

This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leitch et al.

Cloud albedo increase from carbonaceous aerosol

**W. R. Leitch¹, U. Lohmann², L. M. Russell³, T. Garrett⁴, N. C. Shantz¹,
D. Toom-Saunry¹, J. W. Strapp¹, K. L. Hayden¹, J. Marshall⁵, D. Worsnop⁶, and
J. Jayne⁶**

¹Environment Canada, Toronto, Ontario, M3H5T4, Canada

²ETH, Zurich, Switzerland

³Scripps Institute of Oceanography, University of California, San Diego, 92093, USA

⁴University of Utah, Salt Lake City, Utah, 84112-0110, USA

⁵Max Planck Institute for Biogeochemistry, Jena, Germany

⁶Aerodyne Research, Inc., Billerica, MA 01821-397, USA

Received: 20 January 2010 – Accepted: 20 January 2010 – Published: 1 February 2010

Correspondence to: W. R. Leitch (richard.leitch@ec.gc.ca)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Abstract

Atmospheric cooling from the effect of anthropogenic carbonaceous aerosol particles on the reflectivity of sunlight by water clouds remains an uncertainty for climate prediction. Airborne measurements of aerosol and cloud properties as well as light extinction were made below, in and above stratocumulus over the Northwest Atlantic Ocean on consecutive days. On the first day, the history of the below-cloud fine particle aerosol was marine and the fine particle sulphate and organic carbon mass concentrations measured at cloud base were $2.4 \mu\text{g m}^{-3}$ and $0.9 \mu\text{g m}^{-3}$, respectively. On the second day, the below-cloud aerosol was continentally influenced and the fine particle sulphate and organic carbon mass concentrations were $2.3 \mu\text{g m}^{-3}$ and $2.6 \mu\text{g m}^{-3}$, respectively. Correspondingly, the number concentrations of aerosol particles below cloud were approximately a factor of two higher on the second day, while the below-cloud size distributions were similar on both days. The cloud droplet number concentrations (CDNC) on the second day were approximately three times higher than the CDNC measured on the first day, and the vertically integrated cloud light extinction measurements indicate a 6% increase in the cloud albedo principally due to the increase in the carbonaceous components on the second day. Locally, this albedo increase translates to a daytime radiative cooling of $\sim 12 \text{ W m}^{-2}$. This result provides observational evidence that the role of anthropogenic carbonaceous components in the cloud albedo effect can be much larger than that of anthropogenic sulphate, as some global simulations have indicated.

1 Introduction

Climate prediction is challenged by uncertainties in the cooling from the effect of anthropogenic aerosol particles on the reflectivity of sunlight by water clouds. The effect is most notable for lower altitude clouds of modest thickness such as stratocumulus. This cloud albedo effect (also known as the Twomey effect and as first indirect effect of aerosols) is rooted in the control of cloud droplet number concentrations (CDNC) by

Cloud albedo increase from carbonaceous aerosol

W. R. Leitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leaitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

aerosol particles acting as cloud condensation nuclei (CCN) (Twomey, 1977). Twenty-nine estimates of the increase in the global radiative forcing due to the cloud albedo effect range from -0.2 W m^{-2} to -1.85 W m^{-2} (Forster et al., 2007), underscoring the significance of this effect and the uncertainty in its prediction. Scaling of general circulation models with satellite data suggests the cloud albedo effect can be constrained as $-0.7 \pm 0.5 \text{ W m}^{-2}$ (Quaas et al., 2009) and an estimate based on an energy balance approach is consistent with that result (Murphy et al., 2009).

Anthropogenic sources of carbonaceous and sulphate compounds increase the number and mass concentrations of atmospheric aerosol particles and CCN. As a major constituent of the global fine particle aerosol, sulphate components are believed to contribute significantly to the cloud albedo effect due in large part to their relatively high hygroscopicity. Carbonaceous components are also ubiquitous constituents of the global fine particle aerosol (Zhang et al., 2007; Bahadur et al., 2009). Since Novakov and Penner (1993) demonstrated a connection between carbonaceous particles and CCN, many observations have indicated varying degrees of effectiveness of the carbonaceous components at water uptake. Typically carbonaceous components are much less hygroscopic than sulphate components (e.g., McFiggans et al., 2006; Petters and Kreidenweis, 2007) and their effects on CDNC and cloud albedo are highly uncertain. In the absence of significant water activity of the carbonaceous aerosol, the presence of some sulphate influences the ability of the mixed sulphate-carbonaceous particle to act as a CCN (e.g., Lohmann et al., 2004; Petters et al., 2006; King et al., 2007; Prenni et al., 2007). How carbonaceous particles contribute to the aerosol number distribution may represent their most significant CCN influence, since the number distribution is important for the number of CCN (e.g., Russell, 1999; Roberts et al., 2002; Dusek et al., 2006; Pierce et al., 2007) and potentially the CDNC (e.g., McFiggans et al., 2006). Fountoukis et al. (2007) explicitly considered the role of the carbonaceous aerosol in the nucleation of cloud droplets, and found limited sensitivity of their CDNC-aerosol closure study to the solubility of organic mass (OM). On the other hand, ambient CCN observations indicate that the hygroscopicity of the carbonaceous

components takes on greater relative importance as sulphate is reduced (e.g., Roberts et al., 2002; Ming and Russell, 2004; Chang et al., 2007; Prenni et al., 2007; Wang et al., 2008; Gunthe et al., 2009), and Wex et al. (2009) found an increase in the hygroscopicity of secondary carbonaceous components with increasing aerosol water content. Thus, there will be situations for which the typically lower hygroscopicity of carbonaceous aerosols has a significant influence on the CDNC.

The possibility of carbonaceous material, in the form of organic films, impeding water condensation and effectively lowering the mass accommodation coefficient for water vapour condensing on the growing droplets (α_c) has been considered for several years (e.g., Penner et al., 2001). As droplets grow, albeit more slowly, such films are likely to break, thus removing the impedance to water uptake (e.g., Feingold and Chuang, 2002). Johnson et al. (2005) found that volatilization of a low solubility organic that may have coated particles resulted in an increase in their water uptake rate suggesting suppression by the organic material. Kinetic inhibition has also been observed with some anthropogenic aerosols (Ruehl et al., 2008, 2009; Shantz et al., 2010). Shantz et al. (2010) found that values of α_c for concentrated solution droplets needed to be in the range 0.04–0.07 in order to explain observed rates of water uptake by anthropogenic particles. As droplets dilute and the situation approaches water on water condensation, a value of unity is appropriate for α_c (Wagner et al., 1982; Mozurkewich, 1986; Davidovits et al., 2004; Laaksonen et al., 2005). Particularly with field observations, the effects of mass accommodation, surface tension and other physical-chemical properties of the carbonaceous aerosol are not always easily distinguished.

Global simulations of the indirect effect have begun to move away from empirical prediction of CDNC based on a parameterization of sulphate as a surrogate towards the explicit representation of the major global aerosol components. These mechanistic approaches are required for long range prediction; although they have not yet reduced the uncertainty range of the indirect aerosol effect because the aerosol components and their size distributions introduce new uncertainties (Menon et al., 2003; Penner et al., 2006; Lohmann, 2007; Pringle et al., 2009). Some studies that have used mechanistic

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

treatments of the carbonaceous and sulphate aerosols have attributed more radiative cooling via the cloud albedo effect to the carbonaceous aerosol than to the sulphate aerosol. Lohmann et al. (2000) found that the change in the radiative cooling by the cloud albedo effect between present-day and pre-industrial time ranged from near zero to -0.4 W m^{-2} due to anthropogenic sulphate and -0.9 W m^{-2} to -1.3 W m^{-2} due to anthropogenic carbonaceous aerosol. The reduced effect of the sulphate aerosol relative to the carbonaceous aerosol was a combination of a larger increase in the carbonaceous burden and a larger mode radius of the sulphate. Chuang et al. (2002) estimated a global cloud albedo effect of -1.85 W m^{-2} , of which -0.30 W m^{-2} was associated with anthropogenic sulphate, -1.16 W m^{-2} was associated with carbonaceous aerosols from biomass burning, and -0.52 W m^{-2} was associated with carbonaceous aerosols from fossil fuel combustion. Together, these two studies suggest that the impact of cloud albedo by the carbonaceous aerosol is 3–6 times that due to the sulphate. On the other hand, Ghan et al. (2001) found their mechanistic model indicated the cloud albedo effect was dominated by sulphate.

Here, we present observational evidence, analysed with the aid of an adiabatic aerosol-cloud parcel model, to show that the carbonaceous aerosol can indeed enhance the cloud albedo effect as suggested by Lohmann et al. (2000) and Chuang et al. (2002). We contrast airborne observations of aerosol and cloud properties from two flights on back-to-back days over the Atlantic Ocean. These cases are unique in two ways. First, the trajectories suggest a marine character to the aerosol on the first day and a continental/anthropogenic composition to the aerosol on the second day. Second, the number size distribution of the cloud base aerosol sampled on the second day is similar in shape but approximately twice the concentration of that sampled on the first day. The increase in the size distribution is explained by an increase in the submicron carbonaceous aerosol mass concentration relative to the submicron sulphate aerosol mass concentration and the increase is reflected in the observed CDNC and cloud light extinction.

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

2 Experimental

2.1 Overview

The measurement platform was the National Research Council of Canada Convair 580 aircraft. The two flights considered here were conducted in and around stratocumulus on 13 October 2003 and 14 October 2003 within a few hundred kilometers to the east of Nova Scotia; hereafter referred to as flights 1 and 2, respectively. The flights were part of the Canadian SOLAS program. Profile data from flights 1 and 2 were collected near 42°48' N, 62° W and 44°50' N, 57°20' W, respectively. The meteorological back trajectory analyses from the NOAA HYSPLIT model (Draxler and Rolph, 2003; Rolph, 2003) indicate that the aerosol sampled during flight 1 had resided over the Atlantic Ocean to the south-southwest of the sampling area for three days prior to observation (Fig. 1a). The trajectory analysis for flight 2 indicates a significant continental influence to the aerosol (Fig. 1b).

2.2 Instrumentation

Relevant measurements and the corresponding instrumentation are outlined in Table 1. Comprehensive descriptions of the Aerodyne Quadruple Aerosol Mass Spectrometer (Q-AMS) are found elsewhere (Jayne et al., 2000; Jimenez et al., 2003). The Q-AMS was used to measure the average mass concentration and size distributions of non-refractory particulate species every 5-min; 5-min averages were chosen to optimize the signal-to-noise versus the spatial resolution. The temperature of the vaporizer in the AMS, used to volatilize the particles into molecular fragments, was set to about 550°C enabling the measurement of components such as ammonium sulphate and many organics, but not sodium chloride. The transmission of particles into the AMS is approximately 100% in the range 0.1–0.7 μm vacuum aerodynamic diameter (D_{va}), decreasing sharply outside that range (Liu et al., 2007); the upper transmission limit

Cloud albedo increase from carbonaceous aerosol

W. R. Leitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leaitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

for this particular AMS has been demonstrated (Rupakheti et al., 2005). The collection efficiency of particles by the oven is assumed to be 100% for this dataset. This assumption is based on the comparison of the sulphate mass concentrations from the Q-AMS with those from the Particle-In-Liquid-Sampler (PILS; described below), and it is consistent with previous results with this Q-AMS (Rupakheti et al., 2005; Buset et al., 2006; Phinney et al., 2006, 2009); more acidic sulphate, as here, is efficiently sampled. Analysis of AMS data was performed using the Deluxe 1.29 IGOR data analysis package (Allan et al., 2003) with a batch file (used for quantitative calibration) and fragmentation file (used for identification of chemical species present on the aerosol) customized to this data set. The fragmentation file included methanesulphonic acid (Phinney et al., 2009), but MSA was not found to be significant in these observations.

Aerosol particles were collected in water using a PILS and analyzed for their major water soluble inorganic chemical components onboard with two ion chromatographs (IC). The PILS collection system is described briefly here, and it is similar to that described by Orsini et al. (2003); this particular PILS has been previously documented (Buset et al., 2006) and further details are provided in the Supplement (Table S-1: <http://www.atmos-chem-phys-discuss.net/10/2131/2010/acpd-10-2131-2010-supplement.pdf>). Trace gases were denuded from the aerosol prior to entering the PILS, where the particles are grown to supermicron size droplets under supersaturated conditions created by mixing the sample air with steam. Particles larger than 0.03 μm diameter that enter the PILS are activated with a >97% collection efficiency at a sample flow rate of 15 l m^{-1} as used here. The droplets are impacted onto a surface that is washed off with a steady stream of de-ionized water. The water is delivered to trace concentrator columns of the IC for analysis. Samples were averaged for 10 min.

Four instruments were used to measure the size distributions of the aerosol particles. A TSI Scanning Mobility Particle System (SMPS) with a TSI 3010 Condensation Particle Counter (CPC) was used to measure particles from 10 nm to 500 nm geometric diameter (D_g). A TSI Aerodynamic Particle Sizer (APS) was used to measure particles

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

from 0.6 μm to 20 μm aerodynamic diameter (D_a). Both the SMPS and APS were located inboard the aircraft. Two Particle Measuring Systems (PMS) probes, a Passive Cavity Aerosol Spectrometer Probe (PCASP-100x) and a Forward Scattering Spectrometer Probe (FSSP-300) were used to measure particles from 0.14 μm to 1 μm D_g and from 0.3 μm to 20 μm D_g , respectively. The PCASP-100x and the FSSP-300 were suspended from a pylon under a wing of the aircraft. The FSSP is a non-intrusive probe and the particle sizing includes any water that the particles retain. The other three instruments (SMPS, APS and PCASP) are assumed to have measured the nearly dry sizes of the particles due to internal heating (PCASP), the slight warming of air as it entered the aircraft from outside (SMPS and APS), and dried sheath air used (all three). A comparison of this PCASP-100x and FSSP-300 is discussed by Strapp et al. (1992).

A shrouded nearly isokinetic diffuser set 15 cm off the starboard side of the fuselage ahead of the engine was used to bring the aerosol into the cabin. From there, the aerosol was carried by 1 cm OD stainless steel tubing to the SMPS, APS, AMS and PILS. The distance from the intake point to the SMPS and APS was about 1 m and about 5 m to the AMS and PILS. Agreement between the APS and FSSP-300 size distributions was reasonable in the overlap region below 1 μm , and losses at the aircraft intake were found to affect only particles larger than about 1 μm . The particle concentrations measured with the APS above 1 μm were significantly lower than the FSSP-300 (Fig. 2); this can be interpreted as poor transmission of particles $>1 \mu\text{m}$ into and through the inlet system, but drying of the particles may also contribute to the differences. Only size distribution data from the FSSP300 are used for particles $>0.9 \mu\text{m}$.

Cloud liquid water contents (LWC) were measured using a PMS King LWC probe and a Nevzorov LWC probe deployed from a pylon suspended from a wing; comparison of the LWC measurements is given in the Supplement (Fig. S-1: <http://www.atmos-chem-phys-discuss.net/10/2131/2010/acpd-10-2131-2010-supplement.pdf>). The LWC is measured to within 15% and the baseline drift is estimated to be $<0.02 \text{ g m}^{-3}$ (Cober et al., 2001). Correction factors applied for the effects of flow

around the aircraft were between 1.03 and 1.05 (Cober et al., 2001; Drummond and MacPherson, 1985).

Two PMS FSSP100 probes, also suspended from a pylon under a wing, were used to measure the cloud droplet number size distribution and covered the size range of 2–47 μm . Their sample volumes are based on the measured sample area of the probe and the measured airspeed. Sizing calibrations, corrected to the index of refraction of water, were done with glass beads. The data are 1 s average values, representing a sampling path length of 90–100 m. The cloud droplet number concentrations (CDNC) are corrected for coincidence error and probe dead time (Baumgardner et al., 1985). Corrections of the FSSP100 data for flow around the aircraft were not taken into account because the effects of flow on the measurements (ca. 2%) were significantly less than the probe measurement accuracies for the CDNC ($\pm 15\%$) (Cober et al., 2001).

Measurements of the cloud extinction coefficient were made using a Gerber Scientific Inc. Cloud Integrating Nephelometer (CIN; Gerber et al., 2000; Garrett et al., 2001). The CIN probe was suspended from a pylon under a wing. It illuminates a population of cloud particles with laser light at 625 nm wavelength and measures the near-forward and near-backward scattered light intensities from cloud particles 2 μm and larger. The light extinction coefficients from the CIN probe correspond with the light scattering coefficients calculated from the cloud droplet size distributions measured with the FSSP 100 probes (Supplement, Fig. S-2: <http://www.atmos-chem-phys-discuss.net/10/2131/2010/acpd-10-2131-2010-supplement.pdf>). The CIN probe provides a measurement that is independent from the FSSP100 probes, thereby avoiding ambiguities that can occur when comparing the CDNC with cloud light extinction.

3 Observations

Aerosol number concentrations for particles $> 140\text{ nm}$ (APNC_{140}) and CDNC during profiles through the clouds are shown in Fig. 3a and 3b; 140 nm roughly corresponds to the lower size of particles that will activate to cloud droplets in these types of clouds

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leaitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leaitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

(e.g., Leaitch et al., 1996; Russell et al., 1999). The mean APNC₁₄₀ at cloud base during flight 1 was $170\text{ cm}^{-3} \pm 50\text{ cm}^{-3}$ and the mean CDNC through the profile was $190\text{ cm}^{-3} \pm 50\text{ cm}^{-3}$ for cloud liquid water contents (LWC) $>0.15\text{ g m}^{-3}$; smaller LWC are excluded to remove reductions in the CDNC by evaporation at cloud edges. For flight 2, the mean APNC₁₄₀ for cloud base was $390\text{ cm}^{-3} \pm 100\text{ cm}^{-3}$ and the mean CDNC through the profile was $560\text{ cm}^{-3} \pm 110\text{ cm}^{-3}$. The cloud base aerosol and CDNC sampled during flight 2 are higher than those of flight 1 at a confidence level of $>99\%$ (Table 2; time series of the observations are found in the Supplement, Fig. S-3: <http://www.atmos-chem-phys-discuss.net/10/2131/2010/acpd-10-2131-2010-supplement.pdf>).

The LWC profiles were near adiabatic, and the steady increase of the LWC from bottom to top indicates lifting from cloud base (Fig. 3c). Not surprisingly, detection of an aerosol-cloud albedo effect is easier for nearly adiabatic clouds (Kim et al., 2008). The cloud light extinction values were higher for the profile of flight 2 (Fig. 3d). Bifurcation of the profile measurements during flight 2 is the result of the aircraft flying momentarily out of the cloud and then back in. The vertically integrated LWC or cloud liquid water path (LWP) is 30 g m^{-2} for the flight 1 profile and 20 g m^{-2} for the flight 2 profile. Despite the lower LWP, the vertically integrated light extinction (optical depth) through the cloud of flight 2 is higher than that of flight 1 (4.2 versus 3.8) due to the higher CDNC. The cloud droplet size distributions from one of the FSSP-100 probes and averaged through each profile are shown in Fig. 4, indicating a mode diameter of about $12\text{ }\mu\text{m}$ for the distribution of flight 1 and a mode diameter of about $8\text{ }\mu\text{m}$ for the flight 2 distribution. The lower LWP of flight 2 is partially due to flying out of cloud, but the changes in albedo and LWP from flight 1 to flight 2 are consistent with recent studies associating aerosol with changes in LWP and cloud optical depths (Avey et al., 2007; Lebsock et al., 2008).

Profile data of the non-refractory chemical composition of the cloud base fine particle aerosol are shown in Fig. 5. The fine aerosol was dominated by sulphate components on flight 1 and by a mix of organic components (OM: carbonaceous material exclusive of elemental carbon) and sulphate components on flight 2; respective fine particle

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



5 sulphate and OM mass concentrations measured at cloud base were $2.4 \mu\text{g m}^{-3}$ and $0.9 \mu\text{g m}^{-3}$ during flight 1 and $2.3 \mu\text{g m}^{-3}$ and $2.6 \mu\text{g m}^{-3}$ during flight 2 (Table 3). Collection efficiency aside, the maximum uncertainty for the mass concentrations measured with this Q-AMS is estimated at $\pm 20\%$ of the mass concentrations (Rupakheti et al., 2005).

10 The aerosol particle size distributions at cloud base (Fig. 2) differ principally in number concentration; the flight 2 cloud base number concentration is about twice that of flight 1 above 60 nm. The variations of the mass fractions of sulphate relative to sulphate plus OM with particle size from 100 nm to 500 nm are shown in Fig. 6a. The sums of sulphate and OM compare with the total mass distributions estimated from the physical size distributions (Fig. 6b), and no other chemical species of significant concentration were measured in this size range. Detectable sodium and chloride as well as unexplained particle volume increase are evidence for a sea salt component to the particles above 500 nm during flight 2 (Supplement, Fig. S-4: <http://www.atmos-chem-phys-discuss.net/10/2131/2010/acpd-10-2131-2010-supplement.pdf>). Table 4 summarizes the physical properties of the cloud base aerosols observed during the two flights.

4 Modelling

20 An aerosol-cloud adiabatic parcel model is used to model the CDNC for comparison with the observations. The model includes representation of weakly soluble compounds (Shantz et al., 2003) as well as the effects of different surface tensions (Lohmann et al., 2004). The cloud base aerosol size distributions are described by multiple modes each represented as a log-normal function (Supplement, Fig. S-5: <http://www.atmos-chem-phys-discuss.net/10/2131/2010/acpd-10-2131-2010-supplement.pdf>). The chemical representations of each mode are based on the AMS and PILS data for particles > 100 nm. Smaller particles are assumed to be either 100% H_2SO_4 or 70% organic and 30% sulphate by mass, as indicated.

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Table 5 gives the physical and chemical representations for each mode used in the simulations; other initial conditions used in the simulations are given at the bottom of Table 5. Except as discussed, all chemical components within each mode are assumed to be internally mixed. The simulated CDNC are defined as droplets $>2\ \mu\text{m}$ diameter that are still growing after the maximum supersaturation is reached, in accordance with the lower size of the FSSP-100 and the measured distributions (Fig. 4).

Another critical influence on droplet growth rates is the cloud updraft speed. Higher updraft speeds increase the cloud base supersaturation and the resulting droplet growth rates. The method of assessing the appropriate updraft speed to use with an adiabatic parcel model is not well established. In the case of stratiform cloud, as in the present study, it is difficult if not impossible to identify an updraft core. We use the results of the Peng et al. (2005) analysis that indicated a value of one standard deviation of the measured gust velocities provided the best closure of the aerosol and cloud measurements. The physical basis for this is that the smaller positive updrafts sampled when flying level through a stratiform cloud are the result of changes in the stronger vertical motions within cloud, and they are not the updrafts that penetrate from below cloud base up into the cloud. It is expected that there is some distribution of higher updrafts that specify CDNC in the cloud. Based on this reasoning, we expect the updraft speeds higher than one standard deviation are also a factor in determining the CDNC. The updraft speed is represented in the model here by one standard deviation of the vertical gust measurements during level flight: $14\ \text{cm s}^{-1}$ for flight 1 and $50\ \text{cm s}^{-1}$ for flight 2 (Supplement, Fig. S-6: <http://www.atmos-chem-phys-discuss.net/10/2131/2010/acpd-10-2131-2010-supplement.pdf>). Simulations were also conducted with the maximum gusts: $20\ \text{cm s}^{-1}$ for flight 1 and $100\ \text{cm s}^{-1}$ for flight 2 to include the possible range of updrafts.

The sensitivity of the simulated CDNC to the assumption of the composition of the smallest particles that can contribute to the CDNC in this case (mode 2 in Table 5) was considered in the case of flight 2 by changing the composition from H_2SO_4 to 70% OM and 30% H_2SO_4 assuming a solubility of $5\ \text{g l}^{-1}$ for the OM. The sensitivity to the

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leaitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

assumption of an internal mix of the organics and the sulphate was tested by assuming that the OM and the sulphate in modes 2 and 3 of flight 2 were externally mixed. These results are shown under “External mixture” in Table 6. The surface tension used in these calculations is based on that of adipic acid (Ervens et al., 2004). The impact of this surface tension is considered by simulating the flight 2 internally mixed cases for a surface tension of water plus ammonium sulphate for the OM solubilities of 5 and 200 g l^{-1} and for updrafts of 50 and 100 cm s^{-1} . These cases were selected because they are the conditions closest to the observations and should offer the greatest sensitivity to the surface tension due to the higher organic concentrations in solution.

For the reasons discussed in the introduction, we use a value of unity for α_c in the simulations. We expect that the droplets at the time they reach cloud base are sufficiently dilute that $\alpha_c=1$ is appropriate. Fountoukis et al. (2007) used a value of $\alpha_c=0.06$, and we consider the effect of using that lower value; however, the updraft speeds used by Fountoukis et al. (2007) are relatively lower than based on our approach and are mostly for cumulus. The net effect of a lower α_c is to increase the cloud base supersaturation by limiting water uptake (e.g., Leaitch et al., 1986). The net effect of lowering the updraft speed is to reduce the cloud base supersaturation by reducing the rate of cooling. Thus, studies that use lower values of the α_c and the updraft speed can produce results that are comparable to studies that use higher values of the α_c and the updraft speed.

The cloud albedo (A_c) is calculated using

$$A_c = \tau / (7.7 + \tau)$$

where τ is the optical depth calculated from the integration of the extinction measurements through the cloud profile (e.g., Menon et al., 2003). The planetary albedo (A_p) using

$$A_p = A_c + [(1 - A_c)^2 A_s] / (1 - A_c A_s)$$

where in this case A_s is the underlying reflectance from the ocean surface and assumed to be 0.1. The local radiative forcing is simply estimated assuming an increase of -2 W m^{-2} for a planetary albedo increase of 0.5% (Ramanathan, 1988).

5 Results and discussion

5 The CDNC simulated for flight 1 assuming internally mixed sulphate and OM (225 cm^{-3} ; Table 6, 14 cm s^{-1}) are within the range of the observed values (140 – 240 cm^{-3}) and within 18% of the observed mean CDNC. For flight 2, the CDNC was simulated for the three possible unique states of the cloud base aerosol. First, for the assumption that the carbonaceous components of flight 2 were particles externally
10 mixed relative to the sulphate components and either hydrophobic or weakly hygroscopic (Fig. 7a) the simulated CDNC (370 cm^{-3} ; Table 6, red) is outside of the range of the observed CDNC (450 – 770 cm^{-3}) and 34% lower than the mean. Second, for the assumption that the carbonaceous particles were externally mixed and highly hygroscopic (Fig. 7b) the resulting CDNC (560 cm^{-3} ; Table 6, green) is equivalent to the
15 mean of the observed CDNC. Third, for the assumption that carbonaceous particles served primarily as substrates for sulphate condensation (internal mixture, Fig. 7c) and defined the CCN number concentration the simulated CDNC (491 – 579 cm^{-3} ; Table 6, yellow) are within the range of the observed CDNC and the mean (535 cm^{-3}) is within 7% of the observed mean. Reducing the surface tension in these models produced
20 no significant change in the CDNC, and reducing the mass accommodation coefficient to 0.06 (e.g., Fountoukis et al., 2007) increased the CDNC by 12%. The only aerosol model of flight 2 that does not represent the increase in the CDNC of flight 2 relative to flight 1 is that which assumes no effect of the carbonaceous particles on the CDNC (Fig. 7a). Thus, we find that the carbonaceous components of the below-cloud aerosol
25 contributed to the increase in the CDNC.

The updraft speed of flight 2 is applied to the flight 1 aerosol in order to assess the degree to which the higher updrafts of flight 2 were responsible for the higher

Cloud albedo increase from carbonaceous aerosol

W. R. Leaitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

CDNC. The resulting increase in the CDNC (330 cm^{-3} from 225 cm^{-3} ; Table 6) is about 30% of the difference between flights 1 and 2 (560 cm^{-3} from 225 cm^{-3}), leaving 70% of the increase attributable to the change in the aerosol. For 70% of the difference in the optical depths (4.2–3.8), we estimate that the increase in aerosol due to the carbonaceous components produced a local increase in cloud albedo of 5.5%, which equates to a local radiative forcing of approximately -12 W m^{-2} for an ocean surface reflectance of 10%. Because of the reduced vertically integrated cloud liquid water during flight 2, the above forcing estimate is conservative.

The above attribution is predicated on the higher updraft speed during flight 2 as the application of the weaker updraft of flight 1 to the aerosol of flight 2 simulates a CDNC that is slightly lower than that of flight 1. A lower updraft slows the rate of condensation, and only CCN with larger critical diameters will activate or reach cloud droplet size. In that situation, the chemistry has a substantial bearing on the lower activation diameter; CCN observations that point to the importance of the size distribution do not consider supersaturations $<0.1\%$ that are relevant to flight 1. At higher updraft speeds, the cloud base supersaturation is higher (up to 0.2% as modelled for flight 2; Supplement, Fig. S-7: <http://www.atmos-chem-phys-discuss.net/10/2131/2010/acpd-10-2131-2010-supplement.pdf>), the critical diameter of activation is smaller and the number distribution becomes a stronger influence for the CDNC; however the chemistry of the larger CCN still is important due to its control of the cloud base supersaturation via its effect on the water uptake rate. This point is illustrated here by the decrease in the CDNC for the assumption that the aerosol of flight 2 is all sulphate and the updraft speed of 100 cm s^{-1} or greater. It is also consistent with the suggestion that the assumption of organic compounds as hygroscopic affects the sensitivity of the CDNC to the vertical velocity (Rissman et al., 2004; Lance et al., 2004).

Since the flight 2 observations were made at least one day or more downwind of significant carbonaceous aerosol sources (Fig. 1b) the sulphate and carbonaceous components were internally mixed to some degree (e.g., Covert and Heintzenberg, 1984; Heald et al., 2005). Also, previous observations of the residuals of cloud droplets

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leaitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

from marine clouds have found that both components co-exist in cloud water (e.g., Straub et al., 2007; Targino et al., 2007; Hawkins et al., 2008). This is the situation represented in Fig. 7c, in which case the ability of the carbonaceous components to affect the water uptake rate of the flight 2 aerosol has a relatively small effect on the simulated CDNC (Table 6) due to the influence of the sulphate. However, if we assume that the particles are composed completely of sulphate a slight reduction in the CDNC results because the large increase in droplet growth rates reduces the cloud base supersaturation.

The effects of carbonaceous components on the CDNC are inherent in the sulphate-CDNC empirical parameterization (Penner et al., 2001), but they can not be separated from those of sulphate. Menon et al. (2002) developed an empirical relationship for CDNC in an attempt to explicitly identify changes in the CDNC due to changes in sulphate, organic, and sea salt mass concentrations. Application of that formulation to the present observations produces only a 15% increase in the CDNC from flight 1 to flight 2 compared with the 70% increase based on the present observations and analysis. Pringle et al. (2009) finds no reason to suggest mechanistic models are currently better than empirical models, other than the potential for the mechanistic model to better represent regional effects. However, as we begin to consider the effects of changes in emissions (e.g., Kloster et al., 2008), our result emphasises the point that accurate distinction between the carbonaceous and sulphur components is essential for assessing changes in the cloud albedo effect, and continued development of mechanistic approaches is the only way to achieve the needed confidence in our ability to predict effects of emissions changes.

6 Summary and conclusions

A comparison of measurements made above, in and below stratocumulus during flights on two sequential days over the Atlantic Ocean show higher values of the cloud-base fine aerosol particle number concentrations, cloud droplet number concentrations,

cloud light extinction and hence cloud albedo on the second day. The increase in the fine particle cloud base aerosol is principally the result of an increase in the carbonaceous components in the aerosol.

Although we only contrast two cases, the result suggests that the contribution to radiative forcing via the cloud albedo effect from carbonaceous components can be not only substantial but significantly higher than the contribution from sulphate, in agreement with some global models using mechanistic approaches to represent the aerosol and CDNC.

Acknowledgements. We thank C-SOLAS, Rodney Weber, Mohammed Wasey, Mark Couture, Rachel Chang, Rachel Schwartz, Ryan Leitch and the Institute for Aerospace Research. Funding was provided by the Natural Sciences and Engineering Research Council of Canada, the Canadian Foundation for Climate and Atmospheric Science, and Environment Canada. The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and/or READY website (<http://www.arl.noaa.gov/ready.php>) used in this publication.

References

- Allan, J. D., Jimenez, J. L., Coe, H., Bower, K. N., Williams, P. I., Canagaratna, M. R., Jayne, J. T., and Worsnop, D. R.: Quantitative sampling using an aerodyne aerosol mass spectrometer, 1. Techniques of data interpretation and error analysis, *J. Geophys. Res.*, 108, 4090–4100, 2003.
- Avey, L., Garrett, T. J., and Stohl, A.: Evaluation of the aerosol indirect effect using satellite, tracer transport model, and aircraft data from the International Consortium for Atmospheric Research on Transport and Transformation, *J. Geophys. Res.*, 112, D10S33, doi:10.1029/2006JD007581, 2007.
- Bahadur, R., Habib, G., and Russell, L. M.: Climatology of PM_{2.5} organic carbon concentrations from a review of ground-based atmospheric measurements by evolved gas analysis, *Atmos. Environ.*, 43(D21), 1591–1602, 2009.
- Baumgardner, D., Strapp, J. W., and Dye, J. E.: Evolution of the forward scattering spectrometer

Cloud albedo increase from carbonaceous aerosol

W. R. Leitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leaitch et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

probe, part II: corrections for coincidence and dead time errors, *J. Atmos. Ocean. Tech.*, 2, 626–632, 1985.

Buset, K. C., Evans, G. J., Leaitch, W. R., Brook, J. R., and Toom-Sauntry, D.: Use of advanced receptor modeling for analysis of an intensive 5-week aerosol sampling campaign, *Atmos. Environ.*, 40, 482–499, 2006.

Chang, R. Y.-W., Liu, P. S. K., Leaitch, W. R., and Abbatt, J. P. D.: Comparison between measured and predicted CCN concentrations at Egbert, Ontario: focus on the organic aerosol fraction at a semi-rural site, *Atmos. Environ.*, 41, 8172–8182, 2007.

Chuang, C. C., Penner, J. E., Prospero, J. M., Grant, K. E., Rau, G. H., and Kawamoto, K.: Cloud susceptibility and the first aerosol indirect forcing: sensitivity to black carbon and aerosol concentrations, *J. Geophys. Res.*, 107, D21, doi:10.1029/2000JD000215, 2002.

Cober, S. G., Isaac, G. A., Korolev, A. V., and Strapp, W.: Assessing cloud-phase conditions, *J. Appl. Meteorol.*, 40, 1967–1983, 2001.

Covert, D. S. and Heintzenberg, J.: Measurement of the degree of internal external mixing of hygroscopic compounds and soot in atmospheric aerosols, *Sci. Total Environ.*, 36, 347–352, 1984.

Davidovits, P., Worsnop, D. R., Jayne, J. T., Kolb, C. E., Winkler, P., Vrtala, A., Wagner, P. E., Kulmala, M., Lehtinen, K. E. J., Vesala, T., and Mozurkewich, M.: Mass accommodation coefficient of water vapor on liquid water, *Geophys. Res. Lett.*, 31, L22111, doi:10.1029/2004GL020835, 2004.

Draxler, R. R. and Rolph, G. D.: HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) model access via NOAA ARL READY website (<http://www.arl.noaa.gov/HYSPLIT.php>), last access: 18 January 2010, NOAA Air Resources Laboratory, Silver Spring, MD, 2003.

Drummond, A. M. and MacPherson, J. I.: Aircraft flow effects on cloud drop images and concentration measured by the NAE Twin Otter, *J. Atmos. Ocean. Tech.*, 2, 633–643, 1985.

Dusek, U., Frank, G. P., Hildebrandt, L., Curtius, J., Schneider, J., Walter, S., Chand, D., Drewnick, F., Hings, S., Jung, D., Borrmann, S., and Andreae, M. O.: Size matters more than chemistry for cloud-nucleating ability of aerosol particles, *Science*, 312, 1375–1378, 2006.

Ervens, B., Feingold, G., Clegg, S. L., and Kreidenweis, S. M.: A modeling study of aqueous production of dicarboxylic acids: 2. Implications for cloud microphysics, *J. Geophys. Res.*, 109, D15, doi:10.1029/2004JD004575, 2004.

Feingold, G. and Chuang, P. Y.: Analysis of the influence of film-forming compounds on droplet

growth: implications for cloud microphysical processes and climate, *J. Atmos. Sci.*, 59, 2006–2018, 2002.

Forster, P., Ramaswamy, V., Artaxo, P., Bernsten, T., Betts, R., Fahey, D., Haywood, J., Lean, J., Lowe, D., Myhre, G., Nganga, J., Prinn, R., Raga, G., Schulz, M., and Van Dorland, R.: Radiative forcing of climate change, in: *Climate Change 2007: The Physical Science Basis, Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by: Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K. B., Tignor, M., and Miller, H., Cambridge Univ. Press, New York, 129–234, 2007.

Fountoukis, C., Nenes, A., Meskhidze, N., Bahreini, R., Conant, W. C., Jonsson, H., Murphy, S., Sorooshian, A., Varutbangkul, V., Brechtel, F., Flagan, R. C., and Seinfeld, J. H.: Aerosol-cloud drop concentration closure for clouds sampled during the International Consortium for Atmospheric Research on Transport and Transformation 2004 campaign, *J. Geophys. Res.*, 112, D10S30, doi:10.1029/2006JD007272, 2007.

Garrett, T. J., Hobbs, P. V., and Gerber, H.: Shortwave, single-scattering properties of arctic ice clouds, *J. Geophys. Res.*, 106, 15155–15172, 2001.

Gerber, H., Takano, Y., Garrett, T. J., and Hobbs, P. V.: Nephelometer measurements of the asymmetry parameter, volume extinction coefficients and backscatter ratio in clouds, *J. Atmos. Sci.*, 37, 3021–3034, 2000.

Ghan, S., Easter, R. G., Chapman, E., Abdul-Razzak, H., Zhang, Y., Leung, L. R., Laulainen, N. S., Saylorand, R. D., and Zaveri, R. A.: A physically based estimate of radiative forcing by anthropogenic sulfate aerosol, *J. Geophys. Res.*, 106, 5279–5293, 2001.

Gunthe, S. S., King, S. M., Rose, D., Chen, Q., Roldin, P., Farmer, D. K., Jimenez, J. L., Artaxo, P., Andreae, M. O., Martin, S. T., and Pöschl, U.: Cloud condensation nuclei in pristine tropical rainforest air of Amazonia: size-resolved measurements and modeling of atmospheric aerosol composition and CCN activity, *Atmos. Chem. Phys.*, 9, 7551–7575, 2009,

<http://www.atmos-chem-phys.net/9/7551/2009/>.

Hawkins, L. N., Russell, L. M., Twohy, C. H., and Anderson, J. R.: Uniform particle-droplet partitioning of 18 organic and elemental components measured in and below DYCOMS-II stratocumulus clouds, *J. Geophys. Res.*, 113, D14, doi:10.1029/2007JD009150, 2008.

Heald, C. L., Jacob, D. J., Park, R. J., Russell, L. M., Huebert, B. J., Seinfeld, J. H., Liao, H., and Weber, R. J.: A large organic aerosol source in the free troposphere missing from current

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

- models, *Geophys. Res. Lett.*, 32, 18, doi:10.1029/2005GL023831, 2005.
- Jayne, J. T., Leard, D. C., Ahang, X., Davidovits, P., Smith, K. A., Kolb, C. E., and Worsnop, D. R.: Development of an aerosol mass spectrometer for size and composition analysis of submicron particles, *Aerosol Sci. Tech.*, 33, 49–70, 2000.
- 5 Jimenez, J. L., Jayne, J. T., Shi, Q., Kolb, C. E., Worsnop, D. R., Yourshaw, I., Seinfeld, J. H., Flagan, R. C., Zhang, X., Smith, K. A., Morris, J. W., and Davidovits, P.: Ambient aerosol sampling using the aerodyne aerosol mass spectrometer, *J. Geophys. Res.*, 108, 8245–8258, 2003.
- Johnson, G. R., Ristovski, Z. D., D'Anna, B., and Morawska, L.: Hygroscopic behavior of partially volatilized coastal marine aerosols using the volatilization and humidification tandem differential mobility analyzer technique, *J. Geophys. Res.-Atmos.*, 110, D20203, doi:10.1029/2004JD005657, 2005.
- 10 Kim, B.-G., Miller, M. A., Schwartz, S. E., Liu, Y., and Min, Q.: The role of adiabaticity in the aerosol first indirect effect, *J. Geophys. Res.*, 113, D05210, doi:10.1029/2007JD008961, 2008.
- 15 King, S. M., Rosenoern, T., Shilling, J. E., Chen, Q., and Martin, S. T.: Cloud condensation nucleus activity of secondary organic aerosol particles mixed with sulfate, *Geophys. Res. Lett.*, 34, L24806, doi:10.1029/2007GL030390, 2007.
- Kloster, S., Dentener, F., Feichter, J., Raes, F., van Aardenne, J., Roeckner, E., Lohmann, U., Stier, P., and Swart, R.: Influence of future air pollution mitigation strategies on total aerosol radiative forcing, *Atmos. Chem. Phys.*, 8, 6405–6437, 2008, <http://www.atmos-chem-phys.net/8/6405/2008/>.
- 20 Laaksonen, A., Vesala, T., Kulmala, M., Winkler, P. M., and Wagner, P. E.: Commentary on cloud modelling and the mass accommodation coefficient of water, *Atmos. Chem. Phys.*, 5, 461–464, 2005, <http://www.atmos-chem-phys.net/5/461/2005/>.
- Lance, S., Nenes, A., and Rissman, T.: Chemical and dynamical effects on cloud droplet number: Implications for estimates of the aerosol indirect effect, *J. Geophys. Res.*, 109, D22208, doi:10.1029/2004JD004596, 2004.
- 30 Leaitch, W. R., Strapp, J. W., Isaac, G. A., and Hudson, J. G.: Cloud droplet nucleation and cloud scavenging of aerosol sulphate in polluted atmospheres, *Tellus B*, 38, 328–344, 1986.
- Leaitch, W. R., Banic, C. M., Isaac, G. A., Couture, M. D., Liu, P. S. K., Gultepe, I., Li, S.-M., Kleinman, L. I., Daum, P. H., and MacPherson, J. I.: Physical and chemical observations in

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leaitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leaitch et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

marine stratus during 1993 NARE: factors controlling cloud droplet number concentrations, *J. Geophys. Res.*, 101, 29123–29135, 1996.

Lebsock, M. D., Stephens, G. L., and Kummerow, C.: Multisensor satellite observations of aerosol effects on warm clouds, *J. Geophys. Res.*, 113, D15, doi:10.1029/2008JD009876, 2008.

Liu, P. S. K., Deng, R., Smith, K. A., Williams, L. R., Jayne, J. T., Canagaratna, M. R., Moore, K., Onasch, T. B., Worsnop, D. R., and Deshler, T.: Transmission efficiency of an aerodynamic focusing lens system: comparison of model calculations and laboratory measurements for the aerodyne aerosol mass spectrometer, *Aerosol Sci. Tech.*, 41, 721–733, 2007.

Lohmann, U., Feichter, J., Penner, J., and Leaitch, R.: Indirect effect of sulfate and carbonaceous aerosols-A mechanistic treatment, *J. Geophys. Res.*, 105, 12193–12206, 2000.

Lohmann, U., Leaitch, R., Shantz, N., Broekhuizen, K., and Abbatt, J.: How efficient is cloud droplet formation of organic aerosols?, *Geophys. Res. Lett.*, 31, 5, doi:10.1029/2003GL018999, 2004.

Lohmann, U., Stier, P., Hoose, C., Ferrachat, S., Kloster, S., Roeckner, E., and Zhang, J.: Cloud microphysics and aerosol indirect effects in the global climate model ECHAM5-HAM, *Atmos. Chem. Phys.*, 7, 3425–3446, 2007, <http://www.atmos-chem-phys.net/7/3425/2007/>.

McFiggans, G., Artaxo, P., Baltensperger, U., Coe, H., Fachini, M. C., Feingold, G., Fuzzi, S., Gysel, M., Laaksonen, A., Lohmann, U., Mentel, T. F., Murphy, D. M., O'Dowd, C. D., Snider, J. R., and Weingartner, E.: The effect of physical and chemical aerosol properties on warm cloud droplet activation, *Atmos. Chem. Phys.*, 6, 2593–2649, 2006, <http://www.atmos-chem-phys.net/6/2593/2006/>.

Menon, S., Del Genio, A. D., Koch, D., and Tselioudis, G.: GCM Simulations of the aerosol indirect effect: sensitivity to cloud parameterization and aerosol burden, *J. Atmos. Sci.*, 59, 692–713, 2002.

Menon, S., Brenguier, J.-L., Boucher, O., Davison, P., Del Genio, A. D., Feichter, J., Ghan, S., Guibert, S., Liu, X., Lohmann, U., Pawlowska, H., Penner, J. E., Quaas, J., Roberts, D. L., Schuller, L., and Snider, J.: Evaluating aerosol/cloud/radiation process parameterizations with single-column models and Second Aerosol Characterisation Experiment (ACE-2) cloudy column observations, *J. Geophys. Res.*, 108, D24, doi:10.1029/2003JD003902, 2003.

Ming, Y. and Russell, L. M.: Organic aerosol effects on fog droplet spectra, *J. Geophys. Res.*, 109(D10), 4427, doi:10.1029/2003JD004427, 2004.

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leaitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

- Mozurkewich, M.: Aerosol growth and the condensation coefficient for water: a review, *Aerosol Sci. Tech.*, 5, 223–236, 1986.
- Murphy, D. M., Solomon, S., Portmann, R. W., Rosenlof, K. H., Forster, P. M., and Wong, T.: An observationally based energy balance for the Earth since 1950, *J. Geophys. Res.*, 114, D17107, doi:10.1029/2009JD012105, 2009.
- Novakov, T. and Penner, J. E.: Large contribution of organic aerosols to cloud-condensation-nuclei concentrations, *Nature*, 365, 823–826, 1993.
- Orsini, D. A., Ma, Y., Sullivan, A., Sierau, B., Baumann, K., and Weber, R. J.: Refinements to the particle-into-liquid sampler (PILS) for ground and airborne measurements of water soluble aerosol composition, *Atmos. Environ.*, 37, 1243–1259, 2003.
- Peng, Y., Lohmann, U., and Leaitch, W. R.: Importance of vertical velocity variations in the cloud droplet nucleation process of marine stratus clouds, *J. Geophys. Res.*, 110, D21, doi:10.1029/2004JD004922, 2005.
- Penner, J. E., Andreae, M., Annegarn, H., Barrie, L., Feichter, J., Hegg, D., Jayaraman, A., Leaitch, R., Murphy, D., Nganga, J., and Pitari, G.: Aerosols, their direct and indirect effects, in: *Climate Change 2001: The Physical Science Basis, Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change*, edited by: Houghton, J. T., Ding, Y., Griggs, D. J., Noguer, M., van der Linden, P. J., Dai, X., Maskell, K., and Johnson, C. A., Cambridge Univ. Press, New York, 129–234, 2001.
- Penner, J. E., Quaas, J., Storelvmo, T., Takemura, T., Boucher, O., Guo, H., Kirkevåg, A., Kristjánsson, J. E., and Seland, Ø.: Model intercomparison of indirect aerosol effects, *Atmos. Chem. Phys.*, 6, 3391–3405, 2006, <http://www.atmos-chem-phys.net/6/3391/2006/>.
- Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, *Atmos. Chem. Phys.*, 7, 1961–1971, 2007, <http://www.atmos-chem-phys.net/7/1961/2007/>.
- Petters, M. D., Prenni, A. J., Kreidenweis, S. M., DeMott, P. J., Matsunaga, A., Lim, Y. B., and Ziemann, P. J.: Chemical aging and the hydrophobic-to-hydrophilic conversion of carbonaceous aerosol, *Geophys. Res. Lett.*, 33, L24806, doi:10.1029/2006GL027249, 2006.
- Phinney, L., Leaitch, W. R., Lohmann, U., Boudries, H., Toom-Saunry, D., Sharma, S., and Shantz, N.: Characterization of aerosol over the sub-arctic NE Pacific during SERIES, *Deep-Sea Res. Pt. II*, 53, 2410–2433, 2006.
- Phinney, L., Leaitch, W. R., Lohmann, U., Shantz, N. C., and Worsnop, D. R.: Contributions

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

from DMS and ship emissions to CCN observed over the summertime North Pacific, Atmos. Chem. Phys. Discuss., 9, 309–361, 2009,

<http://www.atmos-chem-phys-discuss.net/9/309/2009/>.

Pierce, J. R., Chen, K., and Adams, P. J.: Contribution of primary carbonaceous aerosol to cloud condensation nuclei: processes and uncertainties evaluated with a global aerosol microphysics model, Atmos. Chem. Phys., 7, 5447–5466, 2007, <http://www.atmos-chem-phys.net/7/5447/2007/>.

Prenni, A. J., Petters, M. D., DeMott, P. J., Kreidenweis, S. M., Ziemann, P. J., Matsunaga, A., and Lim, Y. B.: Cloud droplet activation of secondary organic aerosol, J. Geophys. Res., 112, D10223, doi:10.1029/2006JD007963, 2007.

Pringle, K. J., Carslaw, K. S., Spracklen, D. V., Mann, G. M., and Chipperfield, M. P.: The relationship between aerosol and cloud drop number concentrations in a global aerosol microphysics model, Atmos. Chem. Phys., 9, 4131–4144, 2009, <http://www.atmos-chem-phys.net/9/4131/2009/>.

Quaas, J., Ming, Y., Menon, S., Takemura, T., Wang, M., Penner, J. E., Gettelman, A., Lohmann, U., Bellouin, N., Boucher, O., Sayer, A. M., Thomas, G. E., McComiskey, A., Feingold, G., Hoose, C., Kristjánsson, J. E., Liu, X., Balkanski, Y., Donner, L. J., Ginoux, P. A., Stier, P., Grandey, B., Feichter, J., Sednev, I., Bauer, S. E., Koch, D., Grainger, R. G., Kirkevåg, A., Iversen, T., Seland, Ø., Easter, R., Ghan, S. J., Rasch, P. J., Morrison, H., Lamarque, J.-F., Iacono, M. J., Kinne, S., and Schulz, M.: Aerosol indirect effects - general circulation model intercomparison and evaluation with satellite data, Atmos. Chem. Phys., 9, 8697–8717, 2009, <http://www.atmos-chem-phys.net/9/8697/2009/>.

Ramanathan, V.: The greenhouse theory of climate change: a test by an inadvertent global experiment, Science, 240, 293–299, 1988.

Rissman, T. A., Nenes, A., and Seinfeld, J. H.: Chemical amplification (or dampening) of the Twomey effect: conditions derived from droplet activation theory, J. Atmos. Sci., 61, 919–930, 2004.

Roberts, G. C., Artaxo, P., Zhou, J., Swietlicki, E., and Andreae, M. O.: Sensitivity of CCN spectra on chemical and physical properties of aerosol: a case study from the Amazon Basin, J. Geophys. Res., 107, 8070, doi:10.1029/2001JD000583, 2002.

Rolph, G. D.: Real-time Environmental Applications and Display sYstem (READY) website, <http://www.arl.noaa.gov/ready.php>, last access: 18 January 2010, NOAA Air Resources

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leitch et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Laboratory, Silver Spring, MD, 2003.

Ruehl, C. R., Chuang, P. Y., and Nenes, A.: How quickly do cloud droplets form on atmospheric particles?, *Atmos. Chem. Phys.*, 8, 1043–1055, 2008, <http://www.atmos-chem-phys.net/8/1043/2008/>.

5 Ruehl, C. R., Chuang, P. Y., and Nenes, A.: Distinct CCN activation kinetics above the marine boundary layer along the California coast, *Geophys. Res. Lett.*, 36, L15814, doi:10.1029/2009GL038839, 2009.

Rupakheti, M., Leitch, W. R., Lohmann, U., Hayden, K., Brickell, P., Lu, G., Li, S.-M., Toom-Saunty, D., Bottenheim, J. W., Brook, J. R., Vet, R., Jayne, J. T., and Worsnop, D. R.: An
10 intensive study of the size and composition of submicron atmospheric aerosols at a rural site in Ontario, Canada, *Aerosol Sci. Tech.*, 39, 722–736, 2005.

Russell, L. M., Seinfeld, J. H., Flagan, R. C., Ferek, R. J., Hegg, D. A., Hobbs, P. V., Wobrock, W., Flossmann, A., O'Dowd, C., Nielsen, K. E., and Durke, P. A.: Aerosol dynamics in ship tracks, *J. Geophys. Res.*, 104, 31077–31095, 1999.

15 Shantz, N. C., Leitch, W. R., and Caffrey, P.: Effect of organics of low solubility on the growth rate of cloud droplets, *J. Geophys. Res.*, 108, 4168–4177, 2003.

Shantz, N. C., Chang, R. Y.-W., Slowik, J. G., Vlasenko, A., Abbatt, J. P. D., and Leitch, W. R.: Slower CCN growth kinetics of anthropogenic aerosol compared to biogenic aerosol observed at a rural site, *Atmos. Chem. Phys.*, 10, 299–312, 2010,
20 <http://www.atmos-chem-phys.net/10/299/2010/>.

Straub, D. J., Lee, T., and Collett Jr., J. L.: Chemical composition of marine stratocumulus clouds over the Eastern Pacific Ocean, *J. Geophys. Res.*, 112, D4, doi:10.1029/2006JD007439, 2007.

25 Strapp, J. W., Leitch, W. R., and Liu, P. S. K.: Hydrated and dried aerosol size distribution measurements from the particle measuring systems FSSP-300 probe and the de-iced PCASP-100X probe, *J. Atmos. Ocean. Tech.*, 9, 548–555, 1992.

Targino, A. C., Noone, K. J., Drewnick, F., Schneider, J., Krejci, R., Olivares, G., Hings, S., and Borrmann, S.: Microphysical and chemical characteristics of cloud droplet residuals and interstitial particles in continental stratocumulus clouds, *Atmos. Res.*, 86, 225–240, 2007.

30 Twomey, S.: The influence of pollution on the shortwave albedo of clouds, *J. Atmos. Sci.*, 34, 1149–1152, 1977.

Wagner, P.: Aerosol growth by condensation, in: *Aerosol Microphysics II: Chemical Physics of Microparticles*, edited by: Marlow, W. H., Springer-Verlag, Berlin, 129–178, 1982.

Wang, J., Lee, Y.-N., Daum, P. H., Jayne, J., and Alexander, M. L.: Effects of aerosol organics on cloud condensation nucleus (CCN) concentration and first indirect aerosol effect, *Atmos. Chem. Phys.*, 8, 6325–6339, 2008, <http://www.atmos-chem-phys.net/8/6325/2008/>.

5 Wex, H., Petters, M. D., Carrico, C. M., Hallbauer, E., Massling, A., McMeeking, G. R., Poulain, L., Wu, Z., Kreidenweis, S. M., and Stratmann, F.: Towards closing the gap between hygroscopic growth and activation for secondary organic aerosol: Part 1 - Evidence from measurements, *Atmos. Chem. Phys.*, 9, 3987–3997, 2009, <http://www.atmos-chem-phys.net/9/3987/2009/>.

10 Zhang, Q., Jimenez, J. L., Canagaratna, M. R., Allan, J. D., Coe, H., Ulbrich, I., Alfarra, M. R., Takami, A., Middlebrook, A. M., Sun, Y. L., Dzepina, K., Dunlea, E., Docherty, K., DeCarlo, P. F., Salcedo, D., Onasch, T., Jayne, T. J., Miyoshi, T., Shimo, A., Hatakeyama, S., Takegawa, N., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Williams, P., Bower, K., Bahreini, R., Cottrell, L., Griffin, R. J., Rautiainen, J.,
15 Sun, Y. J., Zhang, Y. M., and Worsnop, D. R.: Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere midlatitudes, *Geophys. Res. Lett.*, 34, 13, doi:10.1029/2007GL029979, 2007.

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Cloud albedo increase from carbonaceous aerosol

W. R. Leaitch et al.

Table 1. Instrumentation list.

Measurement	Instrument
Aerosol particle number and size	TSI SMPS (10–500 nm) – inboard – 2.5 min ave. PMS PCASP100X (0.14–3 μm) – outboard under wing – 1 s ave. TSI APS 3321 (0.54–20 μm) – inboard – 1 min ave. PMS FSSP-300 (0.5–20 μm) – outboard under wing – 1 s ave.
Cloud droplet number and size	2 PMS FSSP-100 (2–45 μm) – outboard under wing – 1 s ave.
Cloud liquid water content	PMS King Probe – outboard under wing – 1 s ave.
Cloud light Extinction	Gerber Cloud Integrating Nephelometer (CIN) – outboard under wing – 1 s ave.
Aerosol particle chemistry	Aerodyne Mass Spectrometer (AMS) – inboard – 5 min ave. Particle-In-Liquid sampler (PILS) with Dionex ICS 2000 Ion Chromatographs – inboard – 10 min ave.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Cloud albedo increase from carbonaceous aerosol

W. R. Leaitch et al.

Table 2. Simple statistics of one second observations of the APNC_{140} (units of cm^{-3}) under cloud base for flights 1 and 2. Thirteen minutes was spent directly under cloud base during flight 1 and 6 min during flight 2. These periods are also those used to define the chemistry of the cloud base aerosol as given in Table 3.

Statistic	Flight 1 – 13 Oct 2003 19:07–19:21 UT	Flight 2 – 14 Oct 2003 12:36–12:42 UT
Mean	166	393
Median	166	393
Std Dev	13.8	32
> 99%	166±42	393±96
Range	124–208	297–489

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Cloud albedo increase from carbonaceous aerosol

W. R. Leaitch et al.

Table 3. Aerosol chemistry at cloud base. The mass concentrations of the chemical compounds measured at cloud base with the Q-AMS and the Particle In Liquid Sampler (PILS), for the times given in Table 2. The PILS average from flight 2 is shown as a range of two samples that cover segments of the five-minute Q-AMS average. The Q-AMS non-refractory mass concentrations are slightly different than in the profiles of Fig. 1c because they are for level flight only. The m/z 43, 44 and 57 values are mass concentrations of the corresponding non-refractory carbonaceous ion fragments measured with the Q-AMS; higher m/z 44 indicates organic mass (OM) that is more oxygenated.

Flt.	Sulphate ($\mu\text{g}/\text{m}^3$)	OM ($\mu\text{g}/\text{m}^3$)	Nitrate ($\mu\text{g}/\text{m}^3$)	Ammonium ($\mu\text{g}/\text{m}^3$)	Sodium ($\mu\text{g}/\text{m}^3$)	m/z 43 ($\mu\text{g}/\text{m}^3$)	m/z 44 ($\mu\text{g}/\text{m}^3$)	m/z 57 ($\mu\text{g}/\text{m}^3$)
1	2.4 (2.1)	0.86	0.08 (0.0)	0.0 (0.18)	0.0 (0.02)	0.1	0.0	0.0
2	2.3 (1.78–2.65)	2.6	0.2 (0.25–0.48)	0.0 (0.32–0.40)	0.02 (0.40–0.72)	0.19	0.46	0.03

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Cloud albedo increase from carbonaceous aerosol

W. R. Leaitch et al.

Table 4. Summary of aerosol physical size distribution measurements at cloud base.

Mode	Flight 1 Size range GD (nm)	Number Cn. (cm^{-3})	Mass estimate ($\rho=1.7 \text{ g cm}^{-3}$) ($\mu\text{g m}^{-3}$)	Flight 2 Size range GD (nm)	Number Cn. (cm^{-3})	Mass estimate ($\rho=1.5 \text{ g cm}^{-3}$) ($\mu\text{g m}^{-3}$)
1	10–115	408	0.09	10–115	537	0.17
2	115–500	246	2.6	115–500	498	6.4
3	500–900	13.9	2.4	500–900	20	3.8
4	900–18000	1.0	5.4	900–18000	4.2	57.5
All		668	10.5		1059	67.9

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)

Table 5. Model initial conditions.

	Geo. Mean Dia.	Geo. Std. Dev.	Num. Cn.	Composition
Flight 1	(nm)		cm ⁻³	
Mode 1	56	1.48	395	H ₂ SO ₄
Mode 2	200	1.40	250	25%Org and 75%H ₂ SO ₄
Mode 3	560	1.22	14	25%Org and 75%H ₂ SO ₄
Mode 4	1200	1.45	1.8	NaCl
Total			661	
Flight 2				
Mode 1	30	1.40	79	H ₂ SO ₄
Mode 2	74	1.32	461	H ₂ SO ₄ or 70% Organic and 30% H ₂ SO ₄
Mode 3	200	1.47	503	70% Organic and 30% H ₂ SO ₄
Mode 4	540	1.30	17.7	25% Organic and 75% H ₂ SO ₄
Mode 5	3000	1.60	1.7	NaCl
Total			1062	

- Pressure 960 mb, both flights
- Temperature, 17.2 °C (Flt 2), 12.5 °C (Flt 1)
- Starting RH, 98%
- Accommodation coefficient for water, 1.0; sensitivity to 0.06.
- Organic osmotic coefficient, 1.0
- Organic dissociation factor, 1.0
- Organic MW, 150 g mole⁻¹
- Organic surface tension adipic acid; sensitivity to pure water tested.

Cloud albedo increase from carbonaceous aerosol

W. R. Leaitch et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Cloud albedo increase from carbonaceous aerosol

W. R. Leitch et al.

Table 6. Simulated Cloud Droplet Number Concentrations (CDNC, cm^{-3}). Values are for smaller particles ($<100\text{ nm}$) represented as H_2SO_4 ; values in parentheses are for smaller particles represented as 70% organic and 30% H_2SO_4 . The coloured numbers are referred to in the text and Fig. 7 (red: 7a; green: 7b; yellow: 7c).

Updraft (cm/s)	Flight 1 – organic solubility (g l^{-1})			All H_2SO_4	Flight 2 – organic solubility (g l^{-1})			All H_2SO_4
	0.01	5	200		0.01	5	200	
	Internal mixture				Internal mixture			
14	225	225	225	225	215	215	215	256
20	253	253	253	249				
50	330	330	330	330	538	(491)–579	579	533
100	401	401	401	401	771	(555)–771	771	681
					External mixture			
14						148	206	
50						370	560	

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Cloud albedo increase from carbonaceous aerosol

W. R. Leitch et al.

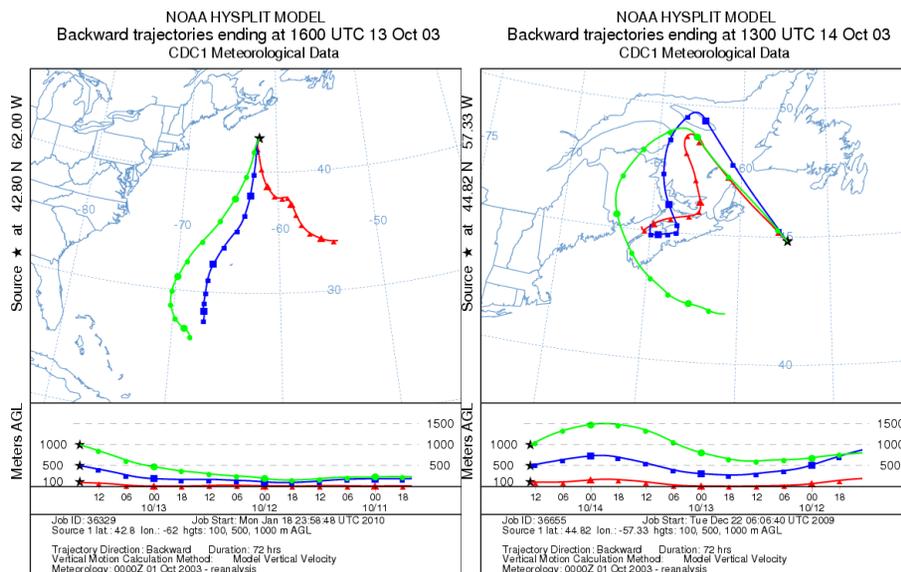


Fig. 1. Meteorological 3-day back trajectories for end points at the approximate time and location of vertical profiles through the clouds (Draxler and Rolph, 2003; Rolph, 2003).

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leitch et al.

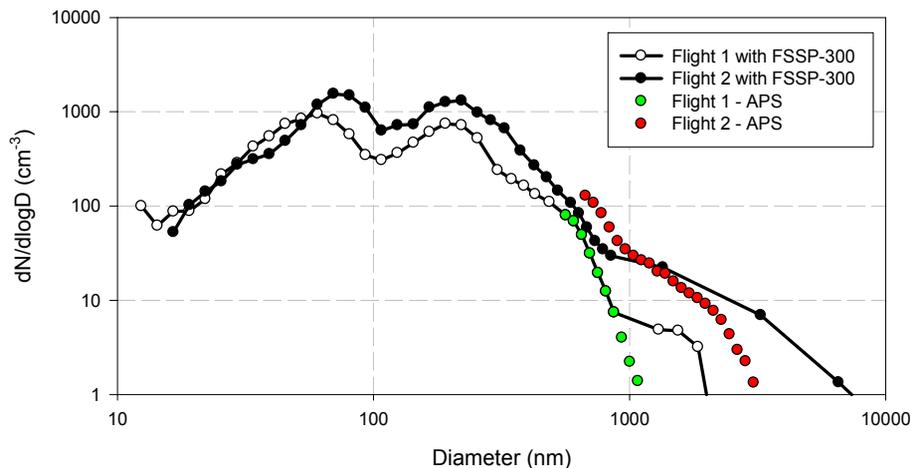


Fig. 2. Size distributions combining data from the Scanning Mobility Particle System (SMPS; 10–400 nm) and the outboard Forward Scattering Spectrometer Probe for coarse aerosol particles (FSSP-300; 0.3–20 μm) as well as data from the inboard Aerodynamic Particle Sizer distributions (APS; sizes converted to a geometric diameter assuming a density of 1.5 g cm^{-3}). Divergence of the FSSP-300 and APS above 1 μm is believed to be due to losses at the aircraft intake. In constructing the size distribution for the model calculations (Fig. 2a), we used the SMPS, the APS up to 0.9 μm and the FSSP-300 above 0.9 μm .

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Cloud albedo
increase from
carbonaceous
aerosol

W. R. Leitch et al.

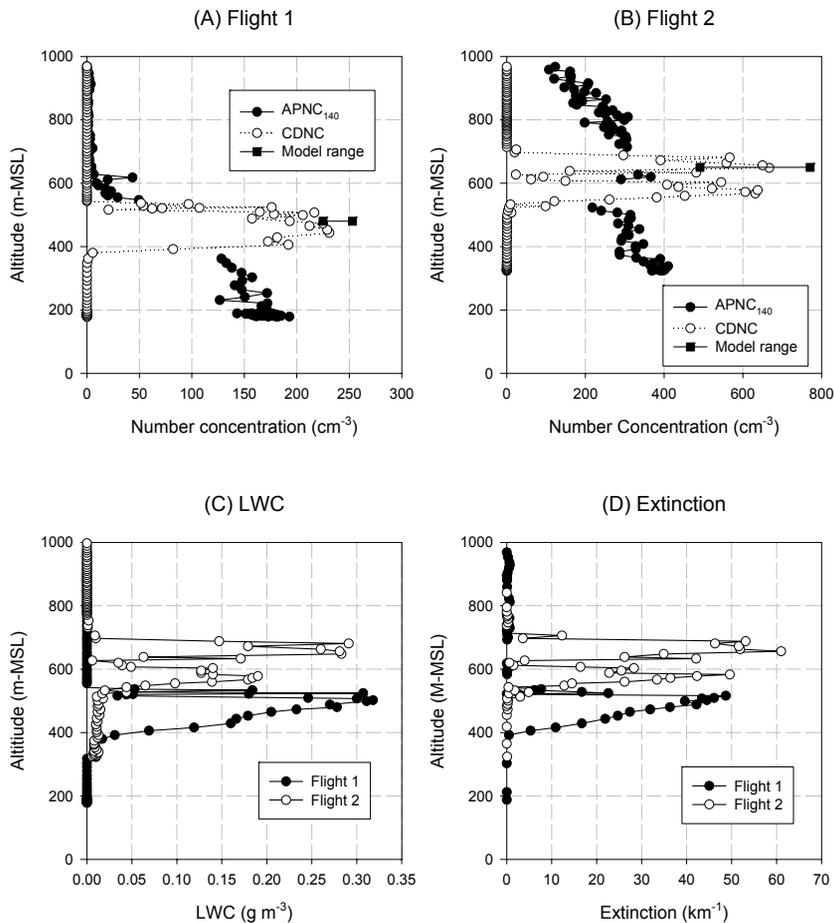


Fig. 3. (A) Vertical profiles of APNC_{>140} and CDNC through cloud during flight 1; (B) as (A) but for flight 2; (C) profiles of LWC for flights 1 and 2; (D) profiles of cloud light extinction for flights 1 and 2.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leaitch et al.

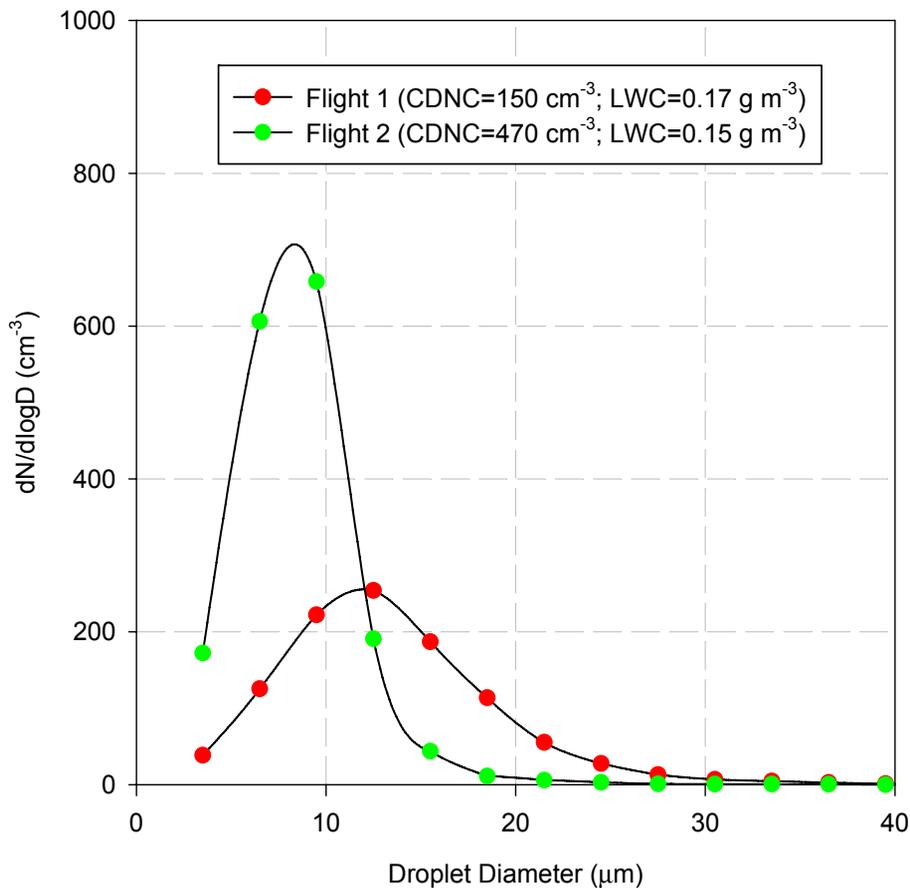


Fig. 4. Droplet size distributions averaged through the profile from each flight from one of the FSSP-100 probes.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leitch et al.

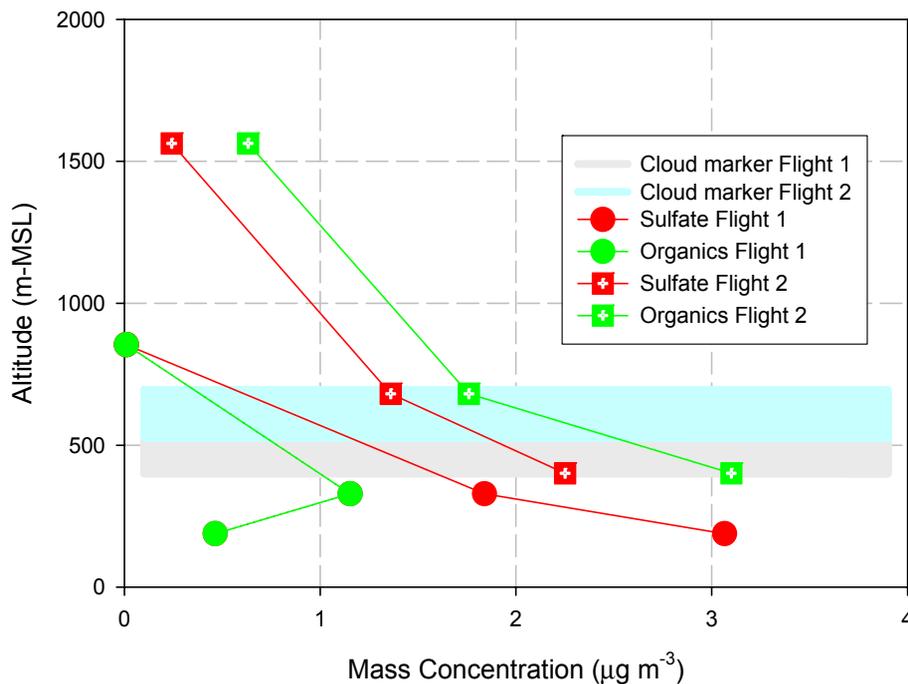


Fig. 5. Vertical profiles of sulphate and total organic mass concentrations from the Q-AMS. Each data point is a 5 min average. The shading indicates the approximate region of cloud.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leitch et al.

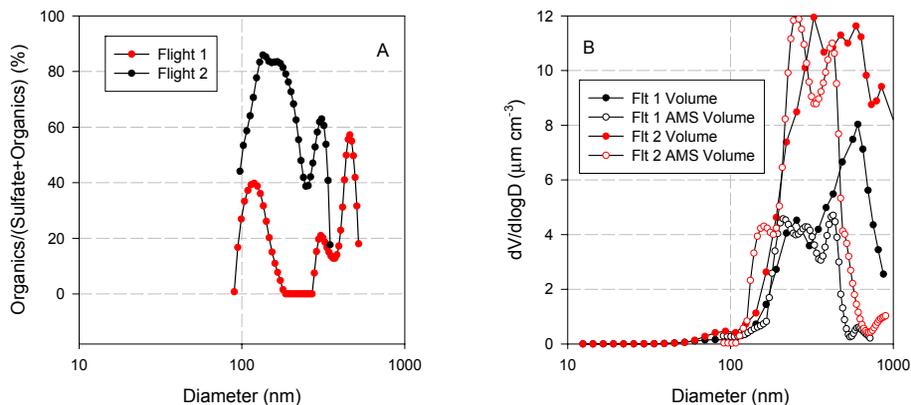


Fig. 6. (A) Below-cloud ratios of sulphate to sulphate plus organics from the Quadrupole Aerosol Mass Spectrometer (Q-AMS) as a function of size; (B) below-cloud volume distributions from Q-AMS and physical measurements.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

**Cloud albedo
increase from
carbonaceous
aerosol**

W. R. Leaitch et al.

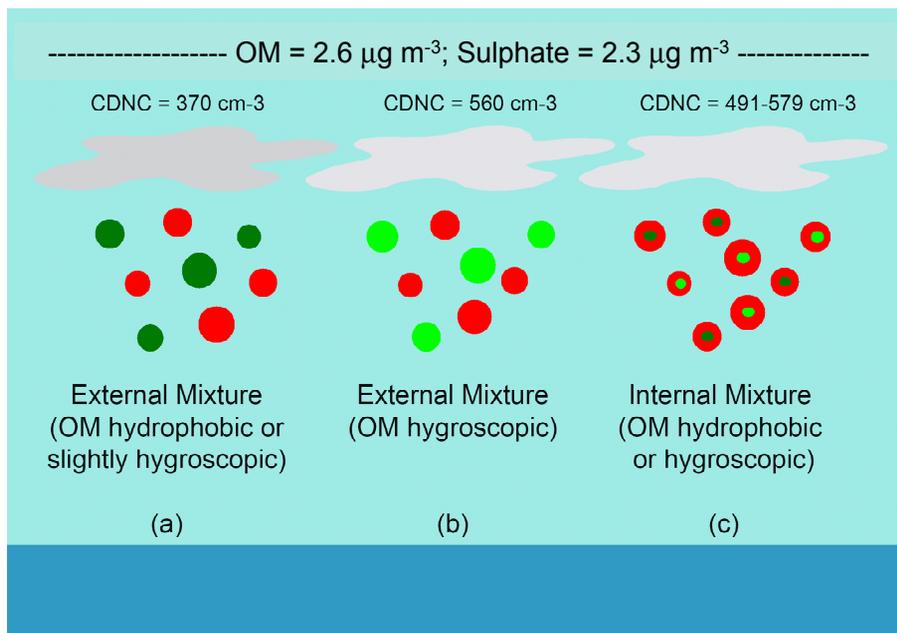


Fig. 7. Scenarios of aerosol component mixing and modelled cloud droplet number concentrations for flight 2. **(a)** The carbonaceous (green) and sulphate (red) components are externally mixed with no water uptake by the carbonaceous particles; **(b)** externally mixed and the carbonaceous particles strongly influence the water uptake; **(c)** internally mixed carbonaceous and sulphate components with no effect of the carbonaceous components on water uptake.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)