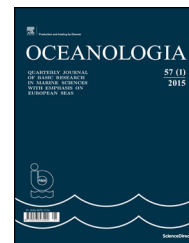




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ORIGINAL RESEARCH ARTICLE

Significant increase of aerosol number concentrations in air masses crossing a densely trafficked sea area[☆]

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Summary In this study, we evaluated 10 months data (September 2009 to June 2010) of atmospheric aerosol particle number size distribution at three atmospheric observation stations along the Baltic Sea coast: Vavihill (upwind, Sweden), Utö (upwind, Finland), and Preila (downwind, Lithuania). Differences in aerosol particle number size distributions between the upwind and downwind stations during situations of connected atmospheric flow, when the air passed each station, were used to assess the contribution of ship emissions to the aerosol number concentration (diameter interval 50–400 nm) in the Lithuanian background coastal environment.

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A clear increase in particle number concentration could be noticed, by a factor of 1.9 from Utö to Preila (the average total number concentration at Utö was 791 cm^{-3}), and by a factor of 1.6 from Vavihill to Preila (the average total number concentration at Vavihill was 998 cm^{-3}). The simultaneous measurements of absorption Ångström exponents close to unity at Preila supported our conclusion that ship emissions in the Baltic Sea contributed to the increase in particle number concentration at Preila.

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1. Introduction

Aerosol particle emissions from global shipping might contribute to 60,000 premature deaths according to a health impact assessment (Corbett et al., 2007), and the emissions are thought to lead to a global temperature decrease due to an increased emission of sulfate particles (Bieltdvedt Skeie et al., 2009). The contribution of particles is generally not extensively quantified, for instance how much the ships contribute to the number of particles and the soot concentrations. One study shows that when air is transported across one major shipping lane in the North Sea, it can increase the daily averaged particle number concentration by 11–19% at a coastal station 1 h downwind of the shipping lane (Kivekäs et al., 2014).

This study has a slightly different focus, and examines the influence that ships have on the particle number size distribution (PNSD) when the air travels along several shipping lanes and encounters multiple ship plumes. For this purpose, particle concentrations have been compared between two field stations upwind of the shipping lanes, and one station downwind of the shipping lanes several hundred kilometers from the upwind stations. In the Baltic Sea, two suitable upwind stations were found, Vavihill in southern Sweden, and Utö, an island in the Finnish archipelago. Preila in Lithuania was chosen as downwind station.

In Section 3, it will be quantified how much the particle number concentration in the size range 50–400 nm diameter increased during transport from the upwind stations to the downwind station. However, not only ship emissions affect the particles in this size range as the air is transported several hundred kilometers over the Baltic Sea. There could be in total 5 or more factors contributing to the increase: (1) Boundary layer evolution could affect PNSD; (2) Condensational growth of new or pre-existing particles can contribute to the particle concentration in the focused size ranges of the PNSD; (3) Land based emissions between the upwind stations and downwind station; (4) Emissions of sea spray aerosol particles; (5) Aerosol particle emissions from ships. In Section 4, the influence of each of the first 4 factors will be discarded as potentially significant for the observed increase in particle number concentration. It will instead be shown that the particle size distribution properties match well with those of aged ship emissions as indicated by the trajectory pathways. Measurements of the absorption Ångström exponent will further confirm the influence of ship emissions.

2. Material and methods

2.1. Measurement stations

Two background stations upwind of the shipping emissions were used, Utö in Finland and Vavihill in Sweden. The background station downwind of the shipping lanes was Preila in Lithuania (Fig. 1).

The Utö station ($59^{\circ}47'N$, $21^{\circ}23'E$, 8 m above sea level, Hyvärinen et al., 2008) is located on a small island in the Baltic Sea about 60 km from the Finnish south-west coast and more than 10 km from the nearest inhabited islands. Turku, the closest town, is about 90 km to the north-east. The island is almost tree-free. Engler et al. (2007) has shown that at the Utö site, air masses with trajectories prevailing from northern sectors (320° – 40°) are considered to be unaffected by anthropogenic land-based sources.

The Vavihill station ($56^{\circ}01'N$, $13^{\circ}09'E$, 172 m above sea level) (Kristensson et al., 2008), is located in southern Sweden,

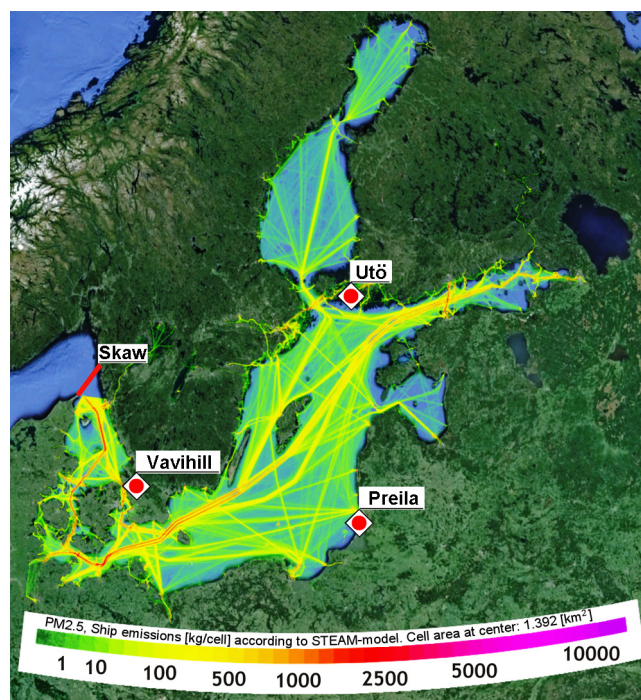


Figure 1 Shipping lanes and Skaw line in the Baltic Sea using Automatic Identification System (AIS, HELCOM, 2013), as well as location of the stations used in this study.

where the surroundings are dominated by deciduous forest. The densely populated areas of Helsingborg and Malmö town and the city of Copenhagen are located 25 km to the west, 50 km to the south, and 45 km to the south-east, respectively.

The Preila station (55°55'N, 21°00'E, 5 m above sea level, Ulevicius et al., 2010) is located in western Lithuania on the Curonian Spit, which is a narrow sandy strip that separates the Baltic Sea and the Curonian Bay. The dunes, up to 50 m height, as well as natural coniferous and dwarf pine trees in low-lying lands dominate in the region. The nearest towns are Klaipėda, 40 km to the north, and Kaliningrad, 90 km to the south.

All stations in this study can be considered to be representative for regional background aerosol measurements because the air is not affected by significant industrial or residential activities in the vicinity of the station. This means that measured aerosol particles are from long range transport, or produced by natural processes. At Preila only the air arriving from the Baltic Sea sector was studied excluding the nearby land based sources. At all stations, the few cases when local pollution sources were evident (high aerosol number concentration increase in certain size bins with short duration) were excluded from the data analysis. The particle number size distribution was measured both before crossing the Baltic Sea, at upwind stations and after the air had crossed the Baltic Sea, at the downwind station. This made it possible to separate the emissions taking place over the Baltic Sea from the pre-existing particle populations at the upwind sites.

Measurement data were collected between September 2009 and June 2010 on these three stations, since there was simultaneous overlap during longer periods in this date interval. PNSD and PM_{0.4} (particle mass concentrations for particles between 50 and 400 electrical mobility diameter) as well as the absorption Ångström exponent (only Preila and Utö) were measured and calculated using recorded light attenuation at the stations, respectively.

2.2. PNSD measurements and PM_{0.4} concentrations

Three mobility particle size spectrometers, operating on the same measurement principle (Wiedensohler, 1988), were used to obtain particle number size distributions at the three stations: a Differential Mobility Particle Sizer (DMPS) in Utö (Hyvärinen et al., 2008), a Twin Differential Mobility Particle Sizer (TDMPs) in Vavihill (Kristensson et al., 2008), and a TROPOS-type Scanning Mobility Particle Sizer (SMPS) in Preila. The inter-comparability of the data set is improved by the fact that the measurements were performed within the frame of the European research infrastructure projects EUSAAR

(European Supersites for Atmospheric Aerosol Research) and ACTRIS (Aerosols, Clouds, and Trace gases Research Infrastructure Network) as well as the EMEP (European Monitoring and Evaluation Program). The stations were audited by the respective authorities and the instrument inter-comparison was performed annually to ensure high quality and comparable data as required by the individual projects. The stations are visited repeatedly for regular maintenance checks. The uncertainty in particle number concentration is estimated to be within the ranges reported by Asmi et al. (2011) and Wiedensohler et al. (2012). The Differential Mobility Analyzer (DMA) type and length, condensation particle counter (CPC) type, observable particle size range, number of size bin steps, time resolution, and flow rates are given in Table 1.

In Section 3, only PNSD and total number concentrations in the size range between 50 and 400 nm diameter are reported. The total number concentration in this range was integrated numerically from the measured size distributions. An enhanced number concentration of aged particles in the atmosphere from ship emissions is expected to be found partly in this size range (Kivekäs et al., 2014). Even though particles from ship emissions are found also in sizes below 50 nm, the size range below 50 nm diameter was not analyzed here because of influence of new particle formation and lower instrument inter-comparability due to diffusion losses (Wiedensohler et al., 2012). The counting statistic is poor for particles above 400 nm diameter due to the low ambient number concentration which causes relatively more noise in this size range (compare with Kivekäs et al., 2014), and is the reason the particle size range above 400 nm was not included in the analysis. The PNSD have been fitted into three log-normal modes using a least-squares fitting algorithm described by Birmili (2001).

PM_{0.4} concentrations have been calculated from the PNSD assuming spherical particles and a density of 1500 kg m⁻³. The density is in reasonable agreement with ambient combustion particles measured in Copenhagen, Denmark (Rissler et al., 2014).

2.3. Light absorption measurements

At the Utö and Preila stations, two identical seven wavelength (370, 450, 520, 590, 660, 880, and 950 nm) aethalometers were used to determine the absorption Ångström exponent. The instrument uses the light transmission through a filter with collected aerosol particles to estimate the aerosol absorption coefficient, σ_{abs} at each given wavelength. To compensate for particle filter loading effects, the same empirical algorithm was used for both instruments (Müller et al., 2011; Virkkula

Table 1 Aerosol PNSD measurement instruments and their specifications at sampling sites.

Site	PNSD instrument	DMA type (length [cm]/type)	CPC type	Size range [nm]	Size steps	Time resolution [min]	Flow rates (aerosol/sheath [lpm])
Preila	SMPS	28/Hauke	UF-02proto ^a	8.7–839.6	71	5	1/10
Utö	DMPS	28/Hauke	TSI 3010	7.0–500.0	30	5	1/10
Vavihill	TDMPs	28 and 11/Hauke	TSI 3760 and 3025	3–900	37	10	1.5/19 and 0.91/5.9

^a Custom build CPC (Mordas et al., 2005).

et al., 2007; Weingartner et al., 2003). However, because of possible high uncertainties due to applied corrections we will not report black carbon mass concentration in this study. On the other hand, Sandradewi et al. (2008), Ulevicius et al. (2010) and others have shown that the absorption Ångström exponent is a useful quantity to represent the origin of carbonaceous aerosol particles. As the light absorption coefficient decreases monotonically with wavelength, it can be approximated by a power-law expression, $\sigma_{abs} \sim \lambda^{-\alpha}$, where α is the absorption Ångström exponent. The absorption Ångström exponent was obtained by a power-law fit over all seven aethalometer wavelengths.

2.4. Selection of trajectory cases

The hourly trajectories (Fig. 2) at two different altitudes of 100 and 300 m above sea level, representing the Baltic Sea atmospheric boundary layer (Gryning and Batchvarova, 2002), were calculated using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT4) model (Draxler

and Rolph, 2003) with the Final Analyses (FNL, 2008–2009) and the Global Data Assimilation System (GDAS) meteorological databases at the NOAA Air Resources Laboratory's web server (Rolph, 2003).

To compare the particle properties between the downwind and upwind stations, only those trajectories passing the entire pathway between the stations over the Baltic Sea were selected. A few exceptions, however, were made to this criterion. In Utö to Preila case, some trajectories passed over the island of Gotland (around 40 km in width of rural land) between the upwind and downwind sites. It results in 7% of the integral of the trajectory pathway being over land. For the Vavihill to Preila trajectory cases there is 70–80 km of land directly downwind of Vavihill that constitutes around 16% of overall trajectory distance. The effects of the land based emissions are discussed later in Section 4. The differences of PNSD, PM_{0.4} and α between the stations were calculated for individual trajectory cases by comparing the values measured at Preila to the corresponding values at the upwind sites at the time when the air mass passed the upwind site according to the trajectory. This was done by

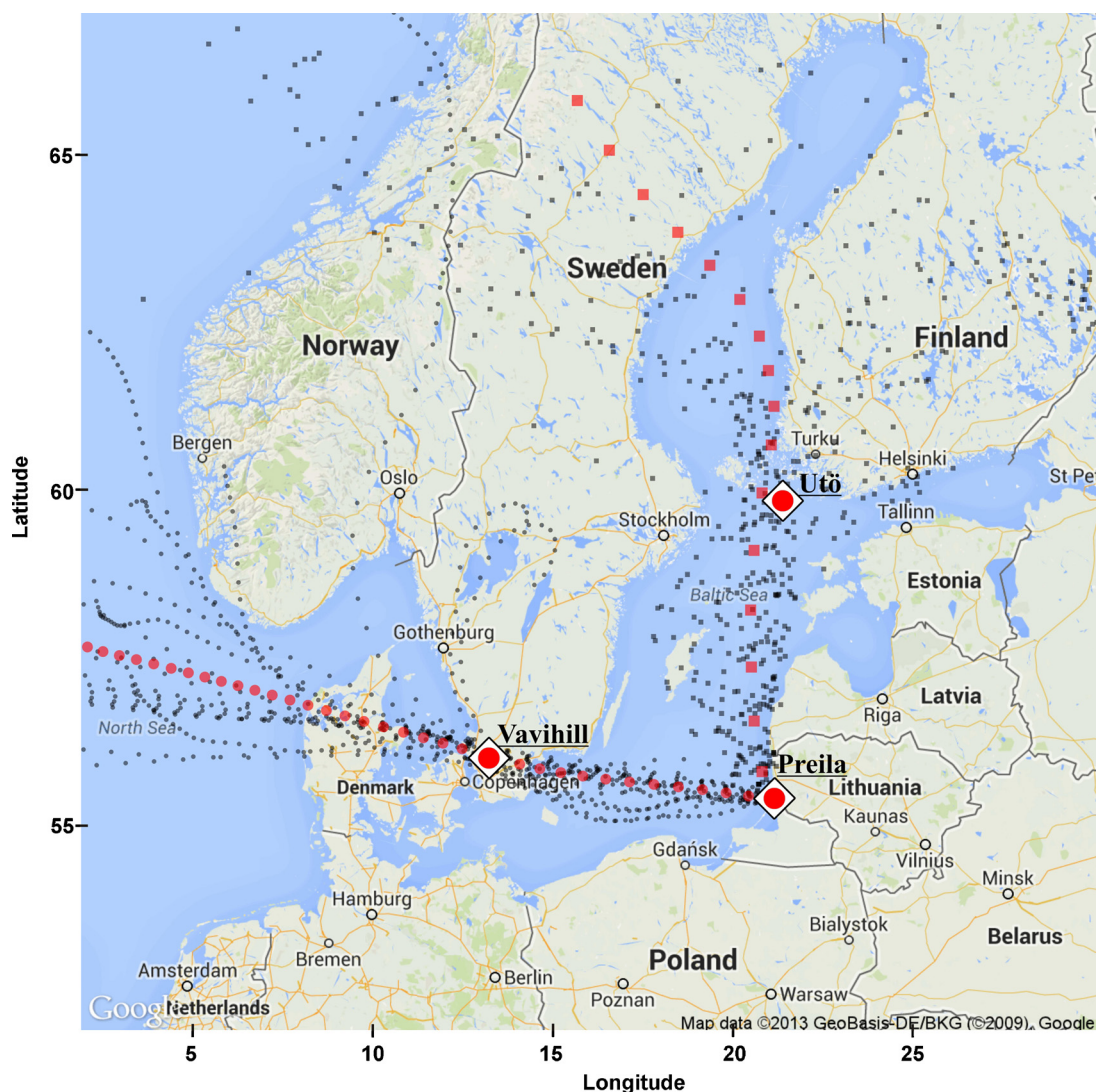


Figure 2 Air mass trajectories passing over the upwind stations and arriving at Preila: smaller dots represent individual hourly trajectories; larger – the mean of trajectories.

calculating the time the air mass needs to travel between the upwind and downwind sites and using this information as a time point at the upwind sites. Also seasonal averages and averages of all trajectory cases were calculated. As the trajectory selection criteria were strict and the simultaneous data coverage of different instruments was not 100%, finding more cases than those described in Section 3 was not

possible. Lowering the criteria for the trajectories, for example allowing longer stretches over land, only resulted in a few more trajectories. These cases were more difficult to analyze, and hence did not improve the analysis and did not improve the statistical accuracy. Moreover, only Vavihill to Preila and Utö to Preila, but not vice versa, air mass transports were analyzed. This is because continental air arriving

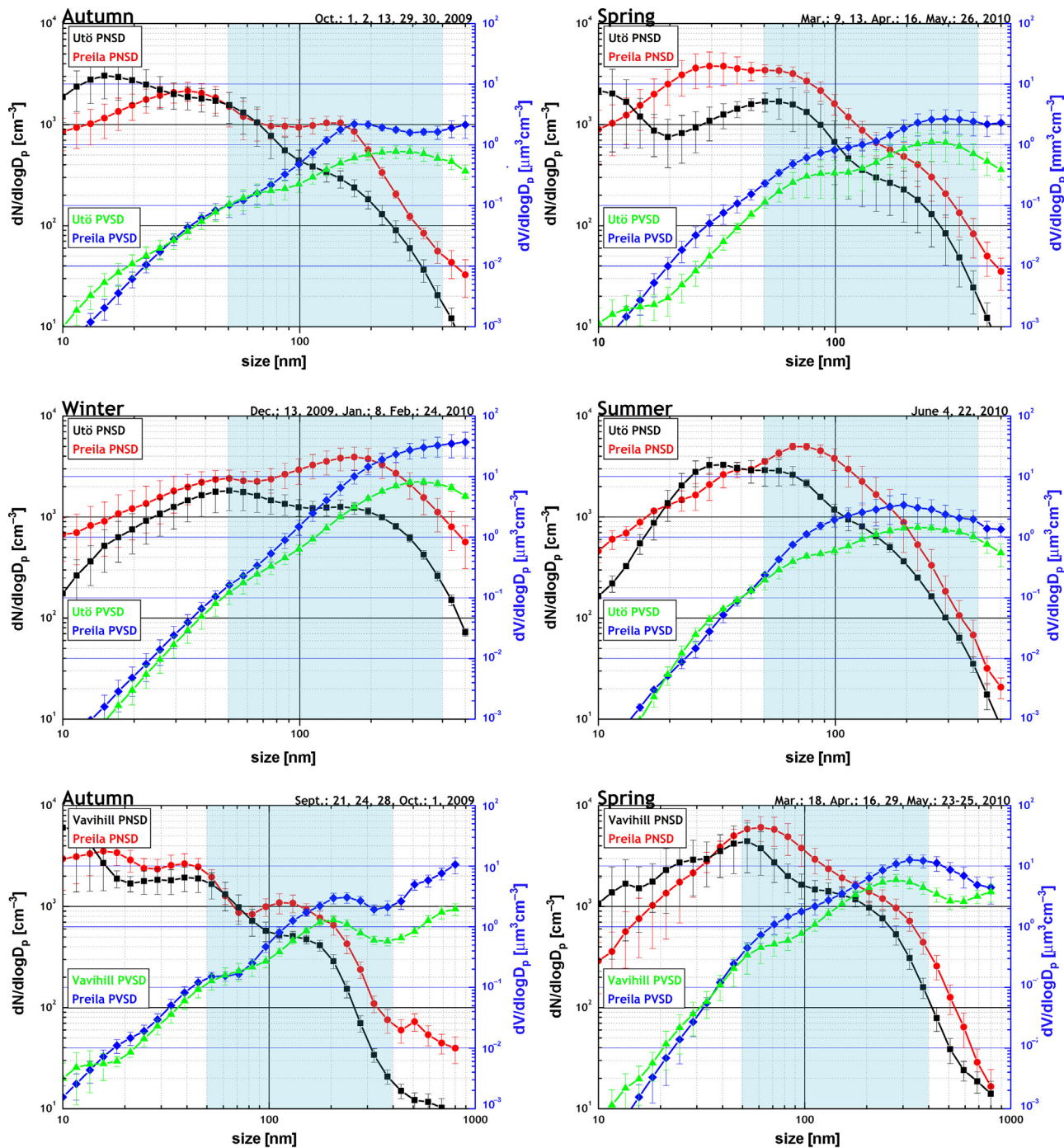


Figure 3 Comparison of average aerosol PNSD and PVSD at Utö, Vavihill, and Preila stations divided by season for our 14 trajectory cases for transport between Utö and Preila and for our 17 trajectory cases for transport between Vavihill and Preila. The 50–400 nm particle diameter range is highlighted with blue color. Here $dN/d\log D_p$ and $dV/d\log D_p$ represent normalized particle number and volume concentrations, respectively. Bars represent a time variability (standard deviation) over the hours sampled. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 2 Average aerosol properties at the three stations by season for the 14 trajectory cases between Utö and Preila and for 17 trajectory cases between Vavihill and Preila.

Parameter	Autumn		Winter		Spring		Summer	
	Utö	Preila	Utö	Preila	Utö	Preila	Utö	Preila
${}^{50-400}N$ [cm^{-3}] ^a	437 ± 260	697 ± 304	1135 ± 446	2219 ± 1350	587 ± 528	1256 ± 411	1006 ± 197	2131 ± 849
${}^{50-400}(N_{\text{Preila}}/N_{\text{Utö}})$		1.6 ± 1.2		1.9 ± 1.4		2.1 ± 2.0		2.1 ± 0.9
PM0.4 [$\mu\text{g m}^{-3}$] ^b	0.7 ± 0.2	1.6 ± 0.3	4.5 ± 0.2	12.7 ± 8.6	0.8 ± 0.8	2.0 ± 1.2	1.3 ± 0.03	2.9 ± 2.2
α ^c	1.2	1.0	1.0	0.9	0.8	1.0	1.0	1.3
Average distance/average time of trajectory [km h^{-1}]		546/18		556/17		575/22		484/15

Parameter	Autumn		Winter		Spring		Summer	
	Vavihill	Preila	Preila	Vavihill	Vavihill	Preila	Preila	Vavihill
${}^{50-400}N$ [cm^{-3}]	508 ± 231	743 ± 238			1489 ± 770	2627 ± 1171		
${}^{50-400}(N_{\text{Preila}}/N_{\text{Vavihill}})$		1.5 ± 0.8				1.8 ± 1.2		
PM0.4 [$\mu\text{g m}^{-3}$]	0.9 ± 0.3	2.0 ± 0.7		No data	3.7 ± 1.1	6.9 ± 2.4		No data
Average distance/average time of trajectory [km h^{-1}]		486/10				502/14		

^a The total aerosol particle number concentration in a range from 50 to 400 nm derived from the PNSD.

^b Integrated total mass concentration (assuming 1500 kg m^{-3} density).

^c Absorption Ångström exponent.

at Preila from inland is highly affected by the continental pollution thus lowering the ratio of Baltic Sea emissions to the total PNSD and thereby making the shipping influence harder to quantify. Also the number of cases when air is transported from Preila to the upwind sites is lower due to the prevailing westerly winds in the region.

Lateral position uncertainties of air mass trajectories are known to vary between 10 and 30% of the trajectory length (Stohl, 1998). The distances from Preila to Vavihill and Preila to Utö are about 490 and 530 km respectively, which results in ± 50 –150 km lateral uncertainty at the distance of the upwind station in trajectories ending at Preila. Within that distance from Vavihill there are several significant centers of population, most notably Copenhagen with a population of almost 2 million people. An aerosol PNSD measured at Preila might carry a significant signal from Copenhagen or other population centers even though the center of the trajectory does not pass through these areas. In such case the same signal would not be present in the Vavihill PNSD. This can cause a difference between the aerosol properties measured at Preila and Vavihill in individual cases. It cannot, however, be assumed that every trajectory calculated to pass over Vavihill passes over Copenhagen in reality. This means that

the influence of Copenhagen can be present in some cases but cannot explain systematic changes on the Vavihill to Preila transport route. The trajectories arriving to Preila from Utö do not have such a problem with trajectory uncertainty and large scale anthropogenic sources, as the anthropogenic sources within 150 km from Utö are minor.

3. Results

After analyzing air mass trajectories and measurement data, 14 and 17 cases were chosen when air was transported directly from Utö to Preila or from Vavihill to Preila, respectively, as shown in Fig. 2. Vavihill to Preila air mass transport cases took place mainly in spring (1 case in March, 2 cases in April, 6 cases in May) and autumn (6 cases in September, 2 cases in October). Utö to Preila cases covered all four seasons: Summer (2 cases in June), Spring (2 cases in March, 1 case in April and 1 case in May), Winter (1 case in December, 1 case in January, 1 case in February) and autumn (5 cases in October). The average PNSD and PVSD (Particle Volume Size Distribution) as well as absorption Ångström exponent for the different seasons at the three sites are shown in Fig. 3 and

Table 3 Aerosol PNSD log-normal fit modal parameters.

Mode parameters	Autumn		Winter		Spring		Summer	
	Utö	Preila	Utö	Preila	Utö	Preila	Utö	Preila
Mode 1								
N_1 [cm ⁻³] ^a	1327 ± 961	695 ± 249	16	311 ± 288	1432 ± 1872	852 ± 864	230 ± 215	–
$\sigma_{g,1}$ ^b	1.6 ± 0.1	1.9 ± 0.2	2.0	1.9 ± 0.1	1.7 ± 0.2	2.0 ± 0.1	2.0	–
$d_{g,1}$ [nm] ^c	12 ± 2	12 ± 1	4	9 ± 2	9 ± 3	9 ± 2	11 ± 2	–
Mode 2								
N_2 [cm ⁻³]	1113 ± 644	1340 ± 682	802 ± 496	1431 ± 791	845 ± 342	2151 ± 1520	1284 ± 253	1588 ± 267
$\sigma_{g,2}$	1.7 ± 0.1	1.7 ± 0.1	1.7 ± 0.1	1.7 ± 0.1	1.7 ± 0.1	1.6 ± 0.1	1.6 ± 0.2	1.6 ± 0.1
$d_{g,2}$ [nm]	36 ± 8	33 ± 7	45 ± 19	43 ± 14	46 ± 17	46 ± 12	49 ± 13	52 ± 2
Mode 3								
N_3 [cm ⁻³]	83 ± 62	366 ± 42	546 ± 43	1854 ± 984	85 ± 114	283 ± 255	113 ± 11	129 ± 47
$\sigma_{g,3}$	1.5 ± 0.3	1.4 ± 0.1	1.6 ± 0.1	1.6 ± 0.1	1.5 ± 0.2	1.5 ± 0.1	1.4	1.4 ± 0.1
$d_{g,3}$ [nm]	179 ± 53	134 ± 12	179 ± 16	169 ± 10	209 ± 35	191 ± 55	186 ± 12	184 ± 4

Mode parameters	Autumn		Winter		Spring		Summer	
	Vavihill	Preila	Vavihill	Preila	Vavihill	Preila	Vavihill	Preila
Mode 1								
N_1 [cm ⁻³]	5665 ± 7547	2922 ± 1359			1053 ± 1569	183 ± 157		
$\sigma_{g,1}$	1.7 ± 0.1	1.5 ± 0.1			1.8 ± 0.2	1.9		
$d_{g,1}$ [nm]	8 ± 1	11 ± 2			10 ± 3	8 ± 2		
Mode 2								
N_2 [cm ⁻³]	875 ± 439	1689 ± 1273			1763 ± 1009	2661 ± 1603		
$\sigma_{g,2}$	1.7 ± 0.1	1.8 ± 0.1		No data	1.6 ± 0.1	1.6 ± 0.1		No data
$d_{g,2}$ [nm]	42 ± 9	33 ± 5			48 ± 8	50 ± 17		
Mode 3								
N_3 [cm ⁻³]	136 ± 69	334 ± 113			526 ± 193	611 ± 265		
$\sigma_{g,3}$	1.4 ± 0.1	1.4 ± 0.1			1.9 ± 0.1	1.5 ± 0.1		
$d_{g,3}$ [nm]	156 ± 22	152 ± 16			170 ± 18	199 ± 36		

^a Mode number concentration.

^b Geometric mode standard deviation.

^c Geometric mode mean diameter.

Table 2. In [Table 2](#) and [Table 3](#) “±” is used for standard deviation and in the ratios $N_{\text{Preila}}/N_{\text{Utö}}$ and $N_{\text{Preila}}/N_{\text{Vavihill}}$ “±” is used as an error from the error propagation formula. The concentrations for each of the 31 cases were calculated from several hour values, which were then used for seasonal averages (in total 69 data points were averaged).

In all seasons and on both transport routes the measured aerosol particle number concentration, in the range from 50 to 400 nm ($^{50-400}N$) was higher by a factor of 1.5–2.1 at Preila than at the upwind station ([Table 2](#)). When looking at individual cases, only 7% of Utö to Preila cases and 10% of Vavihill to Preila cases, respectively, showed a slight decrease in $^{50-400}N$ during the transport. In 67% of Utö to Preila cases and 58% of Vavihill to Preila cases the observed increase in $^{50-400}N$ was more than 50% of the value measured at the upwind site. In absolute terms the (seasonally averaged) increase of $^{50-400}N$ in Utö to Preila air mass transport was 260–1125 cm⁻³, and in the Vavihill to Preila air mass transport 235–1138 cm⁻³. On Utö to Preila transport path PM_{0.4} increased by a factor of 2.5, 2.8, 2.4, and 2.2 for autumn, winter, spring, and summer, respectively. On Vavihill to Preila transport path PM_{0.4} increased by a factor of 2.2 and 1.9 for autumn and spring seasons, respectively. All aerosol PNSDs were also described as a sum of 2 or 3 log-normal modes using an automatic mode fitting algorithm. The modes were defined as: nucleation (particle mean diameter 8–15 nm), Aitken (15–100 nm) and accumulation (100–500 nm) mode. The seasonally averaged modal parameters are presented in [Table 3](#). The increase in N of the accumulation mode is clear in these values.

The Ångström exponent remained around unity (seasonal averages 0.8–1.3) in all cases at both the Utö and Preila sites.

The mobility particle size spectrometers comparability study by [Wiedensohler et al. \(2012\)](#) has shown 5–10% uncertainty in total integrated particle number concentration between the instruments. It can be assumed that a similar range of instrumental uncertainty should be present in this study. However observed increase in $^{50-400}N$ is larger than what the instrumental uncertainty can explain.

The observed changes in $^{50-400}N$ were similar on both transport routes (Utö to Preila and Vavihill to Preila). This was expected because both atmospheric conditions and ship traffic intensity are similar on both routes crossing the main basin of the Baltic Sea.

4. Discussion

4.1. Potential sources of particles. Meteorology: is the change real?

The observed increase in $^{50-400}N$ concentration was significant across both transport routes over the Baltic Sea. A change in observed particle number concentration can, in general, be a result of changed boundary layer (PBL) height (e.g. [Ma et al., 2011](#)). Increased PBL height during daytime mixes the particles near the surface with air above. If the air above the PBL has lower particle concentration, this can result in dilution with time and a decrease in N at surface level and the downwind site. The opposite has also been observed when the air aloft is more polluted ([Clarke et al., 1998](#)). At night the turbulence weakens leaving the particles

equally distributed in the air layer. This does not change N at the surface. New emissions from surface, however, are trapped in the low boundary layer leading to higher increase in N at surface per emission unit compared to the daytime situation. [Gryning and Batchvarova \(2002\)](#) have studied variables which influence boundary layer height over the Baltic Sea. They showed that boundary layer height varies over different time scales, but shows no clear diurnal variation, as seen typically over land cases. This can be explained by the high heat capacity of water making the diurnal variation of sea surface temperature very small and leading to very weak and relatively constant thermal turbulence. Therefore this phenomenon is expected to have an effect only at Vavihill site, which is located inland. In this study the boundary layer height was not examined, but no systematic effects were found when comparing cases in which air had passed the upwind site at daytime and arrived at Preila at night, or vice versa. Also the trajectories at 3000 m altitude showed very similar behavior to those at 100 and 300 m levels, excluding the possibility of significant pollution from the air above. Based on these findings we can assume that the observed systematic changes in $^{50-400}N$ cannot be explained by changes in PBL height only.

The accumulation mode particle number concentrations at both Utö and Preila stations were highest during winter. This might be a result of low boundary layer depths along with stronger inversion conditions and regional scale anthropogenic sources. These sources can be domestic heating in the entire northern Europe. Smaller vertical mixing leads to higher aerosol particle transport efficiency over a longer distance compared to other seasons. This could explain the high values, but not any change during transport between the stations.

4.2. Potential sources of particles. Growth of pre-existing particles and regional new particle formation

Generalized qualitative evidence in particle number concentration changes during transport over the Baltic Sea can be characterized using the relation between the number concentration N of each mode (from [Table 3](#)) in the size distributions and the corresponding modal geometric mean diameter. If each mode is assumed to change as a whole, the particle growth rates required to explain the changes between the upwind stations and Preila can be calculated. This was done for both transport routes.

This study is focused on the particle diameter range from 50 to 400 nm. Therefore, the dynamics of new particle formation and growth to Aitken mode are not examined, even though [Hyvärinen et al. \(2008\)](#) and [Kristensson et al. \(2014\)](#) have shown that there is relatively frequent new particle formation over the Baltic Sea. In case of Utö to Preila transport the nucleation and Aitken modes at Utö, as well as new particle formation along the transport path, could have contributed to the measured accumulation mode at Preila only if particle growth rates of at least 7.4 nm h⁻¹ (for growing the nucleation mode into an accumulation mode) and 5.7 nm h⁻¹ (for Aitken mode to accumulation mode) were assumed (using lowest averaged trajectory speeds). For Vavihill to Preila air mass transport these numbers would have to be as high as 11.9 and 9.3 nm h⁻¹ for

nucleation mode to accumulation mode and Aitken mode to accumulation mode, respectively. Recent studies have shown that mean growth rate (GR) of particles with marine origin is $3 \pm 0.5 \text{ nm h}^{-1}$ (Ehn et al., 2010). This is clearly much less than the growth rates required to explain the changes observed in this study, and therefore the growth of nucleation or Aitken mode particles as the main factor causing the increase in accumulation mode and in $^{50-400}N$ can be ruled out. For new particles formed during the air mass transport the required growth rates would have to be even higher. This rules out the emissions of organics or di-methyl-sulfide from the sea water as an explanation, as well as from new particle formation in ship plumes. The observed changes in the upper Aitken mode and accumulation mode can only be explained by a source of primary emitted particles between the sites.

4.3. Potential sources of particles. Land based emissions between the sites

The air masses advecting over Vavihill pass over a 70–80 km stretch of land between Vavihill and Preila. This can result in significant emissions of primary aerosol particles from land areas east of Vavihill. Even though southern Sweden is densely populated (compared to other parts of Scandinavia), most of the population is located towards west and south of Vavihill. The stretch of land east of Vavihill is mostly used for agriculture and forestry. From this area one can expect emissions from domestic wood combustion and traffic in winter, and from biogenic and traffic sources in summer. These emissions can have an effect on the particle population arriving to Preila from the Vavihill direction. On the transport route from Utö to Preila there is practically no land between the sites, except for the few trajectories passing over Gotland island in the middle of the Baltic Sea. If the land-based emissions were a significant contributor to the observed changes, much smaller changes would be observed on the Utö to Preila transport route. The observed change on Utö to Preila transport path was, however, greater than at Vavihill to Preila transport path in both absolute and relative numbers. Therefore it can be assumed that land based emissions between the sites are not a major or systematic contributor to the observed changes.

4.4. Potential sources of particles. Sea spray

One potential explanation for increased particle number concentration during transport over the Baltic Sea is sea spray; sea salt particles created by breaking waves (Lewis and Schwartz, 2004). Different laboratory methods used to generate surrogate marine aerosols within enclosed tanks confirm the production of accumulation mode particles with geometric mean diameter of 200 nm in marine environment (Stokes et al., 2013).

In this study the contribution of sea salt could not be estimated from chemical composition of the particles, because the only chemical data available was black carbon concentration estimated from absorption measurements. Pettersson et al. (2012) have reported a series of environment fact sheets including monthly wave height variation over a Baltic Sea since 2004. It was shown that the minimum wave

height (0.6 m) is mostly common during the late spring to early fall months. After the height minimum, waves start to grow in height and reach maximum (2.0 m) during winter. Taking into account the relatively low wave heights and low salinity of the Baltic Sea the sea salt emissions are expected to be low.

The influence of sea spray emissions on the number concentration was calculated with a parameterization by Sofiev et al. (2011) using the following assumptions: (a) wind speed at 10 m height is the same as trajectory speed at 100 m altitude; (b) salinity of 9.2‰ (southern Baltic sea); (c) temperature of +5°C in winter, +15°C during other seasons; (d) no deposition or coagulation of sea salt particles; (e) well mixed 300 m boundary layer. Using the average seasonal trajectory speeds (Table 2) the sea salt emissions in a size range between 50 and 400 nm were about 1–2 particles per cm^3 . Using the highest trajectory speed observed in this study, the emissions were slightly above 10 cm^{-3} . To produce the observed increases in aerosol particle number concentration average wind speeds of 50 m s^{-1} would be required. With this in mind, sea salt aerosol particles can explain only a minor fraction of the observed increase in particle number concentration.

4.5. Potential sources of particles. Shipping

The most plausible explanation for the increased $^{50-400}N$ is emissions from ship traffic. The Baltic Sea Skaw line (Fig. 1) was crossed 62,743 times during 2009, so the presence of intensive ship activity is clear. The main shipping lanes on the Baltic Sea can be seen in Fig. 1. Most of the lanes are located at least 100 km from Preila (except the one leading to Klaipeda harbor), so the plumes have had enough time to disperse, and individual peaks (such as in Kivekäs et al., 2014) can no longer be identified. In a recent study of individual ship exhaust plumes measured at a distance of 1 km (Diesch et al., 2013), either uni- or bi-modal aerosol PNSD were found to be common. These modes included an ultra-fine particle mode at around 10 nm consisting mostly of sulfuric acid (González et al., 2011) and an Aitken mode at 40 nm. A notable increase in particle number concentrations at diameter about 150 nm was also found to be common. This carbon-containing mode is made up of mainly soot and absorbed organic materials. This study is focused mostly on the 50–400 nm particle diameter range where the Aitken mode and the carbon-containing mode is the most prominent one. The size of the increased particle number concentrations, as well as the absorption Ångström exponent near unity can be explained by emissions from shipping. The added particles from shipping would probably not be from the ultra-fine mode or new particle formation in the emission plume, as those would require very high growth rates to make it to the observed size range. The observed Ångström exponents at Utö and Preila are consistent with values measured in engine combustion studies, but lower than values associated with wood burning soot (Sandradewi et al., 2008; Ulevicius et al., 2010).

5. General remarks on this study

Most prior particle measurements of ship emissions have focused on laboratory engine studies (e.g. Kasper et al., 2007), single ship plumes (e.g. Petzold et al., 2008), harbor

or single shipping lane influences on coastal areas (e.g. Kivekäs et al., 2014). In a review study by Kumar et al. (2013) there were no studies of measured total ship emissions over a longer stretch of sea. Ship emissions have been modeled on large scales (e.g. Jalkanen et al., 2009) and even globally (e.g. Corbett and Koehler, 2003). When such models are extended past the exhaust pipe of the ship the dispersion, transport, and deposition of the emissions need to be included (e.g. Stipa et al., 2007). For such models long term measurement data of ambient air is needed for comparison.

The main problem with long term measurements of particles downwind of one or several shipping lanes is how to separate the ship emissions from background particle properties. There are two approaches to this. One is to identify the ship plumes in the data at a downwind station and estimate the background particle properties from the non-plume periods (Kivekäs et al., 2014). This approach can only be used close to the shipping lane, where the individual plumes can be separated (Kivekäs et al., 2014; Petzold et al., 2008). The other approach is the one used in this study. If particle properties in the same air mass are measured prior to and after the air trajectory crossing with a ship lane, all changes can be attributed to phenomena taking place between the measurement sites. As discussed above, there are other factors than just ship emissions affecting the particle properties and that need to be taken into account. The longer the air mass travel distance and travel time between the measurement stations, the more uncertainty there is in the results. Also a longer distance between the stations usually leads to fewer cases when air is advected over both stations. This approach is not suitable for detailed process studies, but can reveal the total influence of shipping to a coastal area. This kind of data is needed for validation of model results.

The observed increase in particle number concentration between the upwind stations and Preila was large. In air masses arriving from Utö 37–53% (seasonal averages) of $^{50-400}N$ measured at Preila came from sources and processes during transport between the sites. For air masses arriving from Vavihill to Preila the corresponding range was 30–45%. This increase is the sum of several factors, but as stated above, emissions from ship traffic are likely the dominant contributor. The measured PNSDs at Utö and Vavihill can also be affected by ship emissions outside the main basin of the Baltic Sea, increasing the total contribution from ship emissions even more. The values obtained in this study are larger than the 11–19% contribution observed by Kivekäs et al. (2014) at North Sea. When comparing these studies one should keep in mind that the observed particle diameter range is different, and this study estimates the combined effect of several shipping lanes, whereas Kivekäs et al. (2014) focused on effects from a single shipping lane.

6. Conclusions

This study focuses on the changes in particle properties during transport over the Baltic Sea. All cases with air passing over the Utö measurement station in Finland or the Vavihill measurement station in Sweden, and then later arriving at the Preila measurement station in Lithuania were analyzed. In this way, the same air was measured before and after crossing the main basin of the Baltic Sea. Aerosol particle size

distribution with focus on the 50–400 nm diameter range and absorption Ångström exponent at Preila were compared to those at the upwind sites in the same air mass. The analyzed time period was 10 months (September 2009 to June 2010), allowing qualitative estimation of seasonal variation in the changes. In the authors knowledge no similar study with same air masses measured before and after a long fetch over a heavily trafficked sea area have been published before.

The increase in the number concentration of 50–400 nm particles $^{50-400}N$ was substantial. 26–53% of particles arriving at Preila were generated by processes and emissions taking place between the upwind stations and Preila. The number of analyzed cases was 14 for the Utö to Preila transport path and 17 for the Vavihill to Preila transport path. A clear increase in $^{50-400}N$ was, however, present in almost all studied cases.

The observed increase was the sum of all emissions and processes taking place during the air transport between the sites. These include differences in boundary layer height, growth of pre-existing particles or new particles formed between the sites, land-based emissions between the sites, sea salt emissions, and emissions from ship traffic. The potential contribution of each source was discussed, and shipping was found to be the only source that could explain most of the change. Furthermore, the observed changes were in line with other published ship emission studies.

To reduce the particle emissions from shipping, the International Maritime Organization (IMO) has restricted the sulfur content in ship fuels globally, and especially in defined Sulfur Emission Control Areas (SECAs), which include the Baltic Sea (IMO, 2008). For measuring the total effect of ship emissions on particle properties, and further on health and climate, more long term measurements of ship emissions are needed. The effect of the sulfur regulations on particle properties in large scale also needs to be quantified.

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References

- Asmi, A., Wiedensohler, A., Laj, P., Fjaeraa, A.-M., Sellegri, K., Birmili, W., Weingartner, E., Baltensperger, U., Zdimal, V., Zikova, N., Putaud, J.-P., Marinoni, A., Tunved, P., Hansson, H.-C., Fiebig, M., Kivekäs, N., Lihavainen, H., Asmi, E., Ulevicius, V., Aalto, P.P., Swietlicki, E., Kristensson, A., Mihalopoulos, N., Kalivitis, N., Kalapov, I., Kiss, G., de Leeuw, G., Henzing, B., Harrison, R.M., Beddows, D., O'Dowd, C., Jennings, S.G., Flentje, H., Weinhold, K., Meinhardt, F., Ries, L., Kulmala, M., 2011. Number size distributions and seasonality of submicron particles in Europe 2008–2009. *Atmos. Chem. Phys.* 11, 5505–5538.

- Bjeltvedt Skeie, R., Fuglestedt, J., Berntsen, T., Tronstad Lund, M., Myhre, G., Rypdal, K., 2009. Global temperature change from the transport sectors: historical development and future scenarios. *Atmos. Environ.* 43, 6260–6270.
- Birmili, W., 2001. A moment-preserving parameterization of wide particle size distributions. *J. Aerosol Sci.* 32, S191–S192.
- Clarke, A.D., Varner, J.L., Eisele, F., Mauldin, R.L., Tanner, D., Litchy, M., 1998. Particle production in the remote marine atmosphere: cloud outflow and subsidence during ACE 1. *J. Geophys. Res.* 103 (D13), 16397–16409, <http://dx.doi.org/10.1029/97JD02987>.
- Corbett, J.J., Koehler, H.W., 2003. Updated emissions from ocean shipping. *J. Geophys. Res.* 108 (D20), 4650, <http://dx.doi.org/10.1029/2003JD003751>.
- Corbett, J.J., Winebrake, J.J., Green, E.H., Kasibhatla, P., Eyring, V., Lauer, A., 2007. Mortality from ship emissions: a global assessment. *Environ. Sci. Technol.* 41, 8512–8518.
- Diesch, J.M., Drewnick, F., Klimach, T., Borrmann, S., 2013. Investigation of gaseous and particulate emissions from various marine vessel types measured on the banks of the Elbe in Northern Germany. *Atmos. Chem. Phys.* 13, 3603–3618.
- Draxler, R.R., Rolph, G.D., 2003. HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory). NOAA Air Resources Laboratory, Silver Spring, MD Model access via NOAA ARL READY Website, <http://www.arl.noaa.gov/ready/hysplit4.html>.
- Ehn, M., Vuollekoski, H., Petaja, T., Kerminen, V.M., Vana, M., Alto, P., de Leeuw, G., Ceburnis, D., Dupuy, R., O'Dowd, C.D., Kulmala, M., 2010. Growth rates during coastal and marine new particle formation in western Ireland. *J. Geophys. Res.* 115, <http://dx.doi.org/10.1029/2010JD014292>.
- Engler, C., Lihavainen, H., Komppula, M., Kerminen, V.M., Kulmala, M., Viisanen, Y., 2007. Continuous measurements of aerosol properties at Baltic Sea. *Tellus B* 59, 728–741.
- González, Y., Rodríguez, S., García, J.C.G., Trujillo, J.L., García, R., 2011. Ultrafine particles pollution in urban coastal air due to ship emissions. *Atmos. Environ.* 45, 4907–4914.
- Gryning, S.-E., Batchvarova, E., 2002. Marine boundary layer and turbulent fluxes over the Baltic Sea: measurements and modeling. *Bound.-Lay. Meteorol.* 103, 29–47.
- Hyvärinen, A.-P., Komppula, M., Engler, C., Kivekäs, N., Kerminen, V.M., Dal Maso, M., Viisanen, Y., Lihavainen, H., 2008. Atmospheric new particle formation at Utö, Baltic Sea 2003–2005. *Tellus B* 60, 345–352.
- IMO (International Maritime Organization), 2008. Amendments to the annex of the protocol of 1997 to amend the international convention for the prevention of pollution from ships, 1973, as modified by the protocol of 1978 relating thereto. Available at: <http://www.imo.org/> (March 6th, 2014).
- Jalkanen, J.-P., Brink, A., Kalli, J., Pettersson, H., Kukkonen, J., Stipa, T., 2009. A modelling system for the exhaust emissions of marine traffic and its application to the Baltic Sea area. *Atmos. Chem. Phys.* 9, 9209–9223.
- Kasper, A., Aufdenblatten, S., Forss, A., Mohr, M., Burtscher, H., 2007. Particulate emissions from a low-speed marine diesel engine. *Aerosol Sci. Technol.* 41, 24–32.
- Kivekäs, N., Rusnak, V., Carreno Correa, S., Massling, A., Skov, H., Lange, R., Kristensson, A., 2014. Contribution of ship traffic to aerosol particle concentrations downwind of a major shipping route. *Atmos. Chem. Phys.* 14, 8419–8454.
- Kristensson, A., Dal Maso, M., Swietlicki, E., Hussein, T., Zhou, J., Kerminen, V.-M., Kulmala, M., 2008. Characterization of new particle formation events at a background site in Southern Sweden: relation to air mass history. *Tellus B* 60, 330–344.
- Kristensson, A., Johansson, M., Swietlicki, E., Kivekäs, N., Hussein, T., Nieminen, T., Kulmala, M., Dal Maso, M., 2014. NanoMap: Geographical mapping of atmospheric new particle formation through analysis of particle number size distribution data. *Boreal Environ. Res.* 19 (Suppl. B), 329–342.
- Kumar, P., Pirjola, L., Ketzler, M., Harrison, R.M., 2013. Nanoparticle emissions from 11 non-vehicle exhaust sources – a review. *Atmos. Environ.* 67, 252–277.
- Lewis, E.R., Schwartz, S.E., 2004. Sea salt Aerosol Production: Mechanisms, Methods, measurements, and models – A Critical review. *Geophys. Monogr. Series*, vol. 152. American Geophysical Union, Washington, DC, 413 pp.
- Ma, N., Zhao, C.S., Nowak, A., Müller, T., Pfeifer, S., Cheng, Y.F., Deng, Z.Z., Liu, P.F., Xu, W.Y., Ran, L., Yan, P., Göbel, T., Hallbauer, E., Mildenerger, K., Henning, S., Yu, J., Chen, L.L., Zhou, X.J., Stratmann, F., Wiedensohler, A., 2011. Aerosol optical properties in the North China Plain during HaChi campaign: an in-situ optical closure study. *Atmos. Chem. Phys.* 11, 5959–5973.
- Mordas, G., Kulmala, M., Petäjä, T., Aalto, P.P., Matulevicius, V., Grigoraitis, V., Ulevicius, V., Grauslys, V., Ukkonen, A., Hämeri, K., 2005. Design and performance characteristics of a condensation particle counter UF-02proto. *Boreal Environ. Res.* 10, 543–552.
- Müller, T., Henzing, J.S., de Leeuw, G., Wiedensohler, A., Alastuey, A., Angelov, H., Bizjak, M., Collaud Coen, M., Engström, J.E., Gruening, C., Hillamo, R., Hoffer, A., Imre, K., Ivanow, P., Jennings, G., Sun, J.Y., Kalivitis, N., Karlsson, H., Komppula, M., Laj, P., Li, S.-M., Lunder, C., Marinoni, A., Martins dos Santos, S., Moerman, M., Nowak, A., Ogren, J.A., Petzold, A., Pichon, J.M., Rodriguez, S., Sharma, S., Sheridan, P.J., Teinilä, K., Tuch, T., Viana, M., Virkkula, A., Weingartner, E., Wilhelm, R., Wang, Y.Q., 2011. Characterization and intercomparison of aerosol absorption photometers: result of two intercomparison workshops. *Atmos. Meas. Tech.* 4, 245–268, <http://dx.doi.org/10.5194/amt-4-245-2011>.
- Pettersson, H., Lindow, H., Brüning, T., 2012. Wave climate in the Baltic Sea in 2012. HELCOM Baltic Sea Environment Fact Sheets. Online. 2014. <http://www.helcom.fi/baltic-sea-trends/environment-fact-sheets/>.
- Petzold, A., Hasselbach, J., Lauer, P., Baumann, R., Franke, K., Gurk, C., Schlager, H., Weingartner, E., 2008. Experimental studies on particle emissions from cruising ship, their characteristic properties, transformation and atmospheric lifetime in the marine boundary layer. *Atmos. Chem. Phys.* 8, 2387–2403, <http://dx.doi.org/10.5194/acp-8-2387-2008>.
- Rissler, J., Nordin, E.Z., Eriksson, A.C., Nilsson, P.T., Frosch, M., Sporre, M.K., Wierzbicka, A., Svenningsson, B., Londahl, J., Messing, M.E., Sjogren, S., Hemmingsen, J.G., Loft, S., Pagels, J.H., Swietlicki, E., 2014. Effective density and mixing state of aerosol particles in a near-traffic urban environment. *Environ. Sci. Technol.* 48, 6300–6308, <http://dx.doi.org/10.1021/es5000353>.
- Rolph, G.D., 2003. Real-time Environmental Applications and Display sYstem (READY). NOAA Air Resources Laboratory, Silver Spring, MD Website (<http://www.arl.noaa.gov/ready/hysplit4.html>).
- Sandradewi, J., Prevot, A.S.H., Weingartner, E., Schmidhauser, R., Gysel, M., Baltensperger, U., 2008. A study of wood burning and traffic aerosols in an Alpine valley using a multi-wavelength Aethalometer. *Atmos. Environ.* 42, 101–112.
- Sofiev, M., Soares, J., Prank, M., de Leeuw, G., Kukkonen, J., 2011. A regional-to-global model of emission and transport of sea salt particles in the atmosphere. *J. Geophys. Res.* 116 (D21), <http://dx.doi.org/10.1029/2010JD014713>.
- Stipa, T., Jalkanen, J.-P., Hongisto, M., Kalli, J., Brink, A., 2007. Emissions of NOx from Baltic Shipping and First Estimates of their Effects on Air Quality and Eutrophication of the Baltic Sea. *Univ. Turku, ISBN: 978-951-53-3028-4*, 33 pp.
- Stohl, A., 1998. Computation, accuracy and applications of trajectories – a review and bibliography. *Atmos. Environ.* 32, 947–966.
- Stokes, M.D., Deane, G.B., Prather, K., Bertram, T.H., Ruppel, M.J., Ryder, O.S., Brady, J.M., Zhao, D., 2013. A Marine Aerosol Reference Tank system as a breaking wave analogue for the production of foam and sea-spray aerosols. *Atmos. Meas. Tech.* 6, 1085–1094.
- Ulevicius, V., Bycenkiene, S., Remeikis, V., Garbaras, A., Kecorius, S., Andrijeauskiene, J., Jasineviciene, D., Mocnik, G., 2010.

- Characterization of pollution events in the East Baltic region affected by regional biomass fire emissions. *Atmos. Res.* 2, 190–200.
- Virkkula, A., Makela, T., Hillamo, R., Yli-Tuomi, T., Hirsikko, A., Hameri, K., Koponen, I.K., 2007. A simple procedure for correcting loading effects of Aethalometer data. *J. Air Waste Manage.* 57, 1214–1222.
- Weingartner, E., Saathoff, H., Schnaiter, M., Streit, N., Bitnar, B., Baltensperger, U., 2003. Absorption of light by soot particles: determination of the absorption coefficient by means of aethalometers. *J. Aerosol Sci.* 34, 1445–1463.
- Wiedensohler, A., 1988. An approximation of the bipolar charge distribution for particles in the submicron size range. *J. Aerosol Sci.* 19, 387–389.
- Wiedensohler, A., Birmili, W., Nowak, A., Sonntag, A., Weinhold, K., Merkel, M., Wehner, B., Tuch, T., Pfeifer, S., Fiebig, M., Fjåraa, A. M., Asmi, E., Sellegri, K., Depuy, R., Venzac, H., Villani, P., Laj, P., Aalto, P., Ogren, J.A., Swietlicki, E., Williams, P., Roldin, P., Quincey, P., Hüglin, C., Fierz-Schmidhauser, R., Gysel, M., Weingartner, E., Riccobono, F., Santos, S., Gruning, C., Faloon, K., Beddows, D., Harrison, R., Monahan, C., Jennings, S.G., O'Dowd, C.D., Marinoni, A., Horn, H.-G., Keck, L., Jiang, J., Scheckman, J., McMurry, P.H., Deng, Z., Zhao, C.S., Moerman, M., Henzing, B., de Leeuw, G., Löschau, G., Bastian, S., 2012. Mobility particle size spectrometers: harmonization of technical standards and data structure to facilitate high quality long-term observations of atmospheric particle number size distributions. *Atmos. Meas. Tech.* 5, 657–685.