

**On the potential
contribution of open
lead particle
emissions**

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On the potential contribution of open lead particle emissions to the central Arctic aerosol concentration

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Abstract

During the ice-breaker borne ASCOS expedition (Arctic Summer Cloud Ocean Study) direct eddy covariance measurements of aerosol number fluxes were carried out in August 2008 on the edge of an ice floe drifting in the central Arctic Ocean between 2°–10° W longitude and 87°–87.5° N latitude. The median aerosol transfer velocities over different surface types (open water leads, ice ridges, snow and ice surfaces) ranged from 0.27 to 0.68 mm s⁻¹ during deposition-dominated episodes. Emission periods were observed more frequently over the open lead, while the snow behaved primarily as a deposition surface. Directly measured aerosol fluxes were compared with particle deposition parameterizations in order to estimate the emission flux from the observed net aerosol flux. Finally, the contribution of the open lead particle source to atmospheric variations in particle number concentration was evaluated and compared with the observed temporal evolution of particle number. The direct emission of aerosol particles from the open lead can only explain 5–10% of the observed particle number variation in the mixing layer close to the surface.

1 Introduction

The Arctic region north of 80° N provides a unique setting to investigate the impact of aerosol particles on the climate system. Complex aerosol-cloud-ice-ocean interactions can be studied under very limited anthropogenic influence especially during the summer months (Leck and Persson, 1996). While the influx of polluted mid-latitude air and extended aerosol residence times lead to elevated aerosol concentrations during the winter to early spring Arctic haze period (Heintzenberg and Leck, 1994), minimum effects from continental sources are encountered during summer from June to August when the central Arctic lower atmosphere is effectively isolated from anthropogenic emissions due to the prevailing atmospheric circulation patterns and near-surface processes in the marginal ice zone.

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The complex aerosol-cloud-radiation interactions in the Arctic constitute a warming factor for the regional climate during most of the year (e.g. Intrieri et al., 2002; Tjernström, 2005). This is due partly to the semi permanent ice cover, raising the surface albedo compared to that of the ocean surface, and partly to the very clean air, reducing the cloud albedo. Under clean air conditions as observed in the Arctic summer, even small numbers of ice nuclei (Bigg, 1996) potentially play a key role in cloud development, and thus, the regional climate (e.g. Prenni et al., 2007; Möhler et al., 2007). As long as there are no major intrusions of polluted air (e.g. Carrio et al., 2005), extremely low aerosol particle and cloud condensation nuclei (CCN) concentrations in the high Arctic (e.g. Bigg et al., 1996; Bigg and Leck, 2001; Leck et al., 2002; Lohmann and Leck, 2005) will result in low concentrations of relatively large cloud droplets, leading to a relatively low cloud albedo and to frequent formation of drizzle (e.g. Rasmussen et al., 2002). Previous studies indicate that drizzle formation is likely to affect boundary layer motions on different temporal scales (e.g. Feingold et al., 1999). Optically thin stratiform clouds play a prominent role over the central Arctic Ocean. While they are the single most important factor determining the surface radiation budget, current climate models are unable to yield a realistic description of Arctic clouds and their impact on the surface radiation (e.g. Walsh et al., 2002; Tjernström et al., 2008; Karlsson and Svensson, 2010), and they are far from incorporating the relevant cloud-ice-ocean feedbacks. Model projections suggest that the Arctic regional climate could transition into a new stable regime with no summer sea-ice within only a few decades (Lenton et al., 2008). This would impact considerably on the Arctic Ocean ecosystem and affect large-scale atmospheric and oceanic circulation patterns. Refined parameterizations of aerosol-cloud feedbacks in global and regional climate models require development of novel observational capabilities and extensive field investigations to identify and quantify aerosol sources, sinks, and transport and transformation processes.

The cloud albedo has been shown to be very sensitive to particle concentration changes under clean conditions, and for optically thin clouds (Twomey, 1974). In a changing climate, a small increase in CCN could increase the albedo of the clouds and

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lead to decreased ice melt, while an increase in ice melt would lead to a decreased surface albedo through formation of melt ponds on the ice (Leck et al., 2004). Over the central Arctic Ocean, a local biogenic source of aerosol particles from bursting bubbles at the water-air interface has been suggested (e.g. Leck and Bigg, 1999, 2005a; Leck et al., 2002). It links marine biological activity, clouds and climate through the ejection of organic microcolloids (Wells and Goldberg, 1991) from the surface microlayer of open leads (Bigg et al., 2004) into the atmosphere. Once airborne, some of these particles may act directly as CCN, while others are activated after condensational growth (Leck and Bigg, 2005b). Even though this source of CCN may explain some important aspects of the Arctic aerosol-cloud-climate relationship, many of the controlling mechanisms, e.g. the dependence of the bubble-bursting mechanism on wind, temperature, salinity, and possibly other factors, remain unknown.

This study aims to evaluate the relevance of particles emitted directly from open leads in Arctic aerosol-ice-cloud interactions, and quantifying its contribution to the atmospheric aerosol burden by direct measurements of the net particle flux.

Previously, turbulent particle fluxes have been measured in the high Arctic over the open sea and over the pack ice (Nilsson and Rannik, 2001). However, the measurement footprints over the pack ice were generally large, and Nilsson and Rannik (2001) acknowledge that most measurements were influenced by a mix of open lead and ice surfaces. In this study, turbulence measurements were performed closer to the surface and close to the edge of a lead, thus decreasing the footprint areas and allowing for a better separation of measurements influenced by the open lead and by the ice and snow surfaces, respectively, depending on the wind direction. Still, the problem remains that eddy covariance flux measurements yield a net flux which is a superposition of particle emission and deposition fluxes. In order to derive the emission flux, an independent estimate of the deposition flux is required. This estimate may be obtained from a theoretical parameterization of particle deposition; however, a thorough evaluation of the introduced uncertainties is necessary before any further conclusions can be drawn.

2 Method and site

Turbulent aerosol number fluxes were measured from an ice floe drifting in the central Arctic Ocean between 2°–10° W and 87°–87.5° N from 17 August to 1 September, 2008. An eddy covariance system was set up on the edge of an open lead at 2.5 m above the surface. Depending on the wind direction, the measured turbulent fluxes were influenced by the open lead or by the ice floe. The system consisted of a Gill R3 sonic anemometer (Gill, Lymington, UK) for three-dimensional wind measurements, a Licor LI-7500 gas analyzer (Licor, Lincoln, NE, USA) for carbon dioxide (CO₂) and water (H₂O) vapor concentration measurements, and a condensation particle counter CPC 3760A (TSI, St. Paul, MN, USA) for number concentration measurements of particles greater than 11 nm in diameter. The response time of the particle counter, including the sampling line, was approximately 1.4 s. Wind and CO₂/H₂O data were logged at 20 Hz, while particle number concentration data at 10 Hz, using a MOXA UC7420 computer (Moxa Inc., Brea, CA, USA). All instruments were battery-operated during the entire measuring period.

Aerosol number fluxes were calculated according to standard eddy covariance procedures, with 30 min averaging, after a coordinate rotation using the planar fit method (Wilczak et al., 2001), and linear detrending of the aerosol number time series. Because of the traveling time of the aerosol sample from the sampling point through the inlet tubing to the particle counter, and the traveling time in the particle counter, a constant time lag of 2.6 s was used to synchronize the wind with the aerosol time series. This time lag was confirmed by maximizing the covariance given by the cross-correlation function of the vertical wind speed and particle number concentration. The passage through the sampling line also degraded the response time of the system with regard to ambient aerosol concentration changes. It is important to bear in mind that this eddy covariance setup with a response time of 1.4 s cannot resolve 10 Hz aerosol number concentration fluctuations. The underestimation of the aerosol fluxes due to fluctuation dampening was corrected according to Horst (1997). With typical

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wind speeds of less than 3 m s^{-1} , we found the magnitude of this correction to be typically less than 50%. No additional corrections were applied to the aerosol fluxes.

Spectral analysis of the aerosol number concentration time series confirmed the limited response time of the eddy covariance setup. Figure 1 presents normalized ogive functions, i.e. cumulative cospectra of the vertical wind speed w vs. sonic temperature T and aerosol number c , during two different measurement periods on 20 August and 24 August 2008. The buoyancy flux ogives exhibit a typical shape with flux contributions in the frequency range from 0.03 Hz to 5 Hz. In general, the aerosol ogives show much higher scatter. On 20 August, shown in Fig. 1a, there are only negligible contributions to the aerosol cospectrum at frequencies above 0.2 Hz. In contrast, on 24 August shown in Fig. 1b, we observe high frequency flux contributions even beyond the frequency corresponding with the approximate response time of the particle counter (1.4 s, dashed vertical line). On 24 August, the cospectral shape of aerosol and temperature are rather similar. However, at low frequencies, the buoyancy flux ogive flattens out around 0.03 Hz, whereas the aerosol ogive deviates from this behavior in the range from 0.002 to 0.02 Hz.

Data quality was evaluated by testing the stationarity of the time series according to Foken and Wichura (1996). Data were discarded when the average of six 5 min intervals of the standard deviation of the particle number concentration (or temperature) deviated by more than 70% from the 30 min standard deviation. We also discarded data if the 30 min standard deviation of the particle number concentration was larger than 30 cm^{-3} , indicating particle pollution from, for example, snow mobiles or helicopter flights. Moreover, the integral turbulence characteristic of the vertical wind was calculated as the ratio of the standard deviation of the vertical wind speed, σ_w , and the friction velocity, u_* , and compared to the parameterization recommended by Thomas and Foken (2002). The calculated σ_w/u_* values deviated less than 30% from theory when the friction velocity was larger than $u_* = 0.1 \text{ m s}^{-1}$.

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The surface roughness was characterized by an estimate of the roughness length z_0 which was derived from

$$\ln \frac{z}{z_0} = \frac{u \cdot \kappa}{u_*} \quad (1)$$

with z : measurement height [m], u : wind speed [m s^{-1}], κ : von Karman constant (= 0.40), and u_* : friction velocity [m s^{-1}].

Evaluation of the atmospheric stability conditions was based on the stability parameter z/L , the ratio of the measurement height and the Obukhov length L ,

$$L = - \frac{u_*^3}{\kappa \frac{g}{T} \overline{w'T'}} \quad (2)$$

with g : gravitational acceleration (9.81 m s^{-2}), T : sonic temperature [K], and $\overline{w'T'}$: buoyancy flux [K m s^{-1}] based on the sonic temperature fluctuations.

Turbulent particle exchange is widely quantified by normalizing the turbulent particle flux F_c with the corresponding particle number concentration c ,

$$v_t = - \frac{F_c}{c} \quad (3)$$

The resulting quantity v_t is a normalized flux, often termed “deposition velocity”, or “transfer velocity” in order to include emission and deposition fluxes equally. The uncertainty of the transfer velocity measurements due to counting statistics, Δv_t , was approximated according to Fairall (1984) by

$$\Delta v_t = - \frac{\sigma_w}{\sqrt{N}} \quad (4)$$

with N : number of counted particles in averaging interval [-]. In 90% of the observations, the uncertainty due to counting statistics was less than 30%. Normalizing the flux helps to evaluate the relevance of the turbulent flux with respect to the ambient particle

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number concentration. However, the concept of a transfer velocity according to Eq. (3) is physically unrealistic and contradicts the gradient approach (Foken, 2008). Strictly, the transfer velocity should be defined in terms of the particle number gradient. Then, it can be considered consistent with flux-profile relationships which relate the particle flux and the particle concentration difference between two heights if the effective turbulent exchange between these two heights is known. We restrict the application of v_t to particle deposition, where it can be described as a reciprocal resistance R_t controlling the particle flux to the surface, $v_t = R_t^{-1}$. The total resistance R_t is a combination of the aerodynamic resistance, R_a , the quasi-laminar sublayer resistance, R_b , the surface resistance, R_c , and the gravitational settling velocity, v_g , thus

$$v_t = \frac{1}{R_t} = \frac{1}{R_a + R_b + R_c} + v_g \quad (5)$$

Applying the resistance analogy, size-resolved particle deposition is calculated using four different parameterizations following the parameterizations presented in Nilsson and Rannik (2001) and Zhang et al. (2001), respectively. The exact parameterizations can be found in these references. Here, we only repeat the different parameterizations of the quasi-laminar sublayer resistance, R_b , which exhibit the most prominent differences: Nilsson and Rannik (2001) used a parameterization of R_b given by Schack et al. (1985):

$$R_{b, NR} = \left(AD^{2/3} \left(\frac{u_*}{z_0} \right)^{1/2} \nu^{1/6} + Bd_p^2 \left(\frac{u_*}{z_0} \right)^{3/2} \nu^{-1/2} \right)^{-1} \quad (6)$$

D is the diffusion coefficient, ν is the kinematic viscosity, d_p is the particle diameter, and A and B are empirical parameters depending on the surface type. Nilsson and Rannik (2001) used $A = 0.4$ and $B = 20$, leading to the best agreement of their measured and calculated transfer velocities. For comparison, we also used the original values $A = 0.19$ and $B = 18.8$ given by Schack et al. (1985) for water at $u_* = 0.44 \text{ m s}^{-1}$.

sonic anemometer. While the ice floe was rotating with respect to true North, the given relative wind directions are constant in the local frame of reference as shown in Fig. 2b.

The roughness length varies mainly between 10^{-2} and 10^{-5} m showing a clear dependence on wind direction due to changes in surface type between sectors. These z_0 values are in good agreement with typical z_0 values reported for water and ice surfaces ranging from 10^{-3} to 10^{-5} m (Foken, 2008). Tjernström (2005) estimated a mean value of 3×10^{-3} m from the Arctic Ocean Experiment 2001, with a similar dependence on wind direction and a span from 10^{-5} to 10^{-2} m. Persson et al. (2002) report a mean value of 4.5×10^{-4} m based on measurements from the Surface Heat Budget of the Arctic Ocean (SHEBA) experiment (Uttal et al., 2002) flux tower, while Andreas et al. (2010) obtain a value of 2.3×10^{-4} m based on the SHEBA dataset. Figure 2b displays an aerial view of the measurement site and the surface properties of the six sectors. The lowest surface roughness is found in sector D, a smooth ice surface lacking large roughness elements. In contrast, the ice surface in sector C contains several pressure ridges and large ice blocks. These roughness elements explain somewhat higher z_0 values in sector C compared with sector D. The rugged floe edges in sectors B and E lead to increased surface roughness in these sectors. However, large fractions of smoother ice and lead surfaces contribute to slightly lower z_0 values in sector E. Sector F provides a wide open lead fetch and exhibits relatively low z_0 values. In contrast, the open lead fetch in sector A is rather small and the high surface roughness is dominated by the edge of the opposite ice floe. Also, it is important to note that widening and closing of the lead over the measurement period introduces some variability in the contribution of open lead, floe edge and ice surfaces to the measurement fetch from the lead direction.

Figure 3 shows the cumulative distribution of the net aerosol number fluxes in the six different sectors. Each trace starts at the lowest measured aerosol flux, and then indicates the fraction of measurements below a certain aerosol number flux in each sector. For example, the value for $F_c = 0 \text{ m}^{-2} \text{ s}^{-1}$ indicates the fraction of deposition dominated flux measurements. Clearly, sectors C and D exhibit a high fraction

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of negative (deposition dominated) flux measurements of 80% and 65%, whereas in sectors B, E and F more or less equal fractions of positive and negative fluxes were observed. In sector A, more than 60% of the observed fluxes were dominated by emission. The highest positive (emission dominated) flux measurements were found in sector B, covering rough ice ridges. The rough surface in sector B also leads to very large deposition dominated flux measurements. Strong deposition dominated flux measurements are also found over the ice floe in sectors C and D. These observations cannot be explained by different particle number concentrations, but are also found in the corresponding transfer velocities summarized in Table 1.

For reference, Table 1 also presents the number of 30 min periods dominated by deposition (N_{dep}) or emission (N_{em}), the median values of wind speed and particle number concentration as well as friction velocity u_* and roughness length z_0 . The median values of u_* and z_0 , wind speed and particle number concentration within each sector are very similar during emission and deposition dominated periods, except for sector A. Here, high particle numbers (147 cm^{-3}) and low wind speeds (1.2 m s^{-1}) are observed during an emission dominated period on 18 August, and much lower particle numbers (9 cm^{-3}) yet higher wind speeds (3.1 m s^{-1}) during a deposition dominated period on 25 August. The median values of the transfer velocity range from 0.27 mm s^{-1} to 0.68 mm s^{-1} during deposition dominated periods. The magnitude of the observed deposition velocities, v_t (dep), is in general agreement with earlier estimates of aerosol fluxes over snow and ice surfaces. Ibrahim et al. (1983) report aerosol deposition velocities of $v_t = 0.39 \text{ mm s}^{-1}$ under stable stratification and $v_t = 0.96 \text{ mm s}^{-1}$ under unstable conditions using ^{35}S tagged ammonium sulfate particles. Duan et al. (1988) observed an average aerosol deposition velocity of particles in the diameter range from 150 to 300 nm of $v_t = 0.34 \text{ mm s}^{-1}$ over a partially snow covered field using optical particle counters. Bergin et al. (1995) derived aerosol sulfate deposition velocities ranging from 0.23 mm s^{-1} to 0.62 mm s^{-1} at Summit, Greenland, using surrogate surfaces and impactor data. Grönlund et al. (2002) report median deposition velocities of 0.33 mm s^{-1} over a smooth snow-covered area in Dronning Maud Land, Antarctica. Nilsson and

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Rannik (2001) measured aerosol number fluxes by eddy covariance in a similar setting in the high Arctic. They report median deposition velocities $v_t = 0.26 \text{ mm s}^{-1}$ over smooth ice surfaces, and v_t ranging from 0.40 to 0.73 mm s^{-1} over open lead surfaces. However, the footprints of their flux measurements were considerably larger than in this study, and also possibly a mixture of open lead and ice surfaces. It is interesting to note that in the current measurements the highest particle number concentrations were found when the measurement is influenced by the open lead, in sectors A and F (Table 1).

In order to compare the aerosol number flux above the open lead and the ice surface, Fig. 4a presents the median aerosol number fluxes in 10° wind direction bins over the entire measurement period. In addition, the time fraction of emission dominated periods for different relative wind directions is shown in Fig. 4b. One can easily distinguish two different regimes: for relative wind directions from sectors A and F, i.e. measurements dominated by the open lead, slightly positive median fluxes indicate aerosol emission, and a high fraction of emission periods can be found. In contrast, for relative wind directions from sectors C and D, i.e. the ice-floe dominated fetch, only very few emission dominated periods are observed (Fig. 4b), and the median flux values are mostly negative indicating net deposition (Fig. 4a). Sectors B and E (grey shading) exhibit a transitional behavior.

3.2 Aerosol flux observations – temporal variability

In the following, the temporal variability of the measured fluxes will be discussed. Due to the very low flux estimates, a comparison of cumulative fluxes of momentum, buoyancy and particle number concentration is used. This means that emission fluxes will increase the previously accumulated flux value and deposition fluxes will decrease it. Thus, a positive slope indicates emission, a negative slope indicates deposition, and the steeper the slope, the stronger the flux.

Figure 5 shows the wind speed and direction, the stability parameter z/L , the particle number concentration, and the cumulative fluxes of momentum, buoyancy, and particle

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number concentration from 26 to 29 August. During this four-day period, the lead started to freeze over, and it was continuously covered with a thin layer of ice starting from 27 August. In the night from 26 to 27 August emission dominates and there is weak upwards flux of particles, while particle deposition dominates the flux in the afternoon and evening, especially on 28 and 29 August. However, the particle fluxes are very low throughout 27 August while buoyancy and momentum fluxes exhibit a clear increase in magnitude. On this day, the fetch was mostly over the lead in sector F. However, as noted above, the lead was covered with a thin layer of ice at this time.

The observations discussed above corroborate our findings that the open lead indeed behaved as a source of aerosol particles under certain conditions, yet there is no clear correlation with wind speed or momentum flux. This supports earlier suggestions (e.g. Leck and Bigg, 1999; Leck et al., 2002) that the open lead particle source is not exclusively driven by wind (such as the bubble-bursting mechanism at open sea). Possible non-wind driven sources of bubbles are the release of bubbles trapped in melting sea ice, their transport to the surface by increased turbulence caused by supercooling conditions (Grammatika and Zimmerman, 2001), or bubbles due to respiration of phytoplankton (e.g. Johnson and Wangersky, 1987). Further investigation of alternative bubble source mechanisms can be found in Norris et al. (2010).

3.3 Deposition parameterizations and net flux observations

The net flux estimates derived from our eddy covariance measurements reflect the combined effect of emission and deposition mechanisms on the turbulent vertical exchange of particles. In order to obtain the emission flux, the deposition must be subtracted from the measured net flux. As mentioned earlier, a variety of size-resolved particle deposition parameterizations exist which can be used to estimate the deposition. However, the uncertainties introduced by the parameterizations add to the measurement uncertainties and require careful consideration before robust conclusions can be drawn.

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Figure 6a presents the cumulative particle number flux as measured from 18 to 31 August, and the cumulative deposition fluxes derived from four different particle deposition parameterizations. The large data gap from 21 to 24 August is due to heavy riming on the sonic anemometer. When comparing the temporal behavior of the parameterizations, two different pairs of deposition parameterizations can be found. Nilsson and Rannik (2001) use the original parameterization of the quasi-laminar sublayer resistance R_b by Schack et al. (1985), but with different empirical parameters A and B (cf. Eq. 5). Replacing this formulation with a parameterization suggested by EMEP (2003) for gas-phase species yields deposition patterns similar to the parameterization by Zhang et al. (2001), however different absolute values.

On 18 August, all parameterizations suggest very low particle deposition indicating that the net flux estimates are close to “true” emission values. On 24 August, a deposition period starts. The NR (Nilsson and Rannik) and NR/S (Nilsson and Rannik/Schack) parameterizations show two strong deposition periods during the nights from 25/26 and 28/29 August. The first deposition dominated period can also be found in the observations, whereas the second period is qualitatively different from the observed net fluxes. This leads to a strong deviation of the cumulative flux values of the measurement and the NR parameterization at the end of the considered period.

Obviously, none of the deposition parameterizations are intended to reproduce particle emission events. Therefore, in Fig. 6b emission periods are neglected, while only deposition dominated periods are taken into account and added to the cumulative flux. The shaded areas indicate deviations of 25% (light grey) and 50% (dark grey) from the observed net flux. On 20 August, the Zhang parameterization is slightly larger than the observed flux, while all other parameterizations are smaller than the observed flux. However, during the period starting on 24 August, all parameterizations tend to yield smaller flux values than observed. Since the parameterizations were not designed specifically for our conditions, we are not entirely surprised to find deviations of the parameterizations; however, if we postulate some opposing emission fluxes, we would rather expect the models to overestimate in comparison with the observations.

Qualitatively, the Zhang parameterization tracks the observed net flux best. The EMEP parameterization clearly gives flux values much lower than observed. This may be explained by the fact that the EMEP parameterization is originally designed for gas-phase species.

From this evaluation, there is no individual parameterization that can be considered the “best” description of the deposition flux. All parameterizations show periods when they agree with, and periods when they deviate from, the observed fluxes. However, it is not possible to attribute these deviations to measurement uncertainties, or deficiencies of the parameterizations. In general, the EMEP parameterization may be considered a low estimate of particle deposition.

3.4 Potential contribution of vertical aerosol fluxes to the airborne particle burden

In order to evaluate the significance of direct particle emission and deposition, and its potential contribution to the atmospheric aerosol burden, the change in particle concentration due to turbulent particle emission and deposition fluxes will be considered. The goal is to obtain a rough estimate of the order of magnitude of the aerosol flux contribution to changes in particle number concentrations – can we expect to explain 1% or 50% or even more of the aerosol number variability just by vertical turbulent transport? For these calculations, the measured net fluxes were used to include both emission and deposition processes. In this simplified thought experiment we consider a closed box. The top boundary is given by the mixing height. We neglect horizontal advection because we want to examine the local effect of vertical particle transport by turbulence only. We also neglect processes such as new particle formation and chemical reactions that potentially affect the particle number concentration. Thus, particles enter and leave the box through aerosol emission and deposition at the surface/atmosphere boundary only. We evaluate the changes of particle number concentration over a time interval of 30 min, i.e. the averaging interval of the eddy covariance calculations. At this time resolution, turbulent mixing will reduce concentration differences with height.

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concentration cannot be reproduced by $\Delta_{f,MLH}$. In this case, the best estimate of the mixing layer height is 10 m.

Overall, the evaluation of the aerosol flux contribution to changes in particle number concentrations can only be considered a rough estimate of the order of magnitude of this factor. In particular, the mixed layer depths represent absolute minimum depths over which concentration changes should be evaluated; turbulent mixing will extend above these levels but become increasingly weak. Nevertheless, we find the direct impact of the turbulent particle flux on the atmospheric particle concentration to be minor. In about 85% of the evaluated cases, the flux-derived particle concentration change $\Delta_{f,25}$ (assuming a typical mixing layer height of 25 m, and a 25% area fraction of open leads) is less than $1 \text{ cm}^{-3} \text{ h}^{-1}$, whereas more than 75% of the observed particle concentration changes Δ_c are greater than $1 \text{ cm}^{-3} \text{ h}^{-1}$. While the shape of the cumulative frequency distributions of $\Delta_{f,25}$ and Δ_c are similar, the absolute magnitude of the particle concentration change differs by a factor of about 10 (cf. Table 2). Therefore, additional processes such as horizontal transport, new particle formation, and chemical transformations will strongly affect the particle number concentration.

4 Conclusions

We have successfully carried out direct eddy covariance measurements of particle number fluxes on an ice floe in the central Arctic Ocean and found episodic aerosol emission from open leads. Simultaneous and independent gradient measurements of particle concentrations (Orsini et al., 2010) corroborate our finding that open leads can indeed act as particle sources in the Arctic Ocean. Overall, the direct contribution of the open lead particle emissions to the atmospheric aerosol number concentration appears to be of minor importance, and can only explain a few percent of the observed particle number variability. Additional processes such as advection, chemical transformation and degradation, or vertical mixing from aloft in the upper layers of the marine boundary layer seem at a first approximation to have a significant impact on

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atmospheric particle numbers in the central Arctic (Bigg et al., 1996, 2001; Leck and Persson, 1996). Unfortunately, no information about the size of the emitted particles is available from our direct flux measurements. Thus, it remains unclear if open leads are a significant source of aerosol mass to the Arctic boundary layer. Moreover, we only begin to understand what happens to the emitted particles in the atmosphere. It has been put forward that aerosol particles emitted from open leads in the Arctic are enriched in organic compounds from the marine surface microlayer (e.g. Leck and Bigg, 2005; Bigg and Leck, 2008; Matrai et al., 2008). These gel-like substances found in the aerosol were postulated to have properties consistent with algal and bacterial exopolymer secretions or marine microgels (Decho, 1990). According to Verdugo et al. (2004), the marine microgels span the whole size spectrum from colloidal-size nanogels containing single macromolecules entangled to form single-chain networks to micrometer-size gels (loose matrix associated with the aggregates or granular structures) that can aggregate to tight capsules reaching several 100 μm in diameter. The assembly and dispersion of macromolecules can be affected by environmental parameters, such as UV-B radiation (280–320 nm) dispersing or inhibiting microgel formation, and/or pH and temperature inducing microgel volume phase changes (Orellana and Verdugo, 2003; Chin et al., 1998). Thus, one can speculate that degradation and break-up is a potential atmospheric fate of the open lead-derived aerosol. As suggested by Leck and Bigg (2010) this can lead to a large number of smaller daughter particles derived from a small number of large parent particles emitted from the open lead.

Finally, the melting of sea ice in a changing climate will further increase the fraction of open leads in the Arctic pack ice, and potentially increase the relevance of the open lead particle source. It is very likely that these particles will then play a role as cloud condensation nuclei, and thus provide a direct feedback to the regional Arctic climate.

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search at Stockholm University. ASCOS was made possible by funding from the Knut and Alice Wallenberg Foundation, the Swedish Research Council and the DAMOCLES European Union 6th Framework Integrated Research Project. The Swedish Polar Research Secretariat (SPRS) provided access to the icebreaker *Oden* and logistical support. We are grateful to the Swedish Polar Research Secretariat logistics staff and to Oden's captain Mattias Peterson and his crew. ASCOS is an IPY project under the AICI-IPY umbrella and is an endorsed SOLAS project.

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Table 1. Median values of wind speed, particle number concentration, friction velocity u_* , roughness length z_0 , and transfer velocities v_t , respectively, for six wind sectors with different surface characteristics (L: lead, IR: ice ridge, I: ice floe). N_{dep} is the number of deposition dominated 30 min periods in each sector, N_{em} of emission dominated periods.

sector	from	to	N_{dep}	N_{em}	wind speed	particle conc	u_*	z_0	v_t (dep)	surface
	°	°	–	–	m s^{-1}	cm^{-3}	m s^{-1}	m	mm s^{-1}	
A	15	70	15	24	1.35	138	0.09	2.2E-03	0.41	L/IR
B	70	110	30	27	3.32	50	0.23	6.6E-03	0.51	IR
C	110	190	56	14	3.82	38	0.17	4.0E-04	0.68	I
D	190	230	31	16	3.53	43	0.11	1.7E-05	0.44	I
E	230	305	27	27	2.22	12	0.12	1.9E-03	0.56	IR/ I /L
F	305	15	39	36	3.13	71	0.12	1.0E-04	0.27	L

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Table 2. Percentiles of changes in particle number concentration per hour as expected from measured aerosol number flux (expected), and as observed from direct particle number measurements (observed).

percentile %	expected			observed		
	emission [cm ⁻³ h ⁻¹]	deposition [cm ⁻³ h ⁻¹]	total [cm ⁻³ h ⁻¹]	emission [cm ⁻³ h ⁻¹]	deposition [cm ⁻³ h ⁻¹]	total [cm ⁻³ h ⁻¹]
5	0.1	-3.7	-2.8	0.4	-35.2	-24.5
10	0.1	-2.5	-2.0	0.7	-24.1	-16.7
50	0.6	-0.8	-0.2	6.6	-5.3	-0.3
90	2.5	-0.1	1.3	24.2	-0.6	15.5
95	3.0	-0.1	2.1	34.7	-0.3	23.2

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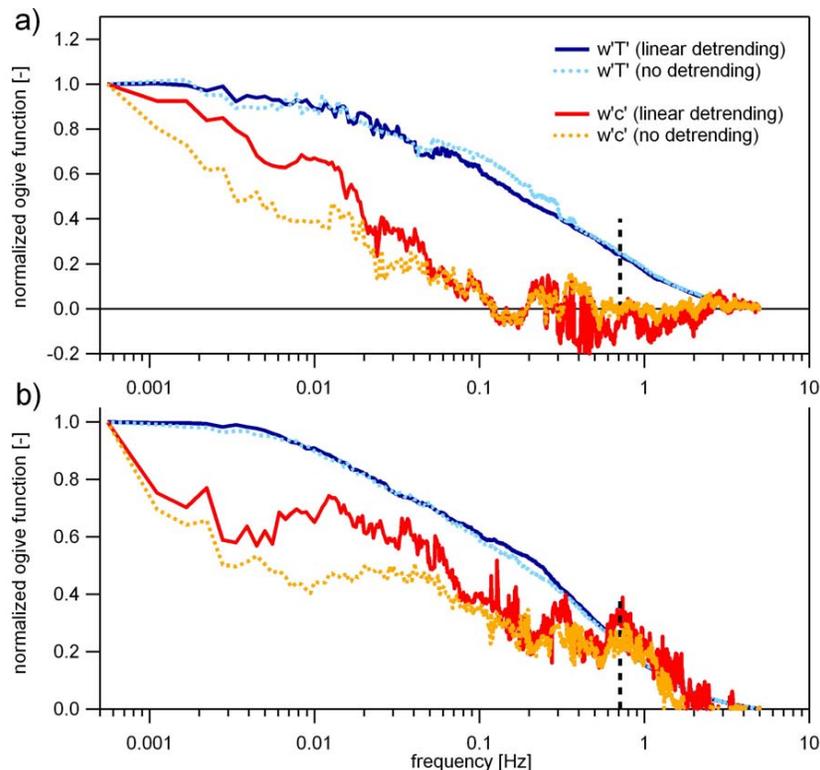


Fig. 1. Normalized ogive functions of vertical wind vs. sonic temperature ($w'T'$, blue) and vertical wind vs. aerosol number concentration ($w'c'$, red). **(a)** Median ogives from 19 August, 22:00–20 August, 19:30. **(b)** Median ogives from 24 August, 00:00–21:30. Broken lines in light colors show ogives without linear detrending.

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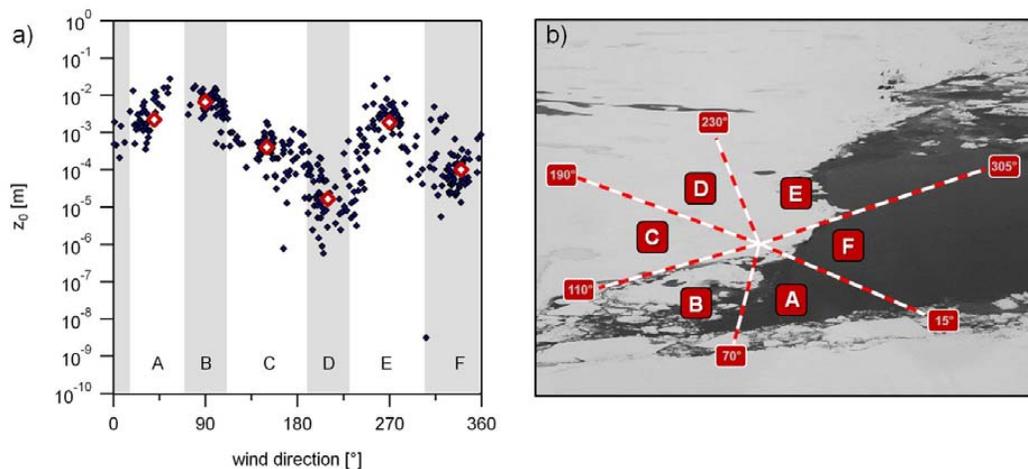


Fig. 2. (a) Roughness length z_0 in six wind direction sectors A–F; red diamonds represent median z_0 values in each wind sector. (b) Aerial view of the measurement site and overlay of the six wind sectors.

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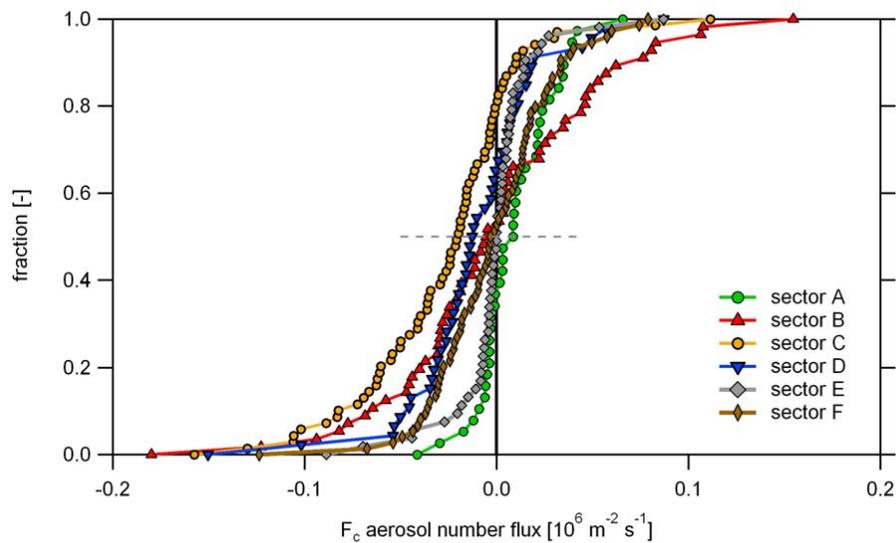


Fig. 3. Cumulative distribution of aerosol number fluxes in six different sectors. Flux value at fraction of 0.5 is the median aerosol number flux.

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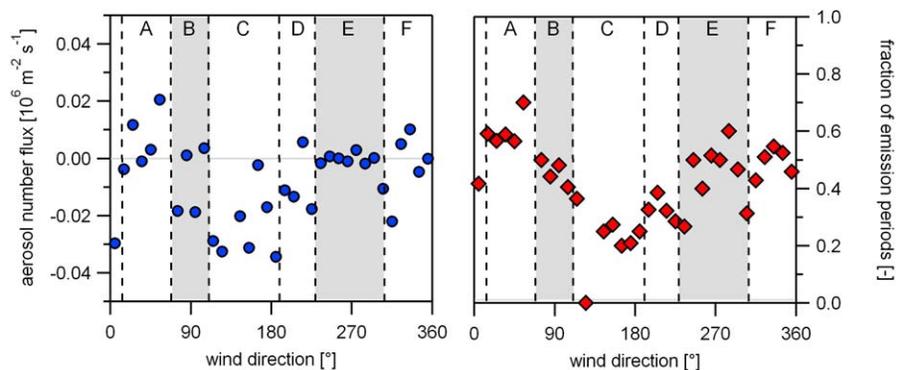


Fig. 4. (a) Median aerosol number fluxes in 10° wind direction bins. (b) Time fraction of emission episodes averaged over 30° wind bins and plotted every 10° . Grey shading indicates the transitional wind sectors B and E.

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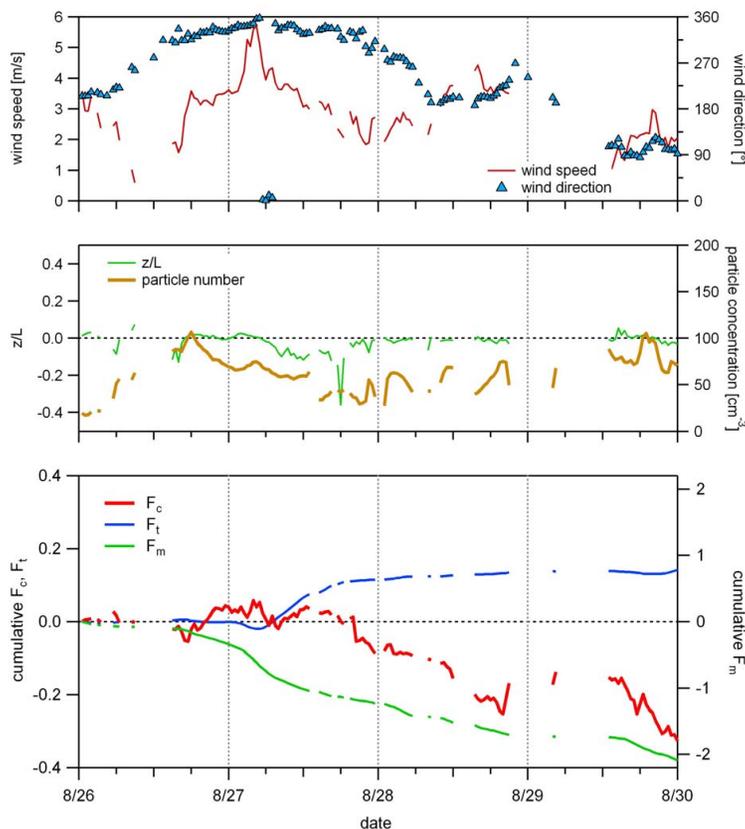


Fig. 5. Top panel shows wind speed (red line) and relative wind direction (triangles) from 26 August through 29 August, 2008; middle panel shows stability parameter z/L (green line) and particle number concentration (orange line); bottom panel shows cumulative fluxes of momentum (F_m , green), buoyancy (F_b , blue), and particle number (F_c , red).

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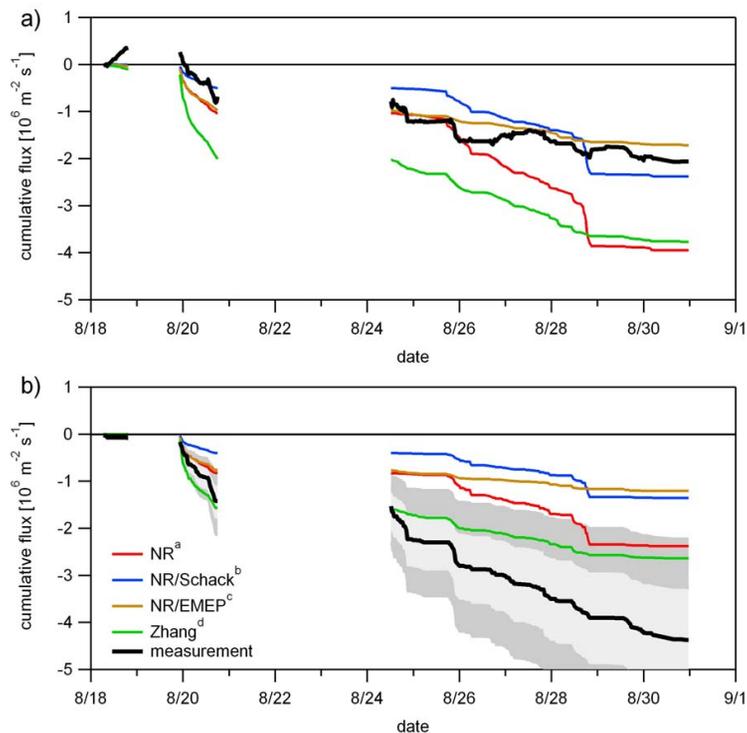


Fig. 6. (a) Measured cumulative particle number flux from 18 to 31 August (black line), and cumulative deposition fluxes derived from four different particle deposition parameterizations according to ^aNilsson and Rannik (2001), ^bSchack et al. (1985), ^cEMEP (2003), ^dZhang et al. (2001). (b) Same as (a), but only during periods when particle deposition was observed. Shaded areas indicate deviations of 25% (light grey) and 50% (dark grey) from the observed deposition flux.

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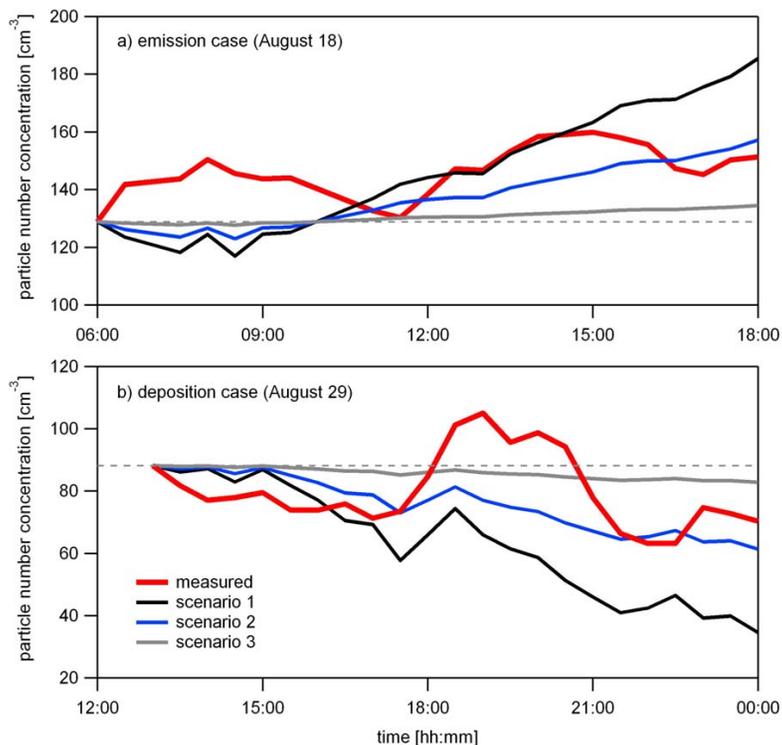


Fig. 7. Temporal change of the particle number concentration as measured and theoretically expected due to turbulent aerosol flux assuming three different scenarios of mixing layer height and area fraction of the open leads. More details in the text.