

Review of Aerosol Research at the Finnish Antarctic Research Station Aboa and its Surroundings in Queen Maud Land, Antarctica

*Aki Virkkula*¹, *Eija Asmi*², *Kimmo Teinilä*², *Anna Frey*², *Minna Aurela*², *Hilkka Timonen*², *Timo Mäkelä*², *Antti Samuli*², *Risto Hillamo*², *Pasi P. Aalto*¹, *Sheila Kirkwood*³ and *Markku Kulmala*¹

¹Department of Physics, University of Helsinki, Finland

²Finnish Meteorological Institute, Research and Development, Helsinki, Finland

³Swedish Institute of Space Physics, Kiruna, Sweden

(Received: May 2009; Accepted: September 2009)

Abstract

All main aspects of aerosols – chemical composition, physical properties, and processes from their origin to removal from the atmosphere to the snow surface – have been studied at Aboa since December 1997. This paper reviews main results of the the work done and presents some selected new results. The travel time from the ocean proved to be an important factor determining the chloride-to-sodium ratio. The contribution of organics may be significant in the Aitken mode particles. Aerosol formation and growth has been observed during all summer campaigns. An automatic particle counter system was run for more than three years using power from solar cells and wind generators, with breaks mainly due to power production. Wind radar data suggests that intrusions from higher atmospheric levels affect clearly surface concentrations of both particles and trace gases.

Key words: aerosols, nucleation, size distribution, chemical composition, light scattering, radar, Antarctica, Aboa

1. Introduction

1.1 General aspects of aerosol research in Antarctica

Aerosols are particles ranging from the size of a large molecule, approximately 1 nm, to coarse supermicron sea-salt and soil dust particles that may be larger than 10 micrometers in diameter. The smallest ones are formed by nucleation of low-vapor-pressure gases, such as sulfuric acid. The fresh nucleation-mode particles, smaller than about 10 nm, grow both by condensation of similar types of gases and by coagulation with other particles into the Aitken mode, at approximately 50–80 nm in diameter, and finally through cloud processing into the accumulation mode at about 100–200 nm. The particles may thus grow from large molecule clusters into sizes that are climatically important, i.e., they scatter or absorb radiation and may act as cloud condensation nuclei.

Aerosol measurements in the cleanest continent, Antarctica, are important for a number of reasons. First they are needed for assessing aerosol climatic effects. Aerosols affect climate both directly by scattering and absorbing radiation and indirectly by their effects on clouds. Although the direct radiative effects are clearly better known than the indirect effects, also their value over Antarctica is much more uncertain than over oceans or areas with vegetation. Even slightly absorbing aerosols may exert a positive forcing, i.e., heating of the atmosphere over regions of high surface albedo, such as the glaciers of Antarctica (e.g., *Haywood and Boucher, 2000*).

Aerosol measurements in Antarctica provide information on natural background concentrations and processes such as particle formation and growth. One of the most important open questions is the role of different nucleation mechanisms (e.g., *Kulmala et al., 2006* and references therein; *Kulmala et al., 2004*). Another open question is how aerosol particles grow after their formation by nucleation, and whether they grow to size ranges where they are able to act as cloud condensation nuclei and further to optically active sizes. Particle formation and growth are therefore also linked with aerosol radiative effects. High-time-resolution size distribution measurements also yield information on gas-phase concentrations since the size-dependent particle growth rate depends on the concentration of the condensing gas (*Kulmala et al., 2005*).

Another important reason for studying Antarctic aerosols is estimating past atmospheric chemistry from ice core samples. To do this it is necessary to establish an unambiguous link between chemical species in the air and snow (*Bergin et al., 1998*). Further, when interpreting the ion chemistry records from deep ice cores drilled at a single site, it is essential to understand the spatial distribution of chemical species in the snow, and the factors controlling the atmospheric transport and deposition of chemical species. Measurements aimed at increasing understanding the interactions between air and snow have been conducted at several coastal sites (e.g., *Wagenbach et al., 1998; Jourdain and Legrand, 2002; Rankin and Wolff, 2003*) and at high plateau sites such as South Pole (e.g., *Bergin et al., 1998; Arimoto et al., 2004; Davis et al., 2004*) and Dome Concordia (e.g., *Udisti et al., 2004*). Large spatial variations have been detected in the physical and chemical properties of the surface snow cover (e.g., *Bertler et al., 2006; Kärkäs et al., 2005*).

1.2 Aerosol research at Aboa

All main aspects of aerosols – chemical composition, physical properties, and processes from their origin to removal from the atmosphere to the snow surface – have been studied at the Finnish Antarctic research station Aboa (73°03'S, 13°25'W) in Queen Maud Land, Antarctica, since December 1997. The station is located on the nunatak Basen 480 m above sea level and approximately 130 km south of the ice edge of the open ocean of Weddell Sea. The general objective of the aerosol research has been to study aerosol formation and other processes in a pristine and clearly natural environment to provide comparison data for measurements made in the polluted and vegetation-covered continents. We have tried to find answers to detailed questions such

as 1) what are the main factors affecting particle concentration, 2) where does the particle formation actually take place, close to the surface or higher up in the atmosphere, above the sea or far away in the middle of the continent, 3) what are the chemical compounds that lead to the growth of the aerosol, 4) do the particles grow into climatically relevant sizes, and 5) how does particle chemical composition change when marine aerosols get transported to inland and how does this relate to the coast-inland gradient in the chemical composition of snow.

The work has been done as a close cooperation between the aerosol research group of the Finnish Meteorological Institute's (FMI) Air Quality Research and the aerosol dynamics group of University of Helsinki (UHEL) Department of Physics. The research on snow chemical composition was a cooperation between FMI and UHEL Division of Geophysics.

Since the FINNARP 2007 expedition cooperation also with the purely meteorological project "Antarctic Meteorology", a consortium of FMI and UHEL meteorological research, has been established to provide deeper understanding of the transport of air masses to the measurement site.

1.3 International connections

Cooperation with other groups making atmospheric research in Queen Maud Land has taken place in some summer campaigns. Dry deposition was studied at the station in cooperation with the Stockholm University in January 2000 (*Grönlund et al.*, 2002). In the austral summer of 2006/2007 Aboa served as one of the ground-based stations against which airborne aerosol measurements were compared during the German-Swedish-Japanese AGAMES (Antarctic Trace Gas and Aerosol Airborne Measurement Study (www.pa.op.dlr.de/aerosol/agames)) campaign. The aerosol optics measurements made in the summers 2006/2007 and 2007/2008 were part of the International Polar Year (IPY) project POLAR-AOD: a network to characterize the means, variability, and trends of the climate-forcing properties of aerosols in polar regions (IPY EoI ID No: 299). In the summers 2006/2007 and 2007/2008 there was a cooperative project with the Swedish Institute for Space Research (IRF, Kiruna) concerning air mass vertical transport. This will be discussed further below. In summer 2007/2008 also trace gas concentrations, for instance surface ozone concentration were measured at Aboa. These data were a contribution to the autonomous surface ozone monitoring network in Dronning Maud Land, Antarctica, a project coordinated by the British Antarctic Survey (*Bauguitte et al.*, 2009).

1.4 Objective of the present work

In this paper we will present a review of the the aerosol research done at Aboa and its surroundings in the years 1997–2008. The methods and the research infrastructure will be presented. Both already published and some selected non-published results of both aerosol chemical composition and physical properties and processes will be presented.

2. Measurements

The first aerosol measurements at Aboa were conducted during the FINNARP 1997 expedition in December 1997 – February 1998 in a small and light cottage made of fibre glass that was just large enough for small-scale sampling but not for additional instruments and activities (*Teinilä et al.*, 2000; *Kerminen et al.*, 2000). The first measurements included filter sampling with 2-stage filter samplers, a 12-stage low pressure impactor and a condensation particle counter (CPC).

In order to make larger campaigns and continuous measurements possible the infrastructure was developed by designing and building a special laboratory container in 1999. It was transported and installed at Aboa during FINNARP 1999. Aerosol measurement campaigns have been conducted with various instrument setups in the Antarctic summers approximately every second year since then.

The most comprehensive setup was used during 1999/2000 and 2000/2001 summers. For details see *Koponen et al.* (2003). Filter samples were taken using a 2-stage low-volume sampler and a high-volume (HV) sampler. Two multistage impactors were used to obtain chemical size distributions. Aerosol physical properties were studied in detail. Number size distributions in the size range 3 nm – 20 µm were measured using a combination of a twin-differential mobility analyser (TDMPS, 3–800 nm) and an aerodynamic particle sizer (APS, 0.5–20 µm). Light scattering and absorption coefficients were measured with a nephelometer and an aethalometer, respectively.

Until 2003 only summertime aerosol measurements were conducted. In order to study annual cycles of aerosol concentrations an autonomous wind-and-solar-powered particle counter system was designed, built and installed in the laboratory container (*Virkkula et al.*, 2004). The actual particle counting was done using a TSI Model 3007 CPC and a Grimm Model 1.108 optical particle counter to get the particle number size distribution in the size range 0.3–20 µm. The system was operational from December 2003 through January 2007. It was attached to the Aboa automatic weather station that made it possible both to control the system and to download data via satellite.

In summer 2004/2005 even smaller particles were studied. For the first time in Antarctica, an air ion spectrometer (AIS) was run. The AIS measures charged particle size distributions in the size range 0.34–40 nm. In addition, a DMPS was used to get the total number concentration in the size range 10–700 nm (*Virkkula et al.*, 2007). During the same season measurements were conducted also outside of the nunatak Basen. A small portable battery-powered particle counter and filter sampler system was built that contained an optical particle counter (OPC, Grimm 1.108) and a CPC (TSI, Model 3007). The goals of the mobile measurements were to study 1) whether the high number concentrations during nucleation events are confined to a small region around Aboa or whether it is a phenomenon observable at a larger geographical range; 2) the sea-salt particle gradient at winds blowing from the sea; 3) the size distribution and transport of dust from the nunatak Basen during high wind speeds. The OPC has an option for filter sampling at a flow rate of 1.2 LPM on a 47 mm diameter Teflon filter. This was used to

get aerosol chemical composition. The mobile system was run at several locations at distances starting from less than 10 km from the coast up to about 300 km from the coast (Table 1). The system was run unequal times at the sampling sites, from about 4 hours to 3 days. The filter samples were stored in Petri slides and analyzed for major inorganic ions at FMI. Even though the mass accumulated on the filters was low in most samples it proved to be above the detection limit for the major ions sulfate, MSA, sodium, and chloride.

Table 1. Locations and sampling times of the mobile aerosol measurements in summer 2004/2005. The latitudes and longitudes are presented as degrees and decimals of degrees. (Negative latitude: south, negative longitude: west)

Sample nr	Site nr	Latitude	Longitude	D = Distance from sea ice edge		
		deg.decimals	deg.decimals	D (km)	Start date & time	End date & time
1	1	-72.532	-16.559	5	20/12/2004 20:17	21/12/2004 09:52
2	2	-72.752	-15.500	48	21/12/2004 14:11	22/12/2004 12:11
3	2	-72.752	-15.500	48	22/12/2004 13:05	22/12/2004 21:00
4	3	-72.752	-14.194	87	02/01/2005 11:01	02/01/2005 14:36
5	4	-73.087	-13.520	122	02/01/2005 22:02	03/01/2005 08:27
6	5	-74.464	-11.539	272	04/01/2005 21:32	05/01/2005 14:17
7	5	-74.464	-11.539	272	05/01/2005 14:46	06/01/2005 13:16
8	6	-73.802	-12.238	203	06/01/2005 22:15	07/01/2005 09:35
9	7	-73.369	-13.135	150	07/01/2005 14:30	10/01/2005 14:09
10	8	-73.041	-13.401	123	31/01/2005 11:50	01/02/2005 10:10

In summer 2006/2007 there was another comprehensive campaign at Aboa. Chemical composition was determined from multi-stage impactor and filter samples, scattering was measured with a nephelometer and absorption with a Particle Soot Absorption Photometer (PSAP). Aerosol optical depth (AOD) was measured with a portable sunphotometer. Air ion spectra were measured with an AIS and total number size distributions with a TDMPS (*Vartiainen et al., 2007*). As a new instrument a custom-made hygroscopicity tandem differential mobility analyzer (HTDMA) was run in Antarctica for the first time. The goal was to study the hygroscopic growth of particles.

In summer 2007/2008 the volatility of aerosols in the size range 10–700 nm were measured with a custom-made volatility differential mobility particle sizer (VDMPS). Aerosol scattering and absorption were measured with a nephelometer and a PSAP, respectively, and AOD with a portable sunphotometer. Filter and impactor samples were taken for aerosol chemical composition. In addition, gas phase measurements were conducted with an ozone monitor (Environnement S.A., Model O3 42M).

The chemical composition of the filter and impactor samples has been analyzed at FMI. For all campaigns the major inorganic and organic ion concentrations have been determined using ion chromatography. For some campaigns elemental and organic Carbon (EC/OC) has been determined and for one campaign (2006/2007) the concentration of water-soluble organic carbon.

Analyses of transport routes of air masses to the sampling site have been done using backtrajectory calculations. A new insight into the air mass transport was obtained when the Swedish Institute for Space Research (IRF, Kiruna) installed a new instrument

MARA (Moveable Atmospheric Radar for Antarctica) in the vicinity of the aerosol laboratory in January 2007. This instrument measures 3D wind fields and turbulence from some hundreds of meters up to several tens of kilometers altitude. MARA is a 54.5 MHz wind-profiler type radar. The radar antenna consists of 48 dipole-antennas, arranged in three square arrays, each with 16 dipoles spaced 4 m apart. Peak transmitter power is 20 kW. The height resolution of the measurements in the lower troposphere is 300 m, and in the upper troposphere, 600 m. Each profile represents an average over 30 seconds, with measurements repeated each 2 minutes. For further details see *Kirkwood et al.* (2007 and 2008).

3. Results and discussion

3.1 Aerosol chemical composition

Aerosol chemical composition has been studied by taking 1 or 2-stage filter samples and multi-stage impactor samples. The samples have been analyzed for major inorganic and organic ion concentrations at FMI. *Teinilä et al.* (2000) and *Kerminen et al.* (2000, 2001) presented the first detailed Antarctic aerosol chemical size distributions, including modal structure of major ionic compound from 40 nm to 15 μm . In these papers it was shown that the sea salt concentrations are clearly lower at Aboa than at coastal stations such as Neumayer and Halley. It was also shown that the aerosol cation-to-anion ratio has a minimum at about 100 nm, these particles were the most acidic.

By comparing chloride-to-sodium ratios it was shown that there was a clear loss of chloride from sea salt particles. It was also shown that the anions responsible for replacing the seasalt chloride are sulfate, methanesulfonate and nitrate. *Virkkula et al.* (2006a) analyzed the chloride loss further by comparing it with backtrajectories and showed that the chloride loss increases as the average transport time from the ice edge increases.

These results were confirmed with the analyses of the mobile filter sampler that was deployed at several distances from the coast in January 2005. Figure 1 shows the concentrations of the major ionic compounds at various distances from the open ocean in mobile filter sampling. The exponential functions were fit to data excluding the first two samples near the shelf ice edge because during their sampling wind blew from the south and the goal of getting a high sea-salt particle concentration near the coast did not succeed.

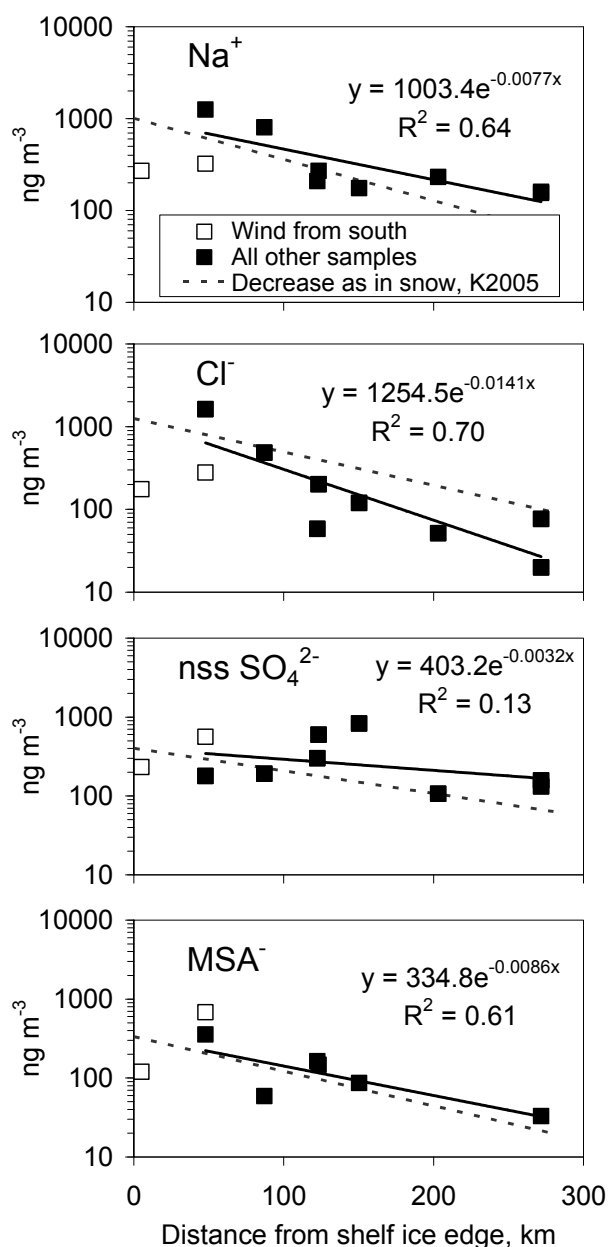


Fig. 1. Major ionic compound concentrations at various distances from the open ocean in mobile filter sampling in January 2005. The exponential functions were fit to data excluding the first two samples near the shelf ice edge. For comparison the exponential functions obtained by *Kärkäs et al.* (2005) from snow samples are presented.

The chloride loss becomes interesting when it is compared with the results obtained from snow samples. *Kärkäs et al.* (2005) took snow samples at various distances from the sea ice towards the polar plateau at locations close to those where the mobile aerosol measurements were conducted. The exponential fits ($C(\text{distance from ice edge}) = C_0 \exp(-k \times \text{distance from ice edge})$) made by *Kärkäs et al.* (2005) were used in Figure 1 so that the C_0 values were taken from the fits to the mobile aerosol measurements and the k factors from Table 4 of *Kärkäs et al.* (2005). Both in aerosol and in snow the concentrations ions originating from the sea – sodium, chloride, nss sulfate, and methane sulfonate (in Antarctica, the source of both MSA and nss sulfate is

oxidation of DMS emitted by phytoplankton) – decrease exponentially with increasing distance from the sea ice edge (Kärkäs *et al.*, 2005; Bertler *et al.*, 2006).

There is a clear difference in the decrease, however. The chloride concentrations decrease faster in aerosol than in snow. When the ratio of chloride to sodium is plotted as a function of distance from the ice edge this becomes more evident. The ratio in snow remains fairly constant – which is also in agreement with the Antarctica-wide study of Bertler *et al.* (2006) – and actually does not decrease with distance from the sea whereas the ratio in aerosol decreases clearly (Fig. 2a). The decrease of the ratio in the mobile system samples is supported by that obtained from the comparison of the chloride-to-sodium ratio and the transport time from the shelf ice edge to Aboa (Fig. 2b). The reason of the discrepancy of the snow and aerosol data remains to be studied.

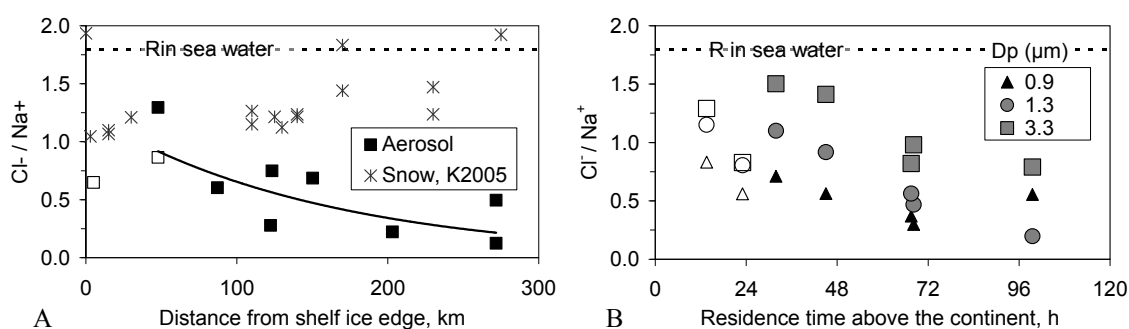


Fig. 2. Chloride-to-sodium mass ratios in aerosol and snow. (A) The ratio at various distances from the open ocean in mobile filter sampling in January 2005 and snow samples of Kärkäs *et al.* (2005). (B) The ratio in coarse mode aerosols as a function of trajectory transport time from the shelf ice edge to Aboa (modified from Virkkula *et al.*, 2006a). R in sea water = 1.7982.

Secondary Organic Aerosols (SOA) are an important and a lot-studied field of aerosol science. However, in Antarctica their role is not yet well known: Virkkula *et al.* (2006c) presented the first Antarctic organic aerosol concentrations. The samples were taken using a high-volume sampler with quartz filters that were analyzed using a thermal-optical organic/elemental carbon analyzer. The contribution of the OC to the sum of all analyzed species was approximately 10–13%. The comparison of DMPS-derived size distributions with mass size distributions calculated from major ion compounds analyzed from 12-stage impactor samples (Virkkula *et al.*, 2006a) suggest that the contribution of SOA may be significant in the Aitken and accumulation modes, and thus contributing also to aerosol growth.

3.2 Aerosol physical properties and processes

3.2.1 The largest campaign in summers 1999/2000 and 2000/2001

The measurements at Aboa in summers 1999/2000 and 2000/2001 were the first detailed analyses of particle size distributions in the size range 3–700 nm, in Antarctica (Koponen *et al.*, 2003). Park *et al.* (2004) reported the first measurements of aerosol size distributions in this size range at South Pole. Koponen *et al.* (2003) observed that

particle formation took place in air masses transported neither from inland nor directly from the open sea but along the coast. These data were also analyzed for particle growth rate which was proven to be the lowest of all sites analyzed by *Kulmala et al.* (2005).

In January 2000 also aerosol dry deposition was studied at the station using the eddy covariance method in cooperation with the Stockholm University (*Grönlund et al.*, 2002). Measurements were performed over a smooth, snow-covered area and over moderately rough, rocky ground. There was a clear diurnal cycle in the dry deposition velocity v_d . The obtained v_d values were somewhat larger than those measured in Greenland over ice.

For modeling the radiative effects of aerosols the refractive index of particles should be known. However, the refractive index of boundary layer aerosol has not been studied much in Antarctica. The measurements conducted at Aboa have contributed to this. The scattering coefficients, number size distributions and chemical size distributions measured during the 1999/2000 campaign were used to derive the first size-fractionated aerosol refractive indices in the size range 40 nm – 15 μ m from Antarctica (*Virkkula et al.*, 2006b). These results were in agreement with the refractive indices derived by *Tomasi et al.* (2007) from spectral AOD measurements.

3.2.2 Seasonal cycles of particle number concentrations

The automatic particle counter system that was running at Aboa in December 2003 through January 2007 showed that there is a clear seasonal cycle in aerosol concentrations (Fig. 3). The maximum monthly average number concentrations ~ 900 cm^{-3} were measured in February. In March number concentration decreased rapidly until it was < 100 cm^{-3} in April and < 10 cm^{-3} in the darkest time of the year. It was also observed that the annual maximum daily-averaged particle concentration is later, in February, than the maximum in solar radiation intensity (Fig. 4). This suggests that the particle concentrations are more closely linked with the ocean temperature than solar radiation, the sea temperature maximum is reached in late summer in polar regions. The most probable explanation is that these particles are derived from dimethyl sulfide (DMS). It is a trace gas produced by its precursor compound, dimethyl sulfoniopropionate (DMSP), which is released by marine phytoplankton in the upper ocean. In the atmosphere, DMS is oxidised to form non-sea-salt sulfate and methane sulfonate (MSA) aerosols, which are a major source of cloud condensation nuclei (CCN) in remote marine air. DMS production in the Southern Ocean is highest when the amount of chlorophyll is highest which is when the sea surface temperature is highest (e.g., *Ayers and Gras*, 1991; *Wright and van den Enden*, 2000; *Gabric et al.*, 2003).

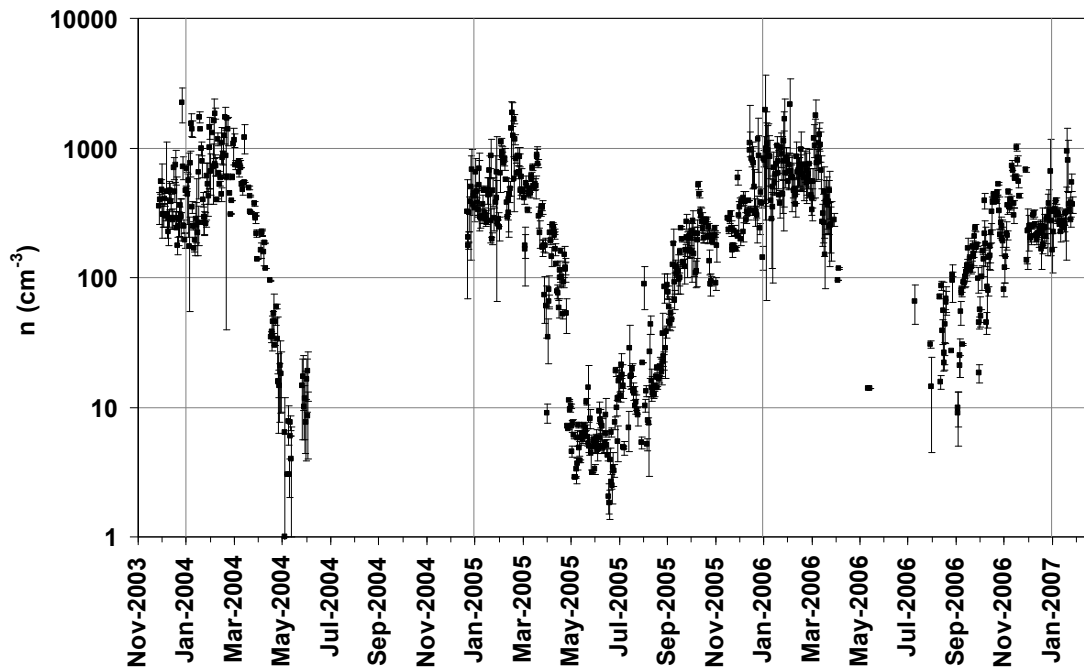


Fig. 3. Daily average particle number concentrations and standard deviations at Aboa in November 2003 – January 2007

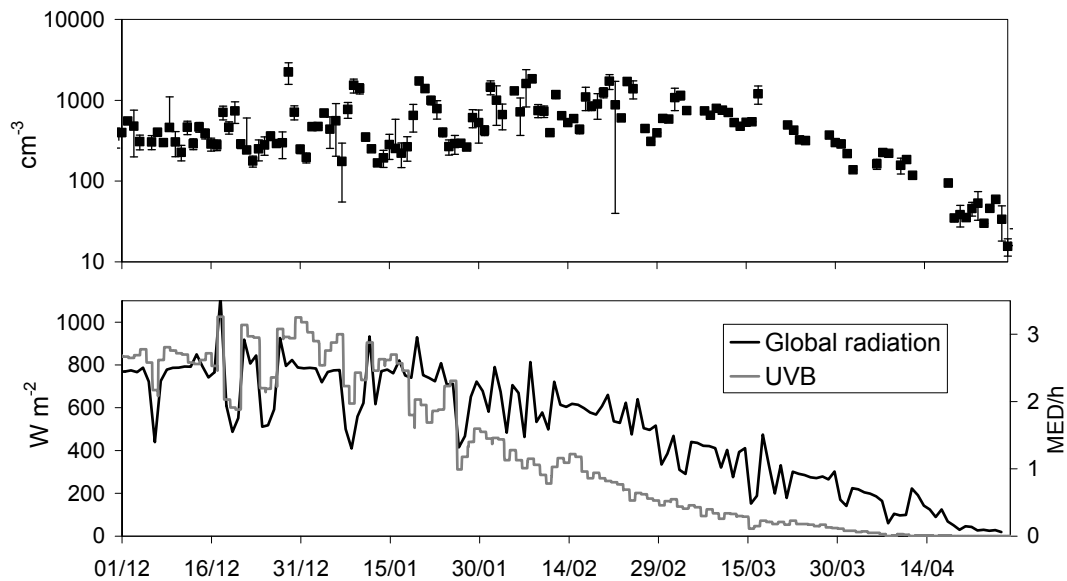


Fig. 4. Daily average particle number concentrations and standard deviations (upper panel) and daily maximum global and UV-B radiation at Aboa in December 2003 – April 2004.

The seasonal cycle observed at Aboa is also in agreement with observations from other Antarctic stations. The multi-annual CN concentration time series from the German Antarctic station Neumayer, the Australian Antarctic station Mawson, and the Japanese station Syowa have shown that the seasonal cycle usually has two maxima, the first one after Antarctic sunrise in October–December and the second and higher in

January–March (*Gras, 1993; Jaenicke et al., 1992; Ito, 1995*). All these are in agreement with the above discussion stating that the particle source is linked with biogenic sulfur emissions that peak in January at several Antarctic coastal stations (*Minikin et al., 1998*).

The seasonal cycle is not a new discovery. However, the novelty is the technical achievement: particle concentrations have not been measured at other unmanned Antarctic stations year-round. The advantage of this method is that the sample air is uncontaminated at all wind directions, contrary to manned stations where diesel generators or vehicle exhaust may contaminate the sample especially at winds blowing from certain sectors. The data in Figure 3 shows that this system is not faultless, there are long breaks in data. The reason for these is breaks in power since in winter the solar panels did not produce power and the wind generators had problems with their mobile parts, possibly due to icing and/or too high wind speeds.

3.2.3 Air ion spectra

The particles that were observed by *Koponen et al. (2003)* were larger than 3 nm in diameter, so the particles had already grown from the size range of stable clusters (~1 nm). In order to study the initial steps of aerosol formation and growth, measurements had to be extended to sizes close to 1 nm. In the summer 2004/2005 air ion size distributions in the size range 0.34–40 nm were measured (*Virkkula et al., 2007*). The purpose of the measurements was also to provide data on air ion concentrations at a site, in which ionization by soil-emitted radon and thoron is negligible. The station is located on top of a nunatak that is surrounded by glaciers. Even though radon concentration has not been measured at the site its contribution to ionization can indirectly be estimated to be low. First, the nunatak consists of basalts (*Luttinen and Furnes, 2000; Luttinen A., personal communication*). Radon is a daughter nuclide of uranium and the uranium contents of basalts is low (e.g., *Abumurad and Al-Tamimi, 2001*). Second, even though the aerosol research laboratory is stationed on rocks, it is close to the ice edge and during typical winds of 10 m/s possibly radon-containing air gets diluted by turbulent mixing with air blown from over the glacier where there definitely are no sources of radon. Therefore it can be assumed that galactic cosmic rays are the primary ionization source. However, the radon emissions from the nunatak have never been measured. In order to quantify its contribution to ionization it should be done.

The 48 measurement days were classified either as a particle formation event day, an undefined day, or a non-event day. Approximately 23 % of the days in summer 2004/2005 were particle formation event days, a figure similar to that at a boreal forest site (*Virkkula et al., 2007*). The respective analysis for the season 2006/2007 (*Vartiainen et al., 2007*) is presently being done. Cluster ions ($D_p < 1.6$ nm) were present during the whole measurement period (*Virkkula et al., 2007*). The ions must have been produced by cosmic rays since no other ionizing source is present according to the reasoning above.

However, the measurements suggest that during strong winds there exists also another ionizing process (Virkkula et al., 2007). There was a clear positive correlation between wind speed and cluster and intermediate size ($D_p < 1.6$ nm and 1.6–8 nm, respectively) ion concentrations. The formation of wind-induced particles was also size-dependent: the increase of cluster size ion number concentrations started at higher wind speeds than that of larger ions (Fig. 5). A plausible explanation is that at strong winds the ions were produced by friction processes in fast moving snow and ice crystals. However, the actual formation mechanism remains unclear.

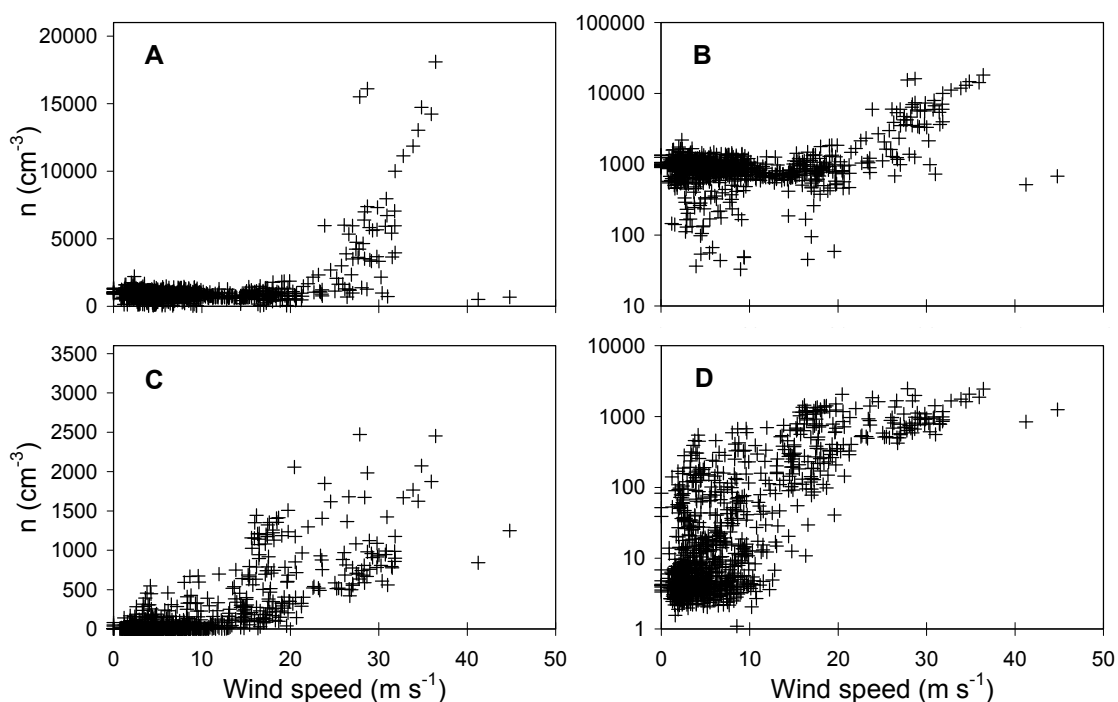


Fig. 5. Total (= sum of positive and negative ion concentrations) cluster ion (A and B) and intermediate ion (C and D) concentrations as a function of wind speed in summer 2004/2005. Left: linear scale; right: logarithmic scale.

3.2.4 Vertical transport of aerosols and ozone

The relationship between strong winds and particle transport and formation was further studied during summer 2007/2008. We analyze here an episode that will shed more light on this. On 10 and 11 January wind was blowing from the clean sector, ozone concentration decreased steadily (Fig. 6). Scattering coefficient varied clearly suggesting variation of source regions. Absorption coefficient was close to detection limit at all times but when wind blew from the contaminated sector (210–270°) for some tens of minutes hours on 10 January. No clear nucleation mode was observed. On 12 January wind speed increased from < 10 close to 20 m/s. At the peak wind speed close to noon a clear step in ozone concentration and scattering coefficient as well as a clear Aitken mode were observed.

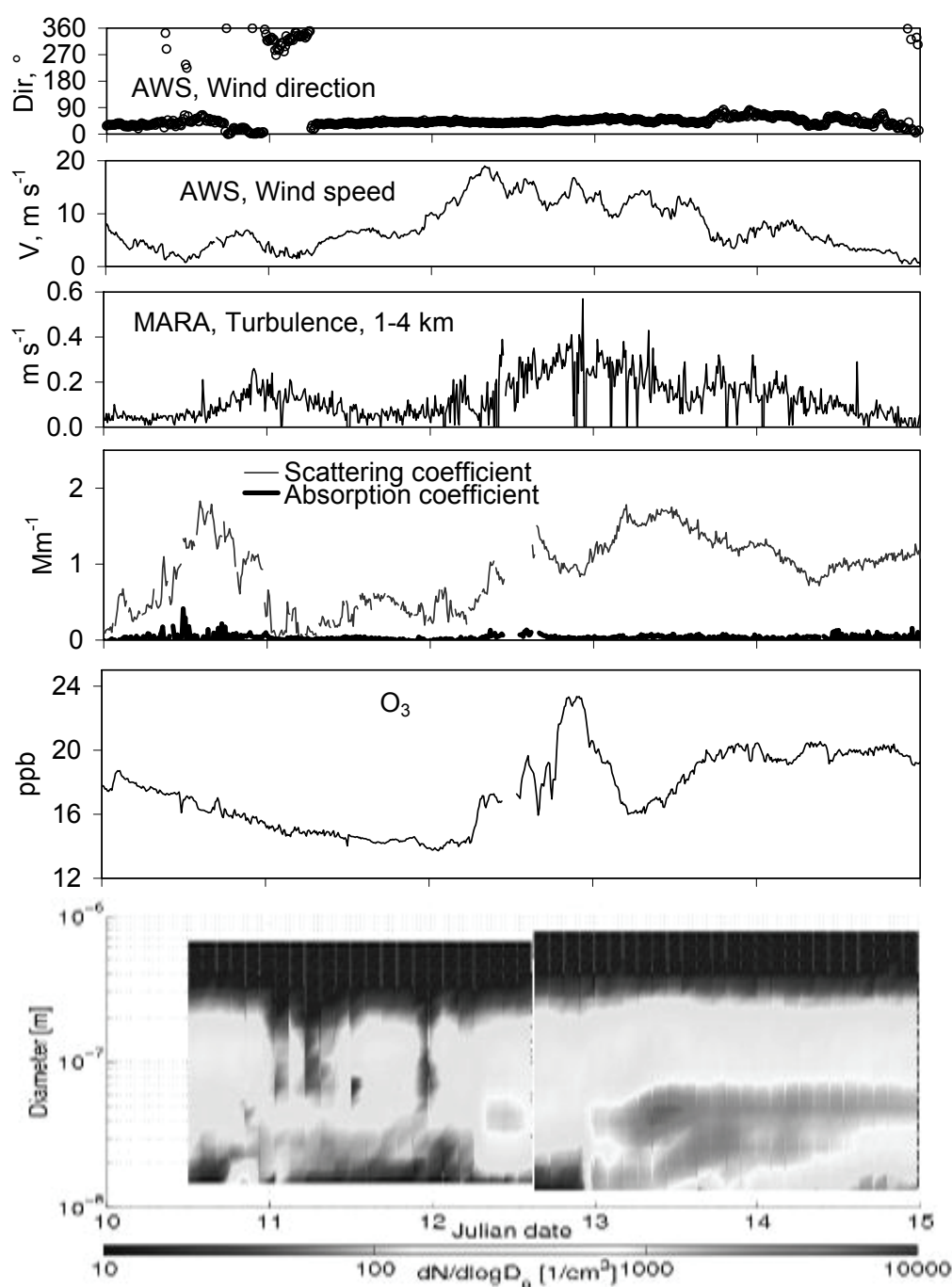


Fig. 6. Surface wind direction and speed measured at the Automatic Weather Station, turbulence obtained from the MARA (averaged over the lowest 4 kilometers), aerosol scattering and absorption coefficient, surface ozone concentration, and aerosol size distributions in January 10–14, 2008.

The highest ozone concentration was observed approximately 12 hours later at the same time as a clear nucleation mode appeared and started growing. The MARA produces, among other data, information on turbulence and vertical wind speed. These were averaged over the lowest 4 km. It is obvious that the peak ozone concentration and the nucleation mode appeared when the turbulence was highest suggesting of an intrusion of air from upper atmospheric levels. The peak turbulence did not occur simultaneously with the highest wind speed but with some time lag. The explanation of

this may only be obtained through a detailed meteorological analysis which is out of the scope of the present paper.

The analysis is supported by the air mass backtrajectories calculated using the NOAA HYSPLIT 4 model (*Draxler and Hess, 1998; Draxler and Rolph, 2003*) and the Global Data Assimilation System (GDAS) 3-hourly archive data. 5-day backtrajectories were calculated that arrive at 100 m, 500 m and 1000 m above ground (Fig. 7). They all show that at the time of the peak ozone concentration and particle formation the trajectories have clear downward motion.

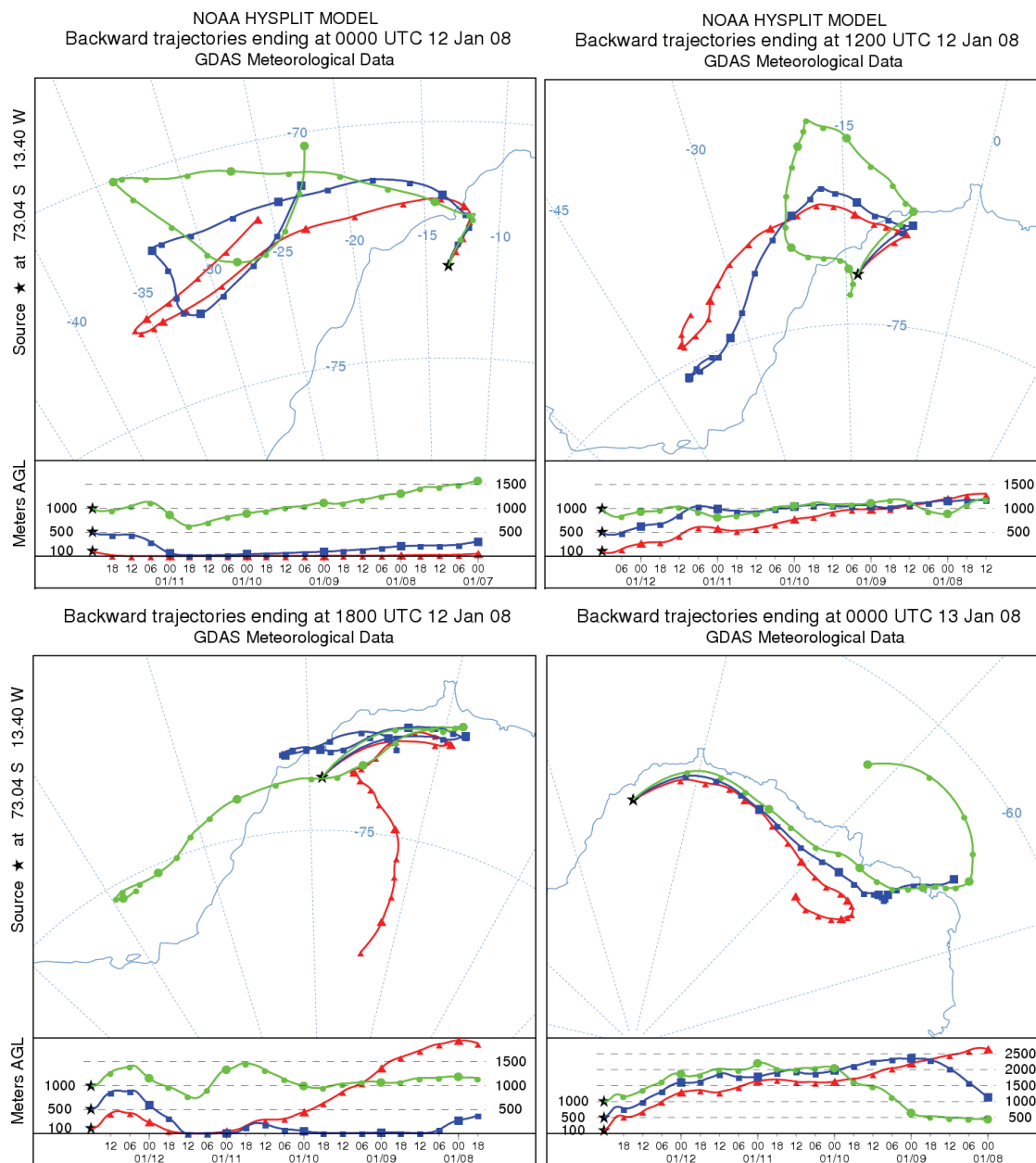


Fig. 7. Air mass back trajectories for arriving at 100 m, 500 m, and 1000 m above ground level at Aboa on 12 January 2008 at 00:00, 12:00 and 18:00 and on 13 January 2008 at 00:00 UTC.

4. Conclusions

Aerosol chemical composition, physical properties and processes have been studied mainly in Antarctic summers at Aboa. The main results of the chemical analyses is that the chemical composition varies greatly as a function of the source region and transport routes. Especially the travel time from the ocean proved to be an important factor determining the acidity, the cation-to-anion ratio and the chloride-to-sodium ratio. In this field there is an important puzzle to be solved: why does the chloride-to-sodium ratio in snow not decrease even though in aerosols it does decrease when sea-salt particles are transported away. Organic aerosols were found in samples taken with a high-volume sampler. These samples were taken with an inlet that passes through even large supermicron particles. If the observed OC particles are secondary organic aerosols they would be in the submicron size range. Therefore it is possible that the contribution of OC to the total submicron aerosol mass be significant. This can be studied by taking either simple submicron filter samples or multistage impactor samples with quartz filters to be analyzed for OC or with some more sophisticated online methods.

Aerosol formation and growth has been observed at Aboa during all summer campaigns. The origin of the nucleation mode particles, i.e., the nucleation mechanism and the gas-phase species that contribute to the growth of the particles are still unknown. *Koponen et al.* (2003) and *Virkkula et al.* (2006b) observed that at Aboa the freshly nucleated particles did grow to sizes larger than 100 nm where they have climatic significance. On the other hand, *Park et al.* (2004) observed that at South Pole nucleated particles did not grow large enough, most probably due to low condensable gas-phase concentrations. These observations suggest that at a range of some hundreds of kilometers from the coast particles do not grow to climatically significant sizes.

The research has proven that a low-power-consuming isopropanol-based particle counter can run automatically in Antarctica with power produced by solar cells and wind generators. Such a setup could easily be enlarged to contain also a DMPS to measure particle number size distributions to study the annual cycle of particle formation. Such instruments could also be installed at more than one location to study the geographical extent of the nucleation episodes and the growth of particles at various distances from the open ocean.

An particle formation event was analyzed using also 3D wind and turbulence data from the recently-installed wind radar. The analysis suggests that during strong winds at Aboa nucleation may be associated with an intrusion of air from higher levels to the surface layer. In earlier campaigns it was observed that often the nucleation episodes occur during high wind speeds. The earlier campaign data should be reanalyzed taking these considerations into account.

References

- Abumurad, K.M. and M. Al-Tamimi, 2001. Emanation power of radon and its concentration in soil and rocks, *Rad. Meas.*, **34**, 423–426.

- Arimoto, R., A. Hogan, P. Grube, D. Davis, J. Webb, C. Schloesslin, S. Sage S. and F. Raccach, 2004. Major ions and radionuclides in aerosol particles from the South Pole during ISCAT-2000, *Atmos. Environ.*, **38** 5473–5484.
- Ayers, G.P. and J.L. Gras, 1991. Seasonal relationship between cloud condensation nuclei and aerosol methanesulphonate in marine air. *Nature*, **353**, 834–835.
- Bauguitte, S.J.-B., A.E. Jones, M.A. Hutterli, P.S. Anderson, D.J. Maxfield, H.K. Roscoe, E.W. Wolff, A. Virkkula, S. Kirkwood, R. Weller, 2009. Preliminary data analysis from the IPY autonomous surface ozone monitoring network in Dronning Maud Land, Antarctica, EGU General Assembly 2009.
- Bergin, M.H., E.A. Meyerson, J.E. Dibb, and P.A. Mayewski, 1998. Relationship between continuous aerosol measurements and firn core chemistry over a 10-year period at the South Pole, *Geophys. Res. Lett.*, **25**, 1189–1192.
- Bertler, N., P.A. Mayewski, A. Aristarain, P. Barrett, S. Becagli, R. Bernardo, Bo S., C. Xiao, M. Curran, D. Qin., D. Dixon, F. Ferron, H. Fischer H., M.Frey, M. Frezzotti, F. Fundel, C. Genthon, R. Gragnani, G. Hamilton, M. Handley, S. Hong, E. Isaksson, J. Kang, J. Ren, K. Kamiyama, S. Kanamori, E. Kärkäs, L. Karlöf, S. Kaspari, K. Kreutz, A. Kurbatov, E. Meyerson, Y. Ming, M. Zhang, H. Motoyama, R. Mulvaney, H. Oerter, E. Osterberg, M. Proposito, A. Pyne, U. Ruth, J. Simões, B. Smith, S. Sneed, K. Teinilä, F. Traufetter, R. Udisti, A. Virkkula, O. Watanabe, B. Williamson, J-G Winther, Y. Li, E. Wolff, Z. Li, A. Zielinski, 2006. Snow chemistry across Antarctica, *Annals of Glaciology*, **41**, 167–179.
- Davis, D., F. Eisele, G. Chen, J. Crawford, G. Huey, D. Tanner, D. Slusher, L. Mauldin, S. Oncley, D. Lenschow, S. Semmer, R. Shetter, B. Lefer, R. Arimoto, A. Hogan, P. Grube, M. Lazzara, A. Bandy, D. Thornton, H. Berresheim, H. Bingemer, M. Hutterli, J. McConnell, R. Bales, J. Dibb, M. Buhr, J. Park, P. McMurry, A. Swanson, S. Meinardi and D. Blake, 2004. An overview of ISCAT 2000. *Atmos. Environ.*, **38**, 5363–5373.
- Draxler, R.R. and G.D. Hess, 1998., An overview of the HYSPLIT4 modeling system for trajectories, dispersion and deposition, *Aust. Meteorol. Mag.*, **47**, 295 – 308.
- Draxler, R.R. and G.D. Rolph, 2003., HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory); www.arl.noaa.gov/ready/hysplit4.html, NOAA Air Resources Laboratory, Silver Spring, MD.
- Gabric, A.J., R. Cropp, T. Hirst and H. Marchant, 2003. The sensitivity of dimethyl sulfide production to simulated climate change in the Eastern Antarctic Southern Ocean, *Tellus*, **55B**, 966–981.
- Gras, J.L., 1993. Condensation nucleus size distribution at Mawson, Antarctica: Seasonal cycle. *Atmos. Environ.*, **27A**, 1417–1425.
- Grönlund, A., D. Nilsson, I.K. Koponen, A. Virkkula A. and M. Hansson, 2002. Aerosol dry deposition measured with eddy-covariance technique at Wasa and Aboa, Dronning Maud Land, Antarctica, *Ann. Glaciology*, **35A**, 355–361.
- Haywood, J. and O. Boucher, 2000. Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: A Review, *Reviews of Geophysics*, **38**, 513–543.

- Ito, T., 1995. Nature and origin of Antarctic submicron aerosols. *NATO ASI Series*, **130**, 23–38.
- Jaenicke, R., V. Dreiling, Lehmann, P.K. Koutsenogui and J. Stingl, 1992. Condensation nuclei at the German Antarctic Station "Georg von Neumayer". *Tellus*, **44B**, 311–317.
- Jourdain, B. and M. Legrand, 2002. Year-round records of bulk and size-segregated aerosol composition and HCl and HNO₃ levels in the Dumont d'Urville, coastal Antarctica. atmosphere: Implications for sea-salt aerosol fractionation in the winter and summer, *J. Geophys. Res.*, **107**(D22., 4645, doi:10.1029/2002JD002471.
- Kerminen, V.-M., K. Teinilä and R. Hillamo, 2000. Chemistry of sea-salt particles in the summer Antarctic atmosphere. *Atmos. Environ.*, **34**, 2817–2825.
- Kerminen, V.-M., R. Hillamo, K. Teinilä, T. Pakkanen, I. Allegrini, R. Sparapani, 2001. Ion balances of size-resolved tropospheric aerosol samples: implications for the acidity and atmospheric processing of aerosols, *Atmos. Environ.*, **35**, 5255–5265.
- Koponen, I.K., A. Virkkula, R. Hillamo, V.-M. Kerminen and M. Kulmala, 2003. Number size distributions and concentrations of the continental summer aerosols in Queen Maud Land, Antarctica, *J. Geophys. Res.*, **108**(D18, 4587, DOI:10.1029/2003JD003614, 2003.
- Kulmala, M., H. Vehkamäki, T. Petäjä, M. Dal Maso, A. Lauri, V.-M. Kerminen, W. Birmili and P. McMurry, 2004. Formation and growth rates of ultrafine atmospheric particles: a review of observations, *J. Aerosol Sci.*, **35**, 143–176.
- Kulmala, M., T. Petäjä, P. Mönkkönen, I. K. Koponen, M. Dal Maso, P.P. Aalto, K. Lehtinen and V.-M. Kerminen, 2005. On the growth of nucleation mode particles: source rates of condensable vapor in polluted and clean environments, *Atmos. Chem. Phys.*, **5**, 409–416.
- Kulmala, M., K.E.J. Lehtinen and A. Laaksonen, 2006. Cluster activation theory as an explanation of the linear dependence between formation rate of 3 nm particles and sulphuric acid concentration, *Atmos. Chem. Phys.*, **6**, 787–793.
- Kirkwood, S., I. Wolf, P. Dalin, H. Nilsson, D. Mikhailova, and E. Belova, 2007., Polar mesosphere summer echoes at Wasa, Antarctica (73°S): First observations and comparison with 68°N, *Geophys. Res. Lett.*, **34**, L15803, doi:10.1029/2007GL030516.1
- Kirkwood, S., H. Nilsson, R.J. Morris, A.R. Klekociuk, D.A. Holdsworth and N.J. Mitchell, 2008. A new height for the summer mesopause: Antarctica, December 2007, *Geophys. Res. Lett.*, **35**, L23810, doi:10.1029/2008GL035915.
- Kärkäs, E., K. Teinilä, A. Virkkula, and M. Aurela, 2005. Spatial variations of surface snow chemistry during two austral summers in western Dronning Maud Land, Antarctica, *Atmos. Environ.*, **39**, 1405–1416.
- Luttinen, A. and H. Furnes, 2000. Flood basalts of Vestfjella: Jurassic magmatism across an archaean-proterozoic lithospheric boundary in Dronning Maud Land, Antarctica, *J. Petrol.*, **41**, 1271–1305.

- Minikin, A., M. Legrand, J. Hall, D. Wagenbach, C. Kleefeld, E. Wolff, E.C. Pasteur and F. Ducroz, 1998. Sulfur-containing species (sulfate and methanesulfonate) in coastal Antarctic aerosol and precipitation, *J. Geophys. Res.*, **103**, 10975–10990.
- Park, J., H. Sakurai, K. Vollmers and P.H. McMurry, 2004. Aerosol size distributions measured at the South Pole during ISCAT, *Atmos. Environ.*, **38**, 5493–5500.
- Rankin, A., and E. Wolff, 2003. A year-long record of size-segregated aerosol composition at Halley Antarctica, *J. Geophys. Res.*, **108**(D24), 4775, doi:10.1029/2003JD003993.
- Teinilä, K., V.-M. Kerminen and R. Hillamo, 2000. A study of size-segregated aerosol chemistry in the Antarctic atmosphere, *J. Geophys. Res.*, **105**, 3893–3904.
- Tomasi, C., V. Vitale, A. Lupi, C. Di Carmine, M. Campanelli, A. Herber, R. Treffeisen, R.-S. Stone, E. Andrews, Sharma, V. Radionov, W. von Hoyningen-Huene, K. Stebel, G.H. Hansen, C.L. Myhre, C. Wehrli, V. Aaltonen, H. Lihavainen, A. Virkkula, R. Hillamo, J. Ström, C. Toledano, V.E. Cachorro, P. Ortiz, A.M. de Frutos, S. Blindheim, M. Frioud, M. Gausa, T. Zielinski, T. Petelski and T. Yamanouchi, 2007. Aerosols in polar regions: A historical overview on the basis of optical depth and in situ observations, *J. Geophys. Res.*, **112**, D16205 doi:10.1029/2007JD008432.
- Udisti, R., S. Becagli, S. Benassai, E. Castellano, I. Fattori, M. Innocenti, A. Migliori, and R. Traversi, 2004. Atmosphere-snow interaction by a comparison between aerosol and uppermost snow-layers composition at Dome C, East Antarctica, *Annals of Glaciology*, **29**, 53–61.
- Vana, M., A. Virkkula, A. Hirsikko, P. Aalto, M. Kulmala and R. Hillamo, 2007. Air Ion Measurements During a Cruise from Europe to Antarctica, in Colin D. O'Dowd and Paul E. Wagner (eds.), *Nucleation and Atmospheric Aerosols*, 368–372.
- Vartiainen, E., M. Ehn, P.P. Aalto, A. Frey, A. Virkkula, R. Hillamo, A. Arneth and M. Kulmala, 2007. Aerosol particle and ion measurements in Queen Maud Land, Antarctica, European Aerosol Conference 2007, Salzburg, Abstract T13A140
- Virkkula, A., R. Hillamo, P.P. Aalto and M. Kulmala, 2004. A modification of TSI model 3007 CPC for unattended use and the test at the Finnish Antarctic research station Aboa. Abstracts of the European Aerosol Conference, Budapest 2004, *J. Aerosol Sci.*, S287–S288.
- Virkkula, A., K. Teinilä, R. Hillamo, V.-M. Kerminen, S. Saarikoski, M. Aurela, I.K. Koponen and M. Kulmala, 2006a. Chemical size distributions of boundary layer aerosol over the Atlantic Ocean and at an Antarctic site, *J. Geophys. Res.*, **111**, D05306, doi:10.1029/2004JD004958, 2006
- Virkkula, A., I. K. Koponen, K. Teinilä, R. Hillamo, V.-M. Kerminen and M. Kulmala, 2006b. Effective real refractive index of dry aerosols in the Antarctic boundary layer, *Geophys. Res. Lett.*, **33**, No. 6, L06805, 10.1029/2005GL024602

- Virkkula, A., K. Teinilä, R. Hillamo, V.-M. Kerminen, S. Saarikoski, M. Aurela, J. Viidanoja, J. Paatero, I.K. Koponen and M. Kulmala, 2006c. Chemical composition of boundary layer aerosol over the Atlantic Ocean and at an Antarctic site, *Atmos. Chem. Phys.*, **6**, 3407–3421.
- Virkkula, A., A. Hirsikko, M. Vana, P.P. Aalto, R. Hillamo and M. Kulmala, 2007. Charged particle size distributions and analysis of particle formation events at the Finnish Antarctic research station Aboa, *Boreal Environ. Res.*, **12**: 397–408.
- Wagenbach, D., F. Ducroz, R. Mulvaney, L. Keck, A. Minikin, M. Legrand, J. S. Hall, and E.W. Wolff, 1998. Sea-salt aerosol in coastal Antarctic regions, *J. Geophys. Res.*, **103**, 10,961–10,974.
- Wright, S.W. and R.L. van den Enden, 2000. Phytoplankton community structure and stocks in the Eastern Antarctic marginal ice zone (BROKE survey, Jan–Mar 1996) determined by CHEMTAX analysis of HPLC pigment signatures. *Deep Sea Res.*, **47**, 2363–2400.