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20714

ACPD

11, 20713–20755, 2011

**Diurnal variations of
condensation particle
concentrations at
Neumayer, Antarctica**

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Abstract

Continuous condensation particle (CP) observations were conducted from 1984 through 2009 at Neumayer Station under stringent contamination control. During this period, the CP concentration (median 258 cm^{-3}) showed no significant long term trend but exhibited a pronounced seasonality characterized by a stepwise increase starting in September and reaching its annual maximum of around 10^3 cm^{-3} in March. Minimum values below 10^2 cm^{-3} were observed during June/July. Dedicated time series analyses in the time and frequency domain revealed no significant correlations between inter-annual CP concentration variations and atmospheric circulation indices like Southern Annular Mode (SAM) or Southern Ocean Index (SOI). The impact of the Pinatubo volcanic eruption and strong El Niño events did not affect CP concentrations. From thermodenuder experiments we deduced that the portion of volatile (at 125°C) and semi-volatile (at 250°C) particles which could be both associated with biogenic sulfur aerosol, was maximum during austral summer, while during winter non-volatile sea salt particles dominated. During September through April we could frequently detect nucleation events which occurred preferentially in the afternoon. Over the year, roughly 20 % of the particles could be assigned to the nucleation mode between 3 nm and 7 nm particle diameter.

1 Introduction

Greenhouse gases and aerosols are atmospheric trace compounds determining decisively the radiation balance of the Earth's atmosphere (Forster et al., 2007), hence thorough knowledge about atmospheric cycling are pivotal to predict their impact on global climate (Haywood and Boucher, 2000; Ramanathan et al., 2001; Hatzianastasiou et al., 2004; Forster et al., 2007). Unlike most of the important greenhouse gases (CO_2 , CH_4 , N_2O), aerosols stand out due to short atmospheric lifetimes typically in the range of a few days, entailing a striking temporal and spatial concentration and

ACPD

11, 20713–20755, 2011

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Diurnal variations of
condensation particle
concentrations at
Neumayer, Antarctica**

R. Weller et al.

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

ultimately radiation forcing variability (Raes et al., 2000; Ramanathan et al., 2001). In this regard the nearly completely ice covered Antarctic continent is a particular interesting case: Primary aerosol sources like sea spray, mineral dust, or soot, as well as aerosol sources due to gas to particle conversion (secondary aerosol) are virtually absent on this continent, apart from some insular rocky regions (on the Antarctic peninsula, in the coastal dry valleys and on high mountain ranges) and volcanic activity of Mt. Erebus. Hence, as pointed out in a noted review about Antarctic aerosol by Shaw (1988), it is remarkable that sub-micrometer particles are present throughout the Antarctic air, in spite of distant source regions and efficient wet depositional removal within the persistent circumpolar low pressure system. In fact, the Southern Ocean remains as being by far the dominant source to the Antarctic aerosol body, making atmospheric sea salt and biogenic sulfur to the major aerosol components (Wagenbach, 1996; Minikin et al., 1998; Preunkert et al., 2007). Nowadays, minor anthropogenic emissions arising from fossil fuel combustion during research and tourism activities might be considered as well (Shirsat and Graf, 2009).

Particle concentrations are usually measured by condensation particle counters (CPC), which optically detect and count each particle above a given particle size (typically 10 nm diameter) after condensation growth in super-saturated butanol vapour (Agarwal and Sem, 1980). Hence, condensation particle (CP) concentration measured in this way represent total particle numbers concentrations above the given cut-off size. So far, continuous long-term studies on particle or CP concentrations in Antarctica were restricted to Amundsen-Scott Base, South Pole (Samson et al., 1990; Bodhaine, 1996) and coastal Georg von Neumayer (70°37' S, 8°22' W; Jaenicke et al., 1992), while from Syowa (69°00' S, 39°35' E) at least year-round CP measurements for the period 1976–1978 are available (Ito, 1985). There is a lack of dedicated investigations concerning the climatology, i.e. long-term trend and inter-annual variability of particle concentrations and their links to climate factors, e.g. the development of southern atmospheric circulation pattern like the Southern Annular Mode (SAM), Southern Ocean Index (SOI) and El Niño teleconnection.

available from Neumayer Station. The CP record is mainly backed-up by concurrent aerosol sampling dedicated for determination of its ionic composition. The results from the latter investigations have been discussed in detail (Weller et al., 2011). We will first focus on long-term changes of CP concentrations and evaluate the Neumayer observations in the time and frequency (spectral) domain along with corresponding time series of sea ice extent, Southern Annular Mode, and Southern Ocean Index. In a second step seasonality and diurnal cycle of CP concentrations will be investigated and tied to the ionic composition of the aerosol. Finally we will examine observed nucleation events, discuss their seasonality, diurnal cycle, as well as their relevance concerning the CP budget at Neumayer.

2 Experimental techniques and data evaluation methods

2.1 Site description and instrumentation

Measurements were conducted at the Air Chemistry Observatory, Neumayer Station (NM, 70°39' S, 8°15' W, http://www.awi.de/en/go/air_chemistry_observatory) which participates in the Global Atmosphere Watch (GAW) programme. To ensure contamination free air sampling, the Air Chemistry Observatory is situated in the clean air district approximately 1.5 km south of NM. Due to the fact that northerly wind directions are very rare, contamination from the base can be excluded for most of the time. Additionally, the power supply (20 kW) is provided by cable from the main station. For a detailed description of the measuring site, meteorological conditions, contamination free sampling, and analysis of the aerosol samples we refer to Wagenbach et al. (1988), König-Langlo et al. (1998) and Weller et al. (2008). A ventilated electro-polished stainless steel inlet stack (total height about 8 m above the snow surface) with a 50 % aerodynamic cut-off diameter around 7–10 μm at wind velocities between 4–10 m s^{-1} (determined with an aerodynamic particle sizer TSI, type APS 3321) supplied the experiments with ambient air. Continuous test measurements with two condensation particle counters of identical

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



technical specification were performed through October 2002, one plugged to the inlet stack the other sampling directly ambient air from outside. No bias or offset between the measured signals (5 min resolution) could be detected and the values agreed within $\pm 5\%$, except few sporadic spikes.

Concerning the question how representative our CP record might be, the characteristics of the boundary layer at this site needs to be considered. As typical for Antarctica, surface inversions are common which could extend up to about 2 km during winter while from November to February inversions are rare and confined to heights less than 1 km (König-Langlo et al., 1998). Frequent stormy weather conditions usually destroy the stratification of the lower troposphere. Due to the stability of the free atmosphere above the boundary layer the typical height of the stable boundary layer (SBL), defined as the lowest altitude above ground where turbulent mixing ceased, ranged between 10 m and 50 m (Handorf, 1996).

Condensation particle measurements started at NM in January 1982. Table 1 gives an overview of the employed particle counters. Up to December 1989 a condensation particle counter (Thermo Systems Inc., TSI, type CPC 3020; nominal 50 % cut-off particle diameter $D_{p(50\%)}$ of 10 nm) provided particle number concentrations typically with hourly resolution (Jaenicke et al., 1992), henceforward in 150 s resolution. From 1990 ongoing, a CPC 3760, CPC 3022, or CPC 3022A (all from TSI) were optionally in use, characterized by a lower $D_{p(50\%)}$ of 7 nm. In order to assess the contribution of particles within the nucleation mode between 3 nm and 7 nm particle diameter (D_p), we employed an ultrafine particle counter (Stolzenburg and McMurry, 1991) with a $D_{p(50\%)}$ of 3 nm (TSI Model CPC 3025A). This instrument was simultaneously run in 150 s resolution with the CPC 3022/3022A from February 1996 ongoing, except for the years 1999, 2009 and between April 2006 and December 2007 (see Table 1).

Particle concentration measurements were complemented by particle volatility studies starting from January 2003 through December 2007. In this case a thermodenuder (TD) was deployed according to Wehner et al. (2002), comprising a controlled heated inlet line coupled to one the CPC 3022 while the other one was connected to the

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

unheated inlet. The TD temperatures were set on a daily basis alternating to 125 °C and 250 °C to characterize particle volatility. This method allowed a classification of the aerosol in volatile, semi-volatile, and non-volatile (refractory) compounds and provided a crude but fast characterization of its chemical habit: sulfuric acid aerosol is volatile above 125 °C and ammonium sulfate above 250 °C (semi-volatile), while soot (black carbon), mineral dust, and sea salt remain non-volatile above 250 °C (Burtscher et al., 2001). For about 15 min every day a diffusion screen separator (Particle Size Selector Model 376060, TSI; further information see Feldpausch et al., 2006) was switched in front of the heated inlet to assess the volatility of particles below a defined diameter (in our case $D_p < 40$ nm). Periodic comparative measurements with both CPC 3022 instruments simultaneously connected with an unheated inlet showed a typical scatter between both signals within 5 % to 10 % but did not indicate any significant bias.

Particle measurements were accompanied by bulk aerosol sampling for chemical analyses. We refer here to Weller et al. (2011) who described the climatology of the ionic composition of the aerosol at Neumayer derived from continuous high volume sampling since 1983 with a temporal resolution of 7–14 days. From October 2003 through December 2010 (except for the period between February 2007 and February 2008), aerosols were also sampled for 24-h time periods using a 2-stage PFA (polyfluoroalkoxy-copolymer) filter holder system, including a teflon and a nylon (Nylasorb) filter in series (all 1 μ m pore size). Generally, samples were analyzed by ion chromatography for methane sulfonate (CH_3SO_3^- , MS), Cl^- , Br^- , NO_3^- , SO_4^{2-} , Na^+ , NH_4^+ , K^+ , Mg^{2+} , and Ca^{2+} . For details concerning aerosol filter handling and analyses we refer to Piel et al. (2006) and Weller et al. (2011).

Throughout this work the time is given in UTC, corresponding to a shift of +32 min relative to the solar time. Condensation particle and ionic concentrations are given in particles cm^{-3} and ng m^{-3} , respectively, referring to SPT (273.16° K and 1013 hPa).

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



2.2 Data evaluation methods

First, all data points recorded under potential contamination conditions, indicated by wind directions within a 330° – 30° sector and/or wind velocities below 2.0 m s^{-1} were removed. From 1996 ongoing, black carbon concentrations were continuously monitored by an aethalometer (Magee Scientific, type AE10, since March 2006 in addition by a Multi Angle Absorption Photometer, MAAP model 5012, Thermo Electron Corp.), providing a supplemental criterion for local pollution. In such cases BC concentrations levels exceeded 100 ng m^{-3} by far. Note that the actual data loss due to potential contamination was less than 2 %.

Analyses of the CP concentration time series in terms of inter-annual variability, long-term trend behaviour, and correlations with atmospheric circulation indices were based on evaluation methods previously applied to our ionic composition records of NM aerosol samples (Weller et al., 2011). The correlations with circulation indices includes Southern Annular Mode (SAM, retrieved from <http://jisao.washington.edu/data/aao/slp/>) and Southern Ocean Index, (SOI, retrieved from <http://www.cdc.noaa.gov/data/climateindices/>) as well as sea ice extent (SIE, retrieved from National Snow and Ice Data Center, NSIDC, <http://nsidc.org/>). Thus, we refer to Weller et al. (2011) and accompanying Supporting Information therein for a more detailed presentation of the application of the used statistical methods. The CP time series was analyzed in the time and frequency domain and trend estimates were based on the nonparametric rank-order Mann-Kendall test (Hirsch et al., 1982). Due the fact that CP concentration variability is best described by a lognormal distribution, all time series analyses were additionally performed using the logarithms of the original data set. We restrict our evaluation to variability on the time scale larger than one year and thus averaged the original signal over non-overlapping “bins” of equal time intervals τ of one month. Furthermore, normalized anomalies $X(\tau)$ for each running time step τ within the observation period were calculated according to Eq. (1):

$$X(\tau) = [x(ij) - \langle x_i \rangle] / \sigma_i \quad (1)$$

20721

ACPD

11, 20713–20755, 2011

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



where $x(ij)$ is the mean of the measured parameter X for the month i for a given year j , $\langle x_i \rangle$ and σ_i are the mean and standard deviation (or its respecting logarithm) over all years for this month. We used Monte Carlo Singular Spectrum Analysis (MC-SSA; e.g. Ghil et al., 2002) to generate data adaptive filtered representations of the CP record which was further analysed by the Multitaper Method (MTM; Percival and Walden, 1993). For all employed statistical methods, we generally refer to $p = 0.05$ significance level if not noted otherwise.

3 Data presentation

Figure 1 summarizes the results of 26 years continuous CP measurements at NM, starting in 1984. There are several points to note: (i) we present daily means of the original data and omitted results before 1984 because for this period data loss by instrumental and data acquisition malfunctions appeared unacceptable for our purpose. (ii) Particularly before 1989 there were still some severe data gaps and the CPC 3020 in operation had a higher cut-off diameter compared to the CP counters employed from then on (Table 1). (iii) From 24 February 2009 through 4 February 2010 only an UCPC 3025A was in operation with a lower particle diameter cut-off. Thus these data are biased to higher values, especially during late summer when ultrafine particle concentrations are highest (see below).

Condensation particle concentrations are well represented by the lognormal probability density function (pdf) with a mean $\mu_{\ln} = 5.455$ and standard deviation $\sigma_{\ln} = 0.925$. To visualize the inter-annual variability, the CP time series is also presented in Fig. 2 along with the corresponding MS record from NM in monthly mean bins. In addition distinct climate related events like strong El Niño periods in 1997 and 2006 or the Pinatubo eruption in 1991 are marked with bars. Here we consider only the period from 1989 through 2009, which is furthermore used for time series analyses. Data gaps in the period before 1989 are unbearable for this purpose. Still missing data (November and

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



December 1989; August–December 1990) were patched with long-term means of the corresponding months.

Apart from distinct inter-annual variations, the CP time series is characterized by a strong seasonality. Figure 3 shows the mean seasonality of CP concentrations for the years 1991–2009, based on daily mean values together with respective data of non-sea salt sulfate (nss-SO_4^{2-}) as well as MS, based on weekly aerosol samples within the same period. Obviously the seasonal CP concentration maximum in March shows up in three stages, starting typically in September and rising again in December and February. Note that this characteristic behaviour is not an artefact of the averaging process, but could be observed more or less pronounced each year (also discernible in Figs. 1 and 2). The relative seasonal amplitude, i.e. $0.5 \times (\text{CP}_{\text{max}} - \text{CP}_{\text{min}}) / \text{CP}_{\text{median}}$ is 1.35 (mean absolute amplitude around 350 cm^{-3}).

In order to estimate the effect of vertical mixing within the planetary boundary layer (PBL) on CP concentrations, we simply used the temperature difference between 2 m and 10 m height, i.e. $\Delta T = T_{10\text{m}} - T_{2\text{m}}$ because turbulence measurements are not available. We sorted CP concentrations into these two cases: (i) well mixed PBL with $\Delta T \leq 0$ and, (ii) suppressed vertical mixing with $\Delta T \geq 2$. Like CP concentrations, the ΔT vertical mixing parameter shows a distinct seasonality with higher values (stronger inversion strength) during winter when CP levels are lowest. Hence we differentiate between summer (December through March) and winter (May through October) to avoid a seasonal bias and used the logarithms of the hourly mean CP concentrations for this analysis. During summer, mean CP concentrations are significantly higher during strong inversions (516 cm^{-3} for $\Delta T \geq 2$ compared to 434 cm^{-3} for $\Delta T \leq 0$; $p < 10^{-40}$, $n = 4000$), while during winter the result is vice versa (110 cm^{-3} for $\Delta T \geq 2$ compared to 170 cm^{-3} for $\Delta T \leq 0$; $p < 10^{-40}$, $n = 12553$).

In Fig. 4 the results of our TD experiments are presented and summarized as box-plots in Fig. 5. Unfortunately only one CPC could be connected with the TD which means that aerosol volatility could not be measured simultaneously at 125°C and 250°C . As a compromise we adjusted the temperature each day around 16:00 to

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



125 °C or 250 °C, respectively. Consequently the time series for CP volatile at 125 °C (CP_{vol} , Eq. 2) or non-volatile at 250 °C (CP_{nonvol} , Eq. 3) is only available in a 2-days rhythm (CP_{RT} , CP_{125} , and CP_{250} are CP concentrations measured at room temperature, 125 °C and 250 °C, respectively):

$$5 \quad CP_{vol} = CP_{RT} - CP_{125} \quad (2)$$

$$CP_{nonvol} = CP_{250} \quad (3)$$

The purpose of these experiments is to characterize particle volatility on longer (seasonal) time scales. Nevertheless, at a given temperature the TD data are available in 15 min averages and thus short-term variations of volatile or non-volatile (but not semi-volatile!) CP concentrations are detectable. To calculate the portion of semi-volatile particles ($CP_{semivol}$, Eq. 4), CP_{vol} and CP_{250} data from different consecutive days have to be considered. We relied on the calculated mean difference using CP_{250} from the day before and after a given CP_{125} was measured:

$$15 \quad CP_{semivol} = CP_{125} - CP_{250} \quad (4)$$

Clearly, this procedure is a compromise and $CP_{semivol}$ data have to be considered with caution. Obviously, the volatile and semi-volatile portion of the particles show a summer maximum, while the non-volatile part dominates during winter (Figs. 4 and 5). Results from our scattered diffusion screen experiments indicate that the relative fraction of non-volatile particles (i.e. most probably sea salt aerosol) with $D_p < 40$ nm is negligible during summer (typically below 10 %) but roughly (30 ± 20) % during winter.

As described above, from the difference of the CP concentrations measured by one of the CPC 3022 (CP_7) and the UCP 3025A (CP_3), the contribution of nucleation mode or ultrafine condensation particles between 3 nm and 7 nm D_p ($UCP_{3,7}$) can be assessed. The raw $UCP_{3,7}$ data set shows an extremely high variability on each time scale. Typically elevated $UCP_{3,7}$ concentrations appear in peaks of some hours duration, which may be assigned to discrete nucleation events. Part of the variability surely

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

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

CP concentrations observed at South Pole were lower by a factor of around two compared to coastal NM, reflecting less efficient transport of the largely marine biogenic CP source (photo-oxidation of algae derived precursor dimethylsulfide, DMS, to nss-SO_4^{2-} and MS aerosol; see discussion below) to the Antarctic Plateau. Year-round observation available from Antarctic coastal sites Mawson ($67^\circ 36' \text{ S}$, $62^\circ 53' \text{ E}$) and Syowa ($69^\circ 00' \text{ S}$, $39^\circ 35' \text{ E}$), showed CP concentrations comparable to NM (Gras, 1993; Ito, 1985; Hara et al., 2011a). Note that due to exceedingly strong and perpetual catabatic winds and low sea salt aerosol concentrations, Mawson has a somewhat continental character (Gras, 1993) which seems, however, not to be reflected in CP concentrations typically observed at this site. Finally, base line sector CP measurements performed at Cape Grim, Tasmania ($40^\circ 42' \text{ S}$, $144^\circ 42' \text{ E}$) during the years 1977–1992 indicated annual median values between 250 and 280 cm^{-3} to be typical for the Southern Ocean (Gras, 1990, 1995).

4.1.2 Inter annual variability and impact of distinctive events

From Fig. 2 it is evident that none of the strong El Niño events consistently influenced the measured CP concentrations. Although so far not explicitly discussed by Bodhaine (1996), this seems also to be valid for the South Pole record for the period 1974–1994 (Bodhaine, 1996; Fig. 3 therein). In contrast, Gras (1990) found a distinct negative correlation between CP concentrations observed at Cape Grim with the SOI. The Pinatubo eruption was the strongest volcanic event in the antecedent century (June 1991 at 15° N) and injected around $18 \pm 2 \text{ Mt}$ (10^{12} g) SO_2 up to the middle stratosphere (Krueger et al., 1995) causing a historical disturbance of the stratospheric sulfate layer (e.g. Russell et al., 1996) associated with an appreciable impact on global climate (McCormick et al., 1995). This event was followed by the much smaller eruption of Cerro Hudson (45° S) in October 1991. Legrand and Wagenbach (1998) extensively discussed the impact of both eruptions and concluded that this event lead only to a relatively small perturbation of the near surface nss-SO_4^{2-} level at NM. Particle concentrations at NM appeared not significantly higher during austral summer 1991/1992, when

the impact of volcanogenic sulfate aerosol should have been most pronounced (Russel et al., 1996; Legrand and Wagenbach, 1998). Consequently the overall relevance of the stratosphere as particle source for coastal boundary layer air at NM seems negligible and also nss-SO₄²⁻ measurements indicated an only ancillary contribution (Minikin et al., 1998; Legrand and Wagenbach, 1998).

For further time series analyses in the time and spectral (frequency) domain, it was important to avoid any spurious influence of the inherent seasonality of the CP data set. Hence, we consider now the anomalies derived from the CP and the SIE records (note that SAM and SOI are anomalies by definition). Generally no significant correlation with circulation indices or SIE could be detected in the time domain. Furthermore, the CP record does not show any significant periodicities > 1 yr. Only a MC-SSA data adaptive filtered representation (i.e. a reconstruction based on the leading four principal components, explaining 35 % of the CP variability) revealed a broadband, i.e. non-harmonic periodicity between 2 yr and 3 yr, which is also evident in the SAM and SOI records. Nevertheless, there is definitely no significant cross correlation between CP anomalies and SAM or SOI, neither in the time nor in the frequency domain. In conclusion, statistical time series analyses revealed that though CP concentrations at NM exhibit a pronounced inter-annual variability, long term trends and correlations with the climate related indices SIE, SAM, and SOI are virtually absent.

In summary, our conclusions drawn from CP time series analyses appear consistent with recent findings regarding the long-term record of the ionic aerosol composition at NM (Weller et al., 2011). The main sources of Antarctica aerosol, as there are biogenic sulfur (i.e. nss-SO₄²⁻ and MS) and sea salt compounds, seem to be essentially dependent on the patchy oceanic source distribution and transport efficiency, the latter governed by regional or even local meteorology, while correlations with climate related indices show weak, mostly inconsistent and barely meaningful correlations (Weller et al., 2011) indicating a minor relevance of these couplings for the observation period and site under discussion.

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

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

4.1.3 Impact of PBL inversion strength

During winter regional DMS sources can be considered as inactive due to sea ice coverage and lack of actinic radiation. In this case, higher CP concentrations are observed when the PBL showed no surface inversion. Such a situation is characteristic for stormy weather under passing low pressure systems which potentially boosts sea salt production but also facilitates long-range transport and down-mixing of free tropospheric air masses (König-Langlo et al., 1998; Hara et al., 2011a). During summer, strong surface inversions are typically associated with bright weather conditions and potentially higher photochemical activity. Now particle concentrations are dominated by regional sulfur aerosol sources and photochemical induced particle nucleation (see Sect. 4.3).

4.2 Link to chemical composition and seasonality

4.2.1 Survey of the chemical composition of coastal Antarctic aerosol

From previous investigations we know that the ionic composition of the aerosol at NM shows a distinct seasonality with high portion of biogenic sulfur during summer (about 36 % by weight referring to ionic species and mineral dust from November through March), decreasing to less than 7 % from April through October while throughout the year sea salt aerosol is clearly dominating with a relative portion of around 50 % during summer increasing to 86 % in winter (Weller et al., 2008). Nevertheless, due to high variability of the ionic aerosol composition, biogenic sulfur frequently constitutes the main ionic aerosol compound (more than 75 %) during summer, while during winter sea spray generally dominates (Weller and Lampert, 2008). Due to the lack of size-segregated aerosol chemistry investigations at Neumayer, we refer to respective measurements from the Finnish station Aboa (73°03' S, 13°25' W) and the French station Dumont d'Urville (66°40' S, 104°01' E) showing a similar mass size distribution for nss-SO_4^{2-} , MS and NH_4^+ with a dominant mode in the accumulation size range at about

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



0.3 μm , while sea salt and nitrate were nearly exclusively in the super micron size range (Teinilä et al., 2000; Jourdain and Legrand, 2001).

4.2.2 Link to chemical composition by TD experiments

Coinciding nss- SO_4^{2-} , MS and CP concentration maxima in summer (Fig. 3), indicate the dominance of biogenic sulfur aerosol in determining particle concentration. This conclusion is consistent with highest non-volatile particles portions during winter, while volatile and semi-volatile particles dominate during summer (Figs. 4 and 5). For the years 2005 and 2006 ionic composition of the aerosol is available in daily resolution and could be compared with the TD experiments. The accordant results presented in Fig. 8 demonstrate the coherence of biogenic sulfur aerosol with volatile and semi-volatile particles and sea salt with the refractory (non-volatile) portion. This agreement is of course only qualitative, because TD experiments refer to particle number concentrations while the chemical composition corresponds to particle mass concentration. Even during summer, when particles are mainly volatile or semi-volatile, aerosol mass can frequently be dominated by (coarse mode) sea salt particles. During winter the situation is more uniform: TD results reveal that roughly 80 % of the particles are refractory suggesting that particle number concentrations and aerosol mass are dominated by sea salt aerosol (Figs. 5 and 8). Nevertheless, non-sea-salt sub-micrometer aerosol (i.e. volatile or semi volatile sulfate and nitrate, as well as non-volatile mineral dust and black carbon), though negligible by mass, may partly contribute to the observed winter-time CP concentrations.

A more detailed inspection of this overall consistent picture revealed some open questions, especially associated with the portion of semi-volatile particles. Measured NH_4^+ concentrations at NM are so low that in general only roughly 15 %, 10 %, or 7.5 % of the nss- SO_4^{2-} could potentially be bound as NH_4HSO_4 (ammoniumbisulfate), $(\text{NH}_4)_3\text{H}(\text{SO}_4)_2$ (letovicite), or $(\text{NH}_4)_2\text{SO}_4$ (ammoniumsulfate), respectively, the most plausible candidates for semi-volatile particles. This has to be compared with the TD results which revealed that about 50 % of the particles are semi-volatile and 25 %

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



volatile. Note, although determination of the semi-volatile part was inherently inaccurate, this caveat could hardly have biased the results all up to the higher end. Thus particle number concentrations seem to be dominated by semi-volatile particles, necessitating a smaller mean diameter compared to the volatile portion to be in agreement with the chemical composition, which is hardly plausible. We may speculate that biogenic sulfur aerosol at NM is to a lesser extent a heterogeneous mixture of H_2SO_4 (volatile) and NH_4HSO_4 (semi-volatile) particles, but rather an internal mixture of partly neutralized sulfuric acid like $\text{H}_2\text{O}-(\text{NH}_4)\text{HSO}_4\text{-H}_2\text{SO}_4$ (including most probably methane sulfonic acid). This is supported by particle volatility measurements at the Finnish Antarctic Station Aboa (Asmi et al., 2010), Syowa (Hara et al., 2011b), and results from combined FTIR spectrometer and sun photometer measurements at NM in January/February 2000 (Rathke et al., 2002). The stoichiometric composition and hence the volatility threshold temperature of this internally mixed aerosol should be variable depending on prevailing atmospheric chemistry of DMS oxidation (gas phase or in cloud processes, presence of NH_3) and meteorological conditions. An open question is the contribution of organic carbon in particles. Measurements of hygroscopicity, volatility and chemical composition of aerosol particles at the Finnish Antarctic station Aboa indicate an organic fraction of around 20–30% in the $<1\ \mu\text{m}$ aerosol mass (Asmi et al., 2010).

4.2.3 Seasonality

Obviously the shape of the CP seasonality with its characteristic stepwise increase in spring is different to the distinct and mostly concurrent seasonal maxima of nss-SO_4^{2-} and MS (Fig. 3) in January. It seems that especially the portion of semi-volatile particles increased during early spring, while the increase of the volatile fraction is more gradually (Figs. 4 and 5). In particular for the early CP rise in September there is no equivalent in the nss-SO_4^{2-} and MS seasonality and the absolute CP maximum in March is accompanied by an only much weaker nss-SO_4^{2-} and MS secondary maximum (Fig. 3). A

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Diurnal variations of
condensation particle
concentrations at
Neumayer, Antarctica**

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



plausible explanation for this finding could be new particle formation (see next section). This photo-chemically initiated process primarily enhanced particle number concentrations (mainly within the nucleation and Aitken modes) but hardly aerosol mass and hence nss-SO_4^{2-} and MS concentrations. Indeed, the observed seasonality of $\text{UCP}_{3,7}$ concentrations supports this perception (Fig. 7). The maximum of $\text{UCP}_{3,7}$ (and CP) concentrations in March may be initiated by enhanced bio-productivity associated with the regular retreat of the last sea ice in the nearby Atka Bay (about 8 km northeast of the station) in late February/March. However, considering the short atmospheric lifetime of nucleation mode particles (coagulation lifetime in remote marine areas according to Heintzenberg et al. (2004) roughly 10 h at 3 nm D_p), observed nucleation during September is difficult to interpret because SIE is at maximum then and only sporadic polynias appear in the adjacencies of NM. One may speculate about the role of springtime photochemical processes associated with halogen activation over sea ice pertaining to DMS oxidation and aerosol formation (e.g. von Glasow and Crutzen, 2004; Simpson et al., 2007; Read et al., 2008). An inspection of our surface ozone time series revealed that several tropospheric ozone depletion events (indicating the impact of halogen chemistry) were accompanied by enhanced CP concentrations, but this finding is not consistent throughout. Nevertheless, supporting our hypothesis, year-round long-pass DOAS at Halley showed maximum boundary layer BrO and IO concentration during spring (Saiz-Lopez et al., 2007).

Similar to NM, CP concentrations at South Pole steadily increase in September to a distinct November peak. In contrast to NM there is a characteristic December sub-minimum, usually followed by short periods with increased CP concentrations from the second half of January through March (Samson et al., 1990, Figs. 1 and 2 therein). Note that South Pole is the sole continental Antarctic site discussed here, i.e. marine aerosol sources are absent and local sulfur aerosol formation seems negligible as demonstrated by the recent ISCAT campaign (Mauldin III et al., 2004). Consequently, CP concentrations at South Pole are governed by atmospheric transport processes (Samson et al., 1990; Mauldin III et al., 2004).

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Mean annual cycles derived from about two years of continuous CP observations at Mawson and Syowa showed enhanced concentrations between September and April (Gras, 1993; Ito, 1985; Hara et al., 2011a), but only at Syowa a distinct absolute maximum in February/March, analogue to NM, was identifiable (Ito, 1985). Regarding the temperate zone of the Southern Ocean, a comparison with CP observations from Cape Grim revealed once again a broad maximum between September and April (Gras, 1990), reflecting the annual cycle of biological activity in the surface water of the Southern Ocean. Finally a recent model approach predicted a broad CP maximum between November and March for NM, but could neither reproduce the early increase in September nor the absolute maximum in March (Spracklen et al., 2010; Fig. 5 therein). This discrepancy indicates that more regional or even local processes not resolved in the present model play a decisive role in determining particle concentrations at this site.

4.3 Diurnal cycle of CP concentrations and nucleation events

From September through April, $UCP_{3,7}$ values commonly showed peaks with a typical duration of 2–10 h, frequently on top of a persistent background $UCP_{3,7}$ level of several tens per cm^3 . We tentatively denote such peaks as nucleation events. Note, however, that according to Dal Maso et al. (2005) a particle formation or nucleation event is only existent if a distinct nucleation mode appears that shows signs of growth. Unfortunately, the latter could not be assessed by our experimental equipment. Figure 9 is a representative observed at 12 February 2006. The temporal resolution of the measured CP concentrations was 150 sec and error bars indicate the estimated accuracy of the derived $UCP_{3,7}$ concentrations. The arrow marks the moment when the diffusion screen was switched on. During these 15 min, $UCP_{3,7}$ concentrations decayed to negligible amounts and the estimated abundance of particles below the diffusion screen cut-off of 40 nm D_p was around 65%. We found that over the year around 20% of the particles can be assigned to the nucleation mode between D_p 3 nm and 7 nm. According to the mean diurnal $UCP_{3,7}$ concentration cycle presented in Fig. 10,

nucleation events occurred primarily during afternoon. Concurrent results from the TD experiments, which were restricted to CP₇ concentrations (and not UCP_{3,7}) indicate a highly variable contribution of volatile and semi-volatile particles (ranging between 20 % and 78 % for each class), while the portion of non-volatile aerosol was generally below 20 % during nucleation events.

In agreement with previous results from the Finnish Antarctic Station Aboa (Koponen et al., 2003), UCP_{3,7} concentration peaks of several hundred cm⁻³ were quite frequent at NM, but did very rarely exceed 1000 cm⁻³. As expected, natural nucleation events could not be detected at continental South Pole (Park et al., 2004). For comparison, much higher UCP_{3,7} concentrations (>10⁴ cm⁻³) were typically observed e.g. during nucleation events in mid-latitude boreal forests (Kulmala et al., 2001) or certain coastal regions with extensive exposed shorelines during low tide (O'Dowd et al., 2002a; McFiggans et al., 2010). We estimated from the ascent branch of the observed UCP_{3,7} concentrations ($N_{3,7}$) in Fig. 9 a nucleation rate $\Delta N_{3,7}/\Delta t$ of about 0.05–0.07 cm⁻³ s⁻¹, a value which could be somewhat biased by advection of unknown spatial UCP_{3,7} concentration variations (the wind speed was around 8 m s⁻¹). According to Kulmala et al. (2004) this simple approach, neglecting coagulation and condensation sinks as well as transport processes, is usually reliable for clean, homogenous air masses. A comparison with other studies emerged partly difficult, because different experimental and evaluation methods were employed to determine nucleation rates. Usually, previous investigations refer to particles within 3–10 nm D_p , often and ideally recorded by continuous size distribution measurements (see Kulmala et al., 2004 for a detailed review). Typically new particle formation rates in the range of 0.01–10 cm⁻³ s⁻¹ were characteristic for remote boundary layer air, reaching in extreme cases (coastal areas and industrial plumes) occasionally 10⁴–10⁵ cm⁻³ s⁻¹ (Kulmala et al., 2004). Finally these reported values as well as our estimate cannot be treated as a true nucleation rate. An assessment of the latter would require the determination of so-called thermodynamically stable cluster (TSC) concentrations in the size range < 2 nm, which has been successfully realized only recently (Kulmala et al., 2007) or would need a considerable

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



additional correction (Kerminen and Kulmala, 2002) which was not feasible on the base of our data. Nevertheless, we may state that nucleation rates observed at NM were rather at the lower end compared to nucleation events typically observed in continental and coastal mid-latitudes.

5 Considering the importance of biogenic sulfur aerosol and the presence of ammonia during summer at NM (Weller and Lampert, 2008), we suggest that ternary nucleation of H_2SO_4 (Kulmala et al., 2000; Sipilä et al., 2010) was the most important nucleation mechanism, probably supported by iodine compounds (O'Dowd et al., 2002a,b; McFiggans et al., 2010). As for the latter, continuous year-round DOAS measurements
10 at NM (Frieß et al., 2001) and Halley (Saiz-Lopez et al., 2007) indicated boundary layer mixing ratios of IO comparable to results from Roscoff (Brittany), a site where iodine induced nucleation events are typical (McFiggans et al., 2010). In addition, as suggested by Asmi et al. (2010), organic compounds participate in new particle formation and particle growth process. We could not ascertain the impact from down
15 mixing of free tropospheric air, but the observed diurnal cycle is consistent with a photochemically induced process. Nucleation events observed at NM were not associated with peculiar general weather situations, the origin of air masses (classification in marine and continental by 5-days backward trajectories), or wind velocity (note however, that at wind speeds $> 10 \text{ m s}^{-1}$ nucleation events above 200 cm^{-3} were rare). Asmi et al. (2010) found some nucleation events linked with high wind speed at Aboa which they attributed to ion production by fast moving ice and snow crystals and subsequent ion-mediated nucleation (Virkkula et al., 2007). Previous results from this site reported by Koponen et al. (2003) indicated new particle formation exclusively in marine/coastal air masses in contrast to our and more recent investigations reported by Asmi et al. (2010).
20

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

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

5 Conclusions

Apart from distinct inter-annual variability, condensation particle concentrations at Neumayer showed no obvious long-term trend. This is remarkable since Antarctica experienced significant climate relevant changes in the past three decades like the development of the ozone hole (Solomon, 1999; Thompson and Solomon, 2002) or warming of the Antarctic Peninsula (Steig et al., 2009). This goes along with a consistent lack of any meaningful correlation with climate factors like sea ice extent or atmospheric circulation indices in our 25 yr long time series of the ionic composition of the aerosol (Weller et al., 2011). In addition, an impact of outstanding events like the Mt. Pinatubo eruption and strong El Niño phases on particle concentrations at Neumayer was not apparent. Particle concentrations at a given site are not only determined by source strength (in our case the bio-productivity of the South Atlantic and wind induced sea salt generation) and transport efficiency governed by regional or even local meteorology, but also by the amount of pre-existing aerosol surface area acting as condensation sink (Heintzenberg et al., 2004). The latter critically affects the relevance of new particle formation and thus ultimately the particle number concentration. We infer from our examinations that eventually these processes determined most probably CP concentration variability at all time scales considered and obliterated the imprint of superordinate climate signals in the Southern Ocean and Antarctic region. Regarding the seasonality of particle concentrations at NM, the stepwise increase to the annual maximum in March is noticeable and evident each year but not identically seen in biogenic sulfur records. Year-round ultrafine particle detection identified particle nucleation as a plausible candidate responsible for the overall seasonality of CP concentrations. These observations emphasize the importance of local particle nucleation within the planetary boundary layer and/or down-mixing of nucleation particle rich air masses from the free troposphere governing the aerosol budget in terms of number concentration at this site.

More detailed conclusions addressing nucleation processes as well as chemical composition and growth dynamics of different particle size modes along with their

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



seasonal and inter-annual variations require additional continuous long-term configured particle size distribution measurements and size segregated aerosol sampling. Moreover, there is still a notorious and serious lack of data regarding the vertical distribution of particles in connection with the general weather situation as well as the dynamics of their vertical exchange in South Polar regions. Future investigations should as well focus on the evaluation of congruent observations at coastal and continental Antarctic sites especially in terms of organic compounds with the objective to gain a more comprehensive picture of Antarctic aerosol dynamics.

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Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Diurnal variations of
condensation particle
concentrations at
Neumayer, Antarctica**

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Diurnal variations of
condensation particle
concentrations at
Neumayer, Antarctica**

R. Weller et al.

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

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Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Table 1. Listing of operation periods of the different particle counters types at Neumayer Station.

period diameter	instrument	temporal resolution	50 % cut-off ($D_{p(50\%)}$)
Jan 1982–Dec 1989	CPC 3020	60 min	10 nm
Jan 1990–Jun 1992	CPC 3760	60 min	7 nm
Jul 1992–Dec 1994	CPC 3760	150 s	7 nm
Jan 1995–Dec 2008	CPC 3022(A)	150 s	7 nm
Feb 1996–Dec 1998	CPC 3025A	150 s	3 nm
Jan 2000–Apr 2006	CPC 3025A	150 s	3 nm
Jan 2008 ongoing	CPC 3025A	150 s	3 nm

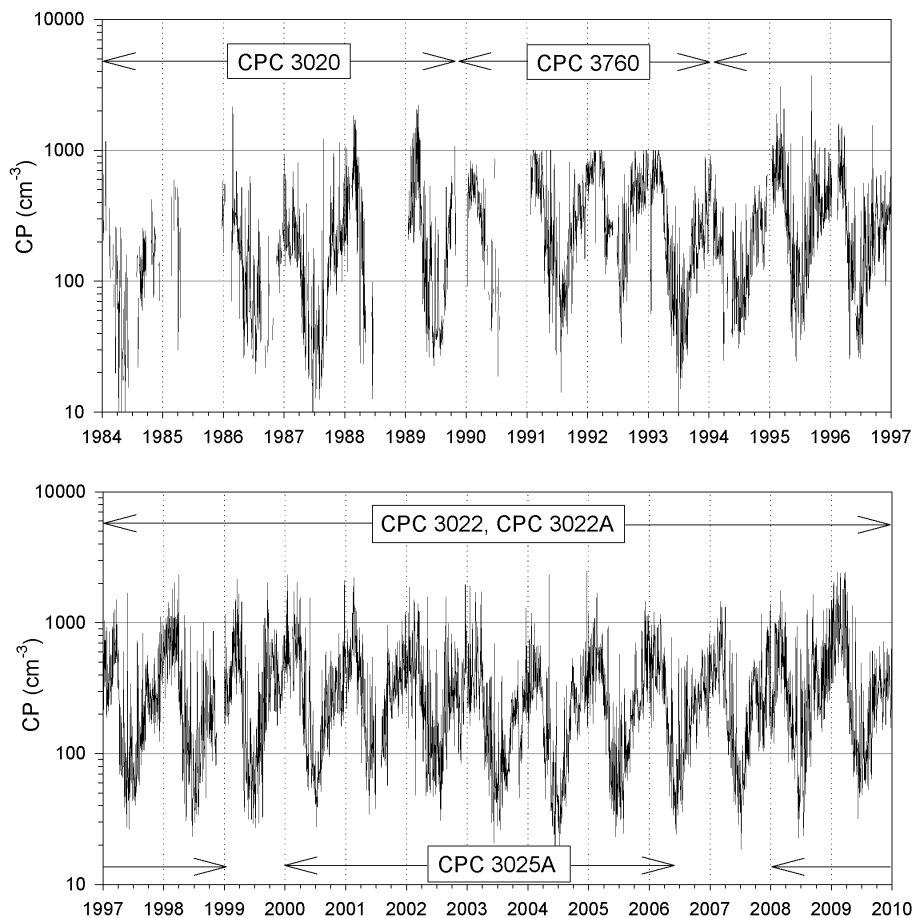


Fig. 1. Time series of the measured CP concentrations (daily means). Also indicated are the operation periods of the different particle counters types.

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



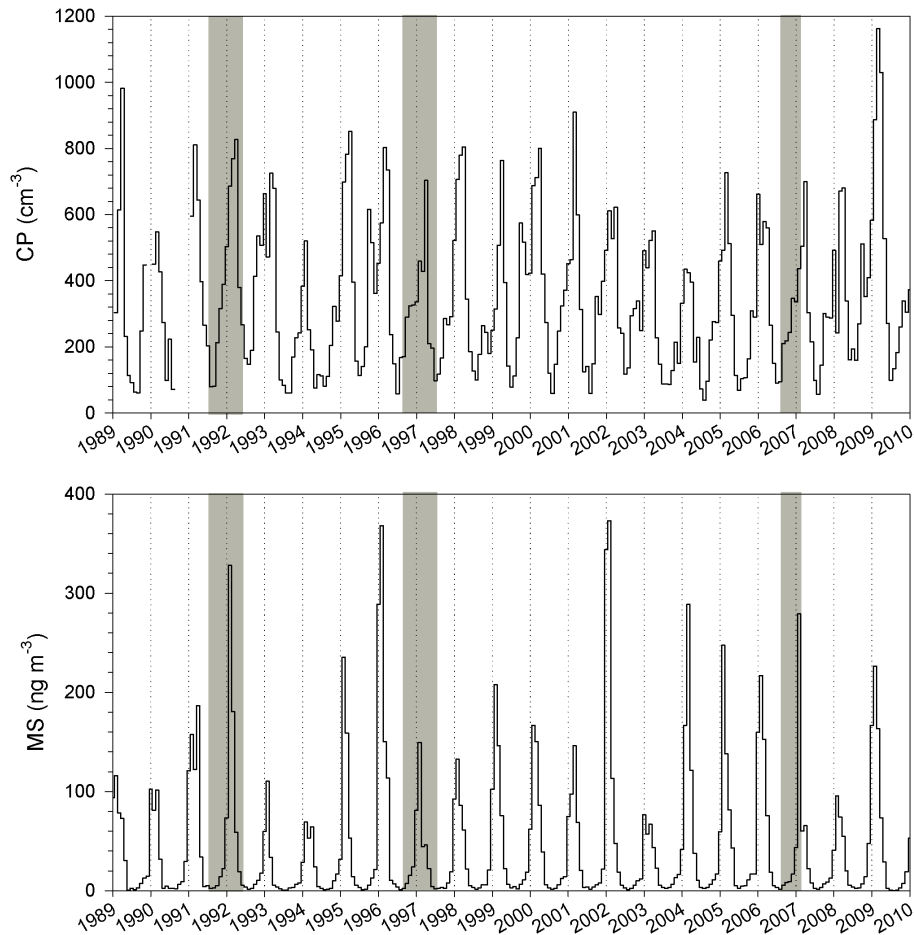


Fig. 2. Condensation particle time series (top panel) and methane sulfonate (MS) record from NM (bottom panel), both displayed in monthly mean bins. Shaded grey areas: El Niño events (1997 and 2006) and eruption of Mt. Pinatubo (1991).

20747

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

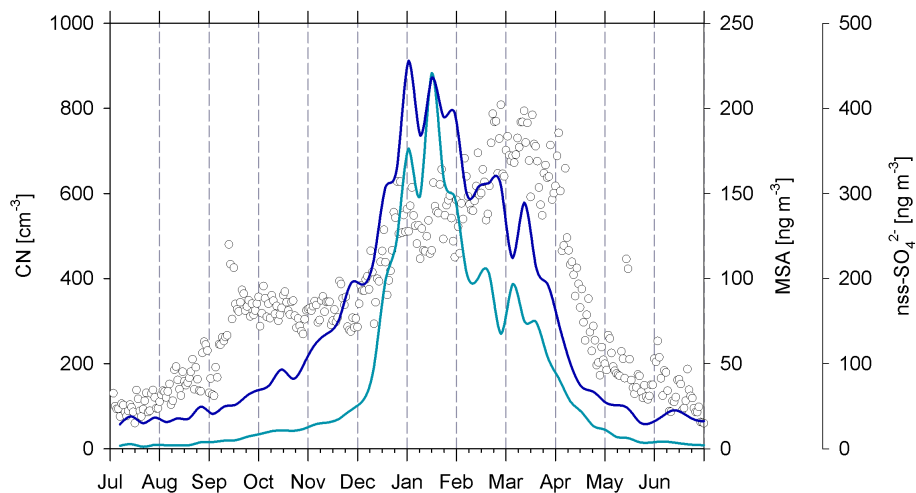


Fig. 3. Mean annual cycle of the CP concentrations (circles, based on daily mean values for the period 1991 through 2009), nss-SO₄²⁻ and MS (solid blue and cyan line, respectively, both based on weekly samples taken during the same period).

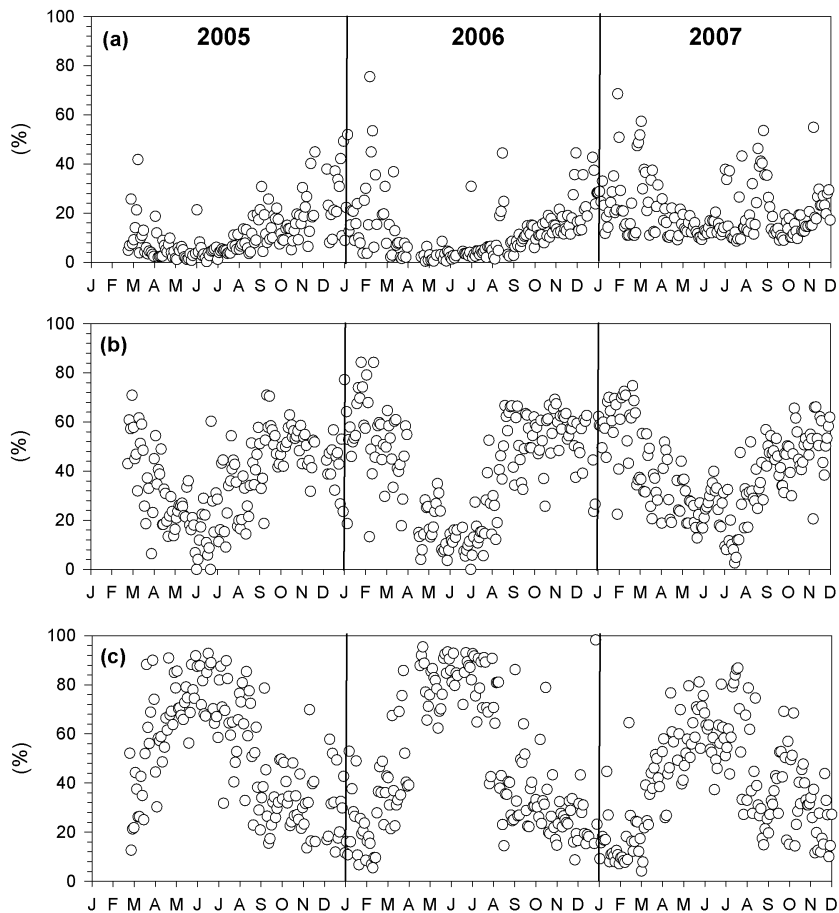


Fig. 4. Results of the thermodenuder experiments: portion of volatile **(a)**, semi-volatile **(b)** and refractory **(c)** particles based on daily mean values.

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

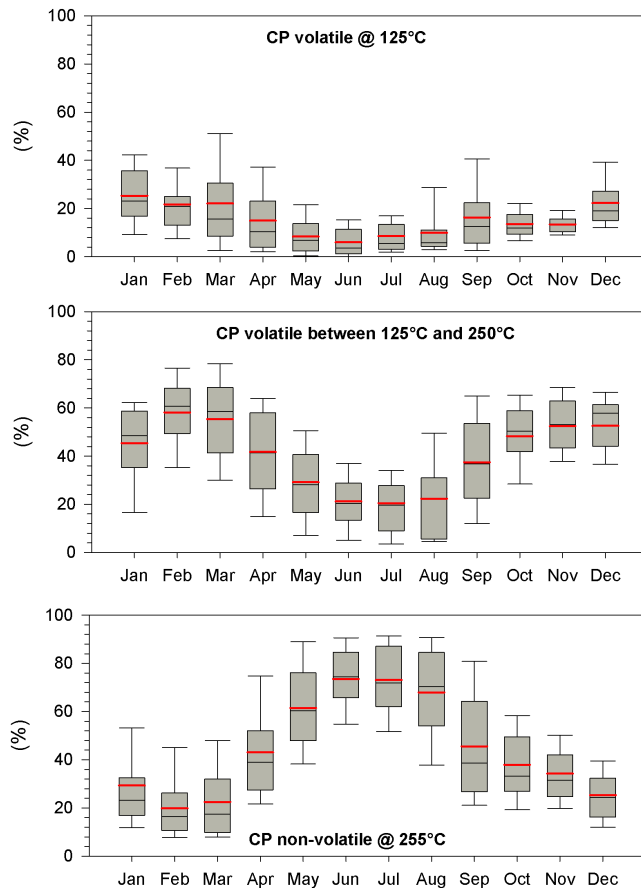


Fig. 5. Box plots of the mean seasonality of the volatile, semi-volatile and refractory portion of the measured particle concentrations for the years 2005–2007. Lines in the middle of the boxes represent sample medians (mean: red line), lower and upper lines of the boxes are the 25th and 75th percentiles, and whiskers indicate the 10th and 90th percentiles.

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

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

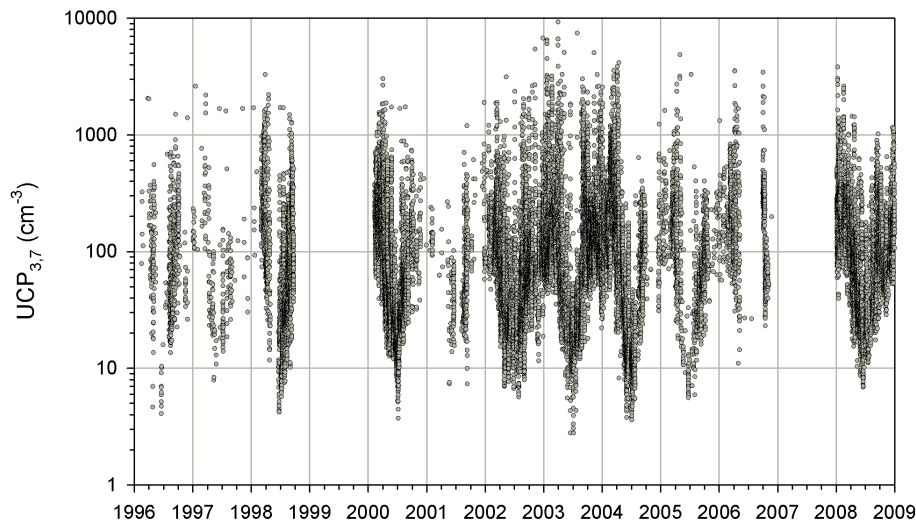


Fig. 6. Measured concentrations of ultrafine particles, $UCP_{3,7}$ (D_p between 3 nm and 7 nm) based on hourly means. Note that the larger data gaps from late 1989 until early 2000 and fall 2006 through 2007 were caused by malfunction of the UCP 3025A.

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

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

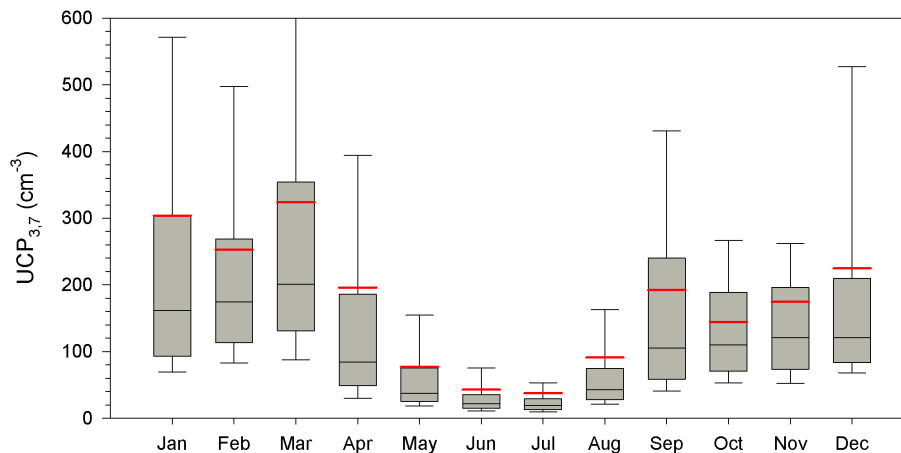


Fig. 7. Notched box plots of the mean seasonality of UCP_{3,7} concentrations. For description of the symbols see Fig. 5.

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

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

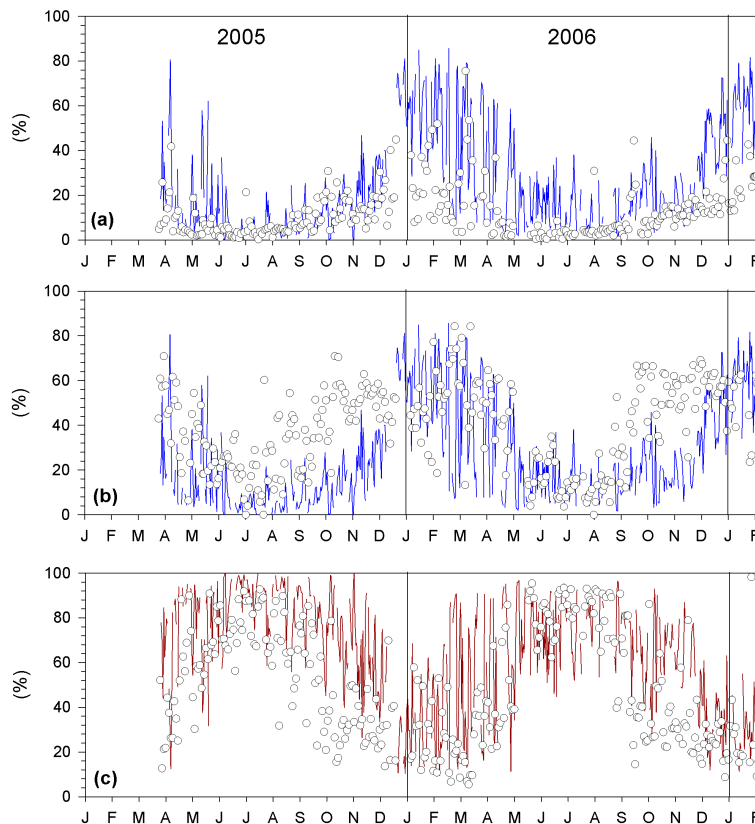


Fig. 8. Results of the thermodenuder experiments: portion of volatile **(a)** and semi-volatile **(b)** particles based on daily mean values along with the portion of biogenic sulfur (i.e. the sum of nss-SO_4^{2-} and MS concentrations, blue line) as well as refractory particles **(c)** along with the portion of sea salt aerosol (red line). The ionic concentrations were derived from daily aerosol samples taken during the same period.

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

**Diurnal variations of
condensation particle
concentrations at
Neumayer, Antarctica**

R. Weller et al.

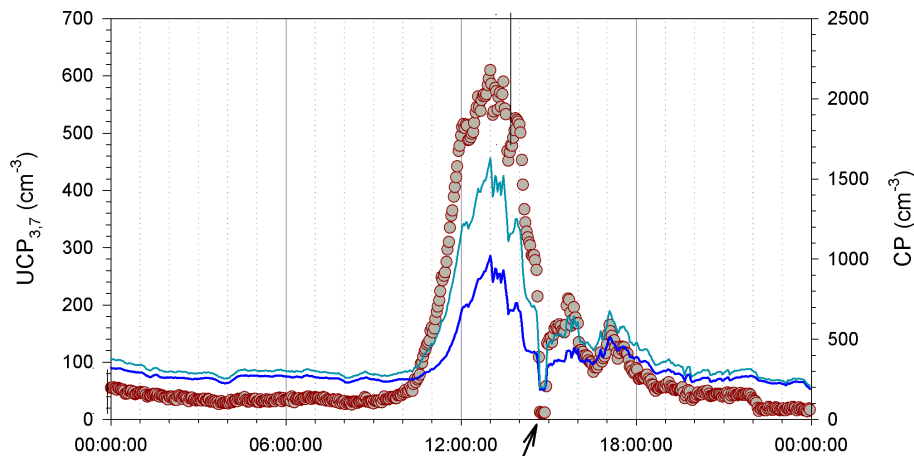


Fig. 9. Nucleation event observed at 12 February 2006. $UCP_{3,7}$ concentrations (circles, left hand scale) derived from the CPC 3022A and UCPC 3025A instruments (green and blue line, respectively; right hand scale) in the highest available temporal resolution (150 s). The two error bars visualize the typical uncertainty of the calculated $UCP_{3,7}$ concentrations (see main text). The arrow marks the moment when the diffusion screen was switch on for 15 min.

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

Diurnal variations of condensation particle concentrations at Neumayer, Antarctica

R. Weller et al.

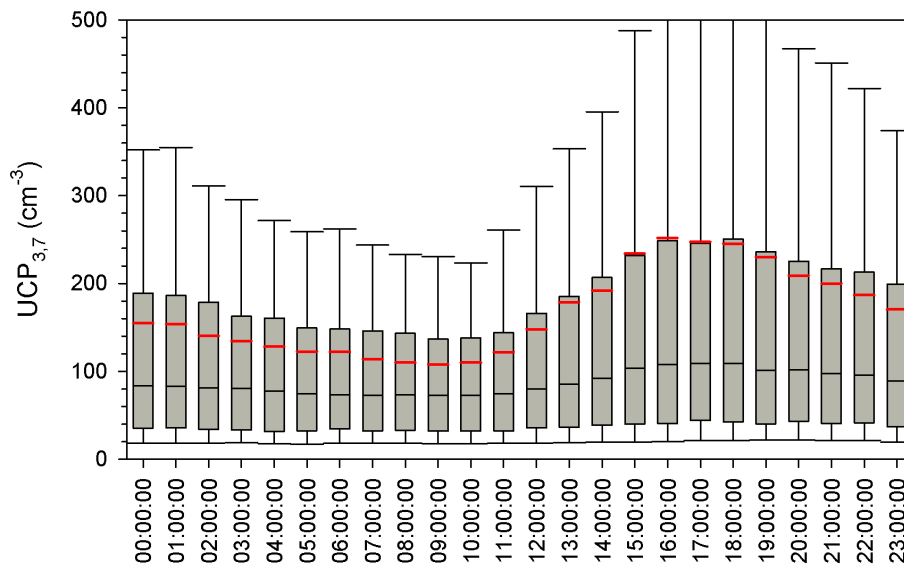


Fig. 10. Mean diurnal cycle of the $UCP_{3,7}$ concentrations. For description of the symbols see Fig. 5.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion