Performance of Operational Chemical Transport Models for Particulate Matter Concentrations in Bulgaria

Hristina Kirova, Nadya Neykova, and Emilia Georgieva

Abstract The main objective of this study is to evaluate the performance of some well-known and widely used operational air quality modelling systems (EMEP-MSC-W, and the models at the Copernicus Atmosphere Monitoring Service (CAMS)) for simulations of ground-level particulate matter in Bulgaria. The analysis is focused on two months—a summer one (August 2017) and a winter one (February 2019). The comparison of models to observations from regular air quality background stations is based on statistical indicators and various plots (box plots, kernel density estimations, and scatter plots). The EMEP and CAMS regional models underestimate the observed concentrations, on average by about 50% for PM_{10} and by about 22% for $PM_{2.5}$. These models perform better at a rural remote (mountain) site than at urban background stations indicating that the outputs of the models could be used for indicative values of PM background concentrations. The model inter-comparison consists of an analysis of the spatial distribution of monthly mean concentrations and values for domain averaged model concentrations. The CAMS global model simulates in summer different spatial distribution due to the assimilation of satellite data providing information for dust storms and wildfires.

Keywords Chemical transport models · Particulate matter · Model validation

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1 Introduction

Particulate matter (PM) are constantly studied because of their effects on the human health and on the environment. While monitoring facilities for surface level concentrations are growing in number and type, the information from observational data is still limited in time and space. Chemical transport models (CTM) have been recognized as valuable tools not only for air quality assessment, but also for policy support in determining abatement measures [1, 2]. The PM modelling, however, still represents a challenging task, as their concentrations are influenced not only by different emission sources, but also by atmospheric processes and chemical transformation mechanisms taking place over various spatial scales. Some international initiatives for evaluation of CTMs in the last years (e.g. AQMEII (Air Quality Modelling Evaluation International Initiatives [3]), FAIRMODE (Forum for AIR quality MODeling [4]) have allowed better understanding of the weaknesses and strengths of the models and have contributed to their improvement and further development. Nowadays, CTMs are the backbone of many comprehensive air quality forecasting systems designed for different scales—from country, to European and global ones [5–7].

At the National Institute of Meteorology and Hydrology (NIMH) a chemical weather forecasting system has also been set up, BgCWFS [8, 9]. The system was evaluated on European scale in the framework of AQMEII [10–12]. On national scale, BgCWFS results with 9 km spatial resolution showed underestimation for PM_{10} [13]. On the other hand, exceedances of PM limit values are often observed at many sites in Bulgaria [14, 15], and there is a public and expert interest in the possibilities of the operational modelling systems to predict surface PM concentrations.

For this study we have chosen to look at freely available results (model outputs) of three well-known and widely used operational air quality modelling systems for a case study in Bulgaria. The systems differ in their input data, emissions handling, parameterisation schemes, chemical mechanisms etc., but it is believed that they capture the main characteristics of the surface PM distribution. The available online results that we use in this study are from the following systems.

EMEP MSC-W (denoted further as EMEP)—the model of the Meteorological Synthesizing Centre-West (MSC-W) of the European Monitoring and Evaluation Programme) [16]. The EMEP model is one of the major instruments which is applied for decision and policy making not only because of its coverage but also because its output includes photooxidants, inorganic and organic aerosols and depositions. This is the model used by the European Environmental Agency for annual reporting on the air quality status in Europe. The model domain covers an extended European region with a horizontal resolution of $0.1 \times 0.1°$ and 34 vertical layers (the lowest with a height of approximately 50 m). The meteorological driver of EMEP is the Integrated Forecasting System of the European Centre for Medium-Range Weather Forecasts (IFS-ECMWF). The emissions of EMEP are based on country reporting. Estimates of the anthropogenic emissions for each country should be provided every year as well as spatial distribution to the EMEP grid. Different chemical mechanisms are applied in the model: for inorganic aerosols—equilibrium module is used to the partitioning between gas and fine-mode aerosol phase in the system of $SO_4^{2+} - HNO_3 - NO_3^- - NH_3 - NH_4^+$ [17]; and for secondary organic aerosols EmChem09soa is used [16]. The gas-phase mechanism ("EMEP scheme") comprises of 70 species and 140 reactions [16]. Conditions from global C-IFS are used for the mineral dust and sea salt. Results from version rv4_33 are used.

The second system is the regional (European) ensemble air quality forecasting system at the Copernicus Atmosphere Monitoring Service (CAMS) (denoted further as CAMS-ENS) [18]. Its main characteristics are: coverage—Europe, spatial and temporal resolution: 0.1° and 1 h, vertical levels up to 5000 m. The Ensemble forecast is the median of the forecasts from 9 different state-of-the-art atmospheric modelling systems (CHIMERE, EMEP, EURAD-IM, DEHM, GEM-AQ, LOTOS-EUROS, MATCH, MOCAGE, SILAM). All CTM's use the same CAMS emissions (regional anthropogenic emissions and for biomass burning), the same meteorological driver (ECWMF weather global operational system) and the same boundary conditions. The regional CAMS-ENS system combines model data and in-situ observations to provide air quality forecasts. The chemical mechanisms in the ensemble members depend on the individual model.

The third system is the global CAMS (CAMS-ECMWF) [19]. It provides operational forecast for atmospheric chemistry parameters globally with horizontal resolution of 40 km on 60 vertical levels going up to 0.1 hPa, the temporal resolution is 3 h. CAMS-ECMWF assimilates data from satellite observations. Thus, emissions by dust storms and wildfires are taken into account. Emissions and fluxes in the global CAMS are: anthropogenic emissions from different inventories, biomass burning emissions from Global Fire Assimilation System (GFAS), wind-blown desert dust and sea salt emissions modelled by the IFS depending on the meteorological forecasts, CO_2 fluxes from vegetation modelled by the C-TESSEL surface scheme, natural and biogenic fluxes provided by various climatological data sets. Chemical scheme applied in the model is carbon bond (CB05). For aerosols of natural origin a bin representation is used.

The period for the case study includes two different months—August 2017 and February 2019. August is a typical summer month, characterized by high temperatures and low precipitations. The weather in August 2017 is determined by prevailing anticyclonic pressure systems. The exceptions were 5 days with weak pressure gradient field and 10 days with cyclonic type of atmospheric circulation [20]. Widespread thunderstorms were registered on 4 days. The number of days at NIMH stations with rainfall more than 1 mm was between 1 and 4, and with rainfall above 10 mm was between 0 and 3. High temperatures and low amount of precipitation are favorable conditions for wildfires which occurred in southern Bulgaria at the beginning and in the second part of the month. Saharan dust intrusions towards the country, detected by data from GOME2 instrument on MetOP satellites, were identified in about 14 days throughout the month [21]. As during summer the domestic heating—one of the major emission sources for PM—is missing, the selected month is assumed to be influenced by natural aerosols, which CAMS modelling systems can treat through the data assimilation.

February is usually one of the coldest months in Bulgaria with elevated concentrations of PM over the whole country, in particular when long lasting episodes of anticyclonic weather occur. In 2019, February was characterized by mild temperatures with +0.6° to +3.6 °C deviation from the monthly norm. The precipitation at the NIMH stations was below the climatic norm—by less than 20% up to 83% [20]. The synoptic situation through the month was dynamic, with frequent passages of Mediterranean cyclones south of the country, atmospheric fronts from west, northwestern direction and short periods of anti-cyclonic conditions. The mean cloudiness in February is usually high. In 2019 it was between 4 and 8 of 10th—scale, and the number of clear days was up to 10 which is above the climatic norm. The particulate matter concentrations during this month are usually high, mainly due to domestic heating—emission source that is, generally, poorly represented in CTM systems.

The main goal of this work is to evaluate the performance of EMEP, CAMS-ENS, and CAMS-ECMWF for PM_{10} and $PM_{2.5}$ surface concentrations in Bulgaria during the selected periods, comparing model results to observations and performing model intercomparison.

2 Methods

To check the performance of the models, two main aspects were considered—comparisons of modelled to observed PM daily concentrations, and model-to-model comparison (model intercomparison).

2.1 Model to Observation Analysis

The modelled mean daily PM values were compared to the mean daily concentrations observed at background type of air quality stations in Bulgaria. Only stations with data availability more than 75% were considered. Thus, the number of stations for August 2017 is 24 for PM_{10}, and 8 for $PM_{2.5}$. For February 2019 the number of stations is lower—17 stations for PM_{10} and 4 for $PM_{2.5.}$ The stations are part of the air quality monitoring network managed by the Bulgarian Executive Environment Agency (ExEA), Fig. 1.

The stations are located mainly in urban areas, only two stations are of rural type and they are in mountainous regions—BG0070A (Kopitoto, 1321 m a.s.l) and BG0053R (Rozhen, 1720 m a.s.l.). The measurement methods used for PM_{10} and $PM_{2.5}$ in these stations are beta absorption, beta ray attenuation or gravimetry.

The performance of the EMEP and CAMS-ENS models was evaluated through box plots (which display the minimum, first quartile, median, third quartile, and maximum of the data set), kernel density estimations (kde) of the probability density functions (pdf), scatter plots, and statistical indicators. Here we show and comment this comparison for four selected stations located in different environment: 2 urban

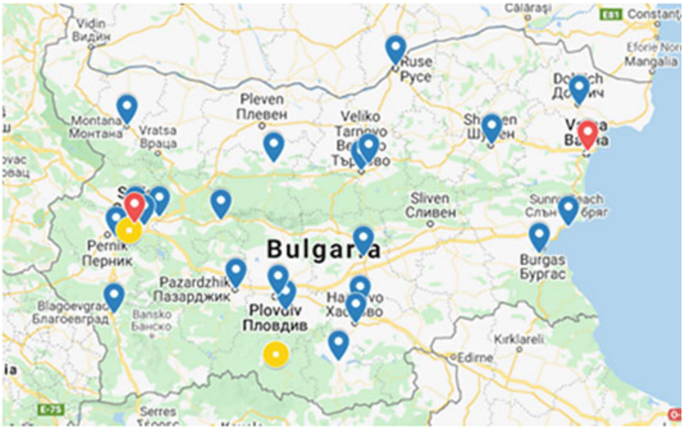

Fig. 1 Map of the stations with available PM_{10} and $PM_{2.5}$ data. Color symbols indicate the selected stations for box and kde plots, red—urban stations, orange—mountain stations

stations—one is in the western part of the country in the city of Sofia, BG0050A (Sofia Hipodruma, 581 m a.s.l.), the second one is at the Black Sea coast in the city of Varna, BG0075A (Varna SOU Angel Kanchev, 83 m a.s.l.). The other 2 stations were the rural stations mentioned above, located in the western and southern part of the country, Fig. 1.

The analysis for all stations was based on scatterplots of the daily concentrations and statistical indicators—the mean observed and modelled concentrations, the mean bias error between model and observation (MBE), the root mean square error (RMSE), the correlation coefficient (Corr), the fractional gross error (FGE), and the normalized mean bias (NMB).

2.2 Model-to-Model Comparison

For this analysis we used qualitative comparison—maps for mean monthly concentrations of particulate matters (PM_{10} and $PM_{2.5}$) were elaborated. As a quantitative measure the values of the domain mean concentrations, as simulated by the different systems for the respective months, were calculated.

3 Results

3.1 August 2017

The comparison of modelled data from EMEP and CAMS-ENS to observed PM_{10} concentrations is visualized in Fig. 2 for the selected three stations (the 2 urban ones, and 1 mountain—Rozhen).

The graphs presenting the kernel density estimations show that PM_{10} concentrations estimated by both models have similar distributions. The box plots indicate that both models have smaller values of the means and the dispersions than the observed data, but the CAMS-ENS model has smaller dispersion than the EMEP model. For the rural remote station Rozhen the models have better resemblance to the observed data than for the urban stations, as expected because this station is not influenced by local emissions typical for the cities. Both models underestimate the observed PM_{10} concentrations. This can also be seen from the statistical indicators of modelled versus observed PM_{10} averaged over all 24 stations presented in Table 1, and on the scatter plots for all 24 stations presented in Fig. 3. The statistical indicators show very similar results for the two models with underestimation on average by about 50%.

The scatter plots indicate that the EMEP model has more often overestimation of some daily values than CAMS-ENS. Overall we can conclude that the EMEP model

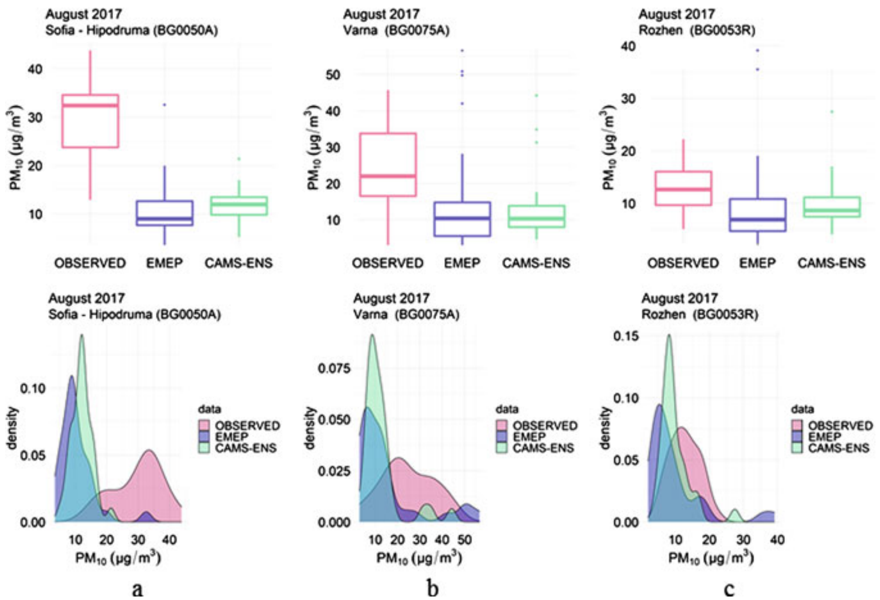

Fig. 2 Observed versus modelled PM_{10} in August 2017: **a** Sofia-Hipodruma, **b** Varna, **c** Rozhen: top—box plots; bottom—kernel density estimations

Table 1 Statistical indicators of modelled versus observed PM_{10} concentrations (μgm^{-3}) for August 2017

PM_{10} $N = 24$	$Mean^{OBS}$	$Mean^{MOD}$	MBE	RMSE	Corr	FGE	NMB (%)
EMEP	25.68	12.90	−12.78	15.82	0.47	0.79	−49.77
CAMS-ENS	25.68	10.99	−14.69	16.11	0.56	0.80	−57.22

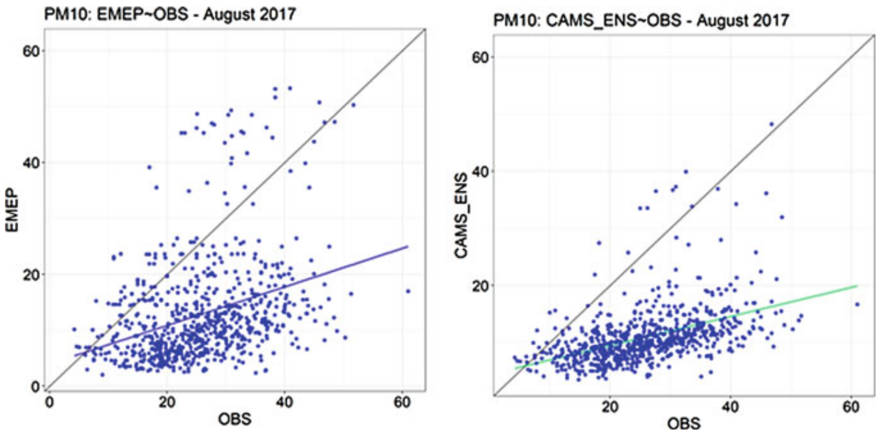

Fig. 3 Scatter plots for the daily PM_{10} at 24 stations in Bulgaria in August 2017: EMEP (left) and CAMS-ENS (right)

underestimates the observed PM_{10} concentrations to a lesser extent than CAMS-ENS model, but the correlation is better for CAMS-ENS.

The distributions of the modelled and observed $PM_{2.5}$ concentrations are presented in the box plots and kernel density estimation plots in Fig. 4. Compared to the results for PM_{10}, the difference between modelled and observed $PM_{2.5}$ concentrations is smaller. The modelled $PM_{2.5}$ concentrations by both models are similarly distributed. Both models have lower values of the means and the dispersions than the observed data, but CAMS-ENS model has smaller dispersion than the EMEP model. As with PM_{10} the performance is better for the rural remote station Rozhen.

Both models underestimate the observed $PM_{2.5}$ concentrations, as indicated by the statistical indicators in Table 2 and the scatter plots in Fig. 5. The underestimation, on average by about 32%, is less than the underestimation for PM_{10}. To note that the number of stations for $PM_{2.5}$ is only 8.

The model to model comparison was based on results for Bulgaria obtained by the three model systems—EMEP, CAMS-ENS and CAMS-ECMWF for August 2017. The spatial distribution of the mean monthly concentrations of PM_{10} and $PM_{2.5}$ is shown in Fig. 6, the averaged for the domain monthly values are provided in Table 3. All models simulate higher concentrations in the eastern part of the domain (Black Sea). EMEP results over land indicate some hot spots in correspondence with big

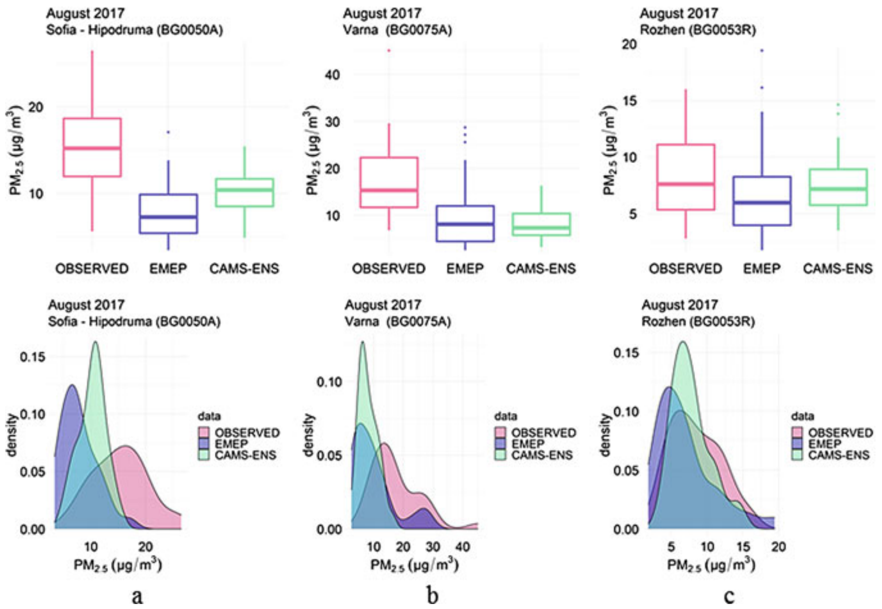

Fig. 4 Observed versus modelled $PM_{2.5}$ in August 2017: **a** Sofia-Hipodruma, **b** Varna, **c** Rozhen: top—box plots; bottom—kernel density estimations

Table 2 Statistical indicators of modelled versus observed $PM_{2.5}$ concentrations (μgm^{-3}) for August 2017

$PM_{2.5}$ ($N = 8$)	$Mean^{OBS}$	$Mean^{MOD}$	MBE	RMSE	Corr	FGE	NMB (%)
EMEP	13.55	9.34	−4.21	7.26	0.48	0.58	−31.09
CAMS-ENS	13.55	8.82	−4.73	5.93	0.66	0.45	−34.90

cities. Higher concentrations in the Lower Danube plain (north-western part of the domain) are simulated by CAMS-ECMWF. This might be a consequence of emission sources that are not included in the other models, but this system is producing due to satellite data assimilation.

The mean monthly map for the Aerosol Optical Depth, as retrieved by MODIS Terra satellite, Fig. 7, also indicates higher aerosol loading north of the country. The character of the spatial distribution by the models is maintained in the maps for $PM_{2.5}$.

The domain mean surface concentrations for PM_{10} and $PM_{2.5}$ are with 9 and 12.5% higher for CAMS-ECMWF compared to EMEP (Table 3). The lowest values are simulated by CAMS-ENS: with 14 and 15.6% lower than the values by CAMS-ECMWF.

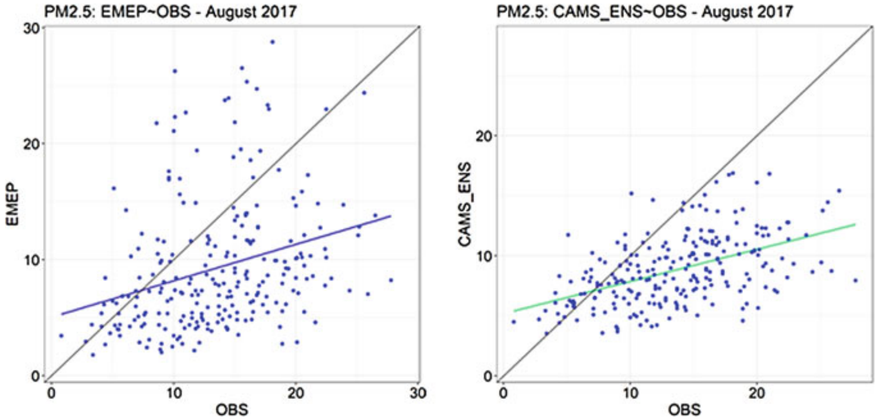

Fig. 5 Scatter plots for the daily $PM_{2.5}$ at 8 stations in Bulgaria in August 2017: EMEP (left) and CAMS-ENS (right)

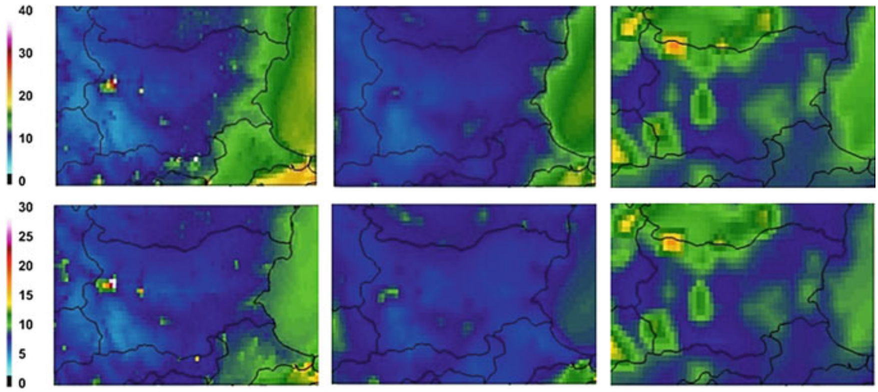

Fig. 6 Monthly mean PM_{10} (top) and $PM_{2.5}$ (bottom) concentrations (μgm^{-3}) for domain Bulgaria (August 2017): EMEP (left), CAMS-ENS (middle) and CAMS-ECMWF (right)

Table 3 Domain mean surface PM concentrations (μgm^{-3}) for August 2017

	EMEP	CAMS-ENS	CAMS-ECMWF
PM_{10}	12	11.3	13.2
$PM_{2.5}$	8.4	8.1	9.6

3.2 February 2019

For the second testing period the performance of the models was carried out following the methodology outlined in Sect. 2. The observational data for the box plots and

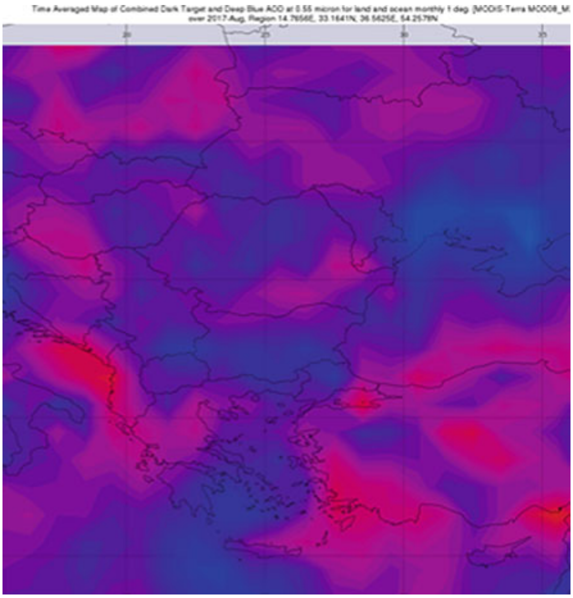

Fig. 7 Mean monthly (August, 2017) map of combined dark target and deep blue AOD at 550 nm for land and ocean MODIS Terra satellite, map created by NASA's Giovanni online system [22]

the kernel density estimations were provided for the same urban stations (Sofia–Hipodruma and Varna SOU Angel Kanchev), while for rural station data from Sofia Kopitoto were used for $PM_{2.5}$ (as not sufficient data were available at Rozhen), and for PM_{10} the data were from Rozhen. Scatter plots and statistical indicators are presented for all stations—17 for PM_{10} and 4 for $PM_{2.5}$.

The comparison of model data from EMEP and CAMS-ENS to observed PM_{10} concentrations is visualized in Fig. 8 for the selected three stations. The graphs presenting the kernel density estimations show that PM_{10} concentrations estimated by both models have similar distributions. The box plots indicate that both models have lower values of the means and the dispersions compared to the observed data, but CAMS-ENS model has smaller dispersion than the EMEP model. For the rural remote station Rozhen the models have slightly better resemblance to the observed data than for the urban stations, similar to what was observed for the summer month.

Both models underestimate the observed PM_{10} concentrations. This conclusion is suggested also by the statistical indicators of modelled versus observed daily PM_{10} concentrations averaged over all 17 stations presented in Table 4 and on the scatter plots, Fig. 9. The statistical indicators show slightly better results for the EMEP model. The underestimation is on average about 45% for EMEP and 53% for CAMS-ENS. Overall we can conclude that EMEP model underestimates the observed PM_{10} concentrations to a lesser extent than CAMS-ENS model.

The available data for $PM_{2.5}$ in Bulgaria in February 2019 are limited to 4 stations. The distributions of the modelled and observed $PM_{2.5}$ concentrations are presented in the box plots and kernel density estimation plots, Fig. 10.

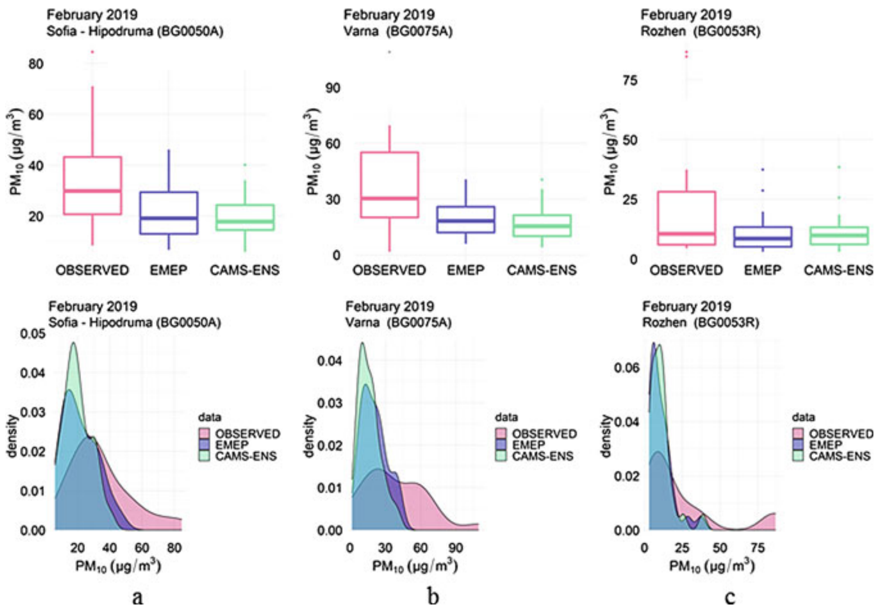

Fig. 8 Observed versus modelled PM_{10} in February 2019: **a** Sofia-Hipodruma, **b** Varna, **c** Rozhen: top—box plots; bottom—kernel density estimations

Table 4 Statistical indicators of modelled versus observed PM_{10} concentrations (μgm^{-3}) for February 2019

PM_{10} $N = 17$	$Mean^{OBS}$	$Mean^{MOD}$	MBE	RMSE	Corr	FGE	NMB (%)
EMEP	35.30	19.52	−15.77	24.00	0.60	0.63	−44.69
CAMS-ENS	35.30	16.45	−18.85	26.13	0.54	0.73	−53.39

Compared to the results for PM_{10}, the difference between modelled and observed $PM_{2.5}$ concentrations is smaller at the urban sites in Sofia and Varna. Both models have similar mean values but the EMEP model has wider spread for the 2 stations. At the mountain station Kopitoto, located near the city of Sofia, both models overestimate the observed monthly mean $PM_{2.5}$ by a factor more than two. This behaviour does not correspond to model data discussed so far for the mountain stations. The low values of the mean observed $PM_{2.5}$ and their spread at Kopitoto station indicate possible measurements error and suggest to check further the accuracy of the monitored data for this period.

The statistical indicators of modelled versus observed PM_{10} concentrations averaged over all 4 stations are presented in Table 5, and the scatter plots in Fig. 11. The averaged MBE and FGE for EMEP are lower than those of CAMS-ENS, the

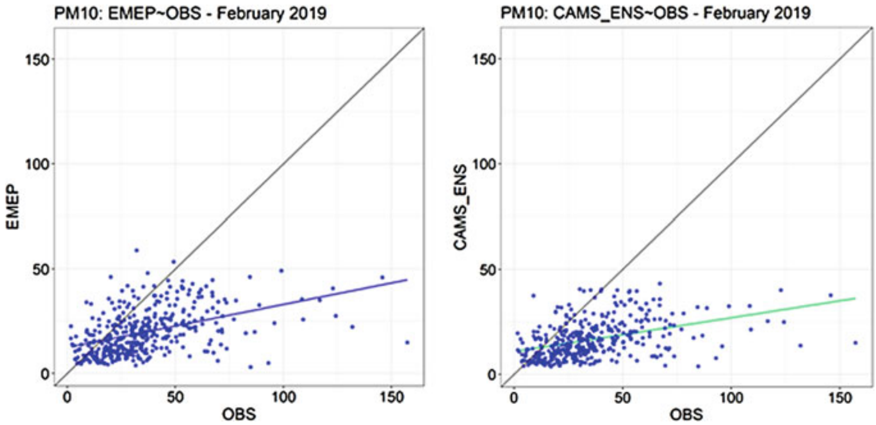

Fig. 9 Scatter plots for the daily PM_{10} at 17 stations in Bulgaria in February 2019: EMEP (left) and CAMS-ENS (right)

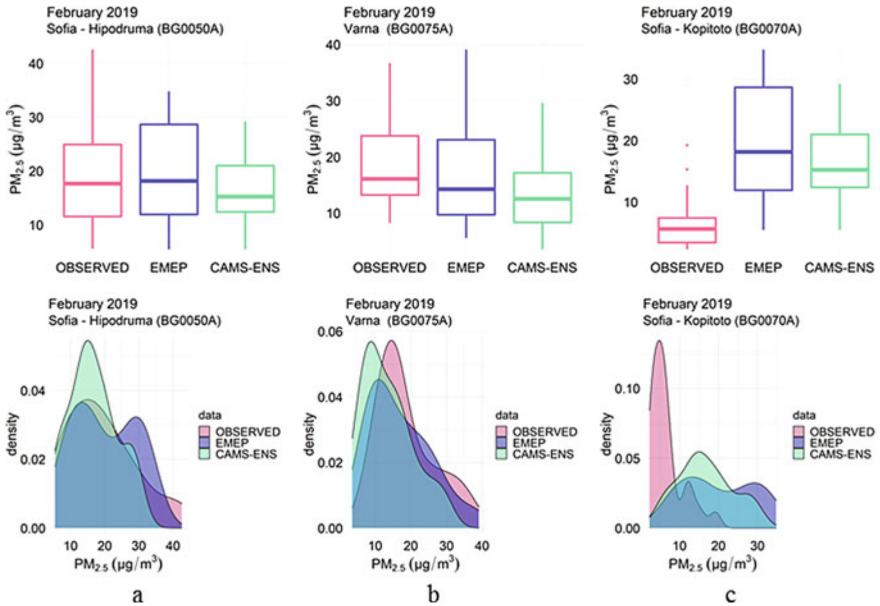

Fig. 10 Observed versus modelled $PM_{2.5}$ in February 2019: **a** Sofia-Hipodruma, **b** Varna, **c** Sofia-Kopitoto; top—box plots; bottom—kernel density estimations

correlation of EMEP is higher than that of CAMS-ENS model. The statistical indicators have to be interpreted with caution due to the very limited number of stations providing data.

Table 5 Statistical indicators of modelled versus observed $PM_{2.5}$ concentrations (μgm^{-3}) for February 2019

$PM_{2.5}$ ($N = 4$)	$Mean^{OBS}$	$Mean^{MOD}$	MBE	RMSE	Corr	FGE	NMB (%)
EMEP	18.55	19.65	1.11	13.46	0.48	0.56	5.96
CAMS-ENS	18.55	15.71	−2.84	13.20	0.66	0.58	−15.29

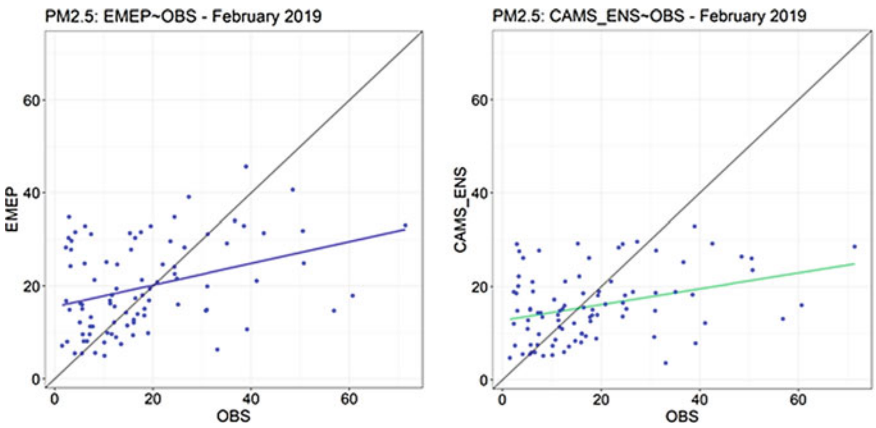

Fig. 11 Scatter plots for the daily $PM_{2.5}$ at 4 stations in February 2019: EMEP (left) and CAMS-ENS (right)

Maps of mean monthly concentrations of PM_{10} and $PM_{2.5}$ for February 2019 are presented in Fig. 12, the averaged for the domain monthly values are given in Table 6. The highest PM_{10} concentrations are simulated in the northern part of the domain

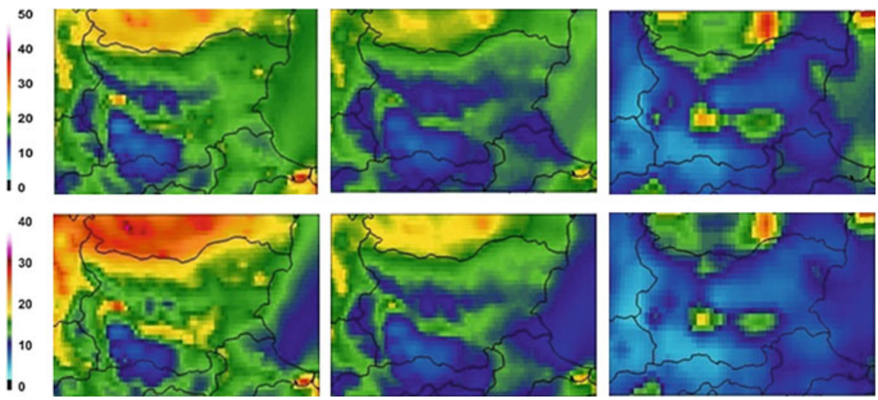

Fig. 12 Monthly mean PM_{10} (top) and $PM_{2.5}$ (bottom) concentrations (μgm^{-3}) for domain Bulgaria (February 2019): EMEP (left), CAMS-ENS (middle) and CAMS-ECMWF (right)

Table 6 Domain mean surface concentrations (μgm^{-3}) for February 2019

	EMEP	CAMS-ENS	CAMS-ECMWF
PM10	19.1	17.3	16.8
PM2.5	16.8	13.3	13.5

(Lower Danube Plain). All models indicate elevated concentrations near big cities in Romania. In Bulgaria the highest concentrations are in the region of Sofia and in the lowland and near the Black Sea. The presence of thermal inversions in winter in these regions can explain this behavior of the models. Similar features were identified in the $PM_{2.5}$ spatial distribution, except for the Black Sea area. The highest domain mean PMs concentrations are simulated by EMEP—for PM_{10} by 14 and 10.4% higher than the mean values by CAMS-ENS and CAMS-ECMWF, respectively. For $PM_{2.5}$ the domain mean EMEP concentrations by 24 and 26% higher.

Summarizing the performance of the models EMEP and CAMS-ENS (that have similar horizontal resolution) we noticed similar behavior for the two months. Both models underestimate PMs observed at background stations in Bulgaria, but the underestimation of the EMEP model is less than that of the CAMS-ENS model. At the same time the correlation is better for the second model. On average for the two months the underestimation for PM_{10} is 47% by the EMEP model and 55% by the CAMS-ENS model. For $PM_{2.5}$ the underestimation is lower–less than 20% by the EMEP model and about 25% by the CAMS-ENS model. These figures for $PM_{2.5}$ should be interpreted, however, with caution, as the number of stations with sufficient data was limited (4–8). At the mountain stations the underestimation is less that at the urban sites, for example for PM_{10} at the remote station Rozhen it is about 22% for the summer month. We noticed also overestimation in the winter month at the mountain site near Sofia (Kopitoto). One possible explanation is the models grid scale (about 10 km) which corresponds to the distance between the central part of Sofia and this mountain station. Thus, the models have difficulties to distinguish between urban and mountain part, especially in winter, when the high mountain areas remain well above the polluted urban boundary layer. The PMs simulated by the global model CAMS-ECMWF are much lower than the previous models, as expected due to the coarser grid resolution (40 km). However, this model system shows different spatial distribution in the summer month, most probably due to the assimilation of satellite data for aerosol optical depth.

4 Conclusion

In this study EMEP and CAMS-ENS simulated concentrations for PM_{10} and $PM_{2.5}$ were compared to data from regular air quality stations in Bulgaria for a test period of two months—a summer month (August 2017) and a winter month (February 2019). Both models underestimate the observed concentrations, on average by about 50% for PM_{10} and by about 22% for $PM_{2.5}$. The models perform better at the rural remote (mountain) site Rozhen than for the urban background stations indicating that these model outputs could be used for indicative values of background PMs concentrations. EMEP model underestimated the observed PM_{10} and $PM_{2.5}$ values to a lesser extent than CAMS-ENS. The PM_{10} spatial distribution indicated higher values in the eastern part of domain Bulgaria and hot spots over main cities for the summer month. For the winter month higher values were simulated for the lowlands in Bulgaria and north of the country (Lower Danube plain). Interestingly, the CAMS-ECMWF model that has coarser grid resolution, indicated also in summer higher PM concentrations north of the country. This might be due to sources not accounted for in the other models, e.g. fires or mineral dust. As this system assimilates satellite data, it could forecast influence of such events on surface PM concentrations. Further analysis are ongoing in this direction.

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