



SHEBA

Sustainable Shipping and Environment of the Baltic Sea region

BONUS Research Project

Call2014-41

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Database Deliverable on T2.2 (Current air pollution and deposition in the Baltic Sea area)

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Deliverable

Database of maps on current air pollution and deposition in the Baltic Sea region

Introduction

This Deliverable contains a database with spatial maps of surface air concentrations and atmospheric deposition output from regional model simulations of air pollution in the Baltic Sea region and the contribution of shipping to it. Two scenario runs were performed, one representing the current situation of air quality in the Baltic Sea region (scenario "CURRENT") and one representing the current situation without emissions from shipping in the Baltic Sea and North Sea. One important aspect is the deposition of nitrogen and sulphur compounds into the Baltic Sea. They contribute to eutrophication and acidification, respectively, and will be further used for marine ecosystem modelling in WP3.

The data displayed in the maps of this Deliverable are in netCDF file format. The files can be downloaded from the SHEBA data portal. The files contain hourly averaged concentrations or hourly accumulated deposition fields for various chemical species. One file is available per day of the simulated year.

Air concentration output from current situation (scenario "CURRENT"):

http://sheba.hzg.de/thredds/catalog/restrictedAccess/bigdata/DELI2.3/CONC/CURRENT/catalog.html

Air concentration output from current situation without emissions from shipping in the Baltic Sea and North Sea (scenario "NOSHIP"):

http://sheba.hzg.de/thredds/catalog/restrictedAccess/bigdata/DELI2.3/CONC/NOSHIP/catalog.html

Atmospheric deposition output from current situation (scenario "CURRENT"):

http://sheba.hzg.de/thredds/catalog/restrictedAccess/bigdata/DELI2.3/DEPO/CURRENT/catalog.html

Atmospheric deposition output from current situation without emissions from shipping in the Baltic Sea and North Sea (scenario "NOSHIP"):

http://sheba.hzg.de/thredds/catalog/restrictedAccess/bigdata/DELI2.3/DEPO/NOSHIP/catalog.html

Spatial Maps Metadata

A brief description of the applied regional air quality model systems for simulation of dispersion and chemistry of air pollutants over the Baltic Sea is presented. Three different regional atmospheric chemistry transport model systems are in use in WP2, each applied chemistry transport model (CTM) together with the meteorological model that provides the meteorological fields to drive the CTM will be described. Information on the computational grids for the applied CTM are given with respect to horizontal and vertical resolution, geographic extent and projection, as well as other configuration parameters of the setup such as the default chemical boundary conditions and land-based emission. The three CTM model systems are:

- EMEP (Simpson et al, 2012), used by IVL;
- SILAM (Sofiev et al., 2006), used by FMI;
- CMAQ (Byun and Ching, 1999, Byun and Schere et al., 2006), used by HZG.

The data of this Deliverable currently only includes output from the CMAQ model. It is subsequently planned to submit an ensemble average of the three models to the SHEBA data portal.

The vertical dimension of the CMAQ model extends up to 100 hPa in a sigma hybrid pressure coordinate system with 30 layers. Twenty of these layers are below approximately 2 km; the lowest layer extends to ca. 35 m above ground. The model runs with CMAQ were performed for the entire year 2012. In order to consider the interaction of pollutants from shipping emissions with emissions of pollutants (for example volatile organic compounds, VOC, nitrogen oxides, NO_x, ozone, O₃, and ammonia, NH₃) from natural and other anthropogenic sources in Europe, the CTM systems started with a simulation run on a European domain. The horizontal model resolution is 64 km × 64 km for the entire continent and 16 km × 16 km for Central and Northern Europe. Subsequently, runs for the nested high-resolution domains on 4 km × 4 km covering the northern as well as the southern part of the Baltic Sea region were performed with the CMAQ model.

The models were driven by meteorological data calculated with either WRF (Grell, 2006) or COSMO-CLM (Rockel et al., 2008). CMAQ model simulations were driven by the meteorological fields of the COSMO-CLM version 5.0 using the ERA-Interim re-analysis as forcing data. The meteorological runs were performed on a $0.11^{\circ} \times 0.11^{\circ}$ rotated lat-lon grid using 40 vertical layers up to 20 hPa for the whole of Europe. High-resolution meteorology obtained from COSMO-CLM on a $0.025^{\circ} \times 0.025^{\circ}$ grid resolution was used for the Baltic Sea regional simulations with CMAQ on a 4 km × 4 km grid.

Chemical boundary conditions for the model simulations were provided through hemispheric CTM simulations, from a SILAM model run on a $0.5^{\circ} \times 0.5^{\circ}$ grid resolution, which was provided by FMI. These were then used as initialization and boundaries for the concentrations of relevant chemical species for the model simulation of the outer model domain that covers the whole of Europe.

Land based emissions for the model simulations were calculated at HZG with the SMOKE for Europe (SMOKE-EU) emission model (Bieser, 2011), version 2.4. These emissions are based on national totals provided by EMEP (www.ceip.at) and point source emissions provided through the European point source emission register EPER. SMOKE-EU distributes these emissions in time and space to make them usable for the chemistry transport model systems. Some emissions, like biogenic volatile organic compounds were calculated inside the CTM during runtime.

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Shipping emissions are an important input for the high-resolution simulations with the three CTM systems. Shipping emissions for the Baltic Sea and the North Sea were calculated with the Ship Traffic Emission Assessment Model (STEAM; Jalkanen et al, 2009; 2012, Johansson et al, 2013). They are based on position data of individual ships taken from reports from the Automatic Identification System (AIS). This includes all merchant ships bigger than 300 GT. The shipping emission database consisted of hourly updated emission data on 2 km × 2 km grid resolution, stored in gridded binary files. The STEAM database provides shipping emissions for the compounds NO_x, SO_x, CO, CO₂ and Particulate Matter (PM), which is further divided into EC (Elementary Carbon), OC (Organic Carbon), SO₄ and mineral ash (Ash). Details about the construction of the emissions and where they can be found are given in the SHEBA D1.2 report and the D2.1 report. Leisure boat activity was not considered in the simulation for the "CURRENT" scenario. Shipping emissions were provided in two vertical layers with emissions below 36 m height and above, which were attributed to the corresponding model layers of the CTMs.

Based on the temperature anomalies (Figure 1) and total precipitation anomalies (Figure 2) for the decade 2004-2014 for Baltic Proper the year 2012 was agreed in WP2 as meteorological reference year for the CTM simulations. Year 2012 anomalies for both meteorological variables were closely aligned to the decadal average of the 2004-2014 period (zero-line in the two figures).



Figure 2: Anomaly of the air temperature in 2m height over Baltic Proper from the coastDat2 data set (Geyer 2014) modelled with COSMO-CLM in the decade 2004-2014. Year 2011 is indicated in blue color, year 2012 is indicated in red color. The zero-line corresponds to the decadal average of the 2004-2014 period.



Figure 2: Anomaly of the total precipitation over Baltic Proper from the coastDat2 data set (Geyer 2014) modelled with COSMO-CLM in the decade 2004-2014. Year 2011 is indicated in blue color, year 2012 is indicated in red color. The zero-line corresponds to the decadal average of the 2004-2014 period.

Spatial Maps Description

The geographic extent of this Deliverable covers the entire region from latitude 53.50° N (south) to 66.00° N (north) and longitude 9.00° E (west) to 31.00° E (east). Spatial maps were generated based on the high-resolution output of the CTM models on 4 km × 4 km covering the Baltic Sea region. Figure 1 gives an exemplary description for the high-resolution maps that are included in the Appendix of this Deliverable.

Spatial maps of surface air concentrations and deposition fluxes for a series of pollutants were produced. The deposition fluxes represented the total deposition to the surface as the sum of dry deposition and wet deposition. Particulate matter and gaseous compounds are deposited to the ground by wet and dry deposition, both of which are modelled by the CTMs.

In the models it is assumed that the dry deposition flux is directly proportional to the local concentration of the deposited compound at a certain reference height above the surface, with the dry deposition velocity as proportionality constant, which then bundles all the complexities of the dry deposition process. The process of dry deposition of gases and particles is generally represented as consisting of three steps: (1) aerodynamic transport down through the atmospheric surface layer to a very thin layer of stagnant air adjacent to the surface; (2) molecular (for gases) or Brownian (for particles) transport across this stagnant layer of air (the quasi-laminar sublayer) to the surface itself; (3) uptake at the surface (Seinfeld and Pandis, 2006).



Figure 1: Exemplary structure of spatial maps spanning from latitude 53.50° N (south) to 66.00° N (north) and longitude 9.00° E (west) to 31.00° E (east). Green shaded area is the high-resolution area which shows output from regional model runs with a grid resolution of 4 x 4 km². Blue outline marks the extent of the southern part of the Baltic Sea region and red outline marks the extent of the northern part of the Baltic Sea region, for which model output from two high-resolution nests were used. For the overlap area, the arithmetic mean of results from both nests was used. White areas of the map are covered by the output from the model nest with $16 \times 16 \text{ km}^2$ resolution.

For large particles, the dry deposition transfer is by turbulent air motion and by direct gravitational sedimentation. The dry deposition velocity for particles is calculated from the particle size distribution and from meteorological and land use information. The deposition algorithm includes an impaction term in the coarse and accumulation modes of PM. In wet deposition, particulate matter and gaseous compounds are transferred by rainfall.

Air concentrations of ozone

Monthly averaged surface air concentrations of ozone (O₃) show a seasonal cycle over the Baltic Sea region, with maximum values (40-50 ppbV) in late spring and minimum values (10-20 ppbV) in winter (Figure A1). With the exception of the summer months, shipping emissions cause reduction of ozone concentrations over the Baltic Sea due to regional scale titration of O₃ by NO and NO₂ emitted in the ship plumes. Reduction of O₃ due to shipping mainly happens in the western part of the Baltic Sea, on average by 10-20 %. O₃ is formed through the photolysis of NO₂ in sunlight. In summer, with increased insolation, photochemical ozone production due to NO_x and CO emissions from ships causes an increase by up to 15 %. A limitation of the model results for regional surface concentrations of O₃ over the Baltic Sea region is the lack of emission data on non-methane volatile organic compounds (NMVOC) from shipping in the STEAM inventory. Additional NMVOC emissions from shipping would serve as precursors of O₃ and enhance photochemical ozone production.

Air concentrations of sulfur dioxide

Monthly averaged surface air concentrations of sulfur dioxide (SO₂) are in general low over the Baltic Sea region, with values below 1.4 ppbV (Figure A2). St. Petersburg and Gdansk are cities with peak SO₂ concentrations ranging up to 4 ppbV or higher. During winter, the northern part of Poland also experiences high SO₂ concentrations due to residential heating with high sulfur (S) lignite-type coal. Over the Baltic Sea, shipping emissions have a high contribution to atmospheric SO₂ concentrations, in particular from March to September. In the summer months, shipping contribution is 80 % or more in a wide range around the main shipping routes of the Baltic Sea. The EU has implemented a sulfur emission control area for the North and Baltic seas, which means that fuels burned on ships in these areas must not contain more than 0.1 % S from 1st January 2015, and not more than 1.0 % S until that date. Since the STEAM inventory reflects the situation in 2012, the 1.0 % fuel sulfur content applies to the model simulations of the current situation.

Air concentrations of nitrogen dioxide

Monthly averaged surface air concentrations of nitrogen dioxide (NO₂) over the Baltic Sea in the background areas without shipping are below 4 ppbV, while along the main shipping routes concentrations of up to 8 ppbV are reached (Figure A3). From the model simulations it is evident that shipping emissions are the main contributor to ambient NO₂ concentrations over the Baltic Sea. In the summer months NO₂ air concentrations over the sea are almost entirely explained by shipping emissions. The STEAM inventory prescribes a split of 95:5 for the emission of NO:NO₂, which means that NO_x is mainly emitted as nitrogen oxide (NO). In the air, NO is however quickly converted to NO₂, so atmospheric NO_x will be mainly in the form of NO₂. The injection of reactive nitrogen into the Baltic atmosphere through shipping activities might have consequences with respect to eutrophication of marine and coastal ecosystems.

Air concentrations of particulate nitrate

Monthly averaged surface air concentrations of particulate nitrate (PM-nitrate) over the Baltic Sea shows a seasonal cycle with maximum values (up to $6 \ \mu g/m^3$) in early spring and winter and minimum values (0.8 - 2.2 $\mu g/m^3$) in summer (Figure A4). There is also a south-north gradient of PM-nitrate in early spring and winter, showing 3-4 times higher concentrations in the southern part of the Baltic Sea. PM-nitrate is a secondary aerosol, produced through the photochemical oxidation of NO₂ to gaseous nitric acid (HNO₃). A gas-phase reaction of HNO₃ with ammonia (NH₃), which is mainly emitted from farming activities, produces ammonium nitrate. The equilibrium gas/particle partitioning of ammonium nitrate is controlled by air temperature and humidity, which causes higher PM-nitrate concentrations during periods of the year with cold and wet weather. Shipping emissions are responsible for 40-70 % of the PM-nitrate concentrations during the summer months over the Baltic Sea.

Air concentrations of particulate ammonium

Particulate ammonium (PM-ammonium) over the Baltic Sea shows similar seasonality and spatial gradients as PM-nitrate due to its analogous formation mechanisms, when present as ammonium nitrate aerosol. Monthly averaged surface air concentrations of PM-ammonium has a seasonal cycle with maximum values (up to 4 μ g/m³) in early spring and winter and minimum values (0.4 - 1.1 μ g/m³) in summer (Figure A5). Shipping emissions are responsible for 30-60 % of the PM-ammonium concentrations during the summer months over the Baltic Sea. It should be kept in mind that since NH₃ is not directly emitted by ships, changes of PM-ammonium in the absence of shipping emissions are indirectly caused by changes of PM-nitrate through the gas/particle partitioning of ammonium nitrate.

Air concentrations of particulate sulfate

Monthly averaged surface air concentrations of particulate sulfate (PM-sulfate) over the Baltic Sea are in the range of $0.7 - 2.2 \ \mu g/m^3$, with highest concentrations in early spring and winter (Figure A6). PM-sulfate is a secondary aerosol, produced through the photochemical oxidation of SO₂ to sulfuric acid (H₂SO₄). Condensation of H2SO4 to existing particulate matter will increase PM-sulfate. Production of PM-sulfate also takes place in clouds, when SO2 becomes dissolved in cloud droplets and subsequently is oxidized to sulfate. Deposition of these S-containing species will contribute to the acidification of marine ecosystems and in-land lakes. Shipping emissions are responsible for 20-60 % of the PM-nitrate concentrations during the summer months over the Baltic Sea.

Atmospheric deposition of nitrogen

The chemistry transport model systems calculate atmospheric deposition, both wet and dry, of all substances treated by the chemistry algorithms. Concerning nitrogen deposition, these are aerosol bound nitrate and ammonium as well as gaseous NO, NO₂, NO₃ radical, HNO₃ and PAN. Figure A7 shows the monthly accumulated total nitrogen deposition over the Baltic Sea region. From spring to autumn there is a clear gradient between land and sea, with 2-3 times higher nitrogen deposition over land. In winter months, the picture changes and land and sea receive similar amounts of nitrogen deposition. Over the Baltic Sea, highest nitrogen deposition is predicted for the autumn months (SON), with maximum values of 1.1 kg-N/ha in the area around the island of Bornholm. Relative contribution of shipping emissions to monthly nitrogen deposition is highest from May to July with up to 60 % in the northern part of Baltic Proper, while it is on average 10 % for other parts of the Baltic Sea.

Atmospheric deposition of sulfur

Concerning sulfur deposition, the considered species are aerosol bound sulfate as well as gaseous SO_2 and H_2SO_4 . Figure A8 shows the monthly accumulated total sulfur deposition over the Baltic Sea region. Enhanced sulfur deposition is found in the coastal regions Denmark, Germany and Poland. Over the Baltic Sea, highest sulfur deposition is predicted for the late autumn and winter, with maximum values of 0.8 kg-S/ha in the southern part. Relative contri-

bution of shipping emissions to monthly sulfur deposition is highest from May to August with up to 70 % along the main shipping routes of the Baltic Sea.

Atmospheric deposition of mineral ash particles

In the CMAQ model, primary mineral particles are modelled as one type of accumulation mode aerosol, which includes particulate matter from natural emissions of mineral dust, windblown soil dust, as well as mineral ashes from incineration processes and from shipping activities. Figure A9 shows the monthly accumulated total mineral dust and ash deposition over the Baltic Sea region. Deposition of mineral particles is mainly coupled to the long-range transport of the mineral aerosols and their scavenging through precipitation in the Baltic Sea region. The difference between the scenarios for the current situation with and without shipping emissions will however reveal the contribution of mineral ash coming from emissions of ships. Relative contribution of shipping emissions to monthly mineral ash deposition is found to be highest in February and March with up to 20 % in various parts of the Baltic Sea, most importantly in the sea between the coast of Denmark and southern Sweden. However, total mineral ash deposition in the concerned areas is generally below 0.04 kg/ha. Deposition of mineral ash particles from shipping might provide input of phosphorus (P) to the marine ecosystems. Based on the mineral dust and ash deposition maps, mineral background particles, depending on their P-content, potentially deliver much higher loads of phosphorus to the sea.

On the difference map for March 2012, the limitations of the applied approach to determine the ship contribution becomes visible. Negative deposition changes (blue colour) were calculated for some areas in southern Finland when emissions from shipping are included. This contra-intuitive behaviour is explained by non-linearity of the aerosol growth and deposition processes: additional injection of mineral ash will cause condensation of gas-phase species to these particles, thereby shifting spatial patterns of the particle deposition. For longer accumulation periods (for example seasonal deposition totals) it is expected that such artefacts of the approach will be levelled out.

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Figure Appendix

Figure A1: Air concentration of O_3 from a model simulation of the current situation in the Baltic Sea region for year 2012: (a-l) Monthly concentration averages, January - December, and (m-x) Monthly average percentage contribution of shipping emissions, January - December, calculated as relative difference between the simulation runs for current situation with shipping emissions and current situation with no shipping emissions. Maps show percentage changes > 5%.

Figure A2: Air concentration of SO_2 from a model simulation of the current situation in the Baltic Sea region for year 2012: (a-l) Monthly concentration averages, January - December, and (m-x) Monthly average percentage contribution of shipping emissions, January - December, calculated as relative difference between the simulation runs for current situation with shipping emissions and current situation with no shipping emissions. Maps show percentage changes >10 %.

Figure A3: Air concentration of NO₂ from a model simulation of the current situation in the Baltic Sea region for year 2012: (a-l) Monthly concentration averages, January - December, and (m-x) Monthly average percentage contribution of shipping emissions, January - December, calculated as relative difference between the simulation runs for current situation with shipping emissions and current situation with no shipping emissions. Maps show percentage changes >10 %.

Figure A4: Air concentration of particulate nitrate from a model simulation of the current situation in the Baltic Sea region for year 2012: (a-l) Monthly concentration averages, January - December, and (m-x) Monthly average percentage contribution of shipping emissions, January - December, calculated as relative difference between the simulation runs for current situation with shipping emissions and current situation with no shipping emissions. Maps show percentage changes >10 %.

Figure A5: Air concentration of particulate ammonium from a model simulation of the current situation in the Baltic Sea region for year 2012: (a-l) Monthly concentration averages, January - December, and (m-x) Monthly average percentage contribution of shipping emissions, January - December, calculated as relative difference between the simulation runs for current situation with shipping emissions and current situation with no shipping emissions. Maps show percentage changes >10 %.

Figure A6: Air concentration of particulate sulfate from a model simulation of the current situation in the Baltic Sea region for year 2012: (a-l) Monthly concentration averages, January - December, and (m-x) Monthly average percentage contribution of shipping emissions, January - December, calculated as relative difference between the simulation runs for current situation with shipping emissions and current situation with no shipping emissions. Maps show percentage changes >10 %.

Figure A7: Atmospheric deposition of nitrogen from a model simulation of the current situation in the Baltic Sea region for year 2012: (a-l) Monthly concentration averages, January - December, and (m-x) Monthly average percentage contribution of shipping emissions, January - December, calculated as relative difference between the simulation runs for current situation with shipping emissions and current situation with no shipping emissions. Maps show percentage changes >10 %.

Figure A8: Atmospheric deposition of sulfur from a model simulation of the current situation in the Baltic Sea region for year 2012: (a-l) Monthly concentration averages, January - December, and (m-x) Monthly average percentage contribution of shipping emissions, January - December, calculated as relative difference between the simulation runs for current situation with shipping emissions and current situation with no shipping emissions. Maps show percentage changes >10 %.

Figure A9: Atmospheric deposition of mineral ash and dust (Ash) from a model simulation of the current situation in the Baltic Sea region for year 2012: (a-l) Monthly concentration averages, January - December, and (m-x) Monthly average percentage contribution of shipping emissions, January - December, calculated as relative difference between the simulation runs for current situation with shipping emissions and current situation with no shipping emissions. Maps show percentage changes >2.5 %.



Figure A1 cont.



Figure A1 cont.





Figure A2 cont.



Figure A2 cont.



Figure A3 cont.

Figure A3 cont.

Figure A4 cont.

Figure A4 cont.

Figure A5 cont.

Figure A5 cont.

Figure A6 cont.

Figure A6 cont.

Figure A7 cont.

Figure A7 cont.

Figure A8 cont.

Figure A8 cont.

Figure A9 cont.

Figure A9 cont.

Review Report

by Marine Systems Institute at Tallinn University of Technology

The SHEBA deliverable 2.3 gives an overview on air quality modelling in WP2 and its first results considering additional shipping influence over the Baltic Sea. These results will be used in WP 3 for the deposition rates of nutrients and particles into marine environment. The deliverable includes metadata on used atmospheric chemistry transport models and their setups. Graphical part of the deliverable consist of monthly deposition fields of different parameters for the reference conditions and relative change of those parameters due to shipping. Some of the main findings from the results are also explained in the deliverable.

Few uncertainties concerning phosphorus fraction in ash deposition remain still open.