Atmos. Chem. Phys., 13, 9661–9673, 2013
www.atmos-chem-phys.net/13/9661/2013/
doi:10.5194/acp-13-9661-2013

Atmospheric Chemistry and Physics Open Access

Air quality over Europe: modelling gaseous and particulate pollutants

E. Tagaris, R. E. P. Sotiropoulou, N. Gounaris, S. Andronopoulos, and D. Vlachogiannis

Environmental Research Laboratory, NCSR Demokritos, 15310 Athens, Greece

Correspondence to: E. Tagaris (tagaris@ipta.demokritos.gr) and R. E. P. Sotiropoulou (rsot@ipta.demokritos.gr)

Received: 17 December 2012 – Published in Atmos. Chem. Phys. Discuss.: 13 March 2013
Revised: 8 August 2013 – Accepted: 18 August 2013 – Published: 30 September 2013

Abstract. Air quality over Europe using Models-3 (i.e., CMAQ, MM5, SMOKE) modelling system is performed for winter (i.e., January 2006) and summer (i.e., July 2006) months with the 2006 TNO gridded anthropogenic emissions database. Higher ozone mixing ratios are predicted in southern Europe while higher NO_2 levels are simulated over western Europe. Elevated SO_2 values are simulated over eastern Europe and higher $PM_{2.5}$ concentrations over eastern and western Europe. Regional average results suggest that NO_2 and $PM_{2.5}$ are underpredicted, SO_2 is overpredicted, while $Max8hrO_3$ is overpredicted for low mixing ratios and is underpredicted for the higher mixing ratios. However, in a number of countries observed and predicted values are in good agreement for the pollutants examined here. Speciated $PM_{2.5}$ components suggest that NO_3 is dominant during winter over western Europe and in a few eastern countries due to the high NO_2 mixing ratios. During summer NO_3 is dominant only in regions with elevated NH_3 emissions. For the rest of the domain SO_4 is dominant. Low OC concentrations are simulated mainly due to the uncertain representation of SOA formation.

1 Introduction

Air quality is a focus of attention because of its important role in many areas including human health, atmospheric reactions, acid deposition, and the Earth's radiation budget (e.g., Seinfeld and Pandis, 2006; Peng et al., 2005). Although air quality management strategies have been applied over recent years to reduce atmospheric pollutant concentrations, ozone and particulate matter pollution are still an issue. For this reason, simulating and forecasting gaseous and particle concentrations as accurately as possible is fundamental in air quality planning for more effective adaptation and implementation guidelines.

Air pollution is not just a local issue since the pollutants released in one country can be transported in the atmosphere, affecting air quality in the nearby countries. As such several research groups have started simulations of the gaseous and particulate matter concentrations over the whole of Europe. However, there are a limited number of such studies. In order to explain the European trends in ozone since 1990, Jonson et al. (2006) have used the EMEP regional photochemistry model for the years 1990 and 1995–2002. The increase in winter ozone, partially, and the decrease in the magnitude of high ozone episodes is attributed to the decrease in ozone precursor emissions while emission reductions have resulted in a marked decrease in summer ozone in major parts of Europe. Using the RegCM3/CAMx modelling system for simulating near surface ozone mixing ratios driven either by the ERA-40 reanalysis dataset or the global circulation model ECHAM5, Zanis et al. (2011) found that on a seasonal basis both ECHAM5 and ERA simulations exhibited a seasonally dependent bias, with winter and spring ozone values being generally underestimated; summer and autumn values were slightly overestimated. However, ozone peak mixing ratios in summer (i.e., higher than 80 ppbV) could not be captured. A decadal (i.e., 1991–2000) study also for ozone over Europe suggests that the selection of external meteorological forcing can be as important as the selection of adequate chemical lateral boundary conditions (Katragkou et al., 2010). A modelling set up for the whole Europe with CALIOPE air quality modelling system has been performed by Pay et al. (2010) suggesting satisfactory performance for ozone but poor performance for particles. Largely, this is caused by the inability of the models to correctly capture the concentrations

of organic matter (e.g., Chen and Griffin, 2005). Applying the CAMx modelling system over Europe Nopmongcol et al. (2012) found an underestimation trend for all pollutants examined (i.e., O_3, NO_x, NO_2, CO, PM_{10}) except for SO_2. Appel et al. (2012), using CMAQ, found that the model overestimated winter daytime ozone mixing ratios in Europe by an average of 8.4 % while in the summer slightly underestimated by 1.6 %. $PM_{2.5}$ is underestimated throughout the entire year. Due to the lack of speciated $PM_{2.5}$ data for the EU, they concluded that it is not clear what is driving this bias. Langmann et al. (2008), using the regional scale atmospheric climate chemistry/aerosol model REMOTE, found that the deviation between modelled and measured organic carbon concentrations can be mainly explained by missing formation of secondary organic aerosols (SOA) and deficiencies in emission data. As such, the authors suggest that an updated emission inventory needs to take into account the changing heating practices in Europe. The need for a more detailed treatment of the formation of SOA has also been pointed out by Sartelet et al. (2007) who simulated aerosols and gas-phase species over Europe with the POLYPHEMUS system. Although they found that hourly ozone, sulfate, and ammonium simulation was good, SO_2 and nitrate concentrations were overestimated. Modelling carbonaceous aerosol over Europe using the EMEP modelling system, Simpson et al. (2007) found that the contribution of biogenic secondary organic aerosol far exceeds that of the anthropogenic one. This modelling work confirms the difficulties of modelling SOA in Europe where a severe underestimation of the SOA components was found. The evaluation of the aerosol components in the CALIOPE air quality modelling system over Europe (Basart et al., 2012) also highlights underestimations in the fine fraction of carbonaceous matter (EC and OC) and secondary inorganic aerosols (i.e., nitrate, sulphate, and ammonium). The total amount of secondary inorganic aerosols was on average underestimated by 18–50 % in most regions of Europe (Pay et al., 2012). SO_2 was systematically overestimated by the CALIOPE system, which suggests that SO_4 formation in the modelling system was often limited by oxidant availability and not always by SO_2. NO_3 concentrations were underestimated in about 60 % in winter and more than 100 % in summer. Bessagnet et al. (2008) implemented the isoprene chemistry for SOA formation over Europe in the CHIMERE model. They found a better agreement between long series of simulated and observed particulate matter concentrations, but a clear underestimation by the CHIMERE model was noted in wintertime possibly due to missing wood burning emissions.

The objective of this study is to simulate gaseous (i.e., O_3, NO_2, SO_2) mixing ratios and particle (i.e., $PM_{2.5}$) concentrations over Europe, assessing the magnitude of disparity for each country and estimating errors and biases between observed and predicted values for European countries with available monitoring data. The current analysis provides an opportunity to compare the modelling results with the results obtained by other regional air quality models commonly used in Europe, noting at the same time, the European countries where the predicted values are in agreement with the observed values.

2 Methods

2.1 Modelling setup

Meteorological fields are derived using the Penn State/NCAR Mesoscale Model (MM5) (Grell et al., 1994). Since most meteorological models, such as MM5, are not built for air quality modelling purposes, it is necessary to address issues related to data format, unit conversion, temporal and spatial domains, and numerical grids. For this purpose, the Meteorology Chemistry Interface Processor (MCIP) (Byun et al., 1999) is used to provide the meteorological data from the MM5 outputs needed for the emissions and air quality models.

Gridded yearly averaged anthropogenic emissions for the year 2006 over Europe are provided by TNO (Netherlands Organization for Applied Scientific Research) at a 0.1×0.1 degree resolution (http://www.tno.nl) in the framework of the AQMEII exercise (http://aqmeii.jrc.ec.europa.eu/). The available data include annual total emissions of CH_4, CO, NH_3, NMVOC, NO_x, PM_{10}, $PM_{2.5}$, and SO_2 for both area and point sources in ten (10) Standardized Nomenclature for Air Pollutants (SNAP) categories (i.e., power generation, residential-commercial and other combustion, industrial combustion, industrial processes, extraction distribution of fossil fuels, solvent use, road transport, other mobile sources, waste treatment and disposal, agriculture) (Table 1). According to this emission inventory, the United Kingdom, Spain, Germany, Ukraine, France, and Italy have the highest NO_x emissions, while Ukraine, Spain, and Poland have the highest SO_2 emissions (only a part of the Russian Federation and Turkey belongs to the domain examined). In general, road transport and energy sector-utilities-refineries are the major sources for NO_x emissions, while SO_2 emissions originate mainly from the energy sector-utilities-refineries. Emissions are processed by the Sparse Matrix Operator Kernel Emissions (SMOKE v2.6) modelling system (http://www.smoke-model.org/index.cfm) to convert their resolution to the resolution needed by the air quality model using monthly, weekly, and hourly time profiles provided by TNO (2011). However, TNO has reported that the temporal profiles are a generalisation, not regularly updated and not country specific and could affect emissions over time for air quality modelling. The Biogenic Emission Inventory System, version 3 (BEIS3) is used for processing biogenic source emissions. Gridded land use data at 1 km resolution provided by the US Geological Survey (USGS) (http://edc2.usgs.gov/glcc/glcc.php). BEIS3 default summer and winter emission factors

Table 1. Anthropogenic emissions (kt yr^{-1}).

	CH_4	CO	NH_3	NMVOC	NO_x	PM_{10}	$PM_{2.5}$	SO_2
Albania	179	113	25	33	26	9	6	31
Austria	326	652	65	158	209	45	25	25
Belarus	762	532	143	190	162	36	25	77
Belgium	360	784	71	190	269	39	26	128
Bosn. & Herz.	160	182	17	49	53	42	18	420
Bulgaria	460	697	58	149	211	74	47	798
Croatia	153	296	46	88	64	23	16	55
Cyprus	48	37	5	14	17	3	2	11
Czech Rep.	473	470	67	173	256	34	20	184
Denmark	271	646	86	110	172	45	33	22
Estonia	89	173	10	36	34	26	21	61
Finland	211	493	36	132	182	48	32	83
France	2619	4711	727	1246	1109	479	302	415
F.Y.R.O.M.	90	103	7	26	40	18	9	102
Germany	2089	4017	623	1189	1353	192	109	545
Greece	404	569	71	332	271	67	51	533
Hungary	365	568	81	163	184	51	35	366
Ireland	610	199	109	57	107	21	14	53
Italy	1837	3895	431	1198	1094	161	113	413
Latvia	84	316	15	63	41	15	13	11
Lithuania	162	187	36	78	69	21	17	35
Luxembourg	17	41	5	13	13	3	2	3
Malta	19	0	1	8	11	0.6	0.4	8
Moldova	216	140	28	38	65	42	23	120
the Netherlands	776	568	135	167	299	39	20	48
Norway	217	397	23	189	205	50	43	20
Poland	1823	3282	296	915	631	282	134	1216
Portugal	514	585	68	284	237	45	36	186
Romania	1210	1390	198	381	272	138	97	457
Russia	23394	13019	772	2791	2853	1459	918	2810
Serbia	533	315	68	148	166	82	42	342
Slovakia	197	278	27	74	83	24	16	71
Slovenia	100	71	19	40	55	9	7	28
Sweden	254	585	50	191	195	53	33	36
Switzerland	167	300	55	102	80	19	9	15
Spain	1780	2205	454	1035	1459	209	141	1231
Turkey	2484	2825	426	729	888	365	260	1710
Ukraine	5143	2923	555	753	1279	516	311	1294
Un. Kingdom	2256	2127	309	926	1489	150	94	608

and the MM5 meteorological fields are used to create hourly model-ready biogenic emissions estimates.

The Community Multiscale Air Quality (CMAQ) v4.7 Modelling System with the Carbon Bond mechanism (CB05) is used here for the regional air quality modelling (Byun et al., 2006; Foley et al., 2010) for winter (i.e., January 2006) and summer (i.e., July 2006) months. CMAQ is a multipollutant, multiscale air quality model for simulating all atmospheric and land processes that affect transport, transformation, and deposition of atmospheric pollutants on both regional and urban scales. The modelling domain covers almost all of Europe with 177×217 grid cells of $35\,\text{km} \times 35\,\text{km}$ spatial resolution and 14 vertical layers (Fig. 1). Although a finer grid resolution could affect modelling results, other studies have found that it does not always enhance model performance (e.g., Queen et al., 2008). The default boundary and initial conditions based on Gipson (1999) for gaseous and particulate species have been used. Boundary conditions have a very minor impact on pollutants concentrations, since European land is far away from the domain borders, except along the eastern border. Russian Federation, Ukraine, and Turkey are close to the eastern borders, but these countries are not included in our analysis. Moreover, a spin up time of 10 days was used to minimise errors due to the initial conditions. In the version of CMAQ used, several new pathways for secondary organic

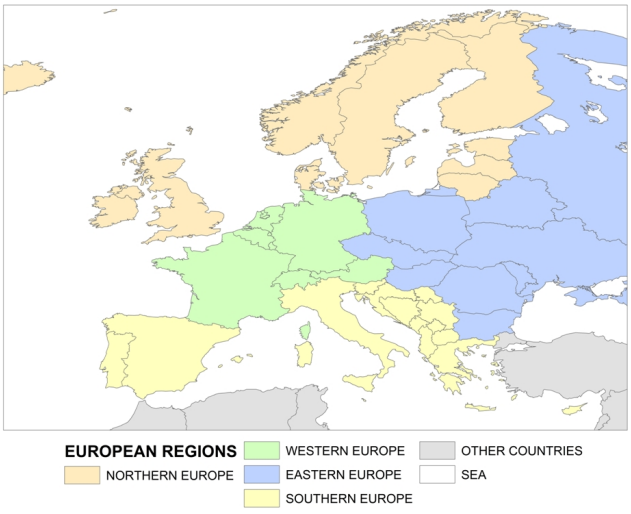

Fig. 1. Modelling domain and the regional European grouping used by the United Nations Statistics Department.

aerosol (SOA) formation have been implemented (Edney et al., 2007; Carlton et al, 2008). The CB05 is a condensed mechanism of atmospheric oxidant chemistry that provides a basis for computer modelling studies of ozone, particulate matter (PM), visibility, acid deposition, and atmospheric concentrations of toxic gases (Yarwood et al., 2005). The core CB05 mechanism has 51 species and 156 reactions. The CB05 has been evaluated against smog chamber data (Jeffries et al., 2002; Carter, 2000) and the results are discussed in detail by Yarwood et al. (2005).

Since an extensive evaluation and discussion of meteorology used has been presented by Vautard et al. (2012), here we focus on discussing the gaseous and particulate pollutant concentrations. Briefly, Vautard et al. (2012) found that the seasonal cycle of the 10 m wind speed is well reproduced although it is overestimated over Europe. The spatial distribution of surface wind speed is fairly well simulated. Wind speed is well simulated along the vertical profile, but markedly overestimated at lower altitudes over Europe. It was also found that the Planetary Boundary Layer (PBL) height at noon is simulated quite well. However, at 18:00 UTC and particularly in the summer months, the modelled PBL height is much lower than the observed. Biases of monthly means of the 2 m temperature are generally small. The diurnal cycle of the 2 m temperature is also fairly well reproduced while the typical vertical temperature profile bias is between ± 1 K. On average, the temperature is slightly underestimated while relative humidity above the surface is overestimated.

2.2 Model evaluation

Comparison between predicted and observed gas and particle concentrations is performed for January and July 2006 using observation data from AirBase, the European air quality database (http://www.eea.europa.eu/data-and-maps/data/airbase-the-european-air-quality-database-2). AirBase is the air quality information system maintained by the European Environment Agency (EEA) through the European topic centre on Air and Climate Change. It contains air quality data delivered annually, offering a reciprocal exchange of information and data from networks and individual stations measuring ambient air pollution within the Member States. Model evaluation is conducted here, for species with sufficient monitoring data all over Europe such as sulfur dioxide (data from 35 countries, in our domain there are 1928 stations for winter and 1883 for summer months), nitrogen dioxide (data from 35 countries, in our domain there are 2591 stations for winter and 2508 for summer months), ozone (data from 35 countries, in our domain there are 1954 stations for winter and 1977 for summer months) and particulate matter < 2.5 µm (data from 30 countries, in our domain there are 266 stations for winter and 267 for summer months). Hourly average data for NO_2 and SO_2 as well as daily average data for maximum 8 h ozone and $PM_{2.5}$ are used in our analysis. Unfortunately, comparison with observed $PM_{2.5}$ components could not be performed since speciated $PM_{2.5}$ data are not readily available for EU; this has also recently been pointed out by other researchers (Appel et al., 2012).

3 Results and discussion

High ozone mixing ratios are predicted in southern Europe where meteorological conditions enhance ozone formation (Fig. 2). The daily average maximum 8 h ozone ($Max8hO_3$) mixing ratio during July is simulated up to 75 ppbV while a large portion of the domain has values higher than 50 ppbV. Higher NO_2 mixing ratios are simulated over western Europe (i.e., Belgium, the Netherlands, Germany, and northern France), northern Italy and the United Kingdom for both seasons. Belgium and the Netherlands have elevated NO_2 values since their small area results in a high emission rate per acre, however, they are not ranked as one of the countries with high NO_x emission rates. Road transport and industry are responsible for the elevated NO_x emissions in northern Italy, while road and non-road transport energy sector and industry are responsible for the high NO_x emission in the United Kingdom. NO_2 mixing ratios are higher during January compared to July for two reasons: energy sector and industry emit more NO_x during winter and NO_2 photolysis is unfavourable during winter. Elevated SO_2 mixing ratios are simulated over eastern Europe, with higher values in Poland and the North Balkan Peninsula. Since power generation and industry are mainly responsible for SO_2 emissions, SO_2 values are very location dependent and also show higher values during winter. Elevated $PM_{2.5}$ levels are simulated over eastern and western Europe (i.e., daily average concentrations up to 30 µg m^{-3} during winter). NO_3 is dominant during winter

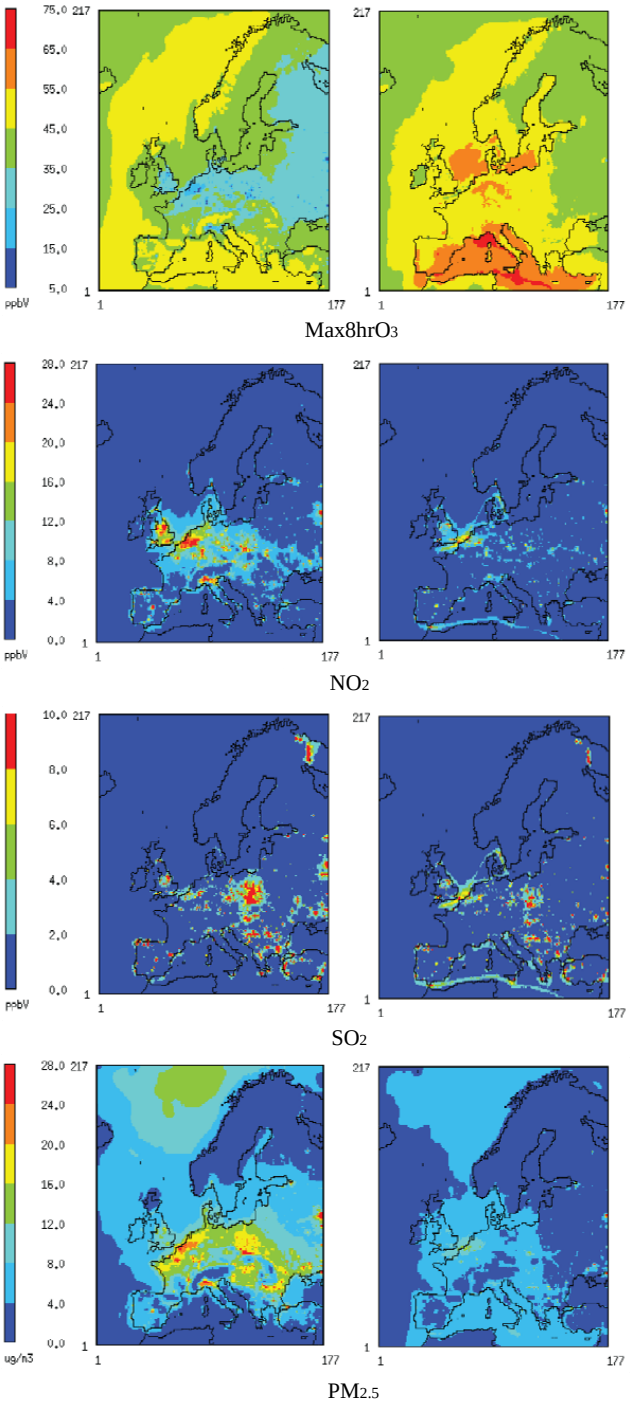

Fig. 2. Simulated daily ($Max8hO_3$, $PM_{2.5}$) and hourly (NO_2, SO_2) average concentrations for January (left column) and July (right column) 2006.

over western Europe and in a few eastern countries due to the high NO_2 mixing ratios (Fig. 3). During summer NO_3 is dominant only in regions with elevated NH_3 emissions (i.e., the Netherlands and northern Italy). For the rest of the domain SO_4 is dominant. Low OC concentrations are simulated in general. Representation of secondary organic aerosol (SOA) formation is uncertain (e.g., Chen et al., 2005; Kroll et al., 2006) and low OC has been noted in the CMAQ approaches (Foley et al., 2010). NH_4 follows SO_4 and NO_3 spatial distribution plots for both seasons, since atmospheric SO_2 is oxidized to sulfuric acid which reacts with ammonia to form ammonium sulfate, while gas-phase NO_x oxidizes to nitric acid which reacts with ammonia to form ammonium nitrate.

Spatial distribution plots presented here for gaseous pollutants and $PM_{2.5}$ are similar with those presented by another study (Pay et al., 2010). Using the WRF-ARW meteorological model, the HERMES-EMEP emission processing model, a mineral dust dynamic model (BSC-DREAM8b), and CMAQ chemical transport model, they provide annual simulations for 2004 over Europe. Both studies have found high ozone mixing ratios over the Mediterranean and the nearby land; elevated NO_2 mixing ratios over western Europe (i.e., Belgium, the Netherlands, Germany, northern France), southern United Kingdom, and northern Italy; elevated SO_2 levels over eastern Europe (i.e., Poland) and northern Balkan Peninsula; higher $PM_{2.5}$ concentrations over western and eastern countries (i.e., Belgium, northern France, Poland, southern Romania) and northern Italy.

Model performance for ozone shows a mixed trend: $Max8hO_3$ is overestimated for low mixing ratios (about 50 ppbV) while it is underestimated for the higher mixing ratios. This trend is in agreement with the CMAQ application performed by Appel et al. (2012) for Europe where daytime ozone mixing ratio is overestimated in winter and underestimated in summer months. The overestimation tendency for the lower mixing ratios gives a much higher mean estimated value during winter compared to the observed one (Table 2), which is diminished during summer where higher ozone mixing ratios are recorded. Both observed and predicted ozone mixing ratios are similarly spread out around mean values during winter (similar simulated standard and mean absolute deviations). During summer the spread of simulated mixing ratios is less and the mean simulated mixing ratios are closer to the observation data; this is related to the overestimation of the lower mixing ratios. At regional scale, according to the grouping used by the United Nations Statistics Department for northern, western, eastern, and southern Europe (http://unstats.un.org/unsd/methods/m49/m49regin.htm#europe; Fig. 1), the model overestimates ozone mixing ratios in all regions during winter; during summer it overestimates ozone mixing ratios in northern and southern Europe and underestimates them in eastern and western Europe where higher mixing ratios have been recorded (Table 2). A consistent bias for NO_2, SO_2, and $PM_{2.5}$ estimations is noted: NO_2 and $PM_{2.5}$ are underestimated while SO_2 is overestimated for both seasons in all regions. The same biases have also been noted by a previous

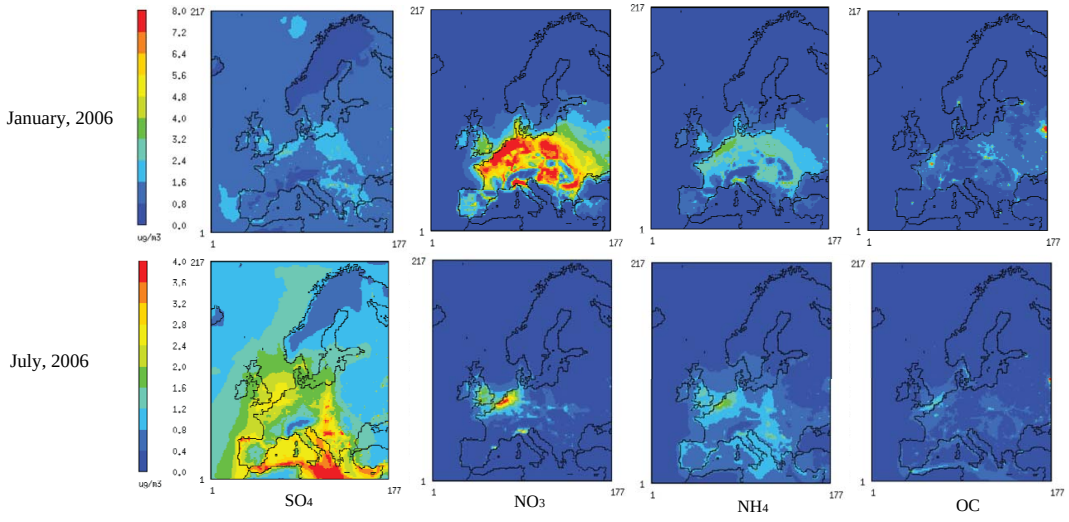

Fig. 3. Simulated $PM_{2.5}$ component daily average concentrations for January and July 2006.

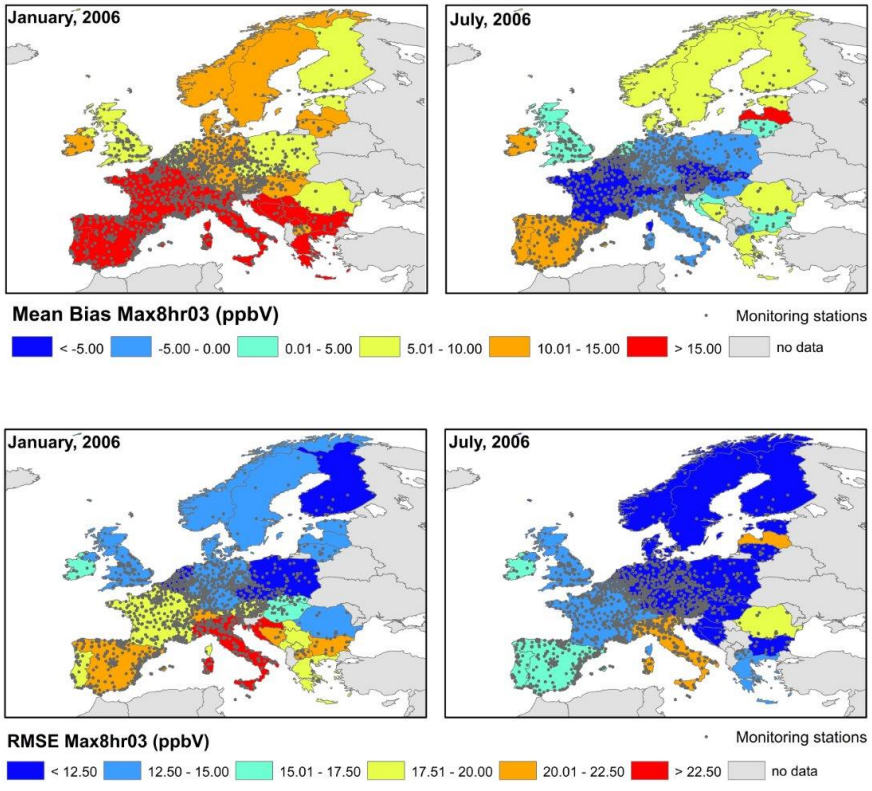

Fig. 4. Mean Bias and RMSE for daily average $Max8hO_3$ mixing ratios for the European countries (dots show the distribution of O_3 monitoring stations).

study (Pay et al., 2010). The consistent underestimation trend for $PM_{2.5}$ has been found also by Appel et al. (2012) using CMAQ modelling system for Europe. At the regional scale, we underestimate NO_2 mixing ratios more in southern Europe and overestimate SO_2 mixing ratios more in eastern Europe where higher underestimation in $PM_{2.5}$ concentrations is found.

Modelling results averaged over large areas (i.e., Europe or European regions) are important for several issues (e.g., model performance, sensitivity studies, and strategic plans for pollutants reduction), but of equally importance are

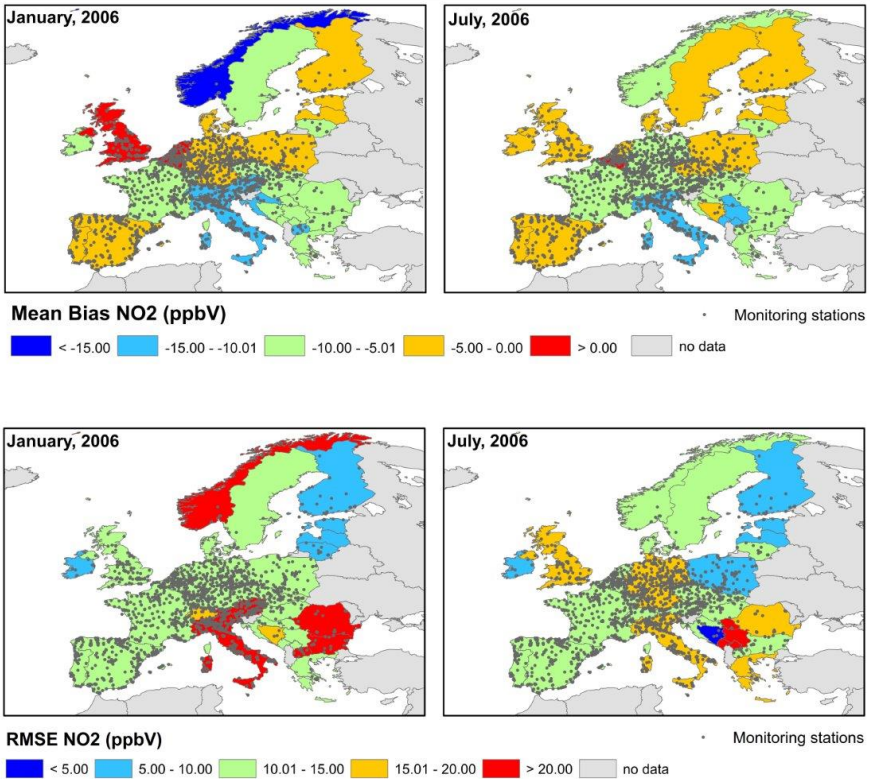

Fig. 5. Mean Bias and RMSE for hourly average NO_2 mixing ratios for the European countries (dots show the distribution of NO_2 monitoring stations).

outcomes at a country level. In an effort to address this we assess the disagreement between mean observed and predicted values for each country based on the ratio $\frac{\text{Observed}_{\text{mean}}}{\text{Predicted}_{\text{mean}}}$ along with the Mean Bias ($\text{MB} = \frac{1}{n}\sum_{i=1}^{n}(\text{P}_{(i)} - \text{O}_{(i)})$) and the Root-Mean-Square Error ($\text{RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(\text{P}_{(i)} - \text{O}_{(i)})^2}$) (where $\text{P}_{(i)}$ stands for predicted data and $\text{O}_{(i)}$ for observed data).

Max8hO_3 is overestimated for all countries during January since low ozone mixing ratios are observed in winter. The observed to predicted ratio is between 0.6 and 0.8 for almost all countries (Table 3). During July, the mean observed values are closer to the mean predicted values for all countries (the ratio is 1.0 ± 0.1 for the majority of the countries). Few countries (i.e., Estonia, Finland, Ireland, Latvia, Norway, and Romania) show similar disagreement for both months. MB for Max8hO_3 mixing ratios is positive for all countries for January since the model overestimates low mixing ratios that recorded during winter (Fig. 4). The calculated MB for the majority of the countries is more than 10 ppbV. Both negative and positive MB values for Max8hO_3 mixing ratios are obtained for July. For many countries (i.e, Bulgaria, Croatia, F.Y.R.O.M., Germany, Hungary, Italy, Lithuania, Luxembourg, the Netherlands, Poland, and the United Kingdom) the calculated MB is 0 ± 5 ppbV for the summer month while higher positive MB values are calculated for Ireland, Latvia, Malta, Portugal, and Spain. Spatial distribution plots with the Normalised MB for the daytime ozone mixing ratios are presented by Appel et al. (2012). Although an exact comparison with Appel et al. (2012) could not be performed, since we present MB for Max8hO_3 mixing ratios averaged per country for January and July 2006 vs. their Normalised MB for daytime ozone mixing ratios averaged per monitoring station for the three winter and the three summer months of 2006, the general trend is similar: better agreement between observed and predicted mixing ratios during summer and model high overestimation during winter. However, Appel et al. (2012) found a tendency to underestimate winter daytime ozone mainly in Czech Republic and Poland. Higher RMSE are calculated for the southern European countries compared to the northern European countries for both months. Lower RMSE are calculated in almost all countries for July compared to January. The countries that seem to have the best agreement between observed and predicted Max8hO_3 mixing ratios for all parameters examined here (i.e., $\frac{\text{Observed}_{\text{mean}}}{\text{Predicted}_{\text{mean}}}$: 1 ± 0.1, MB: 0 ± 5 ppbV, RMSE: ≤ 12 ppbV) in July are: Lithuania, Hungary, Poland, Bulgaria, Germany, the Netherlands, and Croatia, while no country was found to agree well with all the previous parameters for January.

Table 2. Statistical analysis for hourly average NO_2 and SO_2 concentrations and daily average $Max8hO_3$ and $PM_{2.5}$ concentrations over Europe (see Fig. 1 for the definition of the European regions).

		$Max8hrO_3$ (ppbV)					NO_2 (ppbV)					SO_2 (ppbV)					$PM_{2.5}$ ($\mu g/m^3$)				
		January 2006																			
		Europe (Total)	North	South	East	West	Europe (Total)	North	South	East	West	Europe (Total)	North	South	East	West	Europe (Total)	North	South	East	West
MEAN± STANDARD DEVIATION	Observed concentrations	19.5 ± 10.9	22.7± 10.4	20.9± 11.2	22.3± 11.1	17.5± 10.3	19.7± 14.5	16.4± 13.3	19.8± 16.2	18.7± 15.3	20.4± 12.9	4.3± 7.4	2.2± 3.0	3.4± 7.2	9.9± 11.6	3.2± 4.8	27.1± 20.2	14.3± 9.6	23.1± 19.3	44.4± 22.3	28.3± 18.3
	Predicted concentrations	33.8± 11.3	32.6± 11.5	38.9± 10.1	30.6± 10.0	31.3± 11.1	13.4± 11.5	13.4± 12.7	12.3± 12.4	12.8± 10.3	14.4± 10.9	6.6± 15.7	3.7± 7.0	6.6± 17.7	12.9± 25.6	4.6± 7.0	14.1± 12.1	9.4± 6.8	8.4± 7.5	14.1± 8.5	20.2± 14.4
MEAN ABSOLUTE DEVIATION (MAD)	Observed concentrations	8.9	8.4	9.3	8.8	8.4	11.1	10.1	12.8	11.3	9.8	3.7	1.6	3.0	7.4	2.4	16.1	6.9	14.9	18.0	14.5
	Predicted concentrations	9.1	8.9	7.8	8.0	8.3	9.3	10.5	9.7	8.2	8.9	6.9	3.6	7.3	13.4	4.3	8.5	4.9	5.3	6.1	10.0
ROOT MEAN SQUEAR ERROR (RMSE)		18.1	13.2	21.0	13.6	17.5	16.3	13.6	18.2	15.4	15.4	15.7	7.5	18.2	24.9	7.9	24.5	10.4	22.6	36.6	23.7
INDEX OF AGREEMENT (IoA)		0.5	0.7	0.5	0.6	0.5	0.6	0.6	0.6	0.6	0.6	0.3	0.2	0.3	0.4	0.4	0.5	0.6	0.5	0.5	0.4
MEAN ABSOLUTE ERROR (MAE)		15.3	11.0	18.5	10.8	14.9	11.6	9.4	13.2	10.4	11.1	5.9	3.3	6.4	11.1	4.0	17.0	6.8	15.4	30.5	16.7
		July 2006																			
MEAN± STANDARD DEVIATION	Observed concentrations	54.0± 17.1	41.5± 15.9	51.4± 18.9	53.0± 15.2	58.1± 14.9	12.1± 12.0	11.6± 11.9	12.8± 12.2	9.7± 10.2	12.3± 12.3	2.2± 4.8	2.0± 3.3	2.6± 5.6	2.3± 3.9	1.8± 4.2	16.1± 7.7	12.4± 7.2	16.8± 8.2	19.3± 9.3	15.4± 5.8
	Predicted concentrations	53.2± 8.4	47.4± 7.5	56.8± 8.8	50.6± 6.8	52.5± 7.6	6.2± 7.9	7.5± 9.3	6.3± 8.2	5.5± 7.3	6.1± 7.5	4.4± 10.2	3.0± 5.5	4.7± 10.6	7.0± 16.6	3.4± 6.4	6.6± 4.4	5.3± 3.2	5.6± 2.9	6.2± 3.0	8.0± 5.7
MEAN ABSOLUTE DEVIATION (MAD)	Observed concentrations	13.5	12.6	13.9	12.3	12.1	8.6	8.4	9.0	6.9	8.7	1.9	1.5	2.2	1.7	1.6	5.9	5.7	6.4	7.4	4.5
	Predicted concentrations	6.5	6.0	6.8	5.3	5.9	5.3	6.4	5.6	5.0	5.1	4.8	2.9	4.9	7.9	3.6	2.9	2.4	2.1	2.3	4.1
ROOT MEAN SQUEAR ERROR (RMSE)		14.6	13.2	18.7	11.9	12.2	14.0	12.7	14.9	11.2	14.2	11.2	5.9	12.0	17.4	7.3	12.1	9.0	13.4	15.4	10.1
INDEX OF AGREEMENT (IoA)		0.6	0.7	0.5	0.7	0.7	0.5	0.6	0.5	0.6	0.5	0.2	0.3	0.2	0.2	0.3	0.5	0.5	0.5	0.5	0.5
MEAN ABSOLUTE ERROR (MAE)		11.2	11.0	13.9	9.5	9.9	9.0	8.1	10.0	7.0	9.0	4.2	2.6	4.6	6.4	3.1	10.1	7.2	11.4	13.3	8.6

$$\mathrm{RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(\mathrm{P}_{(i)} - \mathrm{O}_{(i)})^2} \qquad \mathrm{MAE} = \frac{1}{n}\sum_{i=1}^{n}|\mathrm{P}_{(i)} - \mathrm{O}_{(i)}| \qquad \mathrm{IoA} = 1 - \frac{\sum_{i=1}^{n}(\mathrm{P}_{(i)} - \mathrm{O}_{(i)})^2}{\sum_{i=1}^{n}(|\mathrm{P}_{(i)} - \overline{\mathrm{O}}| + |\mathrm{O}_{(i)} - \overline{\mathrm{O}}|)^2} \qquad \mathrm{MAD} = \frac{1}{n}\sum_{i=1}^{n}|X_{(i)} - \overline{X}|$$

(X stands for either O or P).

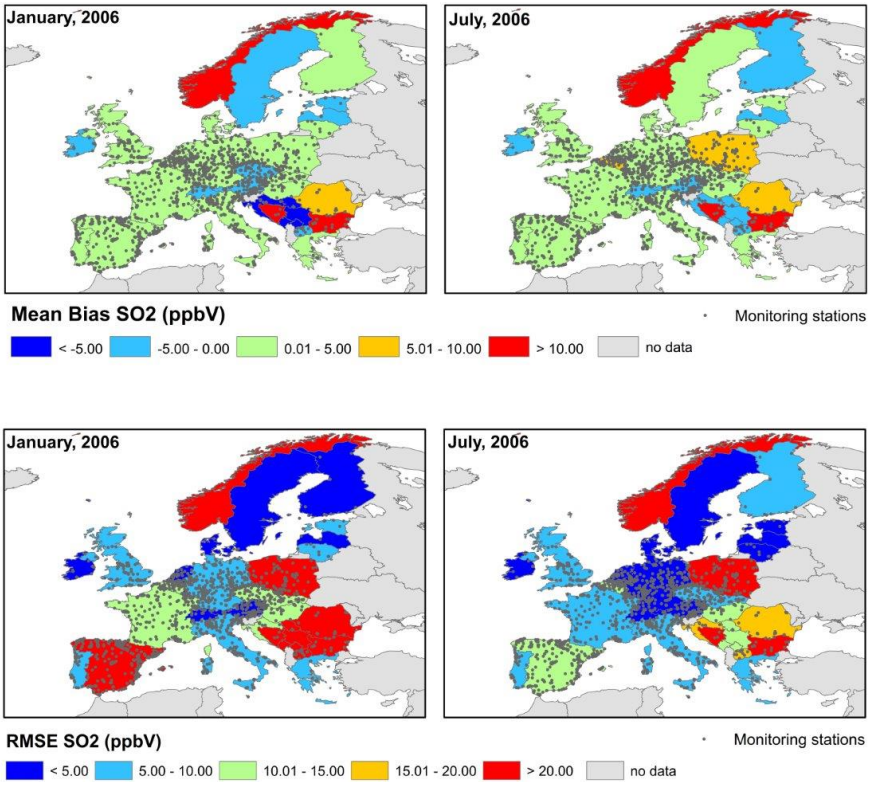

Fig. 6. Mean Bias and RMSE for hourly average SO_2 mixing ratios for the European countries (dots show the distribution of SO_2 monitoring stations).

The mean simulated NO_2 mixing ratios are close to the observed mixing ratios in Belgium, the Netherlands, Spain, and the United Kingdom for both months (Table 3). However, the disagreement is greater during July in almost all countries. MB for NO_2 values is negative for all countries except Belgium for both months as well as for the Netherlands and the United Kingdom in January with values up to 4.2 ppbV (Fig. 5). The largest negative MB is estimated for Norway (< -15 ppbV) in January while estimates for Spain, Estonia, Denmark, Portugal, Finland, Luxembourg, Poland, Germany, and Latvia along with the United Kingdom, the Netherlands, and Belgium have MB 0 ± 5 ppbV. Austria, F.Y.R.O.M., and Norway is estimated to have much higher negative MB in January compared to July, while Serbia, Germany, and Luxembourg is estimated to have much higher negative MB in July compared to January. High RMSE is calculated in Romania, Austria, Italy, F.Y.R.O.M., Norway, and Bulgaria in January while Serbia is estimated to have the highest RMSE in July. The Netherlands has the best agreement between observed and predicted NO_2 mixing ratios for all parameters examined here (i.e., $\frac{\text{Observed}_{\text{mean}}}{\text{Predicted}_{\text{mean}}}: 1 \pm 0.1$, MB: 0 ± 5 ppbV, RMSE: ≤ 12 ppbV) in January. However, model performance for Belgium and the United Kingdom is quite good for the same month. Belgium has the best agreement in July between observed and predicted NO_2 mixing ratios.

The mean simulated SO_2 mixing ratios are close to the observed mixing ratios in F.Y.R.O.M. for both months (Table 3). In few countries (i.e., Czech Republic, Greece, Luxembourg, and Sweden) the average simulated SO_2 mixing ratios are close to the observed mixing ratios in January, while for Finland and Serbia the average mixing ratios show good agreement in July. Three countries (i.e., Croatia, Latvia, and Switzerland) are simulated to have mean SO_2 mixing ratios much lower than the mean observed mixing ratios for both months. However, in three countries (i.e., Greece, Czech Republic, and Sweden) although mean observed mixing ratios are similar to the mean predicted mixing ratios during January they are much lower in July. Estonia shows reverse behaviour for the simulated months i.e., mean observed mixing ratio is much higher compared to the mean predicted in January and much lower in July. MB in SO_2 mixing ratios are estimated to have both positive and negatives values for both months (Fig. 6). The highest positive MB are calculated for Norway, Bulgaria, Bosnia and Herzegovina, and Romania for both months, while the highest negative MB are calculated for Croatia and Serbia in January and for Croatia in July. More than half of the countries have MB in the range 0 ± 2 ppbV for both months. There is no significant change in the MB between the two seasons with the exception of Serbia. High RMSE is calculated for Spain, Romania,

Table 3. $Observed_{mean}/Predicted_{mean}$ concentration ratios for the European countries and the number of monitoring stations (in parenthesis).

	$Max8hO_3$	NO_2	SO_2	$PM_{2.5}$	$Max8hO_3$	NO_2	SO_2	$PM_{2.5}$
Country	January 2006				July 2006			
Austria	0.7 (114)	2.1 (146)	2.2 (115)	4.1 (5)	1.1 (114)	2.2 (147)	1.3 (111)	3.2 (5)
Belgium	0.6 (39)	0.8 (66)	0.5 (61)	1.1 (11)	1.1 (38)	0.9 (65)	0.3 (61)	1.2 (10)
Bosn. & herz.	0.5 (3)	1.9 (3)	0.7 (3)	2.7 (1)	0.9 (3)	1.7 (3)	0.3 (3)	1.7 (1)
Bulgaria	0.5 (13)	2.0 (14)	0.5 (15)	2.7 (4)	0.9 (13)	2.2 (14)	0.3 (15)	2.6 (4)
Croatia	0.2 (1)	2.1 (7)	2.3 (7)	– (–)	1.0 (2)	3.4 (8)	2.2 (7)	– (–)
Czech rep.	0.8 (60)	1.4 (92)	1.1 (89)	3.4 (27)	1.1 (59)	1.4 (91)	0.4 (88)	3.3 (28)
Denmark	0.6 (9)	1.2 (12)	0.4 (2)	1.5 (3)	0.9 (9)	1.5 (11)	0.3 (2)	2.5 (4)
Estonia	0.8 (7)	1.7 (6)	1.9 (7)	– (–)	0.8 (7)	1.9 (6)	0.6 (7)	– (–)
Finland	0.8 (16)	1.8 (30)	0.7 (11)	1.1 (7)	0.8 (17)	2.0 (31)	1.1 (10)	2.1 (7)
France	0.5 (430)	1.6 (483)	0.5 (335)	0.9 (50)	1.1 (441)	2.1 (433)	0.6 (306)	1.7 (50)
F.Y.R.O.M.	0.7 (13)	2.8 (11)	1.1 (13)	– (–)	1.1 (13)	2.9 (13)	1.1 (13)	– (–)
Germany	0.6 (296)	1.3 (425)	0.8 (251)	2.2 (28)	1.1 (296)	2.3 (422)	0.5 (245)	3.0 (31)
Greece	0.6 (20)	1.7 (19)	0.9 (12)	– (–)	0.9 (20)	2.8 (22)	0.5 (12)	– (–)
Hungary	0.6 (17)	1.5 (23)	0.6 (22)	2.6 (3)	1.1 (17)	2.5 (23)	0.3 (23)	3.1 (3)
Ireland	0.7 (10)	1.9 (11)	1.5 (10)	– (–)	0.8 (10)	1.4 (9)	1.5 (7)	– (–)
Italy	0.5 (220)	2.1 (409)	0.6 (273)	4.8 (24)	1.1 (228)	2.9 (398)	0.6 (278)	3.1 (28)
Latvia	0.7 (6)	1.6 (5)	3.0 (4)	– (–)	0.6 (5)	1.7 (5)	1.8 (4)	– (–)
Lithuania	0.7 (11)	2.4 (12)	0.7 (9)	– (–)	0.9 (13)	3.4 (12)	0.6 (9)	– (–)
Luxembourg	0.5 (6)	1.4 (6)	1.1 (6)	1.1 (1)	1.1 (6)	2.8 (6)	1.4 (6)	1.1 (1)
Malta	0.6 (2)	4.7 (2)	1.3 (2)	– (–)	0.8 (4)	1.5 (4)	0.5 (4)	2.9 (2)
the Netherlands	0.6 (38)	0.9 (51)	0.6 (35)	– (–)	1.0 (37)	1.4 (51)	0.5 (35)	– (–)
Norway	0.7 (8)	5.5 (17)	0.2 (1)	2.1 (10)	0.8 (8)	4.0 (15)	0.2 (1)	2.5 (6)
Poland	0.8 (53)	1.4 (93)	0.8 (94)	2.2 (3)	1.1 (54)	1.6 (98)	0.3 (93)	2.2 (3)
Portugal	0.6 (47)	1.3 (59)	0.4 (43)	1.9 (14)	0.8 (44)	1.6 (59)	0.5 (43)	2.2 (13)
Romania	0.8 (23)	1.6 (32)	0.4 (31)	1.7 (2)	0.8 (25)	2.1 (32)	0.3 (31)	3.3 (2)
Serbia	0.5 (1)	1.4 (3)	2.1 (3)	– (–)	– (–)	3.3 (3)	1.1 (3)	– (–)
Slovakia	0.6 (18)	1.9 (27)	0.8 (26)	5.4 (3)	1.1 (17)	2.5 (26)	0.7 (25)	3.7 (3)
Sweden	0.7 (15)	1.7 (14)	1.1 (4)	1.6 (7)	0.9 (15)	1.6 (15)	0.5 (5)	2.5 (8)
Switzerland	0.5 (32)	2.2 (33)	1.9 (13)	3.4 (5)	1.2 (32)	3.3 (33)	1.8 (13)	3.5 (5)
Spain	0.5 (342)	1.1 (363)	0.4 (357)	1.9 (52)	0.8 (342)	1.3 (340)	0.5 (349)	3.3 (47)
United Kingdom	0.7 (83)	0.9 (106)	0.5 (74)	1.1 (6)	0.9 (87)	1.3 (92)	0.6 (74)	2.1 (6)

Poland, F.Y.R.O.M, Serbia, Bulgaria, Bosnia and Herzegovina, and Norway in January, while Poland, Bulgaria, Bosnia and Herzegovina, and Norway are estimated to have high RMSE in July. However, results for countries with a very limited number in monitoring stations (e.g., Norway) may not be representative. Czech Republic, Greece, Luxembourg, and Sweden seem to have the best agreement between observed and predicted SO_2 mixing ratios for all parameters examined here (i.e., $\frac{Observed_{mean}}{Predicted_{mean}}$: 1 ± 0.1, MB: 0 ± 5 ppbV, RMSE: $\leq$12 ppbV) in January as well as Finland and Serbia in July.

Mean observed $PM_{2.5}$ concentrations are higher compared to the predicted concentrations for all countries except France in January (Table 3). For Belgium and Luxembourg mean observed and predicted concentrations are in good agreement for both months while good agreement is also noted for Finland, France, and the United Kingdom in January. MB for $PM_{2.5}$ concentrations is estimated to be negative for all countries for both months except France in January (Fig. 7). The highest (negative) MB and RMSE are calculated for Switzerland, Poland, Bulgaria, Hungary, Czech Republic, Italy, Austria, Slovakia, and Bosnia and Herzegovina in January. However, they are much lower in July. The United Kingdom and Finland in January and Luxembourg in July are estimated to have the lowest MB (i.e., up to -1$\mu g/m^3$) as well as the lowest RMSE (i.e., up to 5.4$\mu g/m^3$). Luxembourg, Finland, and the United Kingdom seem to have the best agreement between observed and predicted $PM_{2.5}$ concentrations for all parameters examined here (i.e., $\frac{Observed_{mean}}{Predicted_{mean}}$: 1 ± 0.1, MB: $0 \pm 5\,\mu g\,m^{-3}$, RMSE: $\leq 12\,\mu g\,m^{-3}$) in January as well as Luxembourg in July. Spatial distribution plots of the seasonal Normalised MB for $PM_{2.5}$ concentrations for 2006 are presented by Appel et al. (2012). Both studies underestimate $PM_{2.5}$ concentrations, and predict lower values of the parameters examined in each study (i.e., MB and Normalised MB) during summer months.

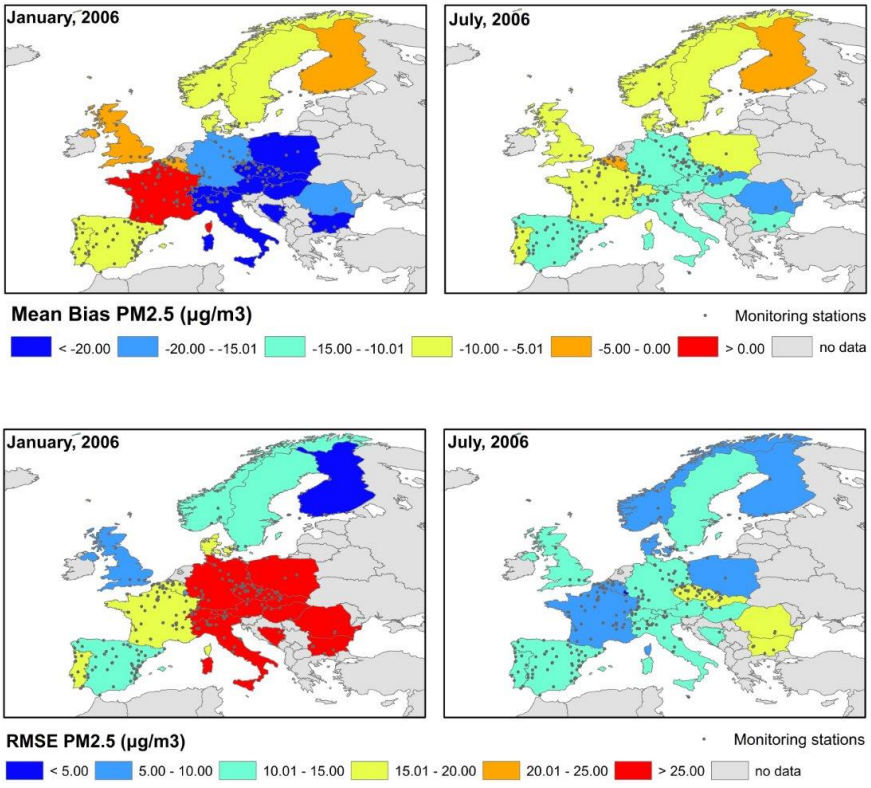

Fig. 7. Mean Bias and RMSE for daily average $PM_{2.5}$ concentrations for the European countries (dots show the distribution of $PM_{2.5}$ monitoring stations).

4 Conclusions

Application of CMAQ modelling system over Europe for January and July 2006 using the TNO gridded anthropogenic emissions database for the year 2006 shows an overestimation trend for low ozone mixing ratios (less than 50 ppbV) while the higher mixing ratios are underestimated. However, spatial distribution plots are reasonably estimated (e.g., higher ozone mixing ratios in southern Europe). Simulated values for NO_2, SO_2, and $PM_{2.5}$ suggest a consistent bias using regional average values: SO_2 is overestimated while NO_2 and $PM_{2.5}$ are underestimated. Speciated $PM_{2.5}$ components give low OC concentrations as a result of the uncertain representation of SOA formation. Statistical analysis (i.e., mean concentrations, MB, RMSE) for each country found that observed and predicted $Max8hO_3$ mixing ratios are in good agreement in Lithuania, Hungary, Poland, Bulgaria, Germany, the Netherlands, and Croatia in July. However, no country was found to be in good agreement in January where low ozone mixing ratios are recorded. The Netherlands has the best agreement between observed and predicted NO_2 mixing ratios in January, although model performance in Belgium and the United Kingdom is quite good for the same month. Belgium was the only country to have good agreement between observed and predicted NO_2 mixing ratios in July. Czech Republic, Greece, Luxembourg, and Sweden had the best agreement between observed and predicted SO_2 mixing ratios in January as well as Finland and Serbia in July. Luxembourg, Finland, and the United Kingdom are estimated to have the best agreement between observed and predicted $PM_{2.5}$ concentrations in January as well as Luxembourg in July.

There are numerous reasons why a bias or an error may exist. This could be related to inaccuracies in emission inventories. Emissions of air pollutants originate from a variety of small and large individual sources (e.g., power plants, industries, motor vehicles) of varying temporal and spatial characteristics. They are subject to significant uncertainties given that they are based on datasets of limited spatiotemporal coverage and that countries do not always estimate emissions in a uniform and transparent manner. However, biases and errors could also be related to discrepancies in the meteorological data and the source locations; incommensurability of grid cell averaged predictions with point measurements, horizontal grid resolution, topographic effects that are not accounted for in the model; or the lack of detail in some of the model parameterizations. An in depth analysis on these issues is required to identify the sources that cause these biases and errors. This will need to consider that small countries are affected by transported pollutants from

neighbouring countries. Assessing (i) the effect of precursor emissions using inverse modelling techniques, (ii) the effect of a finer resolution domain, iii) the effect of other chemical mechanism or even a different air quality model will provide more information for the sources and the magnitude of the uncertainty.

Acknowledgements. This work was supported by the FP7-REGPOT-2008-1grant No. 229773 and the National Strategic Reference Framework (NSRF) 2007–2013 grand No. 09SYN-31-667. We gratefully acknowledge the first Air Quality Model Evaluation International Initiative (AQMEII) activity. The following agencies have prepared the databases used in the study: TNO (European emissions processing), Laboratoire des Sciences du Climat et de l'Environment, IPSL, CEA/CNRS/UVSQ (gridded meteorology for Europe).

Edited by: A. Nenes

References

Appel, K. W., Chemel, C., Roselle, S. J., Francis, X. V., Hu, R. M., Sokhi, R. S., Rao, S. T., and Galmarini, S.: Examination of the Community Multiscale Air Quality (CMAQ) model performance over the North American and European domains, Atmos. Environ., 53, 142–145, 2012.

Basart, S., Pay, M. T., Jorba, O., Pérez, C., Jiménez-Guerrero, P., Schulz, M., and Baldasano, J. M.: Aerosols in the CALIOPE air quality modelling system: evaluation and analysis of PM levels, optical depths and chemical composition over Europe, Atmos. Chem. Phys., 12, 3363–3392, doi:10.5194/acp-12-3363-2012, 2012.

Bessagnet, B., Menut, L., Curci, G., Hodzic, A., Guillaume, B., Liousse, C., Moukhtar, S., Pun, B., Seigneur, C., and Schulz, M.: Regional modeling of carbonaceous aerosols over Europe—focus on secondary organic aerosols, J. Atmos. Chem., 61, 175–202, 2008.

Byun, D. W., Pleim, J. E., Tang, R. T., and Bourgeois, A.: Meteorology-Chemistry Interface Processor (MCIP) for Models-3 Community Multiscale Air Quality (CMAQ) modeling System. In Science algorithms of the EPA Models-3 Community Multiscale Air Quality (CMAQ) modeling System, US Environmental Protection Agency Report, EPA-600/R-99/030, 12-1–12-91, 1999.

Byun, D. W. and Schere, K. L.: Review of the governing equations, computational algorithms, and other components of the Models-3 Community Multscale Air Quality CMAQ) modeling system, App. Mech. Rev., 59, 51–77, 2006.

Carlton, A. G., Turpin, B. J., Altieri, K. E., Seitzinger, S. P., Mathur, R., Roselle, S. J., and Weber, R. J.: CMAQ Model Performance Enhanced When In-Cloud Secondary Organic Aerosol is Included: Comparisons of Organic Carbon Predictions with Measurements, Environ. Sci. Technol., 42, 8798–8802, 2008.

Carter, W. P. L.: Implementation of the SAPRC-99 Chemical Mechanism into the MODELS-3 Framework, Report to the U.S. EPA, available at: http://www.cert.ucr.edu/~carter/absts.htm#s99mod3 and http://www.cert.ucr.edu/~carter/SAPRC99/ (last access: September 2013), 29 January 2000.

Chen, J. and Griffin, R. J.: Modeling secondary organic aerosol formation from oxidation of a-pinene, b-pinene, and d-limonene, Atmos. Environ., 39, 7731–7744, 2005.

Edney, E. O., Kleindienst, T. E., Lewandowski, M., and Offenberg, J. H.: Updated SOA chemical mechanism for the Community Multi-Scale Air Quality model, EPA 600/X-07/025, US EPA, Research Triangle Park, NC, 2007.

Foley, K. M., Roselle, S. J., Appel, K. W., Bhave, P. V., Pleim, J. E., Otte, T. L., Mathur, R., Sarwar, G., Young, J. O., Gilliam, R. C., Nolte, C. G., Kelly, J. T., Gilliland, A. B., and Bash, J. O.: Incremental testing of the Community Multiscale Air Quality (CMAQ) modeling system version 4.7, Geosci. Model Dev., 3, 205–226, doi:10.5194/gmd-3-205-2010, 2010.

Gipson, G. L.: The Initial Concentration and Boundary Condition Processors. In Science algorithms of the EPA Models-3 Community Multiscale Air Quality (CMAQ) Modeling System, US Environmental Protection Agency Report, EPA-600/R-99/030, 12-1–12-91, 1999.

Grell, G., Dudhia, J., and Stauffer, D. R.: A description of the fifth generation Penn State/NCAR mesoscale model (MM5), NCAR Tech. Note, NCAR/TN-398 + STR, Natl. Cent for Atmos. Res., Boulder, Colorado, 1994.

Jeffries, H. E., Voicu, I., and Sexton, K.: Experimental Tests of Reactivity and Reevaluation of The Carbon Bond Four Photochemical Reaction Mechanism, Final report for Cooperative Agreement No. R828906, US EPA, RTP, NC., 2002.

Jonson, J. E., Simpson, D., Fagerli, H., and Solberg, S.: Can we explain the trends in European ozone levels?, Atmos. Chem. Phys., 6, 51–66, doi:10.5194/acp-6-51-2006, 2006.

Katragkou, E., Zanis, P., Tegoulias, I., Melas, D., Kioutsioukis, I., Kruger, B. C., Huszar, P., Halenka, T., and Rauscher, S.: Decadal regional air quality simulations over Europe in present climate: near surface ozone sensitivity to external meteorological forcing, Atmos. Chem. Phys., 10, 11805–11821, doi:10.5194/acp-10-11805-2010, 2010.

Kroll, J. H., Ng, N. L., Murphy, S. M., Flagan, R. C., and Seinfeld, J. H.: Secondary organic aerosol formation from isoprene photooxidation, Environ. Sci. Technol., 40, 1869–1877, 2006.

Langmann, B., Varghese, S., Marmer, E.,Vignati, E., Wilson, J., Stier, P., and O'Dowd, C.: Aerosol distribution over Europe: a model evaluation study with detailed aerosol microphysics, Atmos. Chem. Phys.: 8,1591–1607,2008.

Nopmongcol, U., Koo, B., Tai, E., Jung, J., Piyachaturawat, P., Emery, C., Yarwood, G., Pirovano, G., Mitsakou, C., and Kallos, G.: Modeling Europe with CAMx for the Air Quality Model Evaluation International Initiative (AQMEII), Atmos. Environ., 53, 177–185, 2012.

Pay, M. T., Piot, M., Jorba, O., Gassó, S., Gonçalves, M., Basart, S., Dabdub, D., Jiménez-Guerrero, P., and Baldasano, J. M.: A full year evaluation of the CALIOPE-EU air quality modeling system over Europe for 2004, Atmos. Environ., 44, 3322–3342, 2010.

Pay, M. T., Jiménez-Guerrero, P., and Baldasano, J. M.: Assessing sensitivity regimes of secondary inorganic aerosol formation in Europe with the CALIOPE-EU modeling system, Atmos. Environ., 51, 146–164, 2012.

Peng, R. D., Dominici, F., Pastor-Barriuso, R., Zeger, S. L., and Samet, J. M.: Seasonal Analyses of Air Pollution and Mortality in 100 US Cities, Am. J. Epidemiol., 161, 585–594, 2005.

Queen, A. and Zang, Y.: Examining the sensitivity of MM5–CMAQ predictions to explicit microphysics schemes and horizontal grid resolutions, Part III—The impact of horizontal grid resolution, Atmos. Environ., 42, 3869–3881, 2008.

Sartelet, K. N., Debry, E., Fahey, K., Roustan, Y., Tombette, M., and Sportisse, B.: Simulation of aerosols and gas-phase species over Europe with the POLYPHEMUS system: Part I – Model-to-data comparison for 2001, Atmos. Environ., 41, 6116–6131, 2007.

Seinfeld, J. and Pandis, S. N.: Atmospheric Chemistry and Physics; John Wiley, Hoboken, NJ, USA, 1225 pp., 2006.

Simpson, D., Yttri, K. E., Klimont, Z., Kupiainen, K., Caseiro, A., Gelencsér, A., Pio, C., Puxbaum, H., and Legrand, M.: Modeling carbonaceous aerosol over Europe: Analysis of the CARBOSOL and EMEP EC/OC campaigns, J. Geophys. Res., 112, D23S14, doi:10.1029/2006JD008158, 2007.

TNO: Description of current temporal emission patterns and sensitivity of predicted AQ for temporal emission patterns, TNO Report, 2011

Vautard, R., Moran, M. D., Solazzo, E., Gilliam, R. C., Matthias, V., Bianconi, R., Chemel, C., Ferreira, J., Geyer, B., Hansen, A. B., Jericevic, A., Prank, M., Segers, A., Silver, J. D., Werhahn, J., Wolke, R., Rao, S. T., and Galmarini, S.: Evaluation of the meteorological forcing used for the Air Quality Model Evaluation International Initiative (AQMEII) air quality simulations, Atmos. Environ. 53, 15–37, 2012.

Yarwood, G., Rao, S., Yocke, M., and Whitten, G. Z.: Updates to the Carbon Bond chemical mechanism: CB05. Final Report to the US Environmental Protection Agency, RT-0400675, 2005.

Zanis, P., Katragkou, E., Tegoulias, I., Poupkou, A., Melas, D., Huszar, P., and Giorgi, F.: Evaluation of near surface ozone in air quality simulations forced by a regional climate model over Europe for the period 1991–2000, Atmos. Environ., 45, 6489–6500, 2011.