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Continuous measurements of aerosol properties at the Baltic Sea

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ABSTRACT

Thirty months (March 2003–October 2005) of aerosol size distributions, trace gas and meteorological parameter measurements have been analysed at a background station in Baltic Sea. Log-normal modes have been fitted to the size distributions and a source region analysis using 120 h back trajectories has been accomplished. Seasonal and air mass influences have been studied and compared to other measurement sites. Only weak impacts of meteorological conditions have been found. New particle formation events were observed, but less frequently compared to continental sites, which is concluded being related to the lack of precursor gas sources close to the station and a higher condensation sink. The northeast Atlantic was recognized as an important source of particles in the Aitken mode. Especially in spring and summer, probably primary biogenic particles contribute largely to the particle number concentration. Source regions of the submicron aerosol are strongly dependant of the particle size and so is the contribution to the total number or mass concentration. For this reason, a consideration of the total number or mass concentration was identified not being sufficient in order to describe source areas of aerosol particles.

1. Introduction

Atmospheric aerosols influence climate both directly and indirectly (Haywood and Boucher, 2000; IPCC, 2001). Aerosol particles can modify the radiation budget directly by scattering, absorbing or reflecting the solar radiation back to space, which was estimated in several studies (e.g. Bellouin et al., 2005; Chung et al., 2005). Because of the seasonal and geographical variabilities, the aerosol-driven radiative forcing can locally even exceed the greenhouse effect (Ramanathan et al., 2001). Because of their ability to act as cloud condensation nuclei (CCN), aerosol particles affect the cloud albedo (Twomey, 1977), lifetime of clouds and the formation of precipitation (Ramanathan et al., 2001). Estimations of the climate effect of aerosols are still very uncertain, especially concerning the influence on clouds and the hydrological cycle (IPCC, 2001; Lohmann and Feichter, 2005). Uncertainties in the indirect forcing are mainly due to aerosol and aerosol precursor emissions, aerosol mass concentrations and size distributions and the cloud fraction and are significantly varying regionally (Chen and Penner, 2005). In order to reduce these uncertainties, detailed information on the concentrations, size distributions, optical properties, chemical composition and mixing state of the particles as well as temporal and seasonal distributions are required, but unfortunately only rarely available.

Numerous works have focused on the spatial and temporal characterization of physical aerosol properties in different environments and measurement sites. A comprehensive aerosol phenomenology for various continental sites in Europe was presented by van Dingenen et al. (2004). Continental particle size distributions have been investigated in the Alps (Weingartner et al., 1999), in Finland (e.g. Mäkelä et al., 2000) and in marine and polar environments (e.g. Covert et al., 1996; Heintzenberg et al., 2000; Koponen et al., 2002) with respect to temporal variations. Tunved et al. (2003, 2005) compared the physical aerosol properties of four to five different Scandinavian sites and investigated air mass impacts. Back trajectories were often used in the literature to characterize long range transport (e.g. Seibert et al., 1998), source receptor relations (e.g. Stohl, 1996; Sciare et al., 2003), the role of air masses concerning particle size distributions (Birmili et al., 2001) or the probability of new particle formation (Nilsson et al., 2001; Sogacheva et al., 2005).

All these works concentrate on either continental or marine sites, which points to the question, which behaviour of aerosols we can find at a transition site between continental and marine character. The aim of this work was to characterize the aerosol at a neither continental nor marine site in Baltic Sea in terms of modal structure, seasonal and air mass behaviour. Back trajectories were used with the intention to achieve

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Fig. 1. Location of the wind sectors.

new information on possible pollutant sources all over northern Europe.

2. Experimental and methods

2.1. Measurement site

Measurements were conducted at the EMEP-station (European Monitoring and Evaluation Programme) Utö (59° 47′N, 21° 23′E, 8 m asl; see Fig. 1 for location). Utö is a small island in Baltic Sea some 60 km from the Finnish southwest coast and more than 10 km from the nearest islands around. There are only few local pollutant sources nearby like passing large ships or motor vehicles close to the station. However, the adulterated data resulting from these sources can be excluded easily, since these sources produce high but short time concentrations in certain size classes of the particle size distribution. The closest city, Turku, is situated about 90 km in the northeast of the almost treeless island. The wind direction is dominantly southwesterly in fall and winter and north or southwesterly during spring and summer times, while the wind speed is highest in winter.

2.2. Instrumentation

Continuous measurements of the particle number size distribution were performed since March 2003 using a Differential Mobility Particle Sizer (DMPS) system. The system extracts a small electrical mobility fraction from the sample air using a Differential Mobility Analyzer (DMA, 28 cm long, Hauke-type) and transports it to a Condensation Particle Counter (CPC, TSI model 3010). The sheath flow is provided by a closed loop arrangement, where the relative humidity is kept relatively low (<20%) by using a silica gel drier. The aerosol is charged with a radioactive source before sizing. The used sheath air flow is about 10 l min^{-1} and the aerosol sample flow is 1 l min^{-1}. The temperature difference between the saturator and the condenser of the CPC has been increased to lower the 50% cut off limit and the counting efficiency has also been taken into account in the inversion algorithm. The measurements cover a size range of 7–500 nm which is divided into 30 discrete size bins. One particle size distribution takes about 5 min to measure.

A number of trace gases and meteorological parameters are measured at the station as well. In this study, the SO_2 (Thermo Environmental Instruments, 43S), O_3 (Horiba APOA, 360) and $PM_{2.5}$ (Thermo Anderssen, FH 62 I-R) concentrations and meteorological data (like wind direction, temperature, relative humidity and visibility) were used. The measurements are continuous and 1 h averages were calculated for further use. Five-day back trajectories were calculated with the FLEXTRA model (Stohl et al., 1995), which is based on the wind fields available from the European Center for Medium-Range Weather Forecasts (ECMWF). The trajectories were calculated eight times per day (arrival times of 00, 03, 06, 09, 12, 15, 18 and 21 UTC at the station) for the 950 hPa level.

Data used for this study cover the time period of March 2003–October 2005, but because of some missing data, there are almost 30 month of data available.

2.3. Fittings, wind sector and source region analysis

Fittings of log-normal modes have been realized being useful to compare particle size distributions (e.g. obtained at different stations) objectively. In the size range of <1 μm (fine particles), three modes (nucleation, Aitken and accumulation mode) are used because they are connected to the processes, which lead to their appearance. In order to fit each scan individually, an automated iteration method, presented by Tunved et al. (2003), was used. To produce results with physical relevance, boundary conditions for the calculated fitting parameters had to be given. The used values for nucleation, Aitken and accumulation mode (mode 1–3) can be seen form Table 1.

If no solution could be found within four iterations, a bimodal fitting (D_g 7–90 nm and 90–300 nm, respectively) was suggested. This normally resulted in a fitting consisting of two modes (Aitken and accumulation mode), but a third mode (nucleation) occurred relatively frequently. Though, we have to be aware, that 2 or 3 modes are a very crude simplification of

Table 1. Lower and upper boundaries of the fitting parameters given for the fitting algorithm: Geometric mean diameter (D_g), geometric standard deviation (GSD) and number concentration (N)

	D_g (nm)	GSD	N (cm^{-3})
Mode 1	7–30	1.1–2	0–10^5
Mode 2	30–90	1.1–2	0–10^5
Mode 3	90–300	1.1–2	0–10^5

Table 2. Wind sector definition and their frequency of occurrence (FO)

Name	Sector	FO
North	320°–40°	29.6%
East	70°–130°	13.5%
Central Europe	180°–250°	36.6%
Atlantic	250°–310°	20.2%

the real particle size distribution, which is, however, sufficient to describe the ambient state of the aerosol presented in this work.

In order to investigate advection impacts on the measured aerosol, the data was divided into a subdata set, regarding the wind direction at the station. The limits of the sectors as well as the frequency of occurrence during the measurement period can be seen from Table 2. Figure 1 shows the location of the wind sectors over Europe. The areas between these displayed sectors were left out, since they are not clearly assignable to a certain air mass type and thus representing transition states between the typical air mass properties. Furthermore, most of the data are connected to the considered sectors.

Back trajectories have been recognized as a useful tool to investigate source regions of air pollutants and are successfully used for different receptor sites (e.g. Birmili et al., 2001; Sciare et al., 2003; Tunved et al., 2005). For a comprehensive review of calculation methods and applications, see for example, Stohl (1998). In this work, a method developed by Stohl (1996) was used to determine locations of sources of several pollutants. Calculated back trajectories and ambient pollutant concentrations at the arrival time of the air parcel at a receptor site were associated. In a first step, the concentrations were assigned to each $1^\circ \times 1^\circ$ grid cell crossed by the corresponding trajectory, which results in a first guess concentration field, where potential source areas were spotted out. Using this first guess, an iterative redistribution procedure followed to improve the spatial resolution and get more detailed structures. The iteration ended when the resulting concentration field was not changed by the redistribution anymore. In a last step, the final concentration field was smoothed. In order to ensure the statistical significance, the concentration values were calculated only, if a minimum number of trajectories crossed a grid cell. This minimum number was set to 10 in this work. The resulting concentration field was plotted in a map, where the source areas can be seen from.

We have to be conscious of the fact, that the quality of the results is reduced for aerosols, that go through non-linear processes during transport. For an example sulphate particles are effectively removed by precipitation. In any case, the method was used successfully for particulate sulphate (Stohl, 1996), for CO_2 (Aalto et al., 2002) and for SO_2 and aerosols (Virkkula et al., 1997), and therefore it was applied in this study as well. Beside aerosol properties, SO_2 and O_3 concentrations have also been analysed in this study.

In order to interpret the results in a reasonable way, we have to take the fact into consideration, that the resulting field is not equal to an accurate emission field of the regarded species. This is because it is influenced by all processes, which produce or remove a certain amount of the regarded species. It is instead showing the origin of air parcels and source areas, which the latter result in high concentrations at the measurement site. All non-linear processes influencing the aerosol and regarded species are affecting the results presented here.

3. Results and discussion

3.1. Seasonal variation of aerosol properties

The available data set was analysed regarding seasonal differences, which is described in this section. For this consideration, the data set has been subdivided according to the four seasons spring (March–May), summer (June–August), fall (September–November) and winter (December–February).

3.1.1. Particle size distributions. As pointed out by Tunved et al. (2003), the meteorological conditions in different seasons can have a large impact on the particle size distributions. The median particle size distributions and their fits as well as the 25 and 75% percentiles are displayed in Fig. 2. The results of calculated modal fits based on the median particle size distributions can be seen from Table 3, which shows the number concentration (N_{1-3}), geometric standard deviation (GSD_{1-3}), geometric mean diameter ($D_{g,1-3}$) of the fitted modes and the number concentration ratio between Aitken and accumulation mode (N_2/N_3).

Nucleation days do not appear in the median values of the particle size distribution, which is caused by the large data set in combination with relatively few new particle formation events. In any case, the existence of a nucleation mode in spring is an indicator for new particle formation, which occurs more often during this period compared to the other seasons (see Section 3.1.3).

The highest number concentrations in the Aitken mode were found in spring and summer. A potential explanation for this could be the different strengths of precipitation during the seasons, since wet deposition is the most effective removal process for Aitken mode particles in the atmosphere. But we have to keep in mind, that in order to investigate impacts on the particle concentrations, it is not sufficient to take only the removal processes into account, since the concentrations are always the balance between sources and sinks. New particle formation makes also a contribution to higher Aitken mode concentrations, because of the growth of freshly formed particles into the regarded size range.

The highest accumulation mode particle number concentrations were found in winter and spring (about 450 cm^{-3}), which seems to be connected to the boundary layer conditions. In

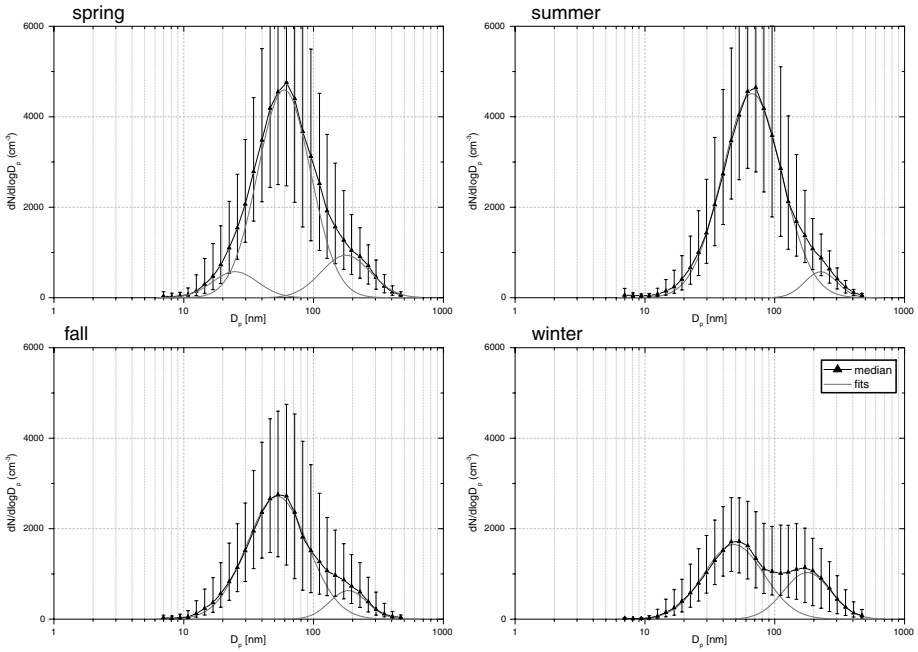

Fig. 2. Median particle size distributions according to different seasons and corresponding log-normal fits. Error bars indicate 25 and 75% percentiles.

winter and spring, when the temperatures were lowest (on average 0.6 and 3.2 °C, respectively), shallow boundary layers with relatively strong inversions were expected. Reduced vertical mixing in combination with stronger horizontal winds during winter means that the particles can be transported in the boundary layer over longer distances compared to the summer seasons. Consequently, the number of accumulation mode particles was much lower in summer (180 cm^{-3}) compared to winter and spring.

The relative contribution of accumulation mode particles with respect to the total number concentrations was highest in winter, where also a more pronounced minimum between Aitken and accumulation modes could be seen. The highest total number concentrations were found in spring, whereas the lowest concentrations were observed in winter. As discussed above, possible explanations for this seasonal behaviour can be the seasonal pattern of solar radiation, precipitation, cloudiness, frequency of new particle formation or even seasonal changes in air masses.

In order to compare the modal parameters resulting from the fittings, Table 3 shows also the modal parameters obtained by Tunved et al. (2003) with the same fitting algorithm as used in this study for two different measurement sites in Finland. Hyytiälä (61°51′N, 24°17′E, 180 m asl) is characterized as a boreal forest site in southern Finland (detailed description is given by, for example, Hari and Kulmala, 2005; Kulmala et al., 2001) and Pallas is situated in Finnish Lapland (68°N, 24° 14′E, 340 m asl) in the sub arctic pine forest (see Hatakka et al., 2003 for a detailed description of the station).

Although the same fitting program was used, the results differ strongly from each other. In Hyytiälä and Pallas, three modes were observed in almost all median size distributions, whereas a nucleation mode was found only in spring in the presented data set. One possible reason for this could be the lack of local emissions of precursors of condensable material and a higher condensation sink, which gets lower the more northern the station is located (Tunved et al., 2005).

The Aitken modes in spring and summer were very similar in Utö, but at the other stations, the number concentrations in spring were significantly lower than in summer. Compared to Hyytiälä and Pallas, higher number concentrations and geometric standard deviations in the Aitken mode were found in Utö for all seasons. Smaller geometric mean diameters than in Hyytiälä could be obtained for all seasons, whereas in comparison to Pallas, only in fall and winter smaller values could be observed. The differences between Utö and Hyytiälä can at least partly be explained by the absence of the nucleation mode in most cases. As already pointed out by Tunved et al. (2005), the particle number concentration is increasing the more southern the station is situated, which is also explaining some of the observed differences.

A different seasonal pattern in the accumulation mode number concentrations was found at all the stations. A similar number of accumulation mode particles was found for winter and spring in Utö and Pallas, but in summer and fall, the values in Utö exceed the number in Pallas by a factor of about three. In comparison to Hyytiälä, higher accumulation mode number concentrations were found in winter and spring, whereas lower values have been observed in summer and fall. But the differences were not

Table 3. Modal parameters from fitting of the median particle number size distributions according to seasons (Hyytiälä and Pallas from Tunved et al., 2003) and wind sectors: Total number concentration (N_{tot}), geometric mean diameter (D_g), geometric standard deviation (GSD) and number concentration (N) for the three modes (Nucleation (1), Aitken (2) and accumulation (3) mode

	N_{tot} (cm^{-3})	N_1 (cm^{-3})	GSD_1	$D_{g,1}$ (nm)	N_2 (cm^{-3})	GSD_2	$D_{g,2}$ (nm)	N_3 (cm^{-3})	GSD_3	$D_{g,3}$ (nm)	N_2/N_3
Season											
Utö Spring	3315	521	1.49	24.8	2347	1.6	59.4	447	1.55	177.1	5.2
Summer	2789	–	–	–	2610	1.7	66.8	179	1.33	226.4	14.6
Fall	1830	–	–	–	1604	1.72	52.5	226	1.39	184.9	7.1
Winter	1424	–	–	–	956	1.7	48.5	468	1.52	177.6	2.0
Hyytiälä Spring	1564	431	1.74	25	849	1.69	67	284	1.44	200	3.0
Summer	1643	429	1.66	34	887	1.58	77	327	1.46	192	2.7
Fall	1298	227	1.63	25	776	1.62	68	295	1.44	200	2.6
Winter	863	204	1.8	25	389	1.62	61	270	1.48	200	1.4
Pallas Spring	382	63	1.54	19	157	1.52	43	162	1.61	168	1.0
Summer	490	93	1.71	29	282	1.49	53	115	1.44	182	2.5
Fall	478	69	1.69	36	179	1.43	74	230	1.61	160	0.8
Winter	275	–	–	–	152	1.85	53	123	1.48	212	1.2
Sector											
Utö North	1805	486	1.55	25.7	969	1.49	53.7	350	1.57	157.1	2.77
East	3018	–	–	–	2725	1.77	71.1	293	1.43	212.6	9.3
Central E.	2642	196	1.51	28.4	1977	1.57	61.7	469	1.51	174.3	4.22
Atlantic	2139	–	–	–	2007	1.77	51.5	132	1.33	199.6	15.2

as significant as in the Aitken mode size range. The number concentrations are caused by the combination of transport and local sources. Horizontal transport is more efficient in winter because of less vertical mixing. Anthropogenic sources are almost evenly distributed all over the year, but slightly increased in winter because of domestic heating in the entire northern Europe. This explains the much higher accumulation mode number concentrations in winter at the station. In Hyytiälä, by contrast, the accumulation mode number concentrations did not show any seasonal pattern. Unlike in Utö, biogenic sources play an important role as well, since the site is located directly in the forest. The seasonal behaviour of biogenic sources is very different compared to anthropogenic emissions, which explains the observed differences between the stations. The variation of the geometric standard deviation showed the same pattern between Utö and Pallas, but was slightly lower in Utö. In Hyytiälä, only a very weak seasonal pattern of the geometric standard deviation and mean diameter could be observed. In Pallas, the highest mean diameter was found in winter, whereas the maximum in Utö was observed in summer.

The shape of the particle size distributions in Utö and Hyytiälä was very similar in summer and winter, but in spring and fall, a more distinct accumulation mode could be seen in Hyytiälä. In winter, a clear minimum between Aitken and accumulation mode was found in Utö, whereas they merged to one mode in summer. At the Finnish Lapland station, a clear minimum between these two modes was found for all seasons except fall.

3.1.2. Relationship to other variables. The aerosol properties are influenced by several atmospheric parameters and processes. In Table 4, the average values of considered meteorological as well as trace gas parameters are summarized. The highest $PM_{2.5}$ and ozone values were found in spring (10 and 81.7 μg m^{-3}), whereas the lowest value could be observed in summer (6.9 μg m^{-3}) and fall (58.5 μg m^{-3}), respectively. Conspicuous were also the much higher temperatures in fall than in spring (9.2 °C versus 3.2 °C).

In order to find some relationships between the different aerosol, trace gas and meteorological parameters, correlation coefficients between some of these parameters were calculated and are presented in Table 5. A relationship between the accumulation mode number, total volume and $PM_{2.5}$ concentration was found throughout the year. Visibility and relative humidity were anti correlated. Additionally, a correlation between temperature and the total number concentration was found in fall. In winter, the SO_2 concentration was related to the accumulation mode number, the total volume and the $PM_{2.5}$ concentration and reversely associated with the temperature. Variabilities according to different advection and wind directions, respectively, were found throughout the seasons (Section 3.2).

3.1.3. Nucleation events. New particle formation, which was found taking place at several locations (see Kulmala et al., 2004 for a comprehensive review), was observed in Utö as well. The measurements were classified in the same way as in former studies (see Mäkelä et al., 2000 for a detailed description of the classification method), including the days with no clear particle formation (non-event days) and those with (at least) some particle formation. Within these formation days we could now distinguish between days with a significant new particle formation and growth (event days) and unclear days with some formation but

Table 4. Average values of meteorological and trace gas parameters for each season and wind sector

	$PM_{2.5}$ ($\mu g\,m^{-3}$)	SO_2 ($\mu g\,m^{-3}$)	NO_x ($\mu g\,m^{-3}$)	O_3 ($\mu g\,m^{-3}$)	T (°C)	RH (%)	Vis. (m)
Season							
Spring	10.0	1.3	7.3	81.7	3.2	84.0	28 700
Summer	6.9	0.9	4.5	74.2	15.9	82.8	31 900
Fall	7.8	0.8	5.6	58.5	9.2	84.9	30 400
Winter	9.0	1.3	4.6	62.2	0.6	86.4	25 600
Annual	8.4	1.0	5.6	70.3	7.9	84.3	29 500
Sector							
North	5.9	1.0	4.4	69.1	6.8	80.1	37 400
East	12.1	1.6	6.8	70.4	8.1	85.7	24 800
Central E.	8.7	0.9	6.0	71.1	9.0	86.8	24 300
Atlantic	4.8	0.7	5.0	70.0	6.8	83.7	34 100

not as clear as for the event days (class 0). According to their intensity and clearness, the event days were categorized into three different classes. If there was a clear formation of nucleation mode particles and a distinct growth to Aitken mode size, the days were named class 1. In class 2 events, the formation or particle growth was less intense and in class 3, formation and growth were even much weaker but there were still some signs of new particle formation. Although clear new particle formation events were observed at the station, the frequency of occurrence was lower than at other sites. During the measurement period, about 12 class 1 and 2 events per year were classified, whereas in the boreal forest station Hyytiälä, 42 (Dal Maso et al., 2005) and in the Finnish Lapland station Värriö, about 29 (Vehkamäki et al., 2004) events per year (using similar classification criteria) were observed. In this work, the correlation between event days in different stations was not analysed in detail. For several but not for all the days, an event could be observed in Utö as well as in Hyytiälä. A detailed analysis of the meteorological conditions at these event days has to be done in order to answer the question after a connection between the two stations concerning the new particle formation events and thus how regional the events are. Figure 3 shows the frequency of new particle formation events (class 1–3) as the ratio of event days divided by the number of days with available data for each month of the measurement duration.

In Utö, new particle formation was most probable during spring times (March–April), although a second maximum in fall occurred as well. Practically, no nucleation events could be observed in winter. The fraction of non-event days showed a reverse pattern: the maximum was found in winter while in spring and fall, a minimum appeared. This behaviour is similar to other observations in southern Finland, where a clear annual bimodal variation in the event occurrence was found as well. In Hyytiälä, the highest number of event days was observed in spring (March–May) and a smaller peak in fall (around September), while a minimum occurred in winter (Mäkelä et al., 2000; Dal Maso et al., 2005). In northern Finland, a similar annual pattern was found, although the new particle formation season is shorter (Komppula et al., 2003a,b; Vehkamäki et al., 2004). Thus, seasonal variations in the nucleation mode concentrations can be explained by the annual behaviour of new particle formation.

3.2. *Influence of air mass types*

3.2.1. Wind sector analysis. Similar to meteorological conditions in different seasons, also the advection of different air masses can have a large impact on the ambient aerosol. The total number and $PM_{2.5}$ concentrations were found being slightly higher during southerly wind direction. In spring, somewhat higher total number concentrations were observed during winds from southeast and higher $PM_{2.5}$ concentrations during winds from east to southeast. The median particle size distributions and their fits as well as the 25 and 75% percentiles are displayed in Fig. 4. In Table 3, the results from calculating modal fits to the median particle size distributions according to the wind sector separation are shown as well.

A nucleation mode was found only during northerly transport or advection from central Europe, but for the central European wind sector the number concentration (196 cm^{-3}) was much lower and the geometric mean diameter (∼30 nm) was slightly larger compared to the northerly sector (486 cm^{-3} and ∼25 nm, respectively). This is probably caused by differences in the aerosol loading between the air masses. New particle formation events occurred mostly in clean, northerly air masses, which caused the nucleation mode, whereas the freshly formed particles during advection from central Europe probably originated from anthropogenically emitted precursor gases. Because of higher condensation sinks, this nucleation mode was less conspicuous than in clean air masses with strong new particle formation events.

The Aitken mode particle number concentrations were highest in eastern air masses, which is due to a big number of sources over eastern Europe. In the northern sector, there are only few sources, which resulted in the lowest Aitken mode concentrations. A detailed discussion of the source regions can be found in Section 3.2.2 Also the geometric mean diameter and

Table 5. Correlation coefficients (r) between aerosol and meteorological parameters in spring, summer, fall and winter. Bold type highlights $|r|$-values ≥ 0.5.

	N_3	N_{tot}	V_{tot}	$PM_{2.5}$	SO_2	vis.	T	RH
Spring								
N_3	1							
N_{tot}	0.45	1						
V_{tot}	**0.63**	0.45	1					
$PM_{2.5}$	**0.5**	0.33	**0.67**	1				
SO_2	0.16	0.21	0.21	0.44	1			
Vis.	−0.26	−0.15	−0.46	−0.39	−0.01	1		
T	0.1	0.23	0.04	0.15	−0.11	−0.09	1	
RH	0.09	0.11	0.18	0.0	−0.17	**−0.62**	0.06	1
Summer								
N_3	1							
N_{tot}	0.44	1						
V_{tot}	**0.5**	0.47	1					
$PM_{2.5}$	0.25	0.23	**0.57**	1				
SO_2	0.26	0.42	0.27	0.26	1			
Vis.	−0.14	−0.08	−0.46	−0.38	−0.02	1		
T	0.22	−0.02	0.37	0.19	0.11	−0.02	1	
RH	0.03	0.05	0.22	0.25	−0.04	**−0.59**	−0.29	1
Fall								
N_3	1							
N_{tot}	**0.51**	1						
V_{tot}	**0.69**	0.47	1					
$PM_{2.5}$	0.28	0.14	0.43	1				
SO_2	0.2	0.21	0.28	0.17	1			
Vis.	−0.38	−0.25	**−0.59**	−0.27	−0.11	1		
T	0.3	**0.57**	0.28	0.07	−0.11	−0.21	1	
RH	0.28	0.15	0.39	0.17	0.06	**−0.69**	0.19	1
Winter								
N_3	1							
N_{tot}	**0.61**	1						
V_{tot}	**0.82**	**0.59**	1					
$PM_{2.5}$	**0.7**	0.41	**0.84**	1				
SO_2	**0.65**	0.45	**0.6**	**0.61**	1			
Vis.	−0.43	−0.22	**−0.51**	−0.49	−0.26	1		
T	−0.2	−0.04	−0.13	−0.12	−0.44	−0.22	1	
RH	0.2	0.01	0.24	0.23	0.08	**−0.69**	0.28	1

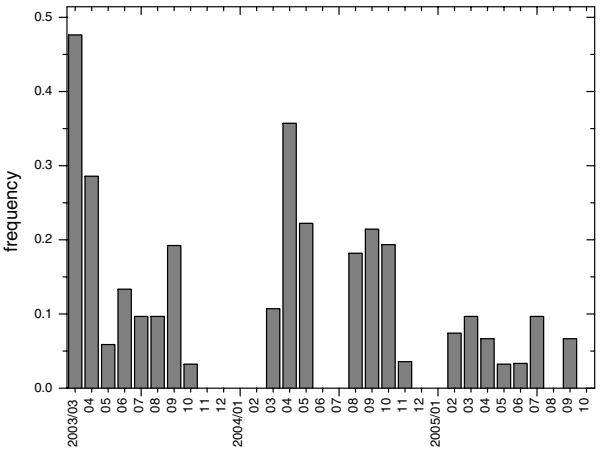

Fig. 3. Frequency of observed new particle formation events (class 1–3) during the measurement period. For June and July 2004 no data is available.

standard deviation showed significant variations between different wind sectors. The smallest mean diameter (54 nm) and standard deviation (1.49) were found in northern air masses, whereas the highest values were found in eastern air masses (71 nm and 1.77). This points to the emission of primary particles and volatile precursor gases in industrial regions. These gases can condense on the available particle surfaces, which results in increased particle sizes. Also in the central European and Atlantic wind sector, a clear Aitken mode could be seen. As in case of easterly advection, also the Aitken mode in central European air masses was probably a result of anthropogenic pollution. The Aitken mode particles in Atlantic air masses, in contrast, are assumed to originate from biogenic activity in the ocean. Especially in spring and summer, the north Atlantic contributes a significant amount of Aitken mode particles (e.g. Cavalli et al., 2004; O'Dowd et al., 2004). Furthermore, the D_g of 51 nm observed in the present study is in fair agreement with the Aitken mode diameter of 46 nm found by O'Dowd et al. (2004) during a period of high biological activity at the marine research station Mace Head.

The highest accumulation mode concentrations were found in the central European wind sector (469 cm^{-3}), whereas the lowest number concentrations were observed in the Atlantic sector (132 cm^{-3}). This seems reasonable, since stronger sources over eastern Europe compared to marine background conditions are expected (see Section 3.2.2). Furthermore, in easterly air masses the Aitken and accumulation modes were more or less merged into one single mode. This is an indication for industrial pollution, since only aged air masses show a typical ('Hoppel') minimum between these two modes due to cloud processing (Hoppel et al., 1985). Air masses from central Europe showed a similar pattern, only the number concentrations in the accumulation mode were higher. Only in northern and Atlantic air masses, a clear accumulation mode shoulder could be observed. Also the difference in the mean diameter ($D_{g,3}$) between eastern (213 nm) and northern (157 nm) advection points to different source types. A possible explanation for the accumulation mode in Atlantic air masses ($D_g = 200$ nm) is the formation of a particle mode with a dry median diameter of about 200 nm by film droplets (O'Dowd et al., 1993), since there are no industrial sources along the trajectories of this sector.

The ratio of Aitken to accumulation mode number concentrations showed big differences concerning the origin of air mass. The smallest value was found in northern air masses (2.8) which is explainable by only few sources for Aitken mode particles in this region. Anthropogenic pollution in both Aitken and

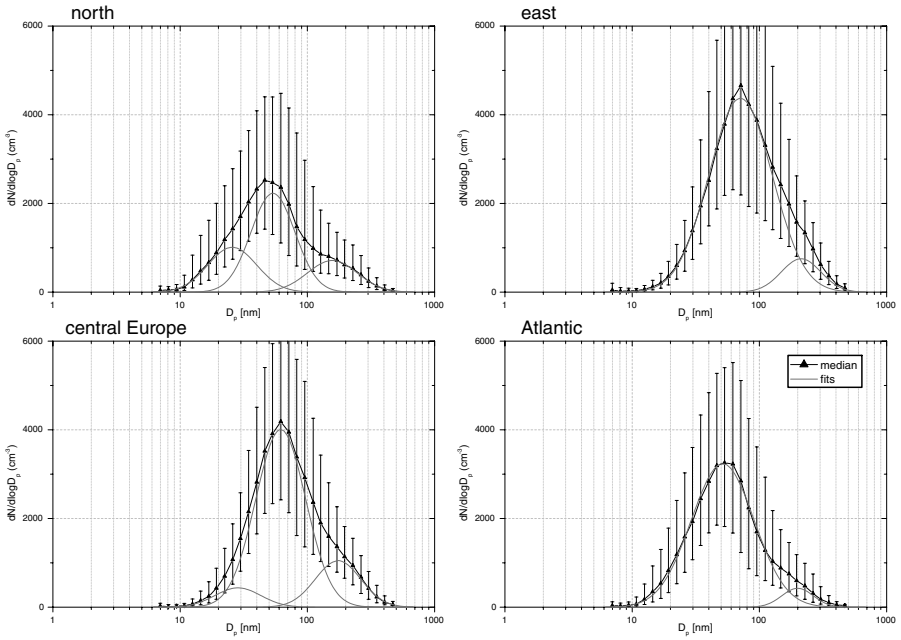

Fig. 4. Median particle size distributions according to different wind sectors and corresponding log-normal fits. Error bars indicate 25 and 75% percentiles.

accumulation mode caused values of 4.2 and 9.3 during central European and eastern advection, respectively. The high value of 15.3 in Atlantic air was maybe caused by the strong biogenic production of Aitken mode particles by the sea (e.g. Cavalli et al., 2004; O'Dowd et al., 2004), which is also consistent with the high N_2/N_3 ratio in summer. Compared to that, the contribution to the accumulation mode was very weak. The total number concentration was lowest in northern (1805 cm^{-3}) and Atlantic (2139 cm^{-3}) air masses and highest during easterly advection (3018 cm^{-3}). The values of central European air masses were situated between these extremes (2642 cm^{-3}).

Regarding the advection direction, also variabilities concerning the average values of meteorological and trace gas parameters were found, which are summarized in Table 4. In eastern and central European air masses, higher values of $PM_{2.5}$ (about 12 $\mu g\,m^{-3}$) and a lower average visibility (about 25 km) were found. As already figured out in Section 3.1.2, there was a connection between these two parameters. The temperature was slightly higher in these air masses. During advection from the north or the Atlantic, the air was much cleaner, which resulted in lower $PM_{2.5}$ (about 4 to 5 $\mu g\,m^{-3}$) and higher average visibility (about 37 km) values. Northerly advection also resulted in slightly lower relative humidities, since the cold polar air is (at least absolutely) dryer than the warmer air from middle latitudes. During the southward transport to the station, the air is heated and thus the relative humidity is decreasing. This cold air advection from the north also resulted in slightly lower temperatures than during advection from the south or east. NO_x as an indication for traffic influence was not surprisingly slightly lower in northern and Atlantic air masses (4.4 $\mu g\,m^{-3}$) compared to advection from the south or east (6.8 $\mu g\,m^{-3}$). Also SO_2 showed the lowest values in Atlantic air masses (0.7 $\mu g\,m^{-3}$) and the highest in eastern air (1.6 $\mu g\,m^{-3}$), which points to industrial activities in this region, since SO_2 is mostly emitted by industries. No variations of the ozone concentration regarding air mass advection were found.

3.2.2. Source region analysis. In order to identify possible sources of pollutants affecting the concentrations at the measurement site, a method assigning the available back trajectories and pollutant concentrations (Stohl, 1996) was applied. The calculations were carried out for different aerosol quantities (number concentration and geometric mean diameter in each fitted mode, total fine number and volume concentrations, $PM_{2.5}$) and trace gas parameters (SO_2 and O_3 concentrations). For the purpose of achieving most detailed results, a separation between spring/summer (March–September) on the one hand, and fall/winter (October–February) on the other hand was made. Unfortunately, a separation into four seasons as in Section 3.1 was impossible because of too few data, since all regarded grid cells had to be crossed by at least 10 trajectories. The resulting concentration charts for the modal number concentrations are displayed in Fig. 5 and for the total number, SO_2 and O_3 concentrations in Fig. 6.

For all considered parameters, there was a clear seasonal variation observable. Figure 5 (chart on top, left) shows, that high nucleation mode particle number concentrations were most probable during advection from three different areas: polar regions, the Atlantic or the Great Britain region. As already shown in

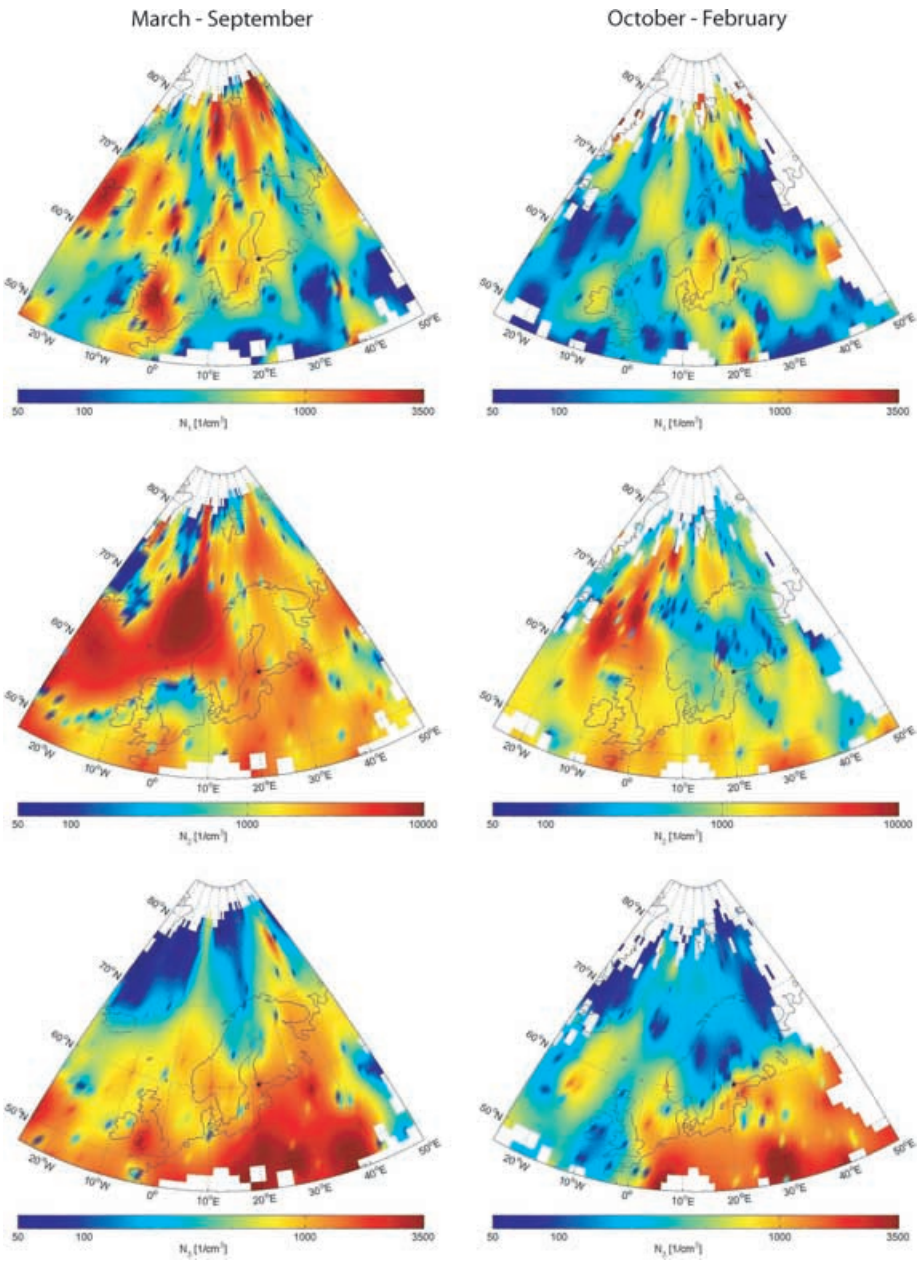

Fig. 5. Spatial distribution of sources according to nucleation (top), Aitken (middle) and accumulation (bottom) mode number concentrations for the warm (left column) and cold (right) season. The colours represent the average concentrations according to back trajectory analysis.

Fig. 3, the frequency of new particle formation events was highest in spring and early fall, which explains the much higher concentrations in the March–September plot. The dominant role of arctic and polar air masses for new particle formation was already shown by Sogacheva et al. (2005) for the forest station Hyytiälä and also for other different continental sites in Finland by Nilsson et al. (2001). Another maximum can be seen in the north Atlantic region, which is probably connected to the much stronger maximum in the Aitken mode in this region, which will be discussed in the following paragraph. In air masses originating from the Great Britain area, a high number of nucleation mode particles was observed as well. A weak maximum in the SO_2 concentration could be seen in this region, too. It seems that both sulphuric acid (Kulmala et al., 2004) and organics like sesquiterpenes (Bonn and Moortgat, 2003) are needed to form new particles under suitable atmospheric conditions. However, the atmospheric residence time of 1–11 d for SO_2 (e.g. Finlayson-Pitts and Pitts, 2000) and up to 6 d for sulphate (Koch et al., 1999) (which is the product of SO_2 cloud-phase reactions) is sufficient to be transported from the considered area to the station.

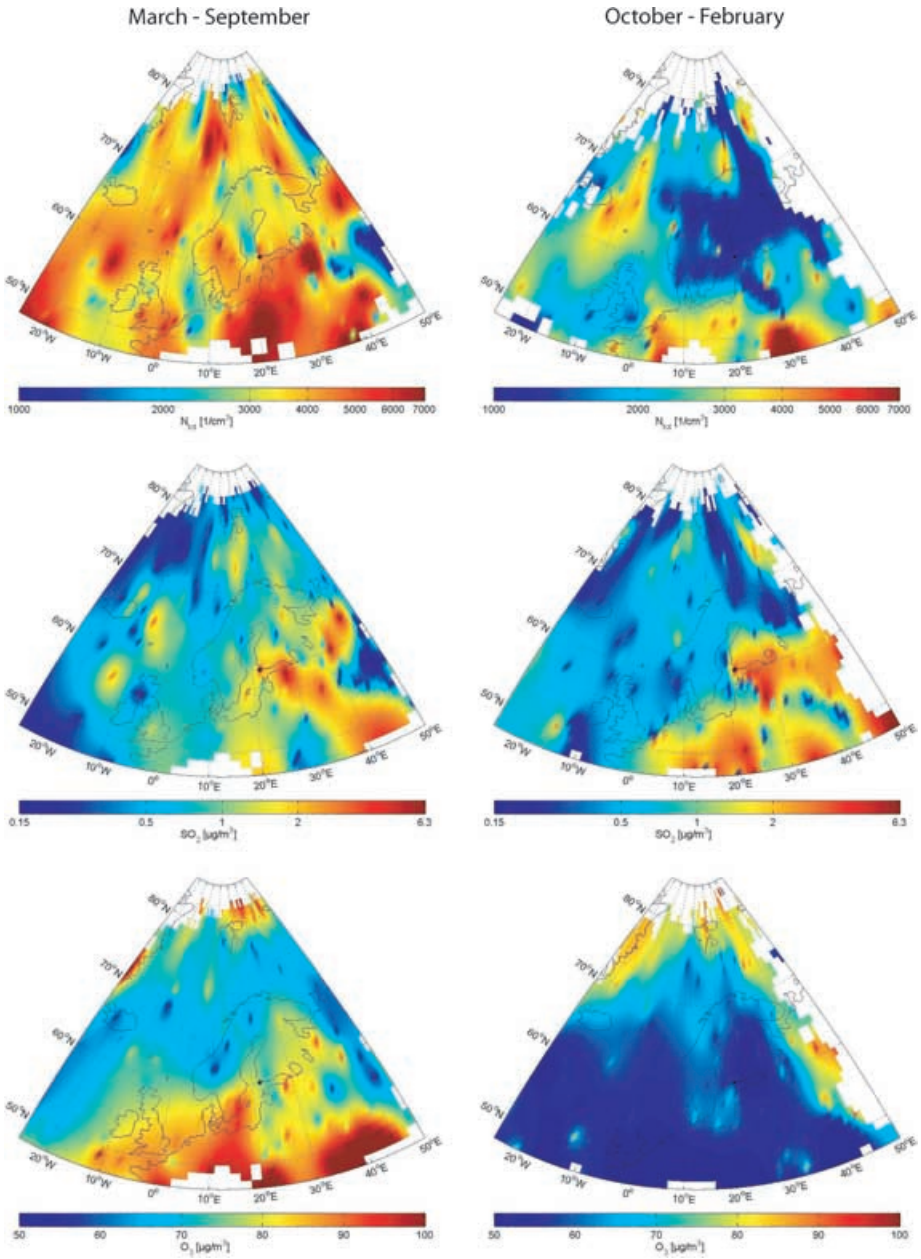

Fig. 6. Spatial distribution of sources according to the total fine particle number concentration (top), SO_2 (middle) and O_3 (bottom) for the warm (left column) and cold (right) season. The colours represent the average concentrations according to back trajectory analysis.

Unfortunately, data for sulphate are only available in an insufficient time resolution, which made a similar analysis impossible. The Scandinavian continent as a forest area is a source of organic compounds, which can be taken up by air masses, originating from the UK region, and thus already containing SO_2 and sulphate. The small nucleation mode concentration maximum over southern Sweden is an indication for that. In this manner, organics, SO_2 and sulphate can be transported to the measurement site and nucleation can take place (by one of the competing processes of biogenic induced or sulphuric acid nucleation or a combination of both), which leads to the observed new particle formation events. New particle formation is a very complex process, which is not yet completely understood and further investigations concerning this topic are still needed. Low nucleation mode number concentrations in the entire regarded area also points to a low probability of new particle formation events during winter.

Concerning the distribution of the Aitken mode particle number concentrations, shown in Fig. 5, a strong maximum over the north Atlantic is most conspicuous and determining the appearance of the Aitken mode at the station. As pointed out by

Mårtensson et al. (2003) and Geever et al. (2005), bursting of air bubbles produced by breaking waves is a major source of primary marine aerosols. Cavalli et al. (2004) and O'Dowd et al. (2004) showed, that the northeast Atlantic is one of the most biologically active oceanic regions and therefore an important biogenic source of particles. Especially in spring and fall, organic matter contributes (together with sea salt) largely to the aerosol particle concentration. Compared to the October–February chart in Fig. 5, slightly larger particle sizes were found (not shown here), which is in agreement with O'Dowd et al. (2004), who found larger particle diameters in the biologically active season as well. Also a smaller maximum over the Baltic Sea was observed, which could be an indication for biological activities in this region. On the other hand, this could also be a result of the very high concentrations transported from the northeast Atlantic region. The weak maximum over the polar sea was caused by new particle formation and thus the maximum of nucleation mode particles in this area. In the northeastern Europe region, a weak maximum was found, too, which was probably linked to the accumulation mode concentration maximum in this area. The winter chart shows a much weaker source in the northeast Atlantic region. Bubble bursting was still a source of primary aerosol particles, but the contribution of organic matter was probably much lower as a result of low biological activity.

The sources of accumulation mode particles were situated in the central to northeast European and Great Britain area. The source maps show a similar pattern for both the summer and the winter part of the year, although the concentrations were generally higher in the warm season. During the warm season there are a lot of wild forest fires in Eastern Europe and Russia (Niemi et al., 2005), which probably increases the sources from those areas. High accumulation mode particle concentrations at the coast of Norway and west and southwest of Great Britain for the warm and west of Great Britain for the cold season are most likely related to the same sources that produce the high Aitken mode particle concentrations over these areas.

The total volume of the fine particle range (V_{tot}) and the mass concentration ($PM_{2.5}$) are determined by the accumulation mode particles. For this reason, the results for the $PM_{2.5}$ and the calculated total volume concentrations are not displayed here, since they show the same pattern as the accumulation mode source field.

In the source area chart of the total fine particle number concentration shown in Fig. 6, all the maxima and minima described above can be retrieved. The effects of the three regarded modes are superposed. For the warm season, the elevated concentrations in the polar region, over the Atlantic and over large parts of Europe, caused by the nucleation, Aitken and accumulation mode particles, respectively, are shown. Figure 6 shows significantly weaker maxima over the northeast Atlantic and eastern Europe for the cold season. Furthermore, very low total fine particle concentrations occur over Fenno-Scandia and especially over Finland for these cold months.

The synopsis of the source maps of the modal and total number concentrations makes clear, that it is not sufficient to analyse only the total number or total mass (like $PM_{2.5}$) concentrations. The latter are not size resolved, whereas the contribution to the number is strongly influenced by the particle size. The mass correlates only with the accumulation mode concentration and the total number is the sum of the modal properties of fine aerosol particles. In order to acquire as detailed information on the aerosol as possible, at least the modal parameters should be considered for further investigations.

As already described above, the atmospheric residence time of a species has a great impact on its transport. Though, a trajectory analysis in order to identify potential source areas only makes sense, if the considered species have a long enough residence time, which is at least in the same order of magnitude as the trajectories used in this study (5 d). Figure 6 also shows the source maps for SO_2 and ozone. The life time of ozone was estimated being about 3 to 18 d (IPCC, 2001) and of SO_2 between several hours and 11 d (Finlayson-Pitts and Pitts, 2000). However, the estimation of the sulphur dioxide concentrations by global models is very complex, since only a certain number of already known processes can be taken into account. Nevertheless, trajectories have been used for studying the behaviour of SO_2 and particulate sulphate by several groups (e.g. Charron et al., 2001; Sciare et al., 2003) and in particular the same method applied in this work was used by Virkkula et al. (1997) for another station. For this reason, we conclude the method being reasonable in order to achieve useful information about the behaviour of these two species as well.

Regarding the SO_2 charts, a seasonal variation does also occur (Fig. 6). Both in the warm and the cold season, a maximum in SO_2 was found over the Baltic states region, which is an indication for industrial activities in this area. In winter, also a second maximum over eastern Europe was observed, which points to industrial emissions as well. Due to blocked vertical mixing and thus favoured horizontal transport in the cold season, this maximum was much stronger during these months compared to the warm season. Even so, this does not mean a stronger source strength in the winter part of the year, but is just a sign for stronger horizontal transport over longer distances compared to the warm season. Also the north Atlantic seemed to make a small contribution to the SO_2 concentration at the station, especially in the warm season. However, it is not completely clear, whether this is caused by anthropogenic pollution, or to DMS emissions of the ocean, which occur stronger in summer (Chin et al., 2000).

DMS is converted into SO_2 by chemical reactions (Finlayson-Pitts and Pitts, 2000) and thus the marine area can (indirectly) be identified as a source of SO_2. But since the contribution of biogenic sulphate in the considered region is relatively low (as reported by Leck et al., 2002; Savoie et al., 2002), anthropogenic pollution seems to be the more probable reason for the slightly elevated SO_2 concentrations over the northeast Atlantic. SO_2 is converted to sulphate/sulphuric acid in clouds, and the

appearance of clouds is very probable in air masses originating from the Atlantic region. This suggests a transport of sulphuric acid from the northeast Atlantic to the present measurement site. Therefore, the elevated SO_2 concentrations in the northeast Atlantic and polar sea area in summer are a possible explanation for the occurrence of nucleation events at the station, because sulphuric acid is supposed to play an initial role in new particle formation (Kulmala et al., 2004).

The source maps of O_3 show a much stronger seasonal variation. The formation of ozone is connected to the abundance of high concentrations of NO_x and volatile organic compounds (VOC) (Seinfeld and Pandis, 1998), while solar radiation is needed as well. High O_3 concentrations were found in air masses originating from western, central or southeastern Europe, which is on the one hand linked to strong emission of NO_x in these highly polluted areas. On the other hand, the southern Scandinavian area is not as polluted as the central European area and thus, elevated ozone concentrations in this region are an indication for the abundance of VOCs—emitted by the forests. However, both VOCs and ozone can be taken up by air masses originating from the Great Britain region and be transported to the station. In those air masses, though, already higher SO_2 and thus sulphate concentrations are present.

As suggested by Bonn and Moortgat (2003), certain organic compounds can initiate nucleation processes and according to Koch et al. (2000) considering new particle formation, the participation of the ozonolysis of terpenes has to be taken into account under suitable atmospheric conditions. Thus, higher SO_2 concentrations from the Great Britain area and higher ozone (and therefore also VOC) concentrations from southern Scandinavia are a possible explanation for high nucleation mode particle concentrations in air masses originating from these regions. In winter, the ozone concentrations were very low in all air masses, since there is less solar radiation and low VOC concentrations available. The source areas in the north could possibly originate from the polar stratospheric ozone intrusions, since the tropopause is very low in this area. In the cold season, there is practically no sunlight available, though the photochemical lifetime of ozone allows the ozone to accumulate (Scheel et al., 1997).

4. Summary and conclusions

Continuous aerosol and trace gas measurements were conducted at the EMEP-station Utö in Baltic Sea from March 2003 to October 2005. Particle number size distributions were measured in the size range of 7–500 nm using a DMPS-system. Meteorological parameters (wind, temperature, relative humidity and visibility), trace gas (SO_2 and O_3) and $PM_{2.5}$ concentrations were determined as well. Log-normal modes were fitted to the size distributions and the data have been separated according to seasons and wind direction sectors. A source region analysis using calculated 120 h back trajectories has been carried out.

Variations concerning seasonal and air mass impacts were studied. Various source types in different directions of the station as well as the seasonal pattern of meteorological parameters were shown to influence many aerosol properties. This might concern parameters like the solar radiation and thus temperature and vertical mixing, cloudiness, precipitation and relative humidity.

New particle formation was observed less frequently than elsewhere in Finland, although conditions leading to particle production seem to be similar to other stations. Nucleation events were found to occur more frequent in spring and fall and during advection from northerly directions. Possibly, the lack of precursor sources close to the station and a higher condensation sink are reasons for the fewer days with new particle formation events compared to the other measurement stations in Finland.

The northeast Atlantic was found to be a considerable source of Aitken mode particles. The Atlantic is a biologically very active region, which can contribute large numbers of primary aerosol particles. Especially in spring and summer, organic matter, produced in this region, is a significant source of primary biogenic aerosol particles.

This study supports that only total particle mass (like $PM_{2.5}$) or number concentrations are not sufficient in order to describe the behaviour of aerosols. The total number is the sum of all particles distributed in the different size modes and the mass is only determined by the aerosols with larger particle sizes. The production of aerosols in the atmosphere is among other things strongly dependent on the particle sizes and thus more detailed analysis of the aerosol properties are required in order to better describe the source and sink regions on earth.

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References

Aalto, T., Hatakka, J., Paatero, J., Tuovinen, J.-P., Aurela, M. and coauthors. 2002. Tropspheric carbon dioxide concentrations at a northern boreal site in Finland: basic variations and source areas. *Tellus* **54B**, 110–126.

Bellouin, N., Boucher, O., Haywood, J. and Reddy, M. S. 2005. Global estimate of aerosol direct radiative forcing from satellite measurements. *Nature* **438**, 1138–1141.

Birmili, W., Wiedensohler, A., Heintzenberg, J. and Lehmann, K. 2001. Atmospheric particle number size distribution in central Europe:

Statitical relations to air masses and meteorology. *J. Geophys. Res.* **106**, 32 005–32 018.

Bonn, B. and Moortgat, K. 2003. Sesquiterpene ozonolysis: Origin of atmospheric new particle formation from biogenic hydrocarbons. *Geophys. Res. Lett.* **30**, 1585, doi:10.1029/2003GL017000.

Cavalli, F., Facchini, M. C., Decesari, S., Mircea, M., Emblico, L. and co-authors. 2004. Advances in characterizationn of size-resolved organic matter in marine aerosol over the North Atlantic. *J. Geophys. Res.* **109**, D24215, doi:10.1029/2004JD005137.

Charron, A., Coddeville, P., Sauvage, S., Galloo, J.-C. and Guillermo, R. 2001. Possible source areas and influential factors for suphur compounds in Morvan, France. *Atmos. Environ.* **35**, 1387–1393.

Chen, Y. and Penner, J. E. 2005. Uncertainty analysis for estimates of the first indirect aerosol effect. *Atmos. Chem. Phys.* **5**, 2935–2948.

Chin, M., Rood, R. B., Lin, S.-J., Müller, J.-F. and Thompson, A. M. 2000. Atmospheric sulfur cycle simulated in the global model GOCART: Model description and global properties. *J. Geophys. Res.* **105**, 24 671–24 687.

Chung, C. E., Ramanathan, V., Kim, D. and Podgorny, I. A. 2005. Global anthropogenic aerosol forcing derived from satellite and ground-based observations. *J. Geophys. Res.* **110**, D24207, doi:10.1029/2005JD006356.

Covert, D. S., Wiedensohler, A., Aalto, P., Heintzenberg, J., McMurry, P. H. and co-authors. 1996. Aerosol number size distributions from 3 to 500 nm diameter in the arctic marine boundary layer during summer and autumn. *Tellus* **48B**, 197–212.

Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T. and co-authors. 2005. Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiälä, Finland. *Boreal Environ. Res.* **10**, 323–336.

Finlayson-Pitts, B. J. and Pitts, J. N. 2000. *Chemistry of the Upper and Lower Atmosphere*, Academic Press, New York.

Geever, M., O'Dowd, C. D., van Ekeren, S., Flanagan, R., Nilsson, E. D. and co-authors. 2005. Submicron sea spray fluxes. *Geophys. Res. Lett.* **32**, L15810, doi:10.1029/2005GL023081.

Hari, P. and Kulmala, M. 2005. Station for measuring ecosystem-atmosphere relations (SMEAR II). *Boreal Environ. Res.* **10**, 315–322.

Hatakka, J., Aalto, T., Aaltonen, V., Aurela, M., Hakola, H. and co-authors. 2003. Overview of the atmospheric research activities and results at Pallas GAW station. *Boreal Environ. Res.* **8**, 365–383.

Haywood, J. and Boucher, O. 2000. Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: a review. *Rev. Geophys.* **4**, 513–543.

Heintzenberg, J., Covert, D. C. and van Dingenen, R. 2000. Size distribution and chemical composition of marine aerosols: a compilation and review. *Tellus* **52B**, 1104–1122.

Hoppel, W. A., Fitzgerald, J. W. and Larson, R. E. 1985. Aerosol size distributions in air masses advecting off the east of the United States. *J. Geophys. Res.* **90**, 2365–2379.

IPCC 2001. *Climate Change (2001): The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change*, (eds.J. T. Houghton, Y. Ding, D. J. Griggs, M. Noguer, P. J. van der Linden, X. Dai, K. Maskell and C.A. Johnson), Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 881 pp.

Koch, D., Jacob, D., Tegen, I., Rind, D. and Chin, M. 1999. Tropospheric sulfur simulations and sulphate direct radiative forcing in the Goddard Institute for Space Studies general circulation model. *J. Geophys. Res.* **104**, 23 799–23 822.

Koch, S., Winterhalter, R., Uherek, E., Kolloff, A., Neeb, P. and Moortgat, K. 2000. Formation of new particles in the gas-phase ozonolysis of monoterpenes. *Atmos. Environ.* **34**, 4031–4042.

Komppula, M., Lihavainen, H., Hatakka, J., Paatero, J., Aalto, P. and co-authors. 2003. Observations of new particle formation and size distributions at two different heights and surroundings in subarctic area in northern Finland. *J. Geophys. Res.* **108**, 4295, doi:10.1029/2002JD002939.

Komppula, M., Dal Maso, M., Lihavainen, H., Aalto, P. P., Kulmala, M. and Viisanen, Y. 2003. Comparison of new particle formation events at two locations in northern Finland. *Boreal Environ. Res.* **8**, 395–404.

Koponen, I. K., Virkkula, A., Hillamo, R., Kerminen, V.-M. and Kulmala, M. 2002. Number size distribution and concentrations of marine aerosols: Observations during a cruise between the English Channel and the coast of Antarctica. *J. Geophys. Res.* **107**, 4753, doi:10.1029/2002JD002533.

Kulmala, M., Hämeri, K., Aalto, P. P., Mäkelä, J. M., Pirjola, L. and co-authors. 2001. Overview of the international project on biogenic aerosol formation in the boreal forest (BIOFOR). *Tellus* **53B**, 324–343.

Kulmala, M., Pirjola, L. and Mäkelä, J. M. 2000. Stable sulphate clusters as a source of new atmospheric particles. *Nature* **404**, 66–69.

Kulmala, M., Vehkamäki, H., Petäjä, T., Dal Maso, M., Lauri, A. and co-authors. 2004. Formation and growth rates of ultrafine atmospheric particles: a review of observations. *J. Aerosol Sci.* **35**, 143–176.

Leck, C., Heintzenberg, J. and Engardt, M. 2002. A meridional profile of the chemical composition of submicrometre particles over the East Atlantic Ocean: regional and hemispheric variabilities. *Tellus* **54B**, 377–394.

Lohmann, U. and Feichter, J. 2005. Global indirect aerosol effects: a review. *Atmos. Chem. Phys.* **5**, 715–737.

Mäkelä, J. M., Koponen, I. K., Aalto, P. and Kulmala, M. 2000. One-year data of submicron size modes of tropospheric background aerosol in southern Finland. *J. Aerosol Sci.* **31**, 595–611.

Mäkelä, J. M., Dal Maso, M., Pirjola, L., Keronen, P., Laakso, L. and co-authors. 2000. Characteristics of the atmospheric particle formation events observed at a boreal forest site in southern Finland. *Boreal Environ. Res.* **5**, 299–313.

Mårtensson, E. M., Nilsson, E. D., de Leeuw, G., Cohen, L. H. and Hansson, H.-C. 2003. Laboratory simulations and parameterization of the primary marine aerosol production. *J. Geophys. Res.* **108**, 4297, doi:10.1029/2002JD002263.

Niemi, J. V., Tervahattu, H., Vehkamki, H., Martikainen, J., Laakso, L. and co-authors. 2005. Characterization of aerosol particle episodes in Finland caused by wildfires in Eastern Europe. *Atmos. Chem. Phys.* **5**, 2299–2310.

Nilsson, E. D., Paatero, J. and Boy, M. 2001. Effects of air masses and synoptic weather on aerosol formation in the continental boundary layer. *Tellus* **53B**, 462–478.

O'Dowd, C. D., Facchini, M. C., Cavalli, F., Ceburnis, D., Mircea, M. and co-authors. 2004. Biogenically driven organic contribution to marine aerosol. *Nature* **431**, 676–680.

O'Dowd, C. D. and Smith, M. H. 1993. Physicochemical properties of aerosols over the northeast Atlantic: Evidence for wind speed related submicron sea salt aerosol production. *J. Geophys. Res.* **98**, 1137–1149.

Ramanathan, V., Crutzen, P. J., Kiehl, J. T. and Rosenfeld, D. 2001. Aerosols, climate, and the hydrological cycle. *Science* **294**, 2119–2124.

Savoie, D. L., Arimoto, R., Keene, W. C., Prospero, J. M., Duce, R. A. and co-authors. 2002. Marine biogenic and anthropogenic contributions to non-sea-salt sulphate in the marine boundary layer over the North Atlantic Ocean. *J. Geophys. Res.* **107**, 4356, doi:10.1029/2001JD000970.

Scheel, H. E., Areskoug, H., Geiß, H., Gomiscek, B., Granby, K. and co-authors. 1997. On the spatial distribution and seasonal variation of lower-troposphere ozone over Europe. *J. Atmos. Chem.* **28**, 11–28.

Sciare, J., Bardouki, H., Moulin, C. and Mihalopoulos, N. 2003. Aerosol sources and their contribution to the chemical composition of aerosols in the Eastern Mediterranean Sea during summertime. *Atmos. Chem. Phys.* **3**, 291–302.

Seibert, P., Kromp-Kolb, H., Kasper, A., Kalina, M., Puxbaum, H., and coi-authors. 1998. Transport of polluted boundary layer air from the Po valley to High-Alpine sites. *Atmos. Environ.t* **32**, 3953–3965.

Seinfeld, J. H. and Pandis, S. N. 1998. *Atmospheric Chemistry and Physics*, John Wiley & Sons, Inc., New York, Chichester, Weinheim, Brisbane, Singapore, Toronto.

Sogacheva, L., Dal Maso, M., Kerminen, V.-M. and Kulmala, M. 2005. Probability of nucleation events and aerosol particle concentration in different air mass types arriving at Hyytiälä, southern Finland, based on back trajectories analysis. *Boreal Environ. Res.* **10**, 479–491.

Stohl, A. 1996. Trajectory statistics - a new method to establish source-receptor relationships of air pollutants and its application to the transport of particulate sulphate in Europe. *Atmos. Environ.* **30**, 579–587.

Stohl, A. 1998. Computation, accuracy and applications of trajectories - a review and bibliography. *Atmos. Environ.* **32**, 947–966.

Stohl, A., Wotawa, G., Seibert, P. and Kromp-Kolb, H. 1995. Interpolation errors in wind fields as a function of spatial and temporal resolution and their impact on different types of kinematic trajectories. *J. Appl. Meteorol.* **34**, 2149–2165.

Tunved, P., Hansson, H.-C., Kulmala, M., Aalto, P., Viisanen, Y., and co-authgors. 2003. One year boundary layer aerosol size distribution data from five nordic background stations. *Atmos. Chem. Phys.* **3**, 2183–2205.

Tunved, P., Nilsson, E. D., Hansson, H.-C., Ström, J., Kulmala and co-authors. 2005. Aerosol characteristics of air masses in northern Europe: Influences of location, transport, sinks, and sources. *J. Geophys. Res.* **110**, D07201, doi:10.1029/2004JD005085.

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

van Dingenen, R., Raes, F., Puteaud, J.-P., Baltensperger, U., Charron, A. and co-authors. 2004. A European aerosol phenomenology - 1: physical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe. *Atmos. Environ.* **38**, 2561–2577.

Vehkamäki, H., Dal Maso, M., Hussein, T., Flanagan, R., Hyvärinen, A. and co-authors. 2004. Atmospheric particle formation events at Värriö measurement station in Finnish Lapland 1998-2002. *Atmos. Chem. Phys.* **4**, 2015–2023.

Virkkula, A., Hillamo, R. E., Kerminen, V.-M. and Stohl, A. 1997. The influence of Kola Peninsula, continental European and marine sources on the number concentrations and scattering coefficients of the atmospheric aerosol in Finnish Lapland. *Boreal Environ. Res.* **2**, 317–336.

Weingartner, E., Nyeki, S. and Baltensperger, U. 1999. Seasonal and diurnal variation of aerosol size distributions ($10 < D < 750$ nm) at a high-alpine site (Jungfraujoch 3580 m asl). *J. Geophys. Res.* **104**, 26 809–26 820.