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Impact of the emissions of international sea traffic on airborne deposition to the Baltic Sea and concentrations at the coastline*

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Abstract

The impact of ship traffic emissions in the Baltic Sea on deposition and airborne concentrations of nitrogen and sulphur compounds in the period 2008–2011 was studied using the Hilatar chemistry transport model with a 0.068° latitude-longitude resolution. An accurate ship emission inventory based on AIS (automatic identification system) security signals was used. The uncertainty of the European emission inventories are discussed, as is an inter-comparison of the Baltic Sea airborne load and concentrations with other model-based estimates and with air quality measurements and the effect of the EU sulphur directive for ship emissions on sulphate concentrations.

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1. Introduction

The state of the Baltic Sea (BS) has been of widespread concern due to the human impact on its ecosystems. The vertical stratification of temperature and salinity of the water column in most sub-basins the whole year round and the low level of water exchange with the Atlantic Ocean make it very vulnerable to external pressures (BACC 2008). Its ecological state and biodiversity are threatened by eutrophication caused by excessive nutrient inputs, by direct pollution, by increasing ship traffic causing illegal spills and increased risk of accidents, by climate change and by direct human actions including overfishing and over-exploitation.

The Baltic Sea is situated between continental and marine climatic zones with the sources of most of the atmospheric nitrogen emissions located in the south. The atmospheric nitrogen and sulphur loads show a high inter-annual and geographical variation with both east-west and north-south gradients. Although the atmospheric load of inorganic nitrogen (N) is only around 31% of the waterborne load of N (HELCOM 2011), it is estimated to be completely bioavailable whereas the fluvial load is not: for example, in Danish waters the bioavailable total nitrogen (TN) fraction varied between 0.25 and 0.8 in winter (January–February) (Carstensen & Henriksen 2009).

The measured and modelled atmospheric load of nitrogen to the BS is reported annually to HELCOM by the EMEP (Co-operative programme for monitoring and evaluation of long-range transmission of air pollutants in Europe) western and eastern centres and by NILU (Norsk institutt for luftforskning) (Bartnicki et al. 2002–2012). In addition, several Nordic and European air pollutant modelling and measurement groups have studied the composition and flux of atmospheric contaminants to the BS (e.g. Schulz et al. 1999, Plate 2000, Hertel et al. 2003, Hongisto & Joffre 2005, Rolff et al. 2008, Langner et al. 2009, Geels et al. 2011).

The BS TN load decreased from 230 kt N in 1995 to 199 kt in 2006 (Bartnicki et al. 2011), but it again exceeded 210 kt in 2008 and 218 kt N in 2010 (Svendsen et al. 2013). The inter-annual variation, ranging from −13 to 17% of the average value, was mainly caused by changing meteorological conditions. The influence of meteorological variability on nitrogen deposition was one of the main goals of the studies of Hongisto & Joffre (2005) and Hongisto (2005, 2011 and 2012). The accumulated deposition was found to be affected by the large-scale circulation type, which determines the main seasonal wind direction with respect to the source areas, the severity of the ice winter, the latitude of the cyclone paths and their frequency of occurrence, the accumulated precipitation, the strength of turbulence and the number of episodes.

The ECOSUPPORT project showed long-term estimates of the past and future development of the Baltic Sea, its external forcing and the ecosystem responses. Those results were published in autumn 2012 in AMBIO 41. Ruoho-Airola et al. (2012) compiled a consistent basin-wise monthly time series of the atmospheric nutrient load to the BS for the period 1850–2006. The modelling part was based mainly on EMEP simulations, but the authors also discovered a wonderful treasure trove of historical measurements.

Models often underestimate the measured wet deposition of nitrogen to the BS as deduced from all model measurement inter-comparison results reported by EMEP annually since 1997. The actual flux of all airborne contaminants to the BS is higher than the measured deposition because the EMEP collectors do not have a wind shield and the dry deposition is not measured. Although the collection efficiency of the rain-collecting instruments situated at windy, coastal sites is rather poor, the measured rain is used as such in flux calculations, presented in units of mass per m^{-2}. The organic nitrogen deposition, which according to Neff et al. (2002) is around a third of the total N load, is not monitored by EMEP. The organic nitrogen might be bioavailable if it disintegrated in water, hence it should be taken into account in eutrophication studies. In estimating the net atmospheric flux to sea areas one should note that in the 1990s many fluxes (CO_2, NH_3) over the sea surface were found to be bidirectional and that deposition should be estimated by a coupled marine-atmospheric model.

The effects of European international shipping on the basis of country-by-country deposition and ozone concentrations have been studied in Jonson et al. (2000). Deposition to the BS caused by European countries and sea traffic is reported annually in EMEP source-receptor matrices.

A review of existing studies on the impacts of shipping emissions of different chemical compounds on air quality in coastal areas is presented and discussed in detail in EEA (2013), along with a summary of the results over the area considered, methodological data and conclusions.

2. Methods and model description

The nitrogen deposition to the BS was calculated with the Hilatar chemistry-transport model (Hongisto 2003). As input, the model uses the forecasts of the FMI operative HIRLAM hydrostatic weather prediction model (HIgh Resolution Limited Area Model, Unden et al. 2002).

The Hilatar, a dynamic Eulerian model covering Europe with a zooming model over the Baltic Sea and its close surroundings (the BS model with 0.068 deg resolution), provides gridded estimates of the fluxes and concentrations of oxidised and reduced nitrogen and sulphur compounds.

Gaseous (g) and particle (p) concentrations are calculated for the following substances: NO_x(g), HNO_3(g), NO_3(p), PAN(g), NH_4NO_3(p), NH_3(g), SO_2(g), SO_4(p) and $(NH_4)_{1.5}SO_4$(p), where PAN is peroxyacetyl nitrate and $NO_x = NO + NO_2$. The chemistry module comprises the EMEP-MSC-W chemistry code (Iversen et al. 1989) with some modifications (Hongisto 2003). The model does not have ozone as a variable, because in photo-oxidant codes the main radical concentrations influencing the chemical transformation of nitrogen and sulphur chemistry are calculated inside the model. Their values are, however, rather seldom verified or even presented. For basic acid chemistry one can use measurement-based functions for all radicals and oxidants needed.

The Hilatar model, run since 1993, has the HIRLAM grid of the current operative model: horizontally rotated spherical coordinates and vertically hybrid sigma coordinates with selected (now 21) layers up to 5–10 km in height. The long-range transported compounds at the borders of the BS model domain, calculated by the 0.15° resolution European-scale model, are included in the advected air with six hour intervals. For the years 2008–2011, both models used the HIRLAM version V71 vertical grid; from the 60 available vertical levels the 18 lowest (up to around 1.5 km) and three additional levels (at around 2 km, 2.8 km and 5.1–5.3 km) are used.

In Hilatar, horizontal advection is solved numerically according to Bott's (1989) method, while chemistry uses the Hesstvedt et al. (1978) algorithm, and vertical diffusion the Tuovinen (1992) algorithm. The time resolution depends on the algorithm and grid resolution, being 56.25 s for all algorithms in the BS model. The dry deposition velocities, used as the lower boundary condition of the vertical diffusion equation, were calculated by resistance analogy. The Lindfors et al. (1991) method was used for calculating the marine atmospheric boundary layer (MABL) parameters for the dry deposition velocities over sea areas. The scavenging rates are based on e.g. the work of Chang (1984, 1986), Scott (1982), Jonsen & Berge (1995) and Asman & Janssen (1987).

3. Emissions

For the European simulations the models use both the EMEP WebDab and the MACC (2011) emission inventories, as well as the FMI inventory for Finnish and north-western Russian sources. The BS model also uses a specific Baltic Sea ship emission inventory (Stipa et al. 2007, Jalkanen et al. 2009, 2012) and Finnish national stack and areal emissions. The time variation for other than ship emissions is based on the GENEMIS project 1990 country-specific emissions and on the diurnal and weekly traffic indices. The initial vertical mixing was estimated by using specific emission height

profiles for each S-emission class of gridded emissions and a plume rise algorithm for stack sources.

The FMI emission inventory for north-west Russia has been maintained because most of the Russian SO_2 emissions near the Finnish borders seem to be very small in the EMEP WebDab official and the expert inventory. The SO_2 emissions of the Kola Peninsula (450–480 kt SO_2 in 2003) were reduced to 32.4 kt SO_2 in 2004 and further to 18.7 kt by 2010. There have also been unexpected stepwise changes in the Russian oxidised nitrogen (NO_x) emissions: the NO_x traffic (S7) emissions, for example, were reduced from about 240 kt to 68.6 kt NO_2 in the EMEP grid 65.80 (St. Petersburg) from the 2009 to the 2010 inventory.

Measurements indicate, however, that there are large sulphur emissions sources on the Russian side of the Finnish border. In the EEA data base on European Air Quality, the measured SO_2 concentrations in northern Norway in 2010 exceeded both the daily limit values for the protection of human health as well as the annual and winter limit values for the protection of ecosystems (EEA 2012). Nikel, Zapoljarnyi, Monchegorsk, Kirovsk, Apatity and Kovdor are also the highest pollution targets, M1–M5, of the environmental hot-spot list of Barentsinfo (2013), and e.g. Norilsk Nikel report directly on the internet their emissions from Nikel and Zapoljarnyi (136 kt SO_2 in 2009) as well as high SO_2 concentrations at Svanvik monitored by themselves (Norils Nikel 2013). Svanvik concentrations can also be followed on-line at http://www.luftkvalitet.info/ and Janiskoski concentrations at http://www.ilmanlaatu.fi/.

In 2007 the total SO_2 emission over the Murmansk region was 21 204 t SO_2 in the EMEP inventory, 289 319 t SO_2 in the MACC inventory and 240 470 t SO_2 in the FMI inventory. The NO_x emissions over the Murmansk region given by MACC, 19 424 t NO_2, were lower than the corresponding EMEP (34 888 t) or FMI emissions (25 626 t NO_2).

For the years after 2007, the MACC emissions were scaled using the emission trends of each country from EMEP. For those emission groups missing from the MACC inventory (natural, marine, volcanic and Iceland emissions) the EMEP emissions were used. For north-western Russia (the Kola Peninsula, Karelia and Leningrad Oblast) the FMI's own inventory is still used, because the locations of the enterprises there are more exact; also there are some well-known sources, e.g. in Karelia, missing from the MACC inventory.

For the Baltic Sea model we use the specific Baltic Sea ship emission inventory. This AIS-signal-based inventory was developed at the FMI in co-operation with researchers from Åbo Akademi University and Turku

University and with the support of the Marine Administration, FMA, and the Finnish State Technical Research Centre, VTT (Stipa et al. 2007).

Each ship over the 300 tons gross tonnage limit sailing the BS has to send AIS-transmitter safety signals at variable time intervals: these signals contain the unique IMO code of the ship and information on the ship's movements, its load, destination and type. These signals are collected by AIS-receiver stations located on the coasts of the Baltic Sea. The FMA collects the AIS signals into a local database and sends this information, as do also the other maritime administration offices surrounding the BS, to the HELCOM database (DB). FMI, having access to the HELCOM DB, decodes the AIS-signals and, using the IMO code, retrieves information on the ship's machinery from the Lloyds data base. The FMI model STEAM (Ship Traffic Emission Assessment Model, Jalkanen et al. 2009) calculates an emission estimate for each individual ship as a function of the ship's type, its engine load, fuel type, speed and emission control technology, using current weather and wave height information, and sums the emissions on a latitude-longitude grid with a selected resolution, then reporting on-line using a $\sim$450 s–1h time-interval. Emissions calculated with STEAM are available from 2006 onwards. That year the temporal coverage of the signals collected was about 93%, while around 16% of ships sailed without an IMO number (Jalkanen et al. 2012). For small pleasure boats and other vessels, we use the VTT emission inventory. When the AIS signal data are missing, the monthly average emission estimate has been used.

The FMI, MACC and EMEP estimates of the BS international ship traffic emissions are compared in Table 1. Over the BS, North Sea and the English Channel the maximum allowable sulphur content of marine fuels decreased due to the EU directive (2005/33/EC) from 1.5 to 1% in July 2010, and to 0.1% in port areas in January 2010. From the year 2009 to 2011, the FMI-estimated ship emissions of SO_2 decreased by 48 kt and the EMEP emissions by 40 kt SO_2. The annual sums of the emissions do not

Table 1. Comparison of the FMI international ship traffic emissions in kt NO_2 and kt SO_2 over the Baltic Sea in the FMI, EMEP and MACC emission inventories. St1 and St2: first and second versions of the FMI emission model STEAM

	FMI NO_x, St1	FMI NO_x, St2	EMEP NO_x	MACC NO_x	FMI SO_2, St1	FMI SO_2, St2	EMEP SO_2	MACC SO_2
2007	400	369	315	350	138	144	167	205
2008	390	377	321		148	132	145	
2009	384	360	327		143	132	122	
2010	402		333		113		99	
2011	414		339		95		82	

differ substantially, considering the overall inaccuracy and errors in emission estimates.

The new version of the STEAM model (St2, STEAM2, Jalkanen et al. 2012) used in this study also calculates emissions of CO, CO_2 and particulate matter (elementary and organic carbon, ash, hydrated SO_4). The main advantage of the new AIS-based inventory is its excellent temporal and spatial resolution.

4. Results

The modelled 2008–2011 average oxidised nitrogen (NO_x), reduced nitrogen (NH_x) and sulphur (S) depositions are presented in Figure 1. The dry deposition share of the total NO_x deposition increases from 10–20% over

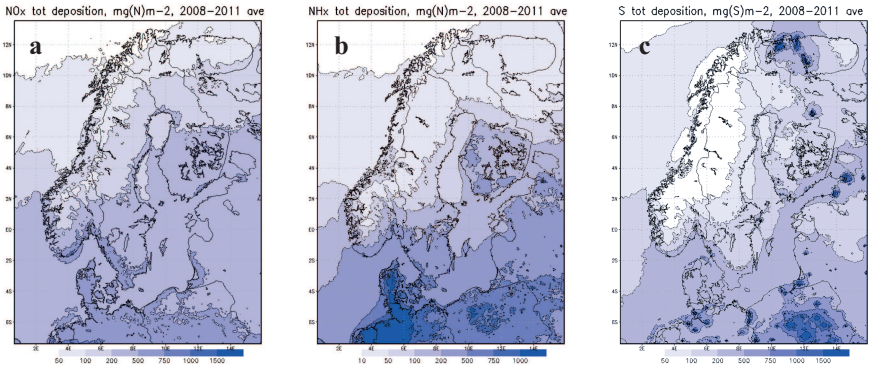

Figure 1. Modelled 2008–2011 average oxidised nitrogen (NO_x), reduced nitrogen (NH_x) and sulphur (S) deposition

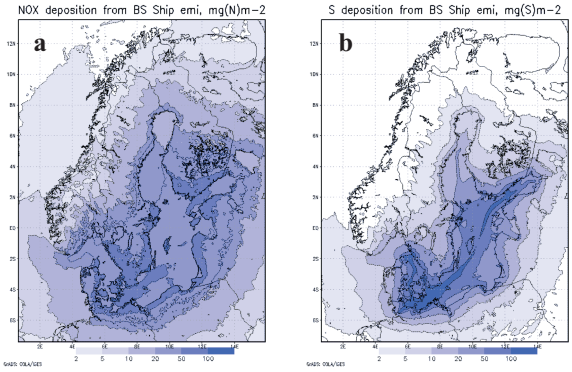

Figure 2. The 2008–2011 average oxidised nitrogen (NO_x) and sulphur (S) deposition caused by international ship traffic in the Baltic Sea

the northern Gulf of Bothnia to 20–30% in the Sea of Bothnia, the Gulf of Finland and the Gulf of Riga, being 30–40% in the central Baltic Proper and in the southern Baltic Sea. The share of reduced nitrogen in the total N deposition was less than 30% north of Åland, increasing gradually southwards to over 50% in the Kattegat and Belt Sea areas. There was a rather sharp dry deposition gradient over the shorelines for both nitrogen compounds.

The 2008–2011 average depositions of NO_x and S caused by the international ship traffic in the BS are presented in Figure 2 and the ship deposition shares of the respective total deposition in Figure 3. The annual sums of the total and ship-emission-originated depositions of sulphur and nitrogen to the BS with a map of BS sub-basins – the Gulf of Bothnia (B1), the Gulf of Finland (B2), the northern Baltic Proper (B3), the southern Baltic Proper (B4) and the Kattegat and the Belt Sea (B5) – are presented in Figure 4.

The ship emission originated deposition of oxidised nitrogen increased between 2008 to 2011 from 12 to 14% of the BS total NO_x deposition, while the respective sulphur deposition declined from 28 to 20% of the total due to the sulphur directive restrictions. Sulphur is effectively dry-deposited into the sea, only 19–25% of the ship emission originated sulphur deposition is wet deposition. The total modelled NO_x deposition to the BS was respectively 6% and 15% lower in 2008 and 2011 but 1% and 5% higher in 2009 and 2010 than the most recent EMEP estimates from HELCOM 2013. The modelled deposition of NH_x was respectively 18, 22, 5 and 15% lower than the EMEP estimate for the years 2008–2011. One reason for the difference is the high deposition gradient at the coastline: in Hilatar, the deposition was integrated only over grid points with 100% open water (372 954 km^2), while the complete 0.068° Hirlam BS mask of 420 325 km^2, also covered non-marine water areas in the BS coastal zone.

Total depositions have a rather high seasonal variation (Figure 5). During spring and early summer when the MABL is usually stably stratified, accumulated precipitation is low and storms are rare, depositions have their minimum values. The May to July NO_x(S) deposition was on average 44(38)% of the November–January deposition of the same year, the minimum monthly deposition being 26(24)% of the respective maximum monthly deposition during the period studied. The temporal variation is higher over the northern sub-basins and for dry and wet deposition separately.

The monthly NO_x deposition originating from BS ship-traffic emissions reached a maximum in the summer months due to higher dry deposition velocities, and a faster chemistry converting NO_2 into scavengable chemical species. The S deposition did not have as high a seasonal variation

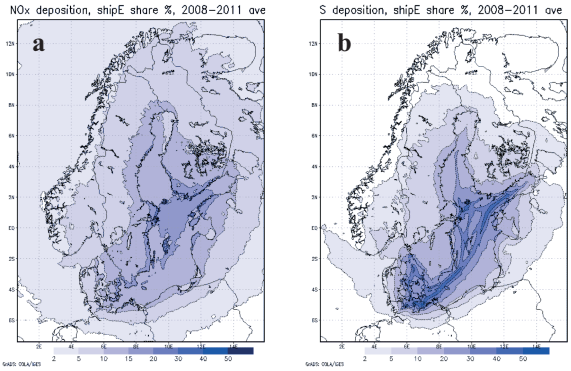

Figure 3. The average 2008–2011 oxidised nitrogen (NO_x) and sulphur (S) deposition shares of the respective total deposition originating in Baltic Sea ship emissions

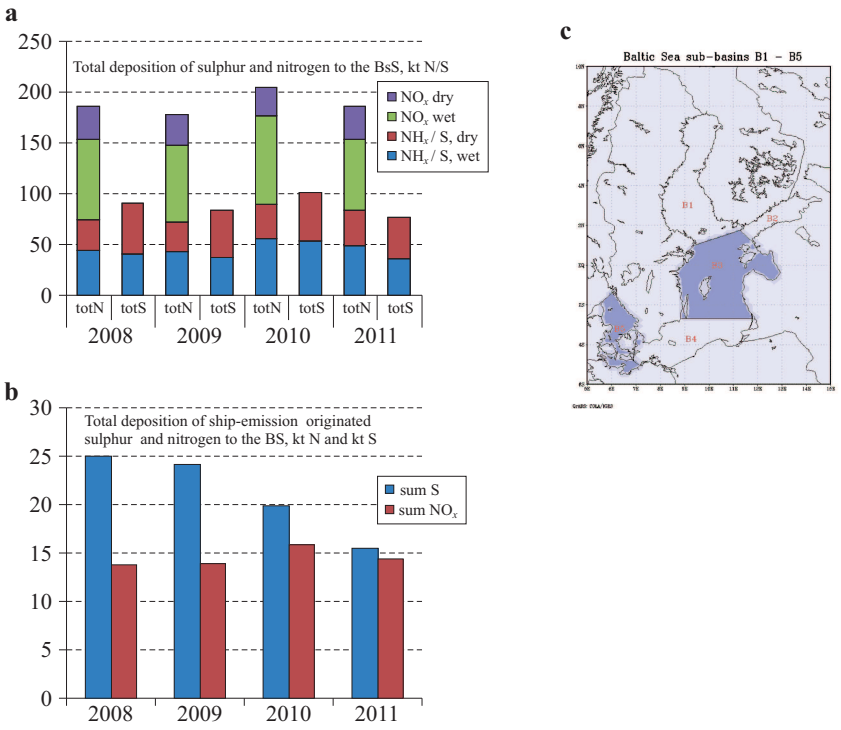

Figure 4. Annual sums of the sulphur [t S] and nitrogen [t N] deposition to the Baltic Sea: a) total deposition, b) deposition emitted by shipping, c) map of the BS sub-basins. The five BS sub-basins: the Gulf of Bothnia (B1), the Gulf of Finland (B2), the Northern Baltic Proper (B3), the Southern Baltic Proper (B4) and the Kattegatt and the Belt Sea (B5)

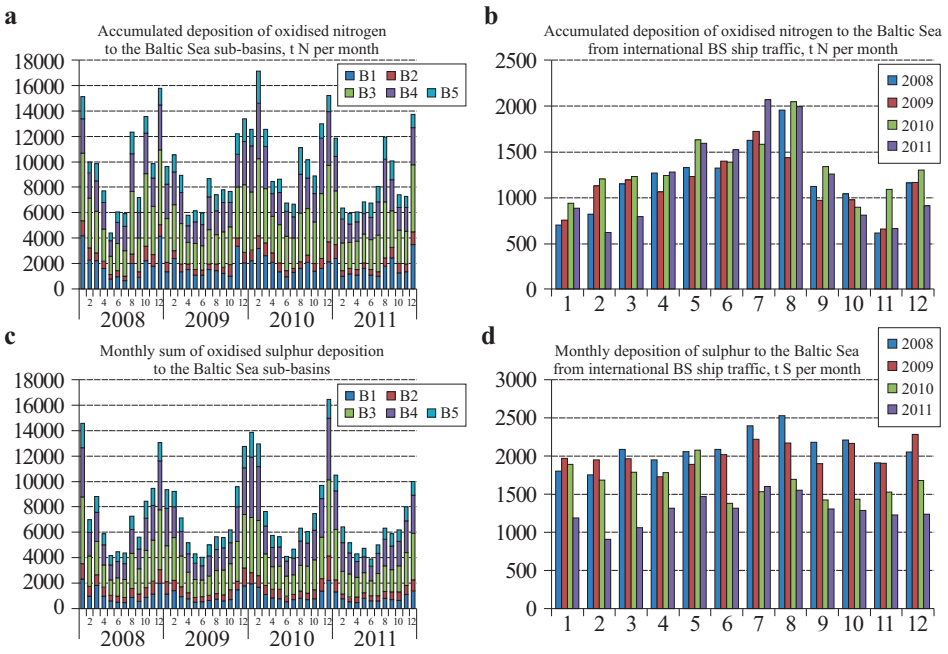

Figure 5. Monthly total accumulated deposition of nitrogen (a) and sulphur (c) to the Baltic Sea and deposition of nitrogen due to shipping (b) and deposition of sulphur due to shipping (d), [t S] and [t N] per month

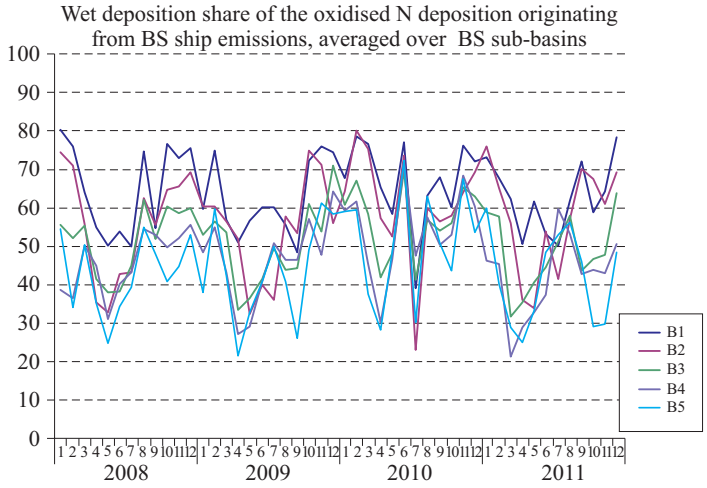

Figure 6. Wet deposition share of the oxidised N deposition originating from BS ship emissions

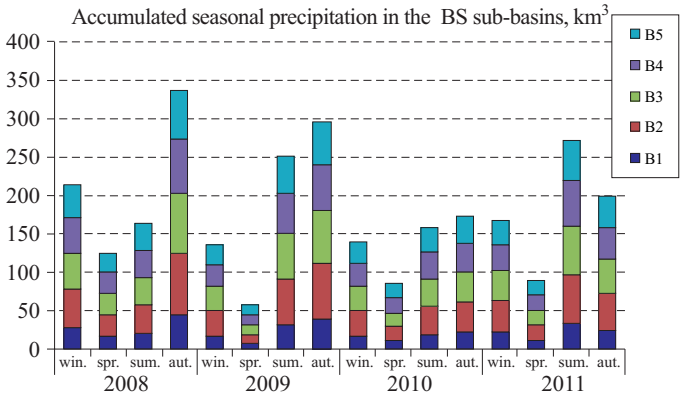

Figure 7. Accumulated seasonal average precipitation to the Baltic Sea sub-basins, [km^3]. Winter (win) = JFM, spring (spr) = AMJ, summer (sum) = JAS, autumn (aut) = OND

(Figure 5). The decline in sulphur emissions due to the EU restrictions regarding fuel S content can be directly seen in the decrease in the S deposition towards 2011.

The monthly average wet deposition share of the NO_x deposition was highest in the northern BS sub-basins in winter (up to 80%) and autumn, and lowest during the spring months in the south (Figure 6). The accumulated seasonal precipitation (Figure 7) and the strength of the ice winter have a direct effect on the dry and wet deposition shares. The contribution of accumulated annual precipitation to the total BS varied from 556 km^3 in 2010 to 839 km^3 in 2008, but the seasonal precipitation sums over sub-pools did display different inter-annual variation. On average, winters were colder at the end of the period, being characterised by more northerly

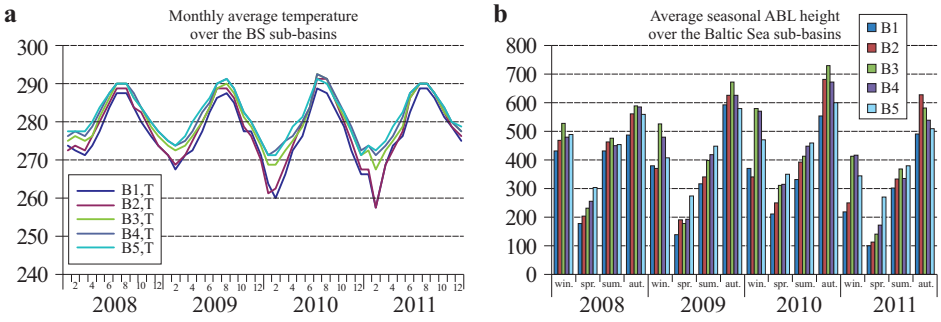

Figure 8. Average monthly temperature [°K] and average height of the atmospheric boundary layer (ABL), [m], over the BS sub-basins during the study period

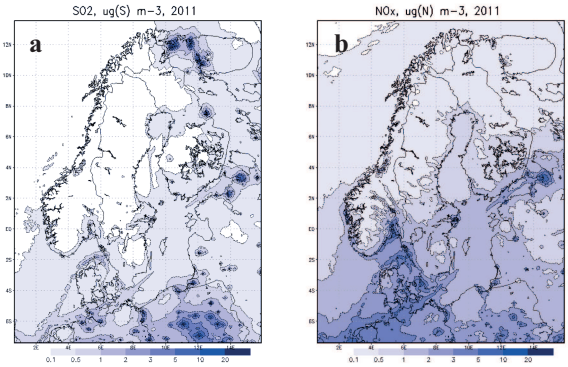

Figure 9. Modelled annual average SO_2 and $NO + NO_2$ concentrations in 2010, [μg (S) m^{-3}] and [μg (N) m^{-3}]

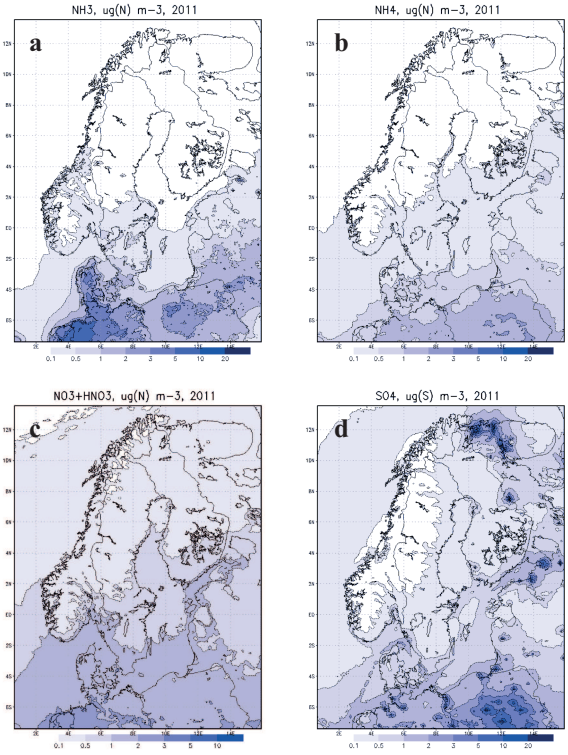

Figure 10. Modelled annual average concentrations of ammonia, ammonium, total nitrate and sulphate in air, 2010

winds from clean areas and lower dry deposition over the ice cover. From 2008 to 2011 the HIRLAM winter (JFM) average MABL height dropped

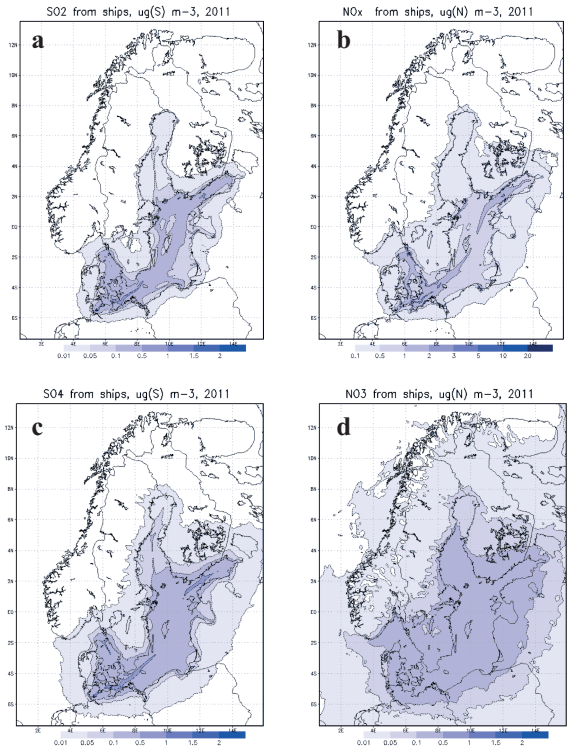

Figure 11. Modelled concentrations of SO_2, NO_x ($NO + NO_2$), sulphate and nitrate in air, originating from Baltic Sea ship emissions in 2010

from 420–450 m to around 200 m over the northern BS sub-basins (Figure 8).

The modelled SO_2 and NO_x ($NO + NO_2$) concentrations in 2011 are presented in Figure 9, the average concentrations of ammonia, ammonium, total nitrate and sulphate in air in Figure 10. Figure 11 shows the modelled concentrations from BS ship emissions of SO_2, NO_x, sulphate and nitrate in air, in 2011, when the marine fuel S content reductions were implemented.

The modelled ship-originated concentrations of sulphate on BS coasts (Figure 11) were 0.1–0.5 μg (S) m^{-3}. The maximum 2010 annual average proportion of ship-originated sulphate, including direct SO_4 emissions and secondary particles, occurred along the shipping routes. Those modelled maximum proportions exceeded 60% of the modelled total SO_4 over the open water areas at the mouth of the GoF, but this ship-emission originated SO_4 share fell generally to 5–30% along the shores of sub-basins B1–B5, exceeding, however, 30% in the coastal areas of the southern BS where the ship routes run close to the coastline.

5. Model validation

For verifying the deposition of this study the monthly average concentrations in precipitation at 22–26 background stations are presented for the years 2008–2011 in Figures 12 and 13 in units of mg l^{-1}. When the intercomparison in units of mg m^{-2} was calculated from daily values (Figure 14) the correlation coefficient was significant (0.6348, N = 5324) and the average annual modelled and measured depositions were close to each other: 0.64 and 0.60 mg (N) m^{-2} respectively. The intercomparison results for one-hour NO_2 concentration at Utö station are presented in Figure 15.

The intercomparison of the concentrations in air with monthly EMEP/NILU measurements is presented in Table 2. The NO_2, SO_2, NH_3, sea-salt corrected SO_4 and sum of NH_3 and NH_4 concentrations in air are rather well simulated; the model overestimates NO_3, HNO_3 and the sum of HNO_3 and NO_3, but underestimates NH_4-concentrations. The correlations are

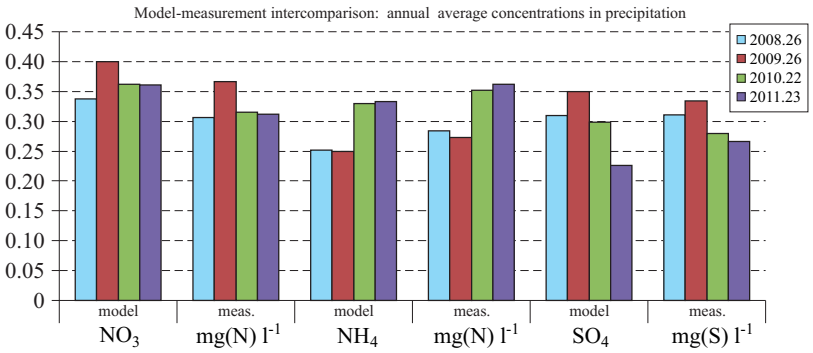

Figure 12. Model-measurement intercomparison: annual average concentrations in precipitation, 2008–2011

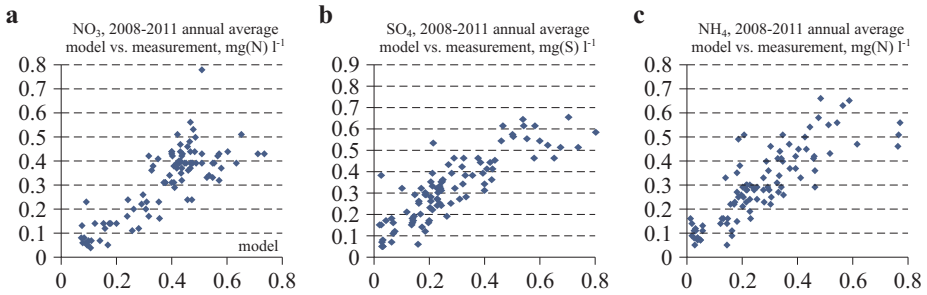

Figure 13. Model-measurement intercomparison: annual average concentrations in precipitation, [mg l^{-1}] 2008–2011

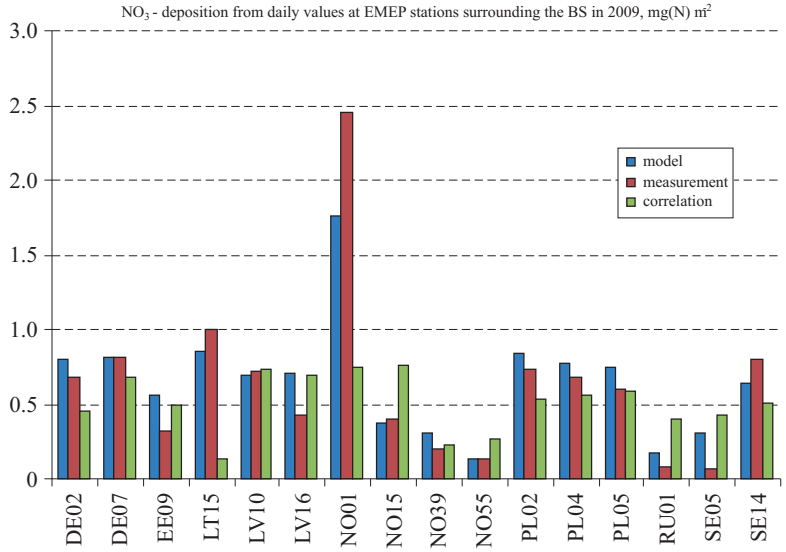

Figure 14. Model-measurement intercomparison: NO_3-deposition at EMEP stations in 2009 in [mg m^{-2}]

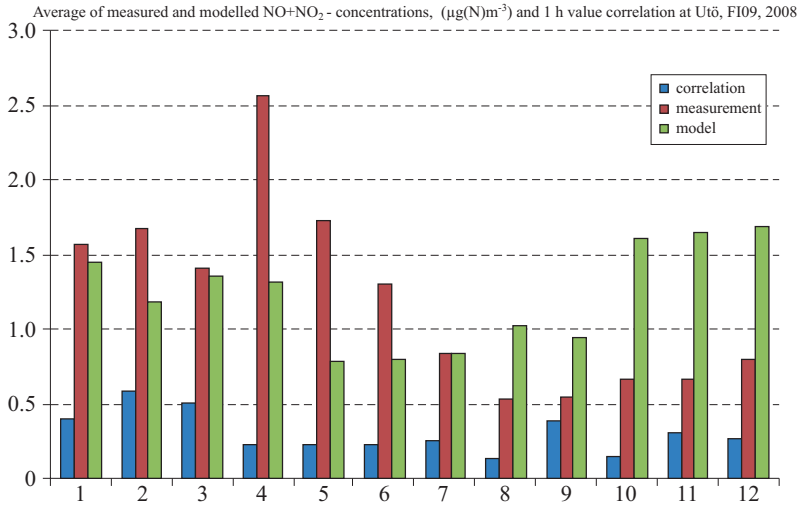

Figure 15. Model-measurement intercomparison: concentration of NO_2 in air at the Finnish EMEP-background station Utö in 2008

rather high and significant, the p-values for each compound being less than 0.001.

The modelled accumulated deposition of oxidised (reduced) nitrogen to the Baltic Sea, which varied between 102–131 (73–90) kt N in 2008–2011, was slightly smaller in comparison to the HELCOM (EMEP) estimates,

Table 2. Model-measurement intercomparison: concentrations in air. Units [μg m^{-3}] (S) or (N)

		Average meas.	Average model.	Correlation	N	Max meas.	Max model.
NO_2	2008–2010	1.32	1.61	0.69	852	6.22	7.91
SO_2	2008–2010	0.44	0.38	0.66	984	4.26	4.30
NH_3	2008–2010	0.68	0.86	0.64	408	3.46	6.14
HNO_3	2008–2010	0.12	0.16	0.47	300	1.11	0.73
NO_3	2008–2010	0.21	0.70	0.64	468	1.61	2.75
SO_4 – corr.	2008–2010	0.34	0.31	0.75	600	1.87	1.73
SO_4 – tot.	2008–2010	0.51	0.38	0.77	972	2.75	3.61
NH_4	2008–2010	0.49	0.25	0.81	780	2.75	1.72
$HNO_3 + NO_3$	2008–2010	0.39	1.00	0.72	912	2.57	6.21
$NH_3 + NH_4$	2008–2010	0.96	0.87	0.81	876	4.75	6.38

but the modelled deposition was summed only over open sea areas. The modelled deposition was rather well simulated when compared with measured concentrations in precipitation (Figure 12). The modelled and measured NO_2 concentrations peaks in air at the Utö coastal station were well reproduced in winter; in spring, however, when the MABL was very stable, the observed concentrations were higher.

According to the data and maps EEA (2012), over the Baltic Sea and its surroundings, in 2009 the annual limit value of NO_x for the protection of vegetation, 30 μg m^{-3}, which should be measured at rural stations (directive 2008/EC/50), was exceeded in southern Norway. The limit values of the annual and winter SO_2 concentrations for the protection of human health and vegetation (20 μg m^{-3}) were also exceeded in northern Norway in 2009 and 2010 (EEA 2012). The modelled concentrations were lower: NO_2 values did not exceed these limits in background areas and SO_2 values near Kola Peninsula were not as high as those measured in Norway. But the modelled concentrations representing a mean value of a ca $7 \times 7 \times 0.03$ km^3 gridbox cannot be directly compared to measured values if there are local sources near the measurement points. In the rather sparse measurement network some stations may have suffered from local industrial or traffic pollution, and if inversion situations are frequent, the concentrations rise. But the measured concentrations are real and the exceeding of the directive values should lead to emission reductions.

6. The effect of particles on human health

One of the aims of this paper was to evaluate the effect of the sulphur directive for protecting people in the BS region from the adverse health effects of the sulphate particles. The modelled annual sulphate

concentration originating from ships' plumes (Figure 11) did not exceed 0.5 μg (S) m^{-3} at any coastal location on the BS in 2010. However, the model results are 7×7 $km^2 \times 30$ m grid averages.

The aerodynamic diameter of the sulphate aerosols is mainly < 2.5 μm. The EU's target annual mean value for particles with diameters < 2.5 μm ($PM_{2.5}$) regarding the protection of human health is 25 μg m^{-3}. Ground-level concentrations of fine particles, PM, < 2.5 μm in aerodynamic diameter are associated with cardiovascular and respiratory mortality. The estimated consequences on human mortality have a rather high variation.

Anenberg et al. (2010) estimated the global burden of human mortality due to the increase in annual average $PM_{2.5}$ concentrations from their preindustrial level on a grid of $2.8^\circ \times 2.8^\circ$ resolution. Concentrations of SO_4, NO_3, NH_4, black carbon BC and anthropogenic organic carbon particles OC were included, but dust, sea salt particles and secondary organic aerosols were excluded. The contribution of SO_4 to the global average $PM_{2.5}$ concentration was 28.3% (the proportion of $(NH_4)_2SO_4$, of which the SO_4 mass makes up 70%, was 40.4%) in Europe in 2000. Those researchers estimated that if there is no low-concentration threshold below which mortality does not increase, then in the year 2000 $PM_{2.5}$ exposure caused 3.7 ± 1 million extra mortalities globally, 633 000 of which were in Europe.

From an average of six PM models Silva et al. (2013) estimated that 2.1 million (1.3 to 3 M) $PM_{2.5}$-related extra deaths occurred globally, 154 000 (105–193 000) of which were in Europe.

A first estimate of the effect of global shipping-related PM emissions on mortality was 60 000 annual deaths in 2002. It was expected to grow by 40% by 2012 (Corbett et al. 2007).

Winebrake et al. (2009) compared the effect of different sulphur control strategies of global ship fuel S content on global mortality rates, and concluded that the 2012 global premature death rate due to ships' emissions, i.e. 87 000, could be reduced by 33 500 persons with a 0.5% sulphur limit and by 43 500 deaths with a 0.1% S limit.

Brandt et al. (2011) developed an integrated model system EVA (Economic Valuation of Air pollution) for assessing the health-related impact of air pollution (O_3, CO, SO_2, SO_4, NO_3 and primary emitted $PM_{2.5}$) from specific emission sources. Their estimate of the total number of premature deaths in Europe due to air pollution, was 680 000 in 2000 and 450 000 in 2020. Of these numbers, 49 500 (2010) and 53 200 (2020) were estimated to be caused by international shipping in the Northern Hemisphere (NH). Brandt et al. estimated that the health effect of all air pollutants from international ship traffic through the North Sea and the

Baltic Sea was 20 377 extra annual deaths in Europe. This is a rather high number, 41% of all deaths caused by NH ship traffic.

The report by Brandt et al. (2011) has been cited by politicians to justify further reductions in the sulphur content of marine fuels (a maximum S content of 0.1% from 1 January 2015). When the sulphur content of the fuel is reduced, PM emissions will also be affected; however, most of the effects can be found in the reduction of secondary sulphate particles, whose ship-originated concentrations calculated in this study were low except close to shipping lanes.

In order to estimate the effect of reduced sulphur emissions from ships on European mortality, the effect of O_3, NO_2 and direct PM emissions should be separated from the overall figure. This requires that the mortality is estimated for each chemical compound and source type separately, but this is generally not the case. According to Anenberg et al. (2010), O_3 caused 6% of the total mortality of $PM_{2.5}$ and O_3 together in Europe, and 15.8% globally. However, this mortality depends on the local relative emission amounts; for example, according to Brandt et al. (2011), the health effect of all Danish emissions on acute deaths in Denmark was negative, because the high NO_x emissions reduced domestic O_3 concentrations.

7. Conclusions

The total deposition of nitrogen to the Baltic Sea open water areas varied between 178 and 205 kt N, and the sulphur deposition from 77 to 101 kt S. The maximum N and S depositions were reached in 2010, the minimum N deposition in 2009 and the minimum S deposition in 2011. The proportions of dry deposition were low in the northern BS, increasing gradually southwards. There was a rather sharp dry deposition gradient over the shorelines. The depositions had a high seasonal variation while in winter and late autumn when the sea is open, high turbulence mixes long-range transported upper concentrations effectively close to the surface, and dry deposition velocities are also high. Additionally, most of the storms occur during these same seasons with stronger precipitation and higher winds. However, the ship emission originated NO_x deposition was highest during the summer due to the higher emissions and the faster chemistry converting compounds into scavengable species. Ship emissions occur near the surface, thus vertical mixing should not play as big a role as for long-range transported compounds. Ship emitted sulphur compounds are mostly in scavengable form, thus their seasonal deposition does not vary as much.

The ship emission originated depositions fraction of the total NO_x deposition to the BS varied during the 2008 to 2011 period from 12 to 14% while the respective contribution of sulphur deposition declined from

28% to 20% of the total modelled S deposition due to the sulphur directive restrictions. Ship emissions contributed from 20 to 40% of the grid average NO_2 concentration and from 10 to 25% of the SO_2 and SO_4 concentrations along BS coasts. In the eastern BS, for example, ship originated SO_4 concentrations fell to $>5\%$ of the modelled total sulphate concentration within 10–100 km of the coast. In general, the proportion of ship emitted concentrations mostly fell quite sharply with distance from the coastline.

The effect of the sulphur directive abatement of ships' sulphur emissions can be deduced indirectly from the proportion of SO_4 concentration in the whole $PM_{2.5}$ mass in Europe. The chemical composition of particulate matter at six urban background sites in Europe was studied during 7-week field campaigns (Sillanpää et al. 2006). The mean concentrations of $PM_{2.5}$ varied from 8.5 to 30 and from 5.4 to 29 μg m^{-3} for $PM_{2.5-10}$, $PM_{2.5}$ being composed mainly of organic matter, elementary carbon (EC), secondary inorganic aerosols and sea salt (SS), while the larger fraction contained soil-derived particles, carbonaceous compounds, SS and nitrate. Non-SS-SO_4 contributed from 14 to 31% to $PM_{2.5}$ and 0.8 to 6.8% to $PM_{2.5-10}$. NO_3 contributed from 1.1–18% to $PM_{2.5}$ and 3.7–14% to $PM_{2.5-10}$; NH_4 7.9–9.3% to $PM_{2.5}$ and 0.06–2.7% to the $PM_{2.5-10}$ fraction.

The model simulations from this study show that the share of ship originated sulphur particles in the modelled total sulphur along BS coastlines in 2010 was around 5% in the northern BS, 5–10% along the Polish coast, 2–5% along the Lithuanian coast, 10–20% north of Stockholm and Turku and along the coast of the eastern GoF, 20–30% on the Swedish coast south of Stockholm and in the south-west corner of Finland; it exceeds 30% only in the coastal areas of the Danish Straits.

The share of the modelled ship originated SO_4 concentration of the total $PM_{2.5}$ on BS coastlines thus varies from 0.3% to 12%, being approximately $<9\%$ along most ($>90\%$) of the coastline and $<5\%$ on ca 70% of the BS coastline. If the aerosol chemical composition of Sillanpää et al. (2006) is used, only 0.15–6% of the total PM mass <10 μm along the BS coastline is BS ship-originated sulphate. This percentage declines sharply with distance from the sea, so in the BS region the contribution of ship originated SO_4 concentrations to PM concentrations is on average very low, and their contribution to the mortality caused by PM concentrations in air should also be low.

The mortality caused by sulphur originating from Baltic Sea ship-emissions was most likely overestimated when the sulphur directive was enacted. The quantitative magnitude of the sulphur-emission effect on mortality should be re-evaluated. The work will continue in that all PM

emissions of BS ships will be modelled, because they produce the majority of the health problems caused by shipping traffic.

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