# THE CONTRIBUTION OF SHIP EXHAUST TO AIR POLLUTION IN NORTH SEA COASTAL AREAS

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## ABSTRACT

*The contribution of benzo(a)pyrene (BaP) ship emissions to the BaP concentration over Europe is investigated in the first part of this study. Subject of the second part is a sensitivity study of the impact of additional $NO_x$, $SO_2$, $PM_{10}$ ship emissions on the BaP concentration and deposition distribution over Europe. The carcinogenic BaP which belongs to the group of polycyclic aromatic hydrocarbons (PAHs) occurs mainly particle bound at temperatures prevailing in the mid-latitudes. Hence, the impact of the aerosol precursor gases $NO_x$ and $SO_2$ as well as $PM_{10}$ on the BaP concentration and deposition distribution is investigated by means of the Community Multiscale Air Quality modelling system (CMAQ). CMAQ is a chemistry transport model which is setup on a 54 x 54 $km^2$ grid for Europe. Two different model runs, one excluding and one including ship emissions were performed for January and July 2000. For the sensitivity study three additional model runs with the other three pollutants were performed for January 2000. It is shown that the contribution of BaP ship emissions to the total BaP emissions is generally small but shows a noticeable seasonal variation. Furthermore, the sensitivity study shows that the other pollutants can cause a higher BaP concentration and deposition change over land than the added BaP ship emissions alone.*

***Keywords:*** *Chemical transport modelling, ship emissions, benzo(a)pyrene, air pollution.*

## 1. INTRODUCTION

Ship emissions can have considerable impact on atmospheric concentrations of several important pollutants especially in coastal areas (Endresen et al., 2003, Isakson et al., 2001, Tsyro and Berge, 1997). The most important ones are $CO_2$, $NO_x$, $SO_2$, CO, hydrocarbons, and particulates because of their role as e. g. greenhouse gas ($CO_2$), their contribution to acid rain ($NO_x$, $SO_2$), and/or their impact on human health (particulates) (Lloyd´s Register Engineering Services, 1995). Corbett et al. (2007) have recently shown that ship emissions lead to an increase in $PM_{2.5}$ ambient air concentration and therewith are responsible for an increase of cardiopulmonary and lung cancer deaths. Micropollutants affecting the human health as e. g. a variety of polycyclic aromatic hydrocarbons (PAHs) with their carcinogenic potential to humans and animals (ATSDR, 1995) have to be considered as well. PAHs mainly originate from incomplete combustion of organic matter like fuel oil. Benzo(a)pyrene (BaP) belongs to the group of PAHs and it is often used as their representative substance. BaP is a semivolatile, persistent organic pollutant (POP) which occurs at temperatures prevailing in middle Europe mainly bound to particles. Therefore, their atmospheric transport is closely linked to aerosols. Bound to particles the chemical degradation of PAHs is very slow (Esteve et al., 2006).

This study focuses on the contribution of $NO_x$, $SO_2$, $PM_{10}$ and BaP ship emissions on the BaP concentration and deposition distribution over Europe especially over North Sea coastal areas. $NO_x$ and $SO_2$ are known as important precursor gases for the formation of secondary aerosols (Seinfeld and Pandis, 2006). That is why they are of interest for the atmospheric transport of

BaP. For the investigation of the ship emission contribution in this study a bottom-up approach on the basis of ship movement data together with average engine loads and emission factors available in literature (Cooper and Gustafsson, 2004) were used to generate a ship emission inventory.

The ship emission inventory served as input for the Models-3 Community Multiscale Air Quality modelling system (CMAQ) (Byun and Ching, 1999, Byun and Schere, 2006). CMAQ is set up on a 54 x 54 $km^2$ grid for Europe. The model is driven by meteorological fields that were calculated with the Fifth-Generation Pennsylvania State University/ National Center for Atmospheric Research (NCAR) Mesoscale Model (MM5) (Grell et al., 1995) and adapted to the chemistry transport model with the Meteorology-Chemistry Interface Processor (MCIP, Otte, 1999). CMAQ has been modified at GKSS to model the transport of PAHs, in particular of BaP (Aulinger et al., 2007). Matthias et al. (2008) have shown that the model is applicable to North Sea coastal regions by evaluating model results by means of measured air concentrations and depositions.

## 2. INPUT DATA

### 2.1. Meteorological fields

The meteorological fields are derived from MM5 (Grell et al., 1995) model runs which were driven by ERA40 6 hourly global reanalysis data on a 1x1degree grid. We used four dimensional data assimilation of the ERA40 fields and applied the more sophisticated physical parameterisation schemes like Reisner 2 (Reisner et al., 1998) for cloud micro-physics, Kain Fritsch 2 (Kain, 2004) for cumulus representation and the MRF (Hong and Pan, 1996) scheme for the boundary layer to produce meteorological data which is as close as possible to wind, temperature and humidity observations.

### 2.2. Initial and boundary conditions for CMAQ

The boundary conditions for the simulations presented here were taken from MOZART (Horowitz et al., 2003) model results for the years 2000. The data has a resolution of 1 x 1° and one day. It includes several gas phase species ($O_3$, O, $O^1D$, CO, NO, $NO_2$, $SO_4$, $HO_2$, OH, PAN, HCOH, isoprene, terpenes and $HNO_3$), but no BaP. The modelled concentrations of these species were interpolated to the boundary of the CMAQ domain, which is one grid cell thick and updated hourly. The boundary conditions for BaP were zero at the western and southern border and at most of the northern border. At the eastern edge of the model domain, monthly average BaP values of a previous model run were assumed to avoid a large gradient in the BaP concentrations in the most eastern grid cells.

### 2.3. Land-based emissions

Emission data for the nitrogen, sulfur and volatile organic compounds as well as for aerosol particles was provided by IER Stuttgart based on EMEP area emissions and EPER point source emissions. The database used for land-based BaP emissions was derived from TNO (Denier van der Gon et al., 2005). The data were provided as annual bulk emissions and spatially distributed on the 50 x 50 $km^2$ stereographic EMEP (Environmental Monitoring Programme) grid. To serve as input data for CMAQ, the emissions were interpolated to the 54 x 54 $km^2$ grid. Furthermore, a temporal variation of the data meeting the seasonal dependence of the emissions with a minimum during summer months and a maximum during winter months was applied by linearly correlating emissions from residential combustion, the dominant source of BaP emissions, to the ambient air temperature. This leads to emissions which are by a factor of 20 higher in January than in July.

## 2.4. Ship Emissions

### 2.4.1 Ship Database

The vessel database was purchased from Lloyds Marine Intelligent Unit (LMIU). It consists of a vessel characteristic database and a vessel movement database and it includes all commercial vessels equal to or greater than 100 gross tonnages (GT). The vessel characteristic database comprises information on e. g. vessel type, engine type, numbers of engines, engine speed at the crankshaft, fuel type, power and maximum speed. The ship movement database provides information on ship movements for the year 2000 in parts of Europe (riparian states of the North and Baltic Sea, Atlantic coast of France, Spain and Portugal). This is covered by 15625 ships which perform 651825 movements on 58324 different routes. The ship movement database consists of previous departure, arrival, departure and next arrival places and dates with a daily time resolution.

### 2.4.2 Ship emission factors

Emission factors (power-based in g/kWh) used in this study are obtained from Cooper and Gustafsson (2004) who compiled these emission factors for Sweden´s international reporting duties. These emission factors rely mainly on guidebooks provided by EMEP (EMEP, 2002) and IPCC (IPCC, 1997) as well as on a report for the European Commission (Whall et al., 2002) and additional ship exhaust measurements. The emissions from ships are affected by several characteristics of the ships as well as the fuel used. The engine type determines the combustion conditions and therefore the emissions of some pollutants. Factors affecting emissions from ships are presented in detail in Whall et al. (2002). Due to a very limited dataset for determining the emission factor for BaP, separate engine and fuel specific factors are not provided. Thus, the BaP emission factor has to be considered as a possibly large error source. Most likely the emission factor of Cooper and Gustafsson (2004) is at the low end of the possible values because it is similar to BaP heavy-duty diesel vehicle emission factors (Doel et al., 2005), although the PAH content of the fuel influences the BaP emissions and the majority of vessels use residual oil with extreme poor quality.

### 2.4.3 Ship emission dataset

The procedure how to obtain the ship emission dataset is displayed in Figure 1. The vessel movement database provides the basis for calculating the routes of the vessels. It is assumed that all vessels take the shortest routes between two ports at sea. The required temporal

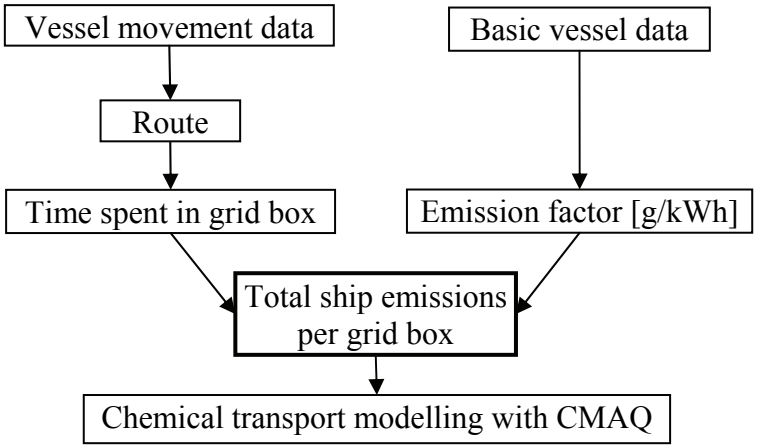


**Figure 1:** Outline of the procedure to generate the ship emission dataset

resolution of the ship movements is computed by means of the assumption that all vessels arrive and depart at 6 a.m. for more than one day travelling time. If they leave the same day they leave the port at 6 p.m. or even earlier if they call another port the same day. The

travelling time is distributed equally to all passed grid boxes on their route. The ship emissions per grid box and time step are calculated for the different pollutants by means of the corresponding emission factors and the engine power of the particular vessel.

The compiled ship emission dataset was compared to $NO_x$ ship emission data from EMEP available at their homepage (http://www.emep.int). For Northwest Europe our $NO_x$ ship emissions (1.56 Tg per year) are 6 % higher than these of EMEP (1.47 Tg per year). Hence, the close agreement in the target area, the North Sea, implies that the ship movements have been treated correctly.

Figure 2 presents monthly averaged land-based (a) and marine (b) BaP emissions for January 2000. The ship emissions account for approximately 0.1 % of the total emissions in coastal areas. Cumulated for the model domain and the whole year 2000 663 t BaP were emitted by land-based emission sources. In comparison ships emitted only 112 kg BaP (0.02 %) in the same time and area. Note that ship movements in the Mediterranean sea were not taken into account.

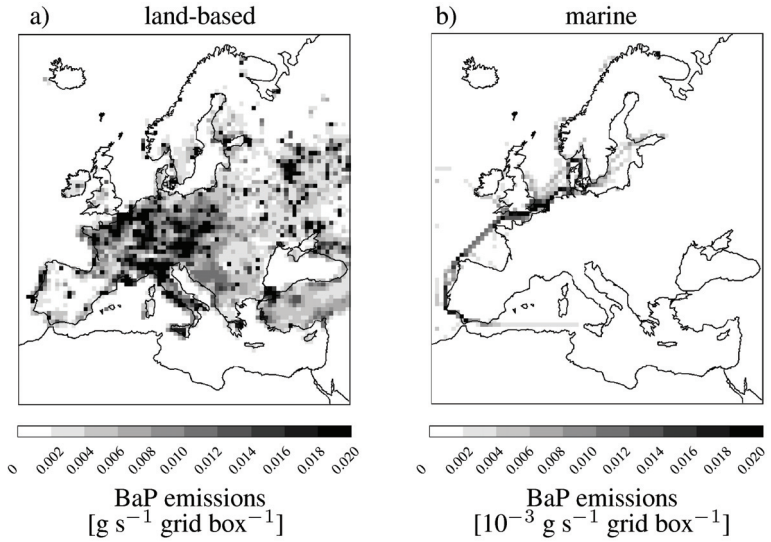


**Figure 2:** Average land-based (a) and marine (b) BaP emissions for the 54 x 54 km$^2$ grid for January 2000

## 3. METHODOLOGY

The land-based and marine emission datasets serve as input into the Eulerian air quality modelling system CMAQ which computes the concentration and deposition distribution over Europe. To investigate the contribution of BaP ship emissions to the BaP air pollution in coastal areas two different model runs were performed with CMAQ, one model run including all land-based emissions and the BaP ship emissions (model run: $s_0$, see Table 1) and the reference model run just including the land-based emissions (model run: lb, see Table 1). The seasonal impact is investigated by comparing January and July monthly average BaP concentrations as representatives for winter and summer. For the sensitivity study three additional CMAQ model runs were performed to investigate the influence of $NO_x$, $SO_2$ and $PM_{10}$ ship emissions on the BaP concentration and deposition distribution over Europe. This sensitivity study is only performed for January because aerosol concentrations are typically much higher in winter than in summer. Table 1 illustrates the setup of the different model runs.

**Table 1:** Description of the different model runs

| Input data | Model run | | | | |
|---|---|---|---|---|---|
| | lb | $s_0$ | $s_{SO2}$ | $s_{NOx}$ | $s_{PM}$ |
| Land-based emissions | + | + | + | + | + |
| Marine emissions | - | BaP | BaP + $NO_x$ | BaP + $SO_2$ | BaP + $PM_{10}$ |

Each model run includes the land-based emissions and different additional marine emissions. The reference model run (lb) contains only land-based emissions. The impact of the ship emissions on the BaP concentration and deposition distribution was calculated by the difference between the $s_0$-, $s_{SO2}$-, $s_{NOx}$-, $s_{PM}$-model run results (Table 1) and the lb-model run results.

## 4. RESULTS

The BaP results of the different model runs for BaP in the lowest model layer are presented and discussed in the following subsections. BaP concentrations refer to the monthly averaged concentrations and the BaP dry and wet depositions refer to the monthly cumulated values.

### 4.1. Reference model run

Figure 3a) shows the BaP concentration, Figure 3b) the dry deposition and Figure 3c) the wet deposition over Europe of the reference model run (lb) for January (1st row) and July (2nd row) 2000.

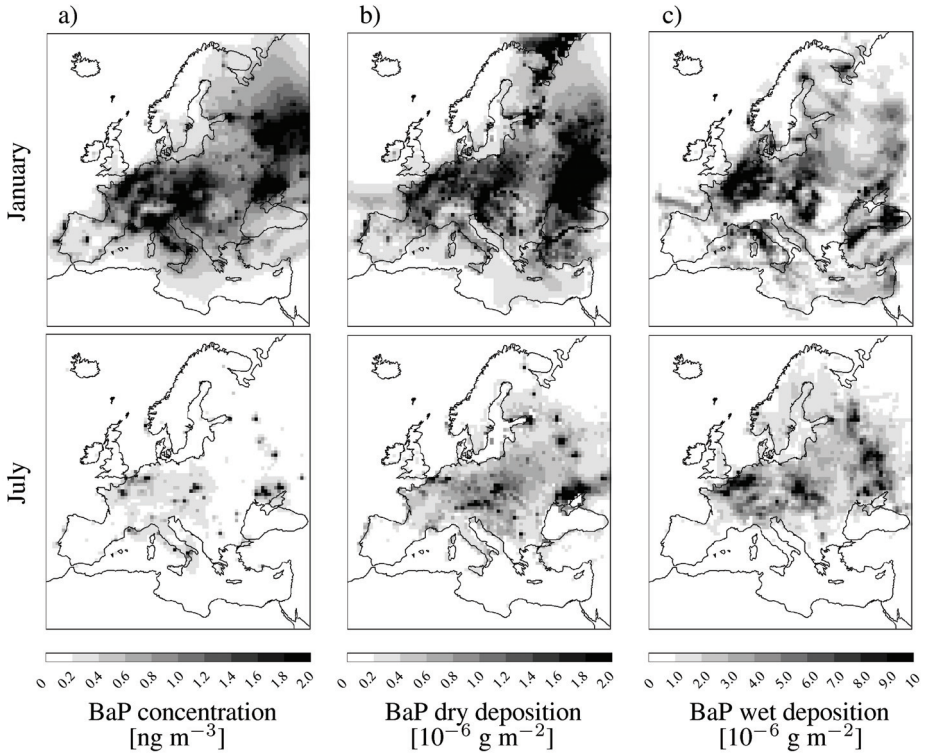


**Figure 3:** Modelled BaP concentration (a), cumulated dry deposition (b) and cumulated wet deposition (c) with land-based emissions for January (1st row) and July 2000 (2nd row)

BaP concentration peaks appear close to the emission sources in regions of congested urban and industrial areas. The deposition is dominated by wet deposition which is by a factor of approximately four higher than the dry deposition. High deposition rates are noticed close to the emission sources except for Russia and the Ukraine in January. Especially the Ukraine

shows comparably higher dry deposition of BaP. The BaP concentration shows a significant difference between the two months. This represents the strong seasonal dependency of the land-based emissions.

### 4.2. BaP ship emissions model run

The impact of the BaP ship emissions on the air pollution of coastal areas is illustrated in Figure 4. In contrast to the results of the lb-model run (Figure 3) it can be seen that BaP concentration changes due to the ship emissions do not vary significantly between January and July (Figure 4a). The January and July BaP concentrations differ by a factor of approximately two which can be ascribed to a decrease of vessel movements during winter. BaP concentrations reach their maximum in the Strait of Dover with average values of $0.6 \cdot 10^{-3}$ ng m$^{-3}$ in January and $1.1 \cdot 10^{-3}$ ng m$^{-3}$ in July. The increase of the BaP concentrations over the North Sea compared to the reference case is 0.2 % in January and 0.8 % in July Figure 4b). The reason for this seasonal difference is that in winter significantly higher land-based emissions are observed than in summer. The impact of the BaP emissions is maybe underestimated because of the highly uncertain emission factor.

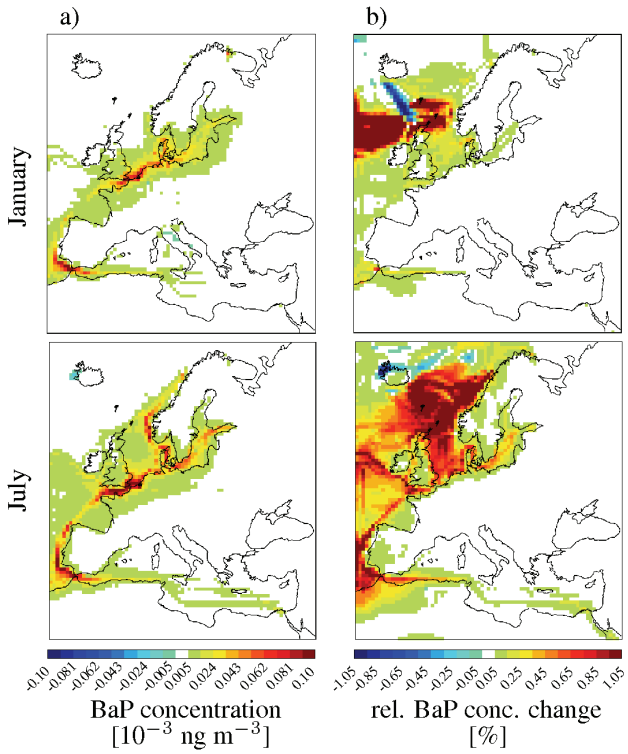


**Figure 4:** BaP concentration change caused by BaP ship emissions (a) and its relative change compared to the reference case (b) for January (1st row) and July 2000 (2nd row)

### 4.3. Sensitivity study – impact of $NO_x$, $SO_2$ and $PM_{10}$ ship emissions on BaP concentration and deposition

The contributions of the ship emissions to the BaP concentration for the four different model runs ($s_0$, $s_{SO2}$, $s_{NOx}$, $s_{PM}$) are shown in Figures 5a$_1$-a$_4$. BaP ship emissions alone increase the BaP concentration at the Belgian, Dutch and German North Sea coastline by around $0.5 \cdot 10^{-3}$ ng m$^{-3}$ with a decreasing gradient towards the open North Sea (Figure 5a$_1$) and on land. The addition of the aerosol precursor gases $NO_x$ and $SO_2$ ($s_{NOx}$, $s_{SO2}$) lead to a pronounced decrease of around $2 \cdot 10^{-3}$ ng m$^{-3}$ in the ground level BaP concentration over land (Figures 5a$_2$-a$_3$) whereas the supplement of $PM_{10}$ ($s_{PM}$) results in a concentration increase of

$2 \cdot 10^{-3}$ ng m$^{-3}$ to $4 \cdot 10^{-3}$ ng m$^{-3}$ over wide areas of Europe (Figure 5a$_4$). In comparison to the total BaP concentration (Figure 3a) the contribution of the ship emissions is small but noticeable Europe-wide. The increase of the BaP concentration with the $s_0$-model run amounts to 0.1 to 0.2 % over the North Sea. With the $s_{PM}$-model run (in Figure 5a$_4$) the BaP concentration is increased by 0.5 % at the North Sea coastline and amounts to 0.1 to 0.2 % over Spain, France and Eastern Europe (Figure 5a$_4$). In contrast, the addition of $NO_x$ and $SO_2$

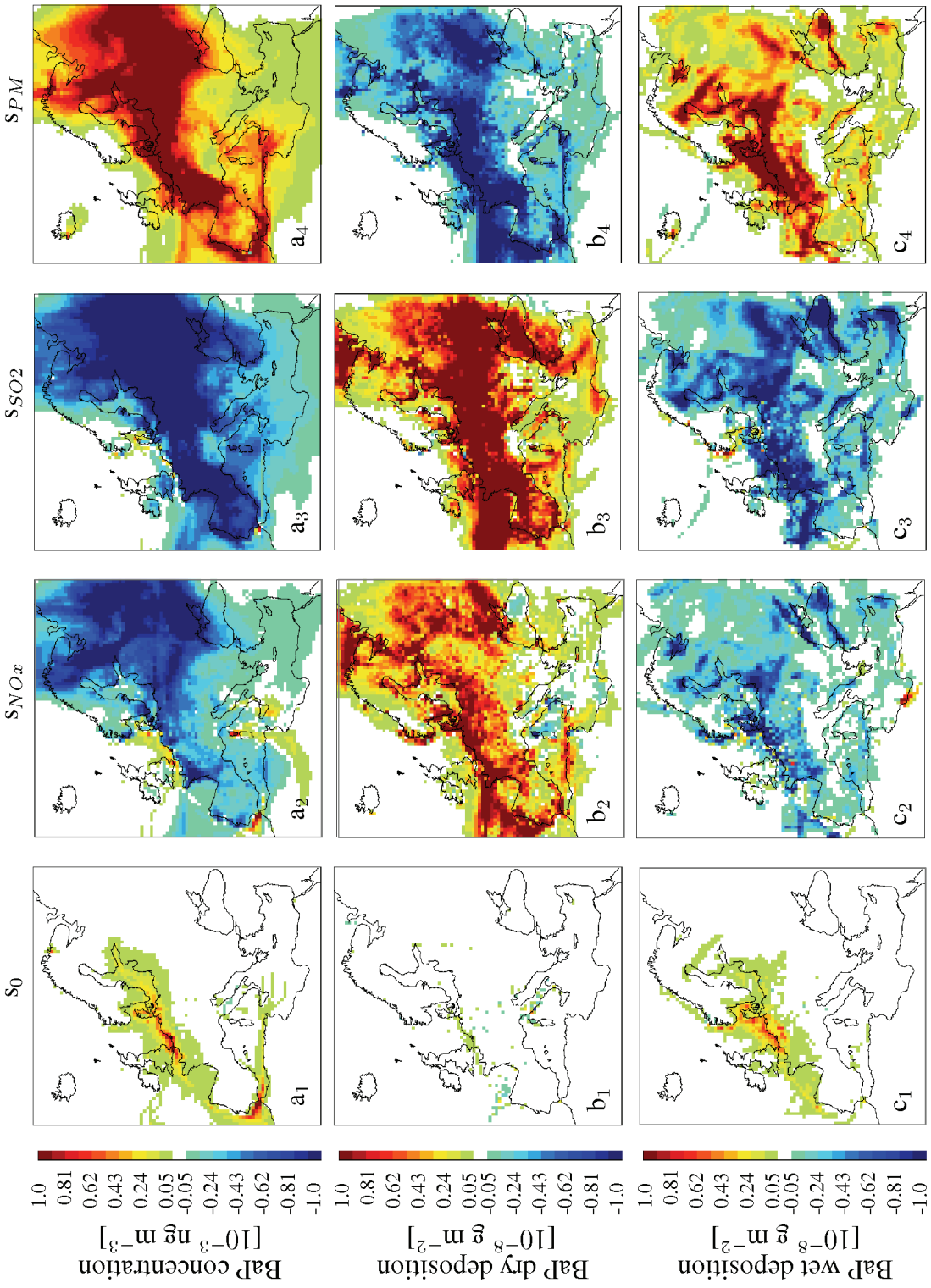


**Figure 5:** Modelled average BaP concentration changes (1st row: a$_1$-a$_4$), modelled accumulated BaP dry deposition changes (2nd row: b$_1$-b$_4$) and modelled accumulated BaP wet deposition changes (3rd row: c$_1$-c$_4$) caused by the addition of the ship emissions for the different model runs $s_0$-$s_3$ for January 2000

ship emissions ($s_{NOx}$, $s_{SO2}$) decreases the BaP concentration by 0.2 to 0.3 %. At the same time the dry deposition increases (Figures $5b_2$-$b_3$) which could be an explanation for the decrease of the BaP concentration.

The dry deposition results of the $s_{NOx}$, $s_{SO2}$ and $s_{PM}$-model runs (Figures $5b_2$-$b_4$) show a decrease in regions where the BaP concentration (Figures $5a_2$-$a_4$) increase and vice versa. The dry deposition of the $s_0$-model run does not change noticeably compared to the reference case. Figure $5b_4$ displays the BaP dry deposition of the $s_{PM}$-model run and shows a Europe-wide decrease of $0.2 \cdot 10^{-3}$ g $ha^{-1}$ (approximately -5 %) at the North Sea coastline. The increase of BaP dry deposition of the $s_{SO2}$-model run is a little smaller than for the $s_{NOx}$-model run With $SO_2$ it amounts to 0.5 to 1 % over Central and Eastern Europe and up to 3 % over Scandinavia whereas with $NO_x$ it amounts to 1 to 2 % over Central and Eastern Europe and 4 % over the Iberian Peninsula.

In contrast to the dry deposition the BaP wet deposition (Figures $5c_1$-$c_4$) increases when the concentration (Figures $5a_1$-$a_4$) increases and the opposite way around. The differences of the dry deposition values between the different model runs are of the same order of magnitude as those for the wet deposition (Figures $5b_1$-$b_4$) but with opposite sign. The BaP dry deposition is slightly larger. This is contrary to the absolute dry and wet deposition ratio of the reference case (Figures 3b-c) where the wet deposition is by a factor of approximately four higher than the dry deposition. The relative change of the BaP wet deposition generated with $s_0$, $s_{NOx}$, $s_{SO2}$ and $s_{PM}$-model runs compared to the result of the reference case shows that the relative change is about a factor of ten smaller than observed for the dry deposition. The $s_0$-model run leads to a relative increase of 0.1 to 0.2 % over the North Sea compared to the lb-model run results. The results of the $s_{NOx}$ and $s_{SO2}$-model runs show a relative decrease of up to 0.1 % for $NO_x$ and 0.1 to 0.3 % for $SO_2$ over wide areas of Europe. The $s_{PM}$-model run results in a BaP wet deposition increase in general of 0.1 to 0.2 % over land and a more pronounced increase of up to 0.5 % over the North Sea and Denmark.

The relative change of the BaP concentration and the dry and wet deposition with ship emissions is always more pronounced over sea than over land because the impact of the high land-based emissions dominates the BaP concentrations over land. This effect is also seen in coastal areas depending on the wind direction and the influence of land-based emissions.

It is remarkable that the BaP concentration is decreasing when adding the aerosol precursor gases $NO_x$ and $SO_2$ ($s_{NOx}$- and $s_{SO2}$-model runs). In contrast the BaP concentration increases in the $s_{PM}$-model run. The loss or gain of the BaP concentrations is found as an increase or decrease of the BaP dry deposition whereas the BaP wet deposition change follows the the concentration change. An explanation is difficult because of the complex chemical reactions, aerosol formation and the aerosol-gas partitioning in CMAQ. The generated secondary aerosols and their interaction with BaP will be further investigated.

## 5. CONCLUSION

The contribution of BaP ship emissions on the BaP concentration in coastal areas was investigated in this study. Secondly, a sensitivity study on the impact of the major ship emissions $NO_x$, $SO_2$ and $PM_{10}$ in addition to BaP ship emissions on the BaP concentration and deposition was performed. It was found that the impact of BaP ship emissions is small but it increases from 0.2 % in January to more than 0.8 % in July because the influence of the strongly seasonal dependent land-based emissions decreases significantly in summer. A higher impact on the concentration change might be underestimated because the BaP emissions are probably underestimated due to the highly uncertain emission factor. The sensitivity study showed that the investigated pollutants have a noticeable impact on the BaP

concentration and deposition Europe-wide even though the absolute concentration and deposition change is small. The aerosol precursor gases $NO_x$ and $SO_2$ which are added with the ship emissions lead to a decrease of 0.1 to 0.3 % in concentration and wet deposition and to an increase of 0.5 to 4 % of the dry deposition over wide areas of Europe. In contrast the additional $PM_{10}$ ship emissions result in an BaP concentration and wet deposition increase and in a decrease of the BaP dry deposition. These different findings are remarkable because all of the three added substances lead to an increase of aerosols in the model. The reason for the different impact of primary and secondary aerosols on the BaP concentration and deposition is still investigated. Furthermore, the added ship-based aerosols affect beside the ship emitted BaP also the land-based emitted BaP. The BaP changes in concentration and deposition are larger than the amount of the additional ship emitted BaP.

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