EURODELTA-CARB EXERCISE: INTERCOMPARISON OF MODELLED ESTIMATES OF BENZO(A)PYRENE (BAP) IN EUROPE

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Abstract: Concentrations of benzo(a)pyrene (BaP) are routinely measured throughout Europe as a marker of the carcinogenic risk of polycyclic aromatic hydrocarbons (PAHs). Due to the low density of measurement locations in many countries, estimates of the spatial distribution of BaP concentrations rely heavily on modelling estimates. In this study, we compare the temporal and spatial distributions of BaP concentrations simulated in Europe during the period December 2017 to December 2018 by four chemistry transport models (CTMs): CHIMERE, GLEMOS, MINNI, SILAM. Modelling results have been evaluated with the observed concentrations from the EMEP monitoring network. The four models used the same EMEP BaP emission inventory but other input data (such as meteorology, temporal emission profiles, land use, etc.) varied from model to model. Although the results show a large variation between the model outputs, both the spatial and temporal distributions of concentrations, similar characteristics in the results (e.g. areas where all models under- or overestimate concentrations) provide an indication of uncertainties in the emission data and/or the observed values.

Keywords: benzo(a)pyrene; chemistry transport models; intercomparison

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) comprise a large group of semi-volatile, hydrophobic organic compounds ubiquitous in the environment. Many PAHs are known or suspected to have carcinogenic, mutagenic, and teratogenic properties that pose a risk to human health and ecosystems (Theakston, 2000; Ravindra et al., 2007). High concentration levels and limited progress in reducing atmospheric concentrations of PAHs in the EMEP (European Monitoring and Evaluation Programme) countries during the last two decades have led to BaP becoming an important issue in the Long-term Strategy of the Convention on Long-range Transboundary Air Pollution for 2020–2030 and beyond (UNECE, 2018). PAHs are mostly released to the environment as unintentional by-products of incomplete combustion of biomass and fossil fuels. The main emission sectors of anthropogenic PAH include domestic burning of wood and coal, industrial activities, road transport, and agricultural sources (Ravindra et al., 2007).

Approaches to the assessment of PAH pollution levels and adverse effects are often based on the use of BaP as a marker compound. High measured annual mean concentrations of BaP, exceeding the EU target value (Directive 2004/107/EC), are common for countries of Central and Southern Europe as well as of some of the EECCA (Eastern Europe, Caucasus and Central Asia) countries. Furthermore, a significant part of the population of EMEP countries lives in areas where the BaP air concentration level exceeds the WHO (World Health Organisation) reference value (WHO, 2010; EEA, 2020). The knowledge of BaP concentrations throughout Europe is important since it is a marker of the carcinogenic risk of PAHs. Due to very few measurement locations in many countries, estimates of the spatial distribution of BaP concentrations rely heavily on modelling estimates.

The Eurodelta-Carb intercomparison of BaP modelling was initiated by the Task Force on Measurement and Modelling of the EMEP program (TFMM) at the beginning of 2021 in the framework of a broader scientific investigation devoted to the modelling of secondary organic aerosol and black carbon pollution levels in Europe. This study evaluates the capabilities of several state-of-the-art chemical transport models

(CTMs), applied in air quality assessments at national and European scales, to simulate BaP pollution. This model intercomparison aims to estimate the spatial distribution of BaP in Europe highlighting the areas that exceed the annual limit value of 1 ng/m^3 set out in Directive 2004/107/EC. Due to measurements' scarcity, currently this can only be done using results from atmospheric dispersion models.

The results of the initial phase of the intercomparison presented here provide indications of the areas with major uncertainties in the simulation of BaP concentrations, favouring the harmonization of models' inputs and processes and of the applied methodologies and reduction of uncertainties. In addition, the model estimates have been evaluated using methods that have been developed by the HARMO community, thus harmonizing the results of this study with many analyses presented in previous conferences.

MODEL SETUP AND INPUT DATA

The Eurodelta-Carb BaP modelling exercise is focused on the time period from the beginning of December 2017 to the end of 2018. Simulations of BaP were performed using four CTMs: CHIMERE, GLEMOS, MINNI and SILAM, developed and applied by the modelling teams of INERIS (France), CIEMAT (Spain), MSC-E (EMEP), ENEA (Italy), and FMI (Finland). All models used the same modelling domain and gridded BaP annual emissions data for 2018, provided by the EMEP Centre on Emission Inventories and Projections (CEIP) (Figure 1). Other input data and parameterizations, such as meteorological input, intra-annual variations of BaP emissions, emissions of other pollutants, boundary conditions, model parameterizations, were specific to each model. The participating models have different degrees of complexity with regard to BaP modelling. In particular, CHIMERE, GLEMOS, and MINNI consider BaP as a reactive semi-volatile substance that undergoes gas-particle partitioning and degradation in the atmosphere due to chemical reactions with OH in the gaseous form. Also, GLEMOS and MINNI include the chemical reaction of BaP with ozone in particulate form. All three models consider deposition of gaseous and particulate BaP. In the case of SILAM, the model simulations were carried out assuming that BaP is an inert substance emitted to the atmosphere in particulate phase and subject only to dry and wet deposition processes.

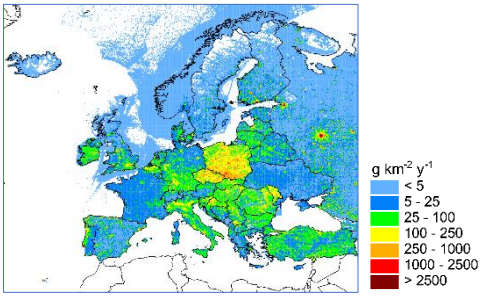

Figure 1. Eurodelta-Carb modelling domain and spatial distribution of annual BaP emission fluxes for 2018.

RESULTS AND DISCUSSION

This section shows the model simulations' results for the base case run for 2018. Spatial distributions of annual mean total BaP air concentrations, simulated by CHIMERE, GLEMOS MINNI and SILAM are shown in Figure 2. The three models that treat BaP as a reactive semi-volatile substance predicted similar patterns of BaP pollution levels for most regions in Europe. SILAM, on the other hand, that treats BaP as an inert substance, estimated substantially higher concentrations. The largest concentrations were estimated by all three models for the countries in Central Europe as well as for Northern Italy and some areas in Eastern Europe. The lowest concentrations were predicted for the countries of Northern Europe and remote areas. In general, relatively higher concentrations were simulated by SILAM, followed by CHIMERE, GLEMOS and MINNI. Other than the fact that SILAM treats BaP as an inert substance, the differences between the simulated BaP concentrations may be attributed to different model parameterizations (e.g. for gas-particle partitioning, degradation, and deposition processes) as well as to different meteorological inputs. Additional contributions could also have been made by different emission temporal profiles and concentrations of reactants such as ozone (O_3) used in the models to estimate BaP chemical transformations.

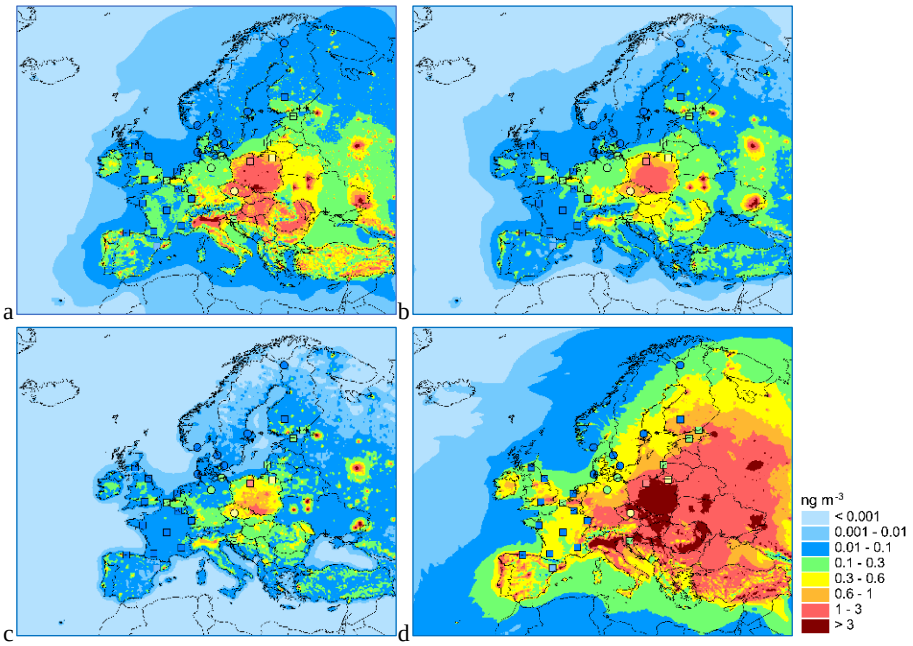

Figure 2. Maps of annual mean modelled total (gaseous + particulate phase) BaP atmospheric concentrations in 2018 simulated by CHIMERE (a), GLEMOS (b), MINNI (c) and SILAM (d) for the base case model run. Observed total and particulate phase BaP concentrations, reported by the EMEP monitoring stations, are shown as coloured circles and squares, respectively, on the same scale as the modelled values.

The evaluation of the model output against measurements was carried out using the data of 29 EMEP monitoring stations for the year 2018. Of these, 9 stations located in Central and Northern Europe measured total BaP concentrations, whilst concentrations in particulate form were measured at 20 stations covering a wider geographical area (Figure 2).

For the model-measurement comparison (excluding SILAM), the daily mean modelled total or particulate phase concentrations, depending on the type of measurement, were extracted from the model output files for the station locations. Modelled values were then averaged to the temporal resolution and periods of the observations (e.g. daily or weekly). A summary of the statistical analysis of the modelled and observed annual mean BaP concentrations is presented in Table 1. All the models reproduced the spatial pattern of observed total and particulate BaP concentrations well with correlation coefficients (R) of 0.84-0.96. CHIMERE and GLEMOS tended to slightly overestimate observed total BaP levels with a mean bias of about 4%, whereas MINNI underestimated the measured values with a mean bias of -53%. For the particulate BaP concentrations, CHIMERE overestimated concentrations with a mean bias about 9%, while GLEMOS and MINNI underestimated the observed concentrations with mean biases of -19% and -52%, respectively. Estimated total BaP concentrations were within a factor of 2 of the measured values for 89%, 78%, and 11% of monitoring stations for CHIMERE, GLEMOS, and MINNI, respectively, whereas for BaP in particulate phase they were within a factor of 2 for 80%, 70%, and 40% of monitoring stations. The fraction of model values that were within a factor of 3 from measurements is larger. In particular, modelled concentrations of CHIMERE and GLEMOS were within the agreement of a factor of 3 for all the stations that measured total BaP concentrations, and for 90% and 85%, respectively, for stations that measured particulate BaP concentrations.

Table 1. Summary of statistical metrics, calculated on the basis of annual mean total and particulate phase BaP air concentrations for 2018, observed at EMEP monitoring stations and estimated by CHIMERE, GLEMOS and MINNI in the base case model run.

Models	**Mean ($ng\ m^{-3}$)**	**NMB[a] (%)**	**R[a]**	**RMSE[a] ($ng\ m^{-3}$)**	**F2[a] (%)**	**F3[a] (%)**
Total BaP concentrations (9 stations), mean observed 0.116 $ng\ m^{-3}$						
CHIMERE	0.120	3.9	0.93	0.058	89	100

GLEMOS	0.121	4.3	0.91	0.087	78	100
MINNI	0.054	-53.3	0.86	0.090	11	22
Particulate BaP concentrations (20 stations), mean observed 0.156 ng m^{-3}						
CHIMERE	0.170	8.9	0.84	0.128	80	90
GLEMOS	0.126	-19.3	0.96	0.095	70	85
MINNI	0.075	-52.1	0.93	0.168	40	70

[a] NMB is normalized mean bias; R is the spatial correlation between modelled and observed concentrations; RMSE is the root mean square error; F2 and F3 represent fractions of sites for which the modelled value is within a factor of 2 and 3, respectively, of the observed value.

The evaluation of annual mean modelled BaP concentrations against the measurements of total and particulate BaP concentrations at EMEP monitoring stations is shown in the scatter plots in Figure 3. An overestimation of observed particulate BaP concentrations was found for two Spanish stations ES0008R and ES0014R for all three models. For other stations, different kinds of discrepancies were obtained. In particular, for CHIMERE, an overestimation by more than a factor of 2 was found for the stations GB0048R, GB1055R, and NL0091R that measured particulate BaP. In the case of GLEMOS, the largest underestimation (more than a factor of 3) was found for total BaP observed at DE0009R and FI0036R, and for particulate BaP at FR0023R and FR0025R. In case of MINNI, the greatest deviations (underestimation by more than a factor of 5) were found for the stations DE0001R, DE0009R, and FI0036R that measured total BaP, and for LV0010R and FR0023R that measured particulate BaP.

The scatter plots indicate that CHIMERE modelling results have the highest regression slope value (0.81) followed by GLEMOS (0.77) and MINNI (0.44). The scattering of modelled-observed pairs is best for GLEMOS (coefficient of determination 0.88) compared with MINNI and CHIMERE (0.83 and 0.80, respectively).

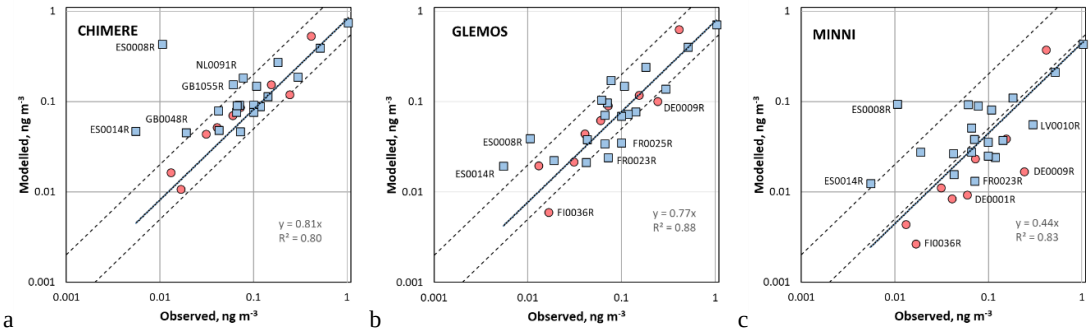

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Figure 3. Scatter plots on a log-log scale of the comparison of modelled BaP concentrations (total and particulate) simulated by CHIMERE (a), GLEMOS (b), and MINNI (c) for the base case model run with measurements of EMEP monitoring stations in 2018. The region between the dashed lines indicates the model estimates within a factor of two of the measured values and the solid line is the linear regression of all data points. Total BaP concentrations are shown as red circles and particulate BaP concentrations are shown as blue squares. The stations, for which the models have the largest deviations from the measurements, are indicated by their names.

Figure 4 shows examples of modelled and observed BaP time series for two EMEP monitoring stations, namely, CZ0003R and PL0009R that measured total and particulate BaP concentrations, respectively. For these stations, the comparison of modelled and observed concentrations shows, in general, a good level of agreement. Model estimates capture the high levels of observed concentrations in the cold season, and low concentrations in the warm season, as well as peak concentrations. However, in some of the episodes, especially in winter months, the models underestimate measured concentrations. Differences between the modelled and measured intra-annual variations of BaP concentrations may be explained both by an underestimate of emissions and by the uncertainties in the temporal disaggregation of BaP emissions that were applied in the model simulations.

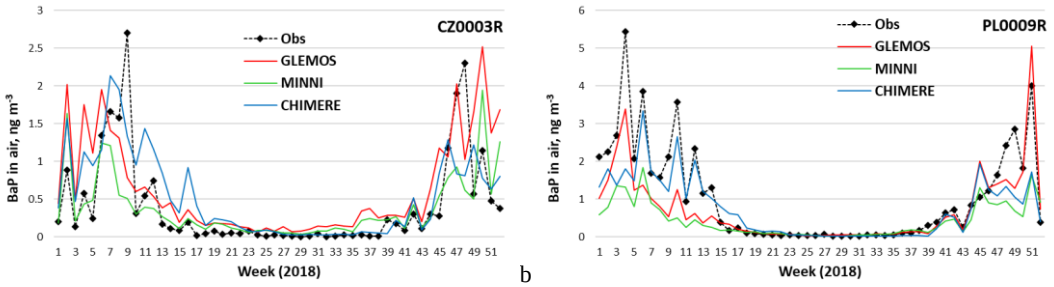

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Figure 4. Intra-annual variations of total BaP concentrations observed at the EMEP station CZ0003R (a) and particulate BaP concentrations observed at the station PL0009R (b) and the total and particulate BaP concentrations simulated by CHIMERE, GLEMOS, and MINNI in the base case model run for 2018.

This preliminary intercomparison of model simulations with prescribed officially reported BaP emissions demonstrates a generally reasonable level of agreement between model estimates. On the other hand, for some of the stations, modelled concentrations significantly deviated from the observed values indicating possible uncertainties in emission estimates, modelling approaches and measurements. More detailed analyses will be, therefore, performed to explore the reasons for the differences between the outputs of participating models and substantial over- and underestimates of observed BaP concentrations for some of the stations. The next stage of the study will focus on the sensitivity analyses, an evaluation of the impact of meteorological drivers and an analysis of other model outputs such as BaP concentrations in precipitation and deposition fluxes and concentrations of species affecting BaP chemical transformations in the atmosphere.

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