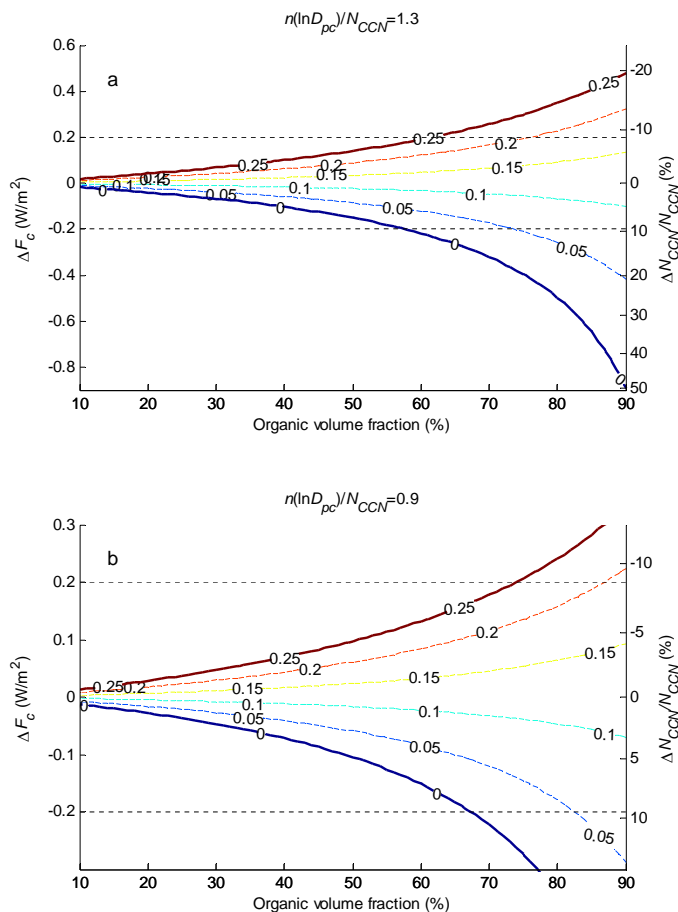


**Fig. 7.** The ratio of predicted to measured  $N_{CCN}$  as a functions of  $\kappa$  of organic species.

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**Fig. 8.** Estimated uncertainty in the first indirect aerosol effect and predicted  $N_{CCN}$  when using an average  $\overline{\kappa_{org}}$  for all organic species. The uncertainty is plotted as a function of  $x_{org}$  for (a)  $n(\ln D_{pc})/N_{CCN}=1.3$  and (b)  $n(\ln D_{pc})/N_{CCN}=0.9$  for marine aerosols. The curves are labeled with  $\kappa_{org}$ .

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