Eurodelta-Carb intercomparison of B(a)P models

The Eurodelta-Carb intercomparison of B(a)P models was initiated by the TFMM in 2021 in the framework of a broader scientific study on modelling of secondary organic aerosol and black carbon. The main objectives of the Eurodelta-Carb study on B(a)P were to analyze performance of air quality models and uncertainties of their results. Besides, the study was aimed to contribute to the refinement of B(a)P emissions from the combustion of fossil fuel and biomass burning and to further improve available B(a)P modelling approaches. Four regional chemistry transport models were applied to simulate the concentrations of B(a)P in Europe. The modelling results were compared with the observed B(a)P concentrations provided by the EMEP monitoring network. Evaluation of the modelled concentrations was performed in close cooperation with national experts in B(a)P modelling.

Model simulations setup and input data

The Eurodelta-Carb B(a)P modelling exercise is focused on the time period from the beginning of December 2017 to the end of 2018. Simulations of B(a)P were performed using four chemistry transport models: CHIMERE, GLEMOS, MINNI and SILAM. These models are being developed by the modelling teams of INERIS (France), CIEMAT (Spain), MSC-E (EMEP), ENEA (Italy), and FMI (Finland) to study air pollution levels on regional and national scales. All the models use prescribed modelling domain and gridded B(a)P annual emissions data for 2018, generated by CEIP. Other input data and parameterizations, such as meteorological input, intra-annual variations of B(a)P emissions, emissions of other pollutants, boundary conditions, model parameterizations, are specific to each model.

The participating models have different approaches to B(a)P modelling. In particular, CHIMERE, GLEMOS, and MINNI consider B(a)P 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 B(a)P with ozone in particulate form. All three models consider deposition of gaseous and particulate B(a)P from the atmosphere. In the case of SILAM, the model simulations were carried out assuming that B(a)P is an inert substance emitted to the atmosphere in the gaseous phase and subject only to degradation process depending on the temperature variation.

The program of model simulations includes a model run for the specified time period with the prescribed B(a)P emission data using independently defined model setups. Preliminary results were published in the previous EMEP status report [*Ilyin et al.*, 2022] and were presented at the HARMO21 conference [*Gusev et al.*, 2022].

In 2023 the B(a)P model intercomparison study was continued. In particular, modelling results of CHIMERE and SILAM were updated. In addition, an analysis of factors affecting differences between the models output on B(a)P was discussed and initiated (e.g. emission temporal profiles,

parametrizations of B(a)P degradation in particulate phase, dry and wet deposition, gas-particle partitioning). In this section updated modelling results and their analysis is described.

Modelling results and analysis

Spatial distributions of annual mean total B(a)P air concentrations, simulated by CHIMERE, GLEMOS MINNI and SILAM for 2018, are shown in Figure 1. The largest concentrations were estimated by all the 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 CHIMERE followed by GLEMOS, MINNI, and SILAM. The differences between the simulated B(a)P concentrations may be attributed to the effect of different model parameterizations applied (e.g. for gas-particle partitioning, degradation, and deposition processes) as well as the different meteorological inputs. Additional contributions could also have been made by different emission temporal profiles and concentrations of reactants used in the models to estimate B(a)P chemical transformations.

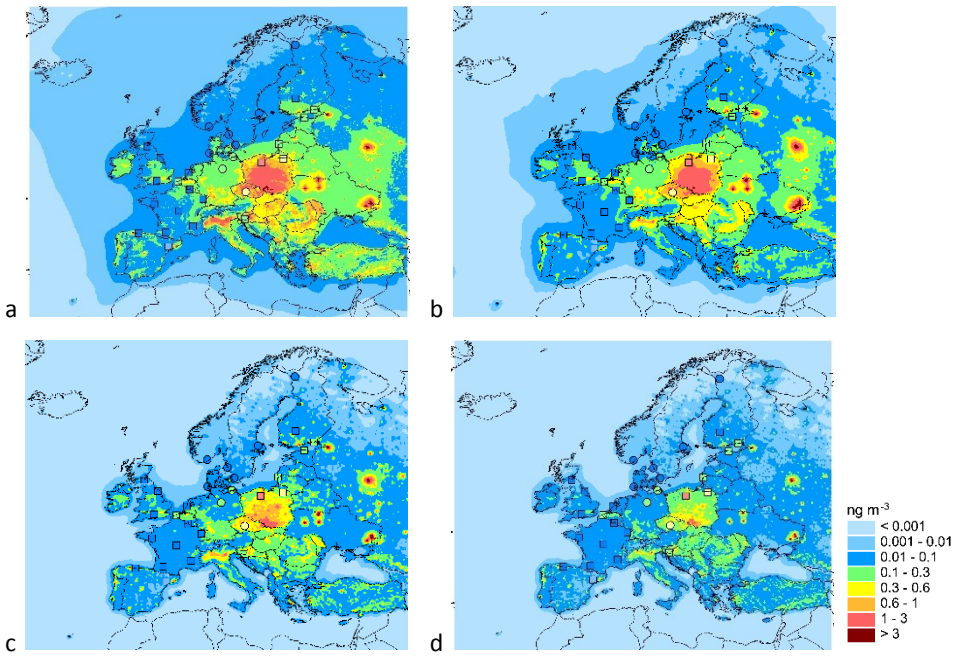

Fig. 1. *Maps of annual mean modelled total (gaseous + particulate phase) B(a)P atmospheric concentrations in 2018 simulated by CHIMERE (a), GLEMOS (b), and MINNI (c) and SILAM (d) for the base case model run. For the comparison, observed total and particulate phase B(a)P concentrations, reported by the EMEP monitoring stations, are shown as colored circles and squares, respectively, on the same scale as the modelled values.*

Evaluation of the model output against measurements for the 2018 was carried out using the data of 29 EMEP monitoring stations. Of these, 9 stations located in Central and Northern Europe measured total B(a)P concentrations, whilst B(a)P concentrations in particulate phase were measured at 20 stations covering a wider geographical area (Fig. 1).

For the model-measurement comparison, 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 B(a)P concentrations is presented in Table 1. All the models reproduced the spatial pattern of observed total and particulate B(a)P concentrations well with correlation coefficients (R) of 0.72-0.96. CHIMERE and GLEMOS tended to slightly overestimate observed total B(a)P levels with a mean bias of about 4%, whereas MINNI underestimated the measured values with a mean bias of -53%. For the particulate B(a)P concentrations, CHIMERE overestimated concentrations with a mean bias about 0.3%, while GLEMOS and MINNI underestimate the observed concentrations with mean biases of -19% and -52%, respectively. Estimated total B(a)P concentrations were within a factor of 2 of the measured values for 89%, 78%, 11%, and 0% of monitoring stations for CHIMERE, GLEMOS, MINNI, and SILAM respectively, whereas for B(a)P in particulate phase they were within a factor of 2 for 80%, 70%, 40%, and 20% of monitoring stations. The fraction of model values that were within a factor of 3 from measurements is larger. In particular, for 100%, 100%, 22%, and 11% of stations, measured total B(a)P concentrations and for 90%, 85%, 70%, and 35% of stations, measured particulate B(a)P concentrations, respectively.

Table 1. *Summary of statistical metrics, calculated on the basis of annual mean total and particulate phase B(a)P 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 B(a)P concentrations (9 stations), mean observed 0.116 ng m^{-3}						
CHIMERE	0.120	3.8	0.93	0.057	89	100
GLEMOS	0.121	4.3	0.91	0.087	78	100
MINNI	0.054	-53.3	0.86	0.090	11	22
SILAM	0.026	-77.5	0.86	0.124	0	11
Particulate B(a)P concentrations (20 stations), mean observed 0.156 ng m^{-3}						
CHIMERE	0.157	0.3	0.88	0.116	80	90
GLEMOS	0.126	-19.3	0.96	0.095	70	85
MINNI	0.075	-52.1	0.93	0.168	40	70
SILAM	0.047	-69.9	0.72	0.226	20	35

[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 B(a)P concentrations against the measurements of total and particulate B(a)P concentrations from individual EMEP monitoring stations is shown in the scatter plots in Fig. 2. An overestimation of observed particulate B(a)P concentrations was found for two Spanish stations ES8 and ES14 for all the models. For other stations, different kinds of discrepancies were obtained. In particular, for CHIMERE, an overestimation about a factor of 2 was found for the stations GB48, GB1055, and NL91 that measured particulate B(a)P. In the case of GLEMOS, the largest underestimation (more than a factor of 3) was found for total B(a)P observed at

DE9 and FI36, and for particulate B(a)P at FR23 and FR25. In case of MINNI, the greatest deviations (underestimation by more than a factor of 5) were found for the stations DE1, DE9, and FI36 that measured total B(a)P, and for LV10 and FR23 that measured particulate B(a)P. In case of SILAM, for most of the stations measured B(a)P concentrations were underestimated by the model.

The scatter plots indicate that CHIMERE modelling results have the highest regression slope value (0.81) followed by GLEMOS (0.77), MINNI (0.44), and SILAM (0.28). The scattering of modelled-observed pairs is best for GLEMOS (coefficient of determination 0.88) compared with MINNI, CHIMERE, and SILAM (0.83, 0.80, and 0.67, respectively).

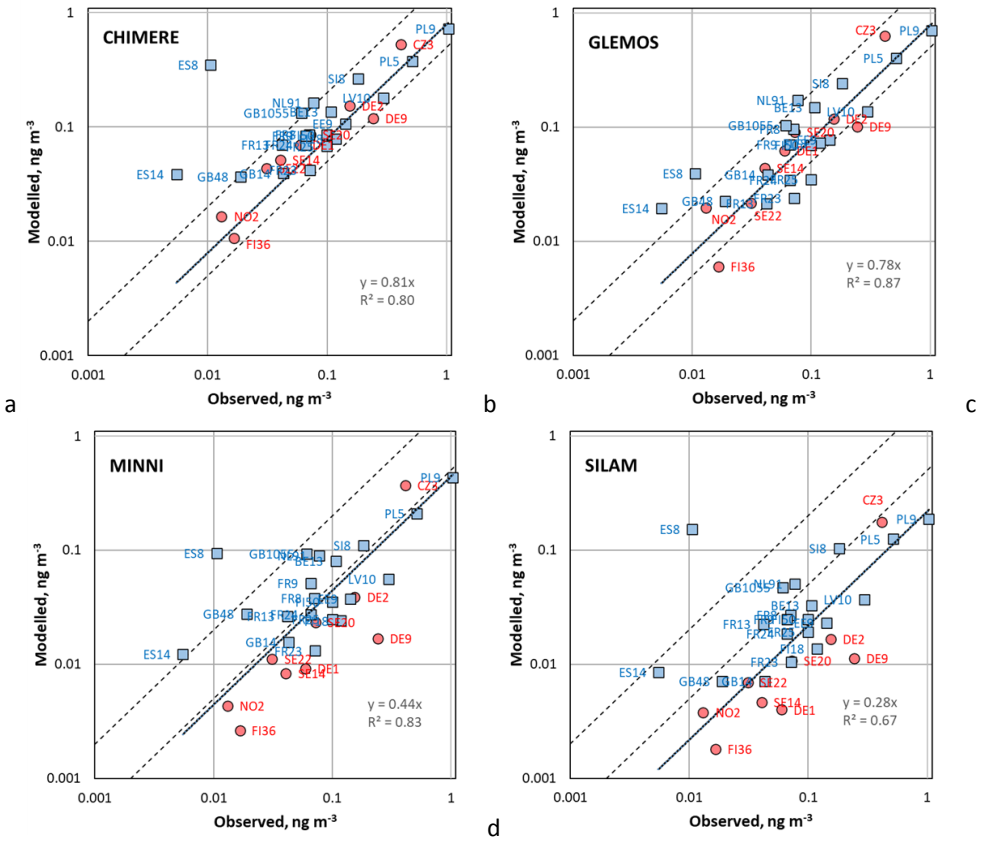

Fig. 2. *Scatter plots on a log-log scale of the comparison of modelled B(a)P air concentrations (total and particulate) simulated by CHIMERE (a), GLEMOS (b), MINNI (c) and SILAM (d) with measurements of EMEP monitoring stations in 2018. The region between the dashed lines indicates the model estimates within a factor of two of the measured values and the solid line is the linear regression of all data points. Total B(a)P concentrations are shown as red circles and particulate B(a)P concentrations are shown as blue squares.*

Figure 3 shows examples of modelled and observed B(a)P time series for two EMEP monitoring stations, namely, CZ0003R and PL0009R that measured total and particulate B(a)P concentrations, respectively. For these stations, the comparison of modelled and observed concentrations for CHIMERE, GLEMOS, and MINNI shows, in general, a good level of agreement. Model estimates capture 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. In case of SILAM, modelled

concentrations show noticeable underestimation of measured B(a)P in cold season compared to other three models. Differences between the modelled and measured intra-annual variations of B(a)P concentrations may be explained both by an underestimation of emissions and by the uncertainties in the temporal disaggregation of B(a)P emissions that were applied in the model simulations.

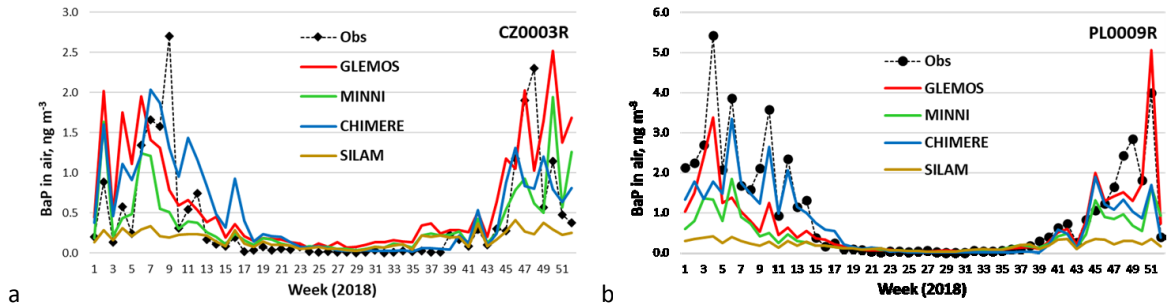

Fig. 3. *Intra-annual variations of total B(a)P air concentrations, observed at the EMEP station CZ0003R (a), and particulate B(a)P concentrations, observed at the station PL0009R (b), and the total and particulate B(a)P concentrations simulated by CHIMERE, GLEMOS, MINNI, and SILAM for 2018.*

In Figure 4 the the model estimates of B(a)P particulate to total ratio are shown for CHIMERE, GLEMOS, and MINNI. The highest ratio of particulate to total (gaseous + particulate) B(a)P concentration for the locations of 29 EMEP stations is obtained by MINNI (about 0.95) followed by CHIMERE (about 0.88) and GLEMOS (about 0.75) (Fig. 4a). In case of B(a)P concentrations in the whole modelling domain (Fig. 4b), MINNI similarly provided the highest ratio of B(a)P in particulate phase (about 0.9). At the same time, CHIMERE and GLEMOS showed more comparable results and slightly lower mean ratio of B(a)P in particulate phase of about 0.55. Differences in modelled particulate and total B(a)P concentrations can be explained by different parameterizations of gas-particle partitioning and degradation processes applied in the models.

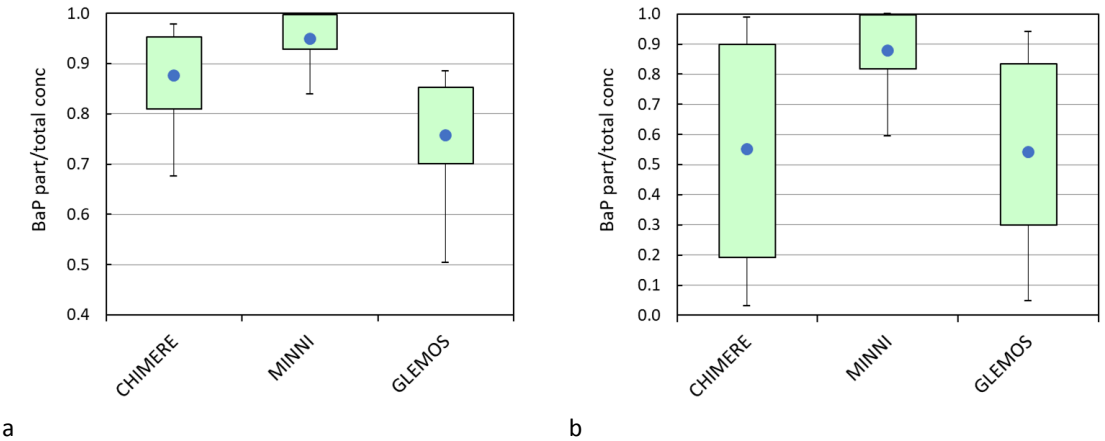

Fig. 4. *Average ratio of annual mean particulate to total B(a)P concentrations and its variability estimated by CHIMERE, GLEMOS, and MINNI for 2018 for the locations of 29 EMEP stations (a) and for the whole modelling domain (b). On the diagrams dots represent median ratio, colored boxes – 25% and 75% percentiles, and whiskers - 5% and 95% percentiles.*

Participated models have shown high spatial correlation of predicted and observed B(a)P concentrations. Besides, most of the models provided high correlation with observed intra-annual variation of B(a)P concentrations. Furthermore, the model simulations indicated overprediction of observed B(a)P concentrations in Spain and underprediction in Northern Europe (Finland, Latvia, Estonia), which is likely explained by the uncertainties of the reported B(a)P emissions.

Significant difference between the modelling results of four participated models is noted. Thus, more detailed analysis is required to explore the reasons of the differences and substantial over- and underestimates of observed B(a)P concentrations for some of the stations. Further activities within the study can be focused on the sensitivity analyses, an evaluation of the meteorological drivers and an analysis of other model outputs such as B(a)P concentrations in precipitation and deposition fluxes and concentrations of species affecting B(a)P chemical transformations in the atmosphere.