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Air quality over Europe: modeling gaseous and particulate pollutants and the effect of precursor emissions

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Abstract

Air quality over Europe using Models-3 (i.e. CMAQ, MM5, SMOKE) modeling 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 concentrations are illustrated in southern Europe while higher NO_2 concentrations are simulated over western Europe. Elevated SO_2 concentrations are simulated over eastern Europe while elevated $PM_{2.5}$ levels are simulated over eastern and western Europe. Results suggest that NO_2 and $PM_{2.5}$ are underpredicted, SO_2 is overpredicted while $Max8hrO_3$ is overpredicted for low concentrations and is underpredicted for the higher ones. Speciated $PM_{2.5}$ components suggest that NO_3 is dominant during winter in western Europe and in a few eastern countries due to the high NO_2 concentrations. 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. The difference between observed and predicted concentrations for each country is assessed for the gaseous and particulate pollutants. The simultaneous precursor emissions change applying scaling factors on NO_x, SO_2 and $PM_{2.5}$ emissions based on the observed/predicted ratio for each country seems to statistically enhance model performance (in gaseous pollutants the improvement in root mean square is up to 5.6 ppbV, in the index of agreement is up to 0.3 and in the mean absolute error is up to 4.2 ppbV while the related values in $PM_{2.5}$ are 4.5 $\mu g\,m^{-3}$, 0.2 and 3.5 $\mu g\,m^{-3}$, respectively).

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 are applied during the last years in order to reduce atmospheric pollutant

ACPD
13, 6681–6705, 2013

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 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 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. A modeling set up for the whole Europe 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 CAMx modeling 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 modeling system found that the model overestimates 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 mentioned that it is not clear what is driving the bias, since speciated $PM_{2.5}$ data are not readily available for EU. Langmann et al. (2008) using the regional scale atmospheric climate chemistry/ aerosol model REMOTE, found that the deviation between modeled and measured organic carbon concentrations can be mainly explained by missing formation of secondary organic aerosols (SOA) and deficiencies in emission data. As such authors suggest that an updated emission inventories need to take into account the changing heating practices in Europe. The need for a more detailed treatment of

ACPD
13, 6681–6705, 2013

the formation of SOA has been also pointed out by Sartelet et al. (2007) while simulated aerosols and gas-phase species over Europe with the POLYPHEMUS system. Although they found out that hourly ozone, sulfate and ammonium simulation was good; SO_2 and nitrate concentrations tend to be overestimated. While modeling carbonaceous aerosol over Europe using EMEP modeling system Simpson et al. (2007) found the contribution of biogenic secondary organic aerosol far exceeds that of anthropogenic one. This modeling work confirms the difficulties of modeling SOA in Europe where a severe underprediction of the SOA components was found. The evaluation of the aerosol components in the CALIOPE air quality modeling 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 objective of this study is to simulate gaseous (i.e. O_3, NO_2, SO_2) and particle (i.e. $PM_{2.5}$) concentrations over Europe assessing their magnitude of disparity for each country and the effect of precursor emissions. The current analysis provides an opportunity to compare the modeling results with the results obtained by other regional air quality models commonly used in Europe and suggests possible uncertainties in precursor emissions for European countries.

2 Methods

2.1 Modeling setup

Meteorological fields are derived using the Penn State/NCAR Mesoscale Model (MM5) (Grell et al., 1994). MM5 is a limited-area, nonhydrostatic, terrain-following sigma-coordinate model designed to simulate or predict mesoscale atmospheric circulation. Since most meteorological models, such as MM5, are not built for air quality modeling purposes, to address issues related to data format translation, conversion of units of parameters, extraction of data for appropriate window domains, and reconstruction

ACPD
13, 6681–6705, 2013

of meteorological data on different grid and layer structures is needed. 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 in a 0.1 × 0.1 degrees resolution (http://www.tno.nl) in the frame 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 UK, 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) modeling 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 (TNO, 2011). However, TNO has reported that the temporal profiles are a generalization, not regularly updated and not country specific and could affect emissions over time for air quality modeling. The Biogenic Emission Inventory System, version 3 (BEIS3) is used for processing biogenic source emissions. Gridded land use data in 1 km resolution provided by USGS (http://edc2.usgs.gov/glcc/glcc.php), the default summer and winter emission factors and meteorological fields are used to create hourly model-ready biogenic emissions estimates.

The Community Multiscale Air Quality (CMAQ) v4.7 Modeling System with the Carbon Bond mechanism (CB05) is used for the regional air quality modeling (Byun et al.,

ACPD
13, 6681–6705, 2013

2006) 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 modeling domain covers almost entire Europe with 177 × 217 grid cells of 35 km × 35 km spatial resolution and 14 vertical layers (Fig. 1). Although a finer domain could affect modeling results studies found that it does not always enhance model performance (e.g. Queen et al., 2008). The default boundary and initial conditions 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 expect the eastern border. However, due to the prevailing wind direction over Europe it does not affect pollutant concentrations. Moreover, a spin up time of 10 days was used to wash out errors in the initial conditions. In the version used, several new pathways for secondary organic 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 modeling studies of ozone, particulate matter (PM), visibility, acid deposition and air toxics issues (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 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 PBL height at noon is simulated quite well. However, at 18 UTC and particularly in the summer months, the modelled PBL height is much lower than the observed. Biases of monthly means of 2 m temperature are generally small. The

ACPD
13, 6681–6705, 2013

diurnal cycle of 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 establishing 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 $< 2.5\,\mu m$ (data from 30 countries, in our domain there are 266 stations for winter and 267 for summer months) (Fig. 2). 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).

2.3 Effect of precursor emissions

Emissions of air pollutants originated from a variety of small and large individual sources (e.g. power plants, industries, motor vehicles) varying temporally and spatially. Therefore, emission inventories are subject to significant uncertainties given that

ACPD
13, 6681–6705, 2013

they are based on data sets of limited spatiotemporal coverage while countries do not always estimate emissions in a uniform and transparent manner. Assessing such uncertainties is an essential step towards the better computation of air pollutants concentrations. In an effort to do so, here, we assess the effect of precursor emissions on air quality applying scaling factors for January and July, 2006 on NO_x, SO_2 and $PM_{2.5}$ emissions based on the ratio $\frac{\text{Observed}_{\text{average}}}{\text{Predicted}_{\text{average}}}$ for each country. This is not just a sensitivity analysis assessing the effect of each precursor separately on air pollution since we examine the effect of the simultaneous emission change for multiple precursors on air quality. The selection of the above precursor emissions is due to the significant number of monitoring data throughout the modeling domain. Although other precursor emissions (e.g. VOCs, NH_3) affect air pollutant concentrations the limited number of their monitoring data throughout the modeling domain does not allow us to include them in the presented analysis.

3 Results and discussion

3.1 Air quality

High ozone concentrations are illustrated in southern Europe where meteorological conditions enhance ozone production (Fig. 3). The daily average maximum 8 h ozone ($Max8hrO_3$) concentration during July is simulated up to 75 ppbV while a big part of the domain faces concentrations higher than 50 ppbV. Higher NO_2 concentrations are simulated over western Europe (i.e. Belgium, the Netherlands, Germany and northern France), northern Italy and UK for both seasons. Belgium and the Netherlands have elevated NO_2 concentrations 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 at northertn Italy while road and non-road transport energy sector and industry are responsible for the high NO_x emission in the UK. NO_2 concentrations are higher during

ACPD
13, 6681–6705, 2013

January compared to July for two reasons: energy sector and industry emit more NO_x during winter and NO_2 photolysis is unfavorable during winter. Elevated SO_2 concentrations are simulated over eastern Europe with higher concentrations over Poland and the North Balkan Peninsula. Since power generation and industry are mainly responsible for SO_2 emissions, SO_2 concentrations are very locally depended showing higher values during winter. Elevated $PM_{2.5}$ levels are simulated over eastern and western Europe (i.e. up to 30 $\mu g\,m^{-3}$ during winter). NO_3 is dominant during winter in western Europe and in a few eastern countries due to the high NO_2 concentrations (Fig. 4). 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, 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.

Model performance for ozone shows a mixed trend: $Max8hrO_3$ is overpredicted for low concentrations (about 50 ppbV) while it is underpredicted for the higher ones. 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 overprediction tendency for the lower concentrations gives a much higher mean concentration during winter (Table 2) which is diminished during summer where higher ozone values are simulated. Both observed and predicted ozone concentrations are similarly spread out around mean values during winter

ACPD
13, 6681–6705, 2013

(similar simulated standard and mean absolute deviations). During summer simulated concentrations are closer to the mean simulated concentration compared to observation data; this is related to the overprediction of the lower concentrations. 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) model overestimates ozone concentrations in all regions during winter while during summer model overestimates ozone concentrations in northern and southern Europe and underestimates them in eastern and western Europe where higher concentrations 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 consistently underpredicted while SO_2 is consistently overpredicted for both seasons. The same biases have been also noticed by another previous study (Pay et al., 2010). The consistent underprediction trend for $PM_{2.5}$ has been found also by Appel et al. (2012) using CMAQ modeling system for Europe. At regional scale model underestimates NO_2 concentrations more in southern Europe and overestimates SO_2 concentrations more in eastern Europe where higher underestimation in $PM_{2.5}$ concentrations is noted.

3.2 Effect of precursor emissions on air quality

The difference between observed and predicted concentrations for each country based on the ratio $\frac{\text{Observed}_{\text{average}}}{\text{Predicted}_{\text{average}}}$ is presented in Table 3. This ratio for $\text{Max8hr}O_3$ is less than 1.0 during January, 2006 in all countries due to the underprediction of low ozone concentration observed in winter. During July, 2006 the average observed concentrations are closer to the average predicted ones for all countries (this ratio is 1.0 ± 0.1 for the majority of the countries). Beside the general overprediction trend for SO_2 concentrations regionally over Europe, the ratio is greater than 1.0 in few countries for both seasons denoting that the average observed concentrations are higher than the average predicted ones for those countries. The ratio for NO_2 and $PM_{2.5}$ concentrations is almost in all countries greater than 1.0. There are numerous reasons why a bias may exist. This

ACPD
13, 6681–6705, 2013

could be related to inaccuracies in emission inventories; a discrepancy with the meteorological data and the source locations; topographic effects that are not accounted for in the model; or the model itself may have built in biases. To explore this, here, we examine modeling results using modified NO_x, SO_2 and $PM_{2.5}$ emissions based on the ratio $\frac{\text{Observed}_{\text{average}}}{\text{Predicted}_{\text{average}}}$ for each country.

The modified emissions improve model's performance for all examined pollutants (Table 2). Better closure is noticed for SO_2 predictions. Mean simulated SO_2 concentrations using scaled emissions is similar to the mean observed concentrations while both simulated and observed values are similarly spread out around means for both seasons (i.e. similar standard deviation and mean absolutely values). Moreover all statistical parameters examined here (i.e. root mean square, index of agreement and mean absolute error) are improved suggesting better model performance using scaled emissions. Statistical analysis suggests that while NO_2 concentrations are better simulated using scaled emissions improvement is minor compared to SO_2 performance. This is attributed to the fact that scaling factors are not applied directly on NO_2 but on NO_x. As such NO_2 to NO_x ratio probably includes additional uncertainties that need to be investigated in the future. Standard and mean absolute deviations suggest that simulated and observed values are similarly spread out around means for both seasons using scaled emissions. An improved performance is also noted for $PM_{2.5}$ for both seasons. Although higher $PM_{2.5}$ concentrations are simulated using scaled emissions and values are spread out far away of the mean value compared to the simulations with original emissions (higher standard deviation and mean absolute error) model still underpredicts $PM_{2.5}$ concentrations. This is probably related to the uncertain representation of secondary organic aerosol formation (e.g. Chen et al., 2005; Kroll et al., 2006). Better performance is also noted for $Max8hrO_3$ concentrations mainly during winter; however, the model seems to overpredict $Max8hrO_3$ concentrations.

The modified emissions used here although improve model's performance for all examined pollutants could not actually address the real issues in the emissions as the simplified method used takes into account chemistry and transport (e.g. small countries

ACPD
13, 6681–6705, 2013

is affected by transported pollutants from neighbor countries). However, this part of our work could suggest possible uncertainties in precursor emissions for European countries although a more robust method to improve the emissions (e.g. inverse modeling on the precursor emissions) is needed for real improvements in the temporal and/or spatial allocation of the emissions. Assessing (i) the effect of other precursor emissions, (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 role of other sources of uncertainty.

4 Conclusions

Application of CMAQ modeling system over Europe for January and July, 2006 using the TNO gridded anthropogenic emissions database for the year 2006 shows an overprediction trend for low ozone concentrations (less than 50 ppbV) while it is underpredicted for the higher ones, although spatial distributions are reasonably estimated (e.g. higher ozone concentrations in southern Europe). Simulated concentrations for NO_2, SO_2 and $PM_{2.5}$ suggest a consistent bias: SO_2 is overpredicted while NO_2 and $PM_{2.5}$ are underpredicted. Speciated $PM_{2.5}$ components give low OC concentrations as a result of the uncertain representation of SOA formation. Assessing the difference between observed and predicted concentrations for each country and scaling the emissions based on that seems to statistically enhance model performance (i.e. root mean square, index of agreement and mean absolute error are improved). Although a number of reasons could affect model performance (e.g. meteorological data, topographic effects or the model itself), results from the current study could suggest possible uncertainties in precursor emissions for the European countries.

ACPD
13, 6681–6705, 2013

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ACPD
13, 6681–6705, 2013

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ACPD
13, 6681–6705, 2013

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ACPD
13, 6681–6705, 2013

Table 1. 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
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	380.1	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
UK	2256	2127	309	926	1489	150	94	608

ACPD
13, 6681–6705, 2013

Table 2. Statistical analysis for hourly average NO_2 and SO_2 concentrations and daily average $Max8hrO_3$ and $PM_{2.5}$ concentrations over Europe.

		$Max8hrO_3$ (ppbV)					NO_2 (ppbV)				
		January 2006									
		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
	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
	Scaled data	24.5 ± 14.2	25.4 ± 13.7	31.4 ± 13.7	20.0 ± 11.8	20.8 ± 13.7	22.9 ± 14.3	20.5 ± 13.5	21.6 ± 16.9	22.5 ± 11.6	24.4 ± 12.8
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
	Predicted concentrations	9.1	8.9	7.8	8.0	8.3	9.3	10.5	9.7	8.2	8.9
	Scaled data	12.4	11.6	11.4	9.8	11.8	12.2	11.6	14.7	9.9	10.9
Root mean squear error (RMSE)	Predicted concentrations	18.1	14.0	20.1	11.7	13.1	16.3	13.6	18.2	15.4	15.4
	Scaled data	14.1	9.9	17.1	11.3	13.0	15.1	13.0	17.3	14.8	15.0
Index of agreement (IoA)	Predicted concentrations	0.5	0.5	0.7	0.6	0.5	0.6	0.6	0.6	0.5	0.6
	Scaled data	0.7	0.8	0.7	0.7	0.7	0.7	0.7	0.7	0.6	0.7
Mean absolute error (MAE)	Predicted concentrations	15.3	11.0	15.9	11.5	14.9	11.6	9.4	13.2	10.4	11.1
	Scaled data	11.1	7.9	11.4	8.8	10.0	10.4	9.0	12.1	9.5	10.6

$$\text{RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(O_{(i)} - P_{(i)})^2}; \quad \text{MAE} = \frac{1}{n}\sum_{i=1}^{n}|P_{(i)} - O_{(i)}|; \quad \text{IoA} = \left|\frac{\sum_{i=1}^{n}(P_{(i)} - O_{(i)})^2}{\sum_{i=1}^{n}(|P_{(i)} - \overline{O}| + |O_{(i)} - \overline{O}|)^2}\right|; \quad \text{MAD} = \frac{1}{n}\sum_{i=1}^{n}|X_{(i)} - \overline{X}|.$$

ACPD
13, 6681–6705, 2013

Table 2. Continued.

		Max8hrO$_3$ (ppbV) July 2006					NO$_2$ (ppbV)				
		Europe (Total)	North	South	East	West	Europe (Total)	North	South	East	West
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
	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
	Scaled data	57.9 ± 10.7	47.8 ± 9.1	59.5 ± 10.6	56.8 ± 8.8	57.7 ± 10.2	11.4 ± 12.2	12.3 ± 12.7	11.5 ± 13.1	8.7 ± 11.0	11.8 ± 12.5
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
	Predicted concentrations	6.5	6.0	6.8	5.3	5.9	5.3	6.4	5.6	5.0	5.1
	Scaled data	8.6	7.1	8.3	7.1	8.2	9.4	9.6	9.9	7.8	9.3
Root mean squear error (RMSE)	Predicted concentrations	14.6	13.2	18.7	11.9	12.2	14.0	12.7	14.9	11.2	14.2
	Scaled data	14.0	14.0	20.1	11.7	10.4	12.8	11.8	13.1	10.4	13.0
Index of agreement (IoA)	Predicted concentrations	0.6	0.7	0.5	0.7	0.7	0.5	0.6	0.5	0.6	0.5
	Scaled data	0.7	0.7	0.6	0.7	0.8	0.6	0.7	0.6	0.6	0.6
Mean absolute error (MAE)	Predicted concentrations	11.2	11.0	13.9	9.5	9.9	9.0	8.1	10.0	7.0	9.0
	Scaled data	10.8	11.7	15.4	9.0	8.2	7.8	7.3	9.0	6.4	8.2

$$\mathrm{RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(O_{(i)} - P_{(i)})^2}; \quad \mathrm{MAE} = \frac{1}{n}\sum_{i=1}^{n}|P_{(i)} - O_{(i)}|; \quad \mathrm{IoA} = \left|\frac{\sum_{i=1}^{n}(P_{(i)} - O_{(i)})^2}{\sum_{i=1}^{n}(|P_{(i)} - \overline{O}| + |O_{(i)} - \overline{O}|)^2}\right|; \quad \mathrm{MAD} = \frac{1}{n}\sum_{i=1}^{n}|X_{(i)} - \overline{X}|.$$

ACPD
13, 6681–6705, 2013

Table 2. Continued.

		SO_2 (ppbV) January 2006					$PM_{2.5}$ ($\mu g\,m^{-3}$)				
		Europe (Total)	North	South	East	West	Europe (Total)	North	South	East	West
Mean ± standard deviation	Observed concentrations	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	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
	Scaled data	4.4 ± 9.7	2.3 ± 3.9	3.7 ± 9.5	9.8 ± 17.5	3.4 ± 4.9	20.5 ± 16.5	13.1 ± 10.3	14.9 ± 17.0	26.8 ± 16.2	25.1 ± 15.4
Mean absolute deviation (MAD)	Observed concentrations	3.7	1.6	3.0	7.4	2.4	16.1	6.9	14.9	18.0	14.5
	Predicted concentrations	6.9	3.6	7.3	13.4	4.3	8.5	4.9	5.3	6.1	10.0
	Scaled data	4.5	2.0	4.0	9.7	3.0	12.2	7.0	10.9	11.9	11.3
Root mean squear error (RMSE)	Predicted concentrations	15.7	7.5	18.2	24.9	7.9	24.5	10.4	22.6	36.6	23.7
	Scaled data	10.1	4.6	10.1	17.5	6.2	20.0	10.5	17.9	27.8	20.6
Index of agreement (IoA)	Predicted concentrations	0.3	0.2	0.3	0.4	0.4	0.5	0.6	0.5	0.5	0.4
	Scaled data	0.5	0.5	0.5	0.6	0.5	0.7	0.6	0.8	0.6	0.7
Mean absolute error (MAE)	Predicted concentrations	5.9	3.3	6.4	11.1	4.0	17.0	6.8	15.4	30.5	16.7
	Scaled data	4.3	2.2	4.1	9.0	3.1	13.5	6.3	12.1	21.3	14.3

$$\mathrm{RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(O_{(i)} - P_{(i)})^2}; \quad \mathrm{MAE} = \frac{1}{n}\sum_{i=1}^{n}|P_{(i)} - O_{(i)}|; \quad \mathrm{IoA} = \left|\frac{\sum_{i=1}^{n}(P_{(i)} - O_{(i)})^2}{\sum_{i=1}^{n}(|P_{(i)} - \overline{O}| + |O_{(i)} - \overline{O}|)^2}\right|; \quad \mathrm{MAD} = \frac{1}{n}\sum_{i=1}^{n}|X_{(i)} - \overline{X}|.$$

ACPD
13, 6681–6705, 2013

Table 2. Continued.

		SO_2 (ppbV) July 2006					$PM_{2.5}$ ($\mu g\,m^{-3}$)				
		Europe (Total)	North	South	East	West	Europe (Total)	North	South	East	West
Mean ± standard deviation	Observed concentrations	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	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
	Scaled data	2.4 ± 4.8	2.1 ± 3.3	2.8 ± 5.8	2.3 ± 4.8	2.0 ± 4.0	9.6 ± 6.3	7.7 ± 5.1	8.7 ± 6.0	9.6 ± 5.4	11.0 ± 6.9
Mean absolute deviation (MAD)	Observed concentrations	1.9	1.5	2.2	1.7	1.6	5.9	5.7	6.4	7.4	4.5
	Predicted concentrations	4.8	2.9	4.9	7.9	3.6	2.9	2.4	2.1	2.3	4.1
	Scaled data	2.4	1.9	2.9	2.5	2.2	4.6	3.8	4.3	4.1	5.2
Root mean squear error (RMSE)	Predicted concentrations	11.2	5.9	12.0	17.4	7.3	12.1	9.0	13.4	15.4	10.1
	Scaled data	6.5	4.2	7.9	6.0	5.3	10.2	7.5	11.6	12.7	8.7
Index of agreement (IoA)	Predicted concentrations	0.3	0.3	0.3	0.2	0.3	0.4	0.5	0.5	0.5	0.4
	Scaled data	0.6	0.5	0.5	0.6	0.6	0.6	0.7	0.6	0.6	0.5
Mean absolute error (MAE)	Predicted concentrations	4.2	2.6	4.6	6.4	3.1	10.1	7.2	11.4	13.3	8.6
	Scaled data	2.6	2.0	3.2	2.7	2.2	8.1	5.5	9.6	10.3	7.1

$$\mathrm{RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(O_{(i)} - P_{(i)})^2}; \quad \mathrm{MAE} = \frac{1}{n}\sum_{i=1}^{n}|P_{(i)} - O_{(i)}|; \quad \mathrm{IoA} = \left[\frac{\sum_{i=1}^{n}(P_{(i)} - O_{(i)})^2}{\sum_{i=1}^{n}(|P_{(i)} - \overline{O}| + |O_{(i)} - \overline{O}|)^2}\right]; \quad \mathrm{MAD} = \frac{1}{n}\sum_{i=1}^{n}|X_{(i)} - \overline{X}|.$$

ACPD
13, 6681–6705, 2013

Table 3. Observed$_{avergae}$/Predicted$_{average}$ concentrations for the European countries.

Country	Max8hrO$_3$	NO$_2$	SO$_2$	PM$_{2.5}$	Max8hrO$_3$	NO$_2$	SO$_2$	PM$_{2.5}$
	January 2006				July 2006			
Austria	0.7	2.1	2.2	4.1	1.1	2.2	1.3	3.2
Belgium	0.6	0.8	0.5	1.1	1.1	0.9	0.3	1.2
Bosn. & Herz.	0.5	1.9	0.7	2.7	0.9	1.7	0.3	1.7
Bulgaria	0.5	2.0	0.5	2.7	0.9	2.2	0.3	2.6
Croatia	–	2.1	2.3	–	1.0	3.4	2.2	–
Czech Rep.	0.8	1.4	1.1	3.4	1.1	1.4	0.4	3.3
Denmark	0.6	1.2	0.4	1.5	0.9	1.5	0.3	2.5
Estonia	0.8	1.7	1.9	–	0.8	1.9	0.6	–
Finland	0.8	1.8	0.7	1.1	0.8	2.0	1.1	2.1
France	0.5	1.6	0.5	0.9	1.1	2.1	0.6	1.7
F. Y. R. O. M	0.7	2.8	1.1	–	1.1	2.9	1.1	–
Germany	0.6	1.3	0.8	2.2	1.1	2.3	0.5	3.0
Greece	0.6	1.7	0.9	–	0.9	2.8	0.5	–
Hungary	0.6	1.5	0.6	2.6	1.1	2.5	0.3	3.1
Ireland	0.7	1.9	1.5	–	0.8	1.4	1.5	–
Italy	0.5	2.1	0.6	4.8	1.1	2.9	0.6	3.1
Latvia	0.7	1.6	3.0	–	0.6	1.7	1.8	–
Lithuania	0.7	2.4	0.7	–	0.9	3.4	0.6	–
Luxembourg	0.5	1.4	1.1	1.1	1.1	2.8	1.4	1.1
Malta	0.6	4.7	1.3	–	0.8	1.5	0.5	2.9
Netherlands	0.6	0.9	0.6	–	1.0	1.4	0.5	–
Poland	0.8	1.4	0.8	2.2	1.1	1.6	0.3	2.2
Portugal	0.6	1.3	0.4	1.9	0.8	1.6	0.5	2.2
Romania	0.8	1.6	0.4	1.7	0.8	2.1	0.3	3.3
Serbia	0.5	1.4	2.1	–	–	3.3	1.1	–
Slovakia	0.6	1.9	0.8	5.4	1.1	2.5	0.7	3.7
Sweden	0.7	1.7	1.1	1.6	0.9	1.6	0.5	2.5
Switzerland	0.5	2.2	1.9	3.4	1.2	3.3	1.8	3.5
Spain	0.5	1.1	0.4	1.9	0.8	1.3	0.5	3.3
UK	0.7	0.9	0.5	1.1	0.9	1.3	0.6	2.1

ACPD
13, 6681–6705, 2013

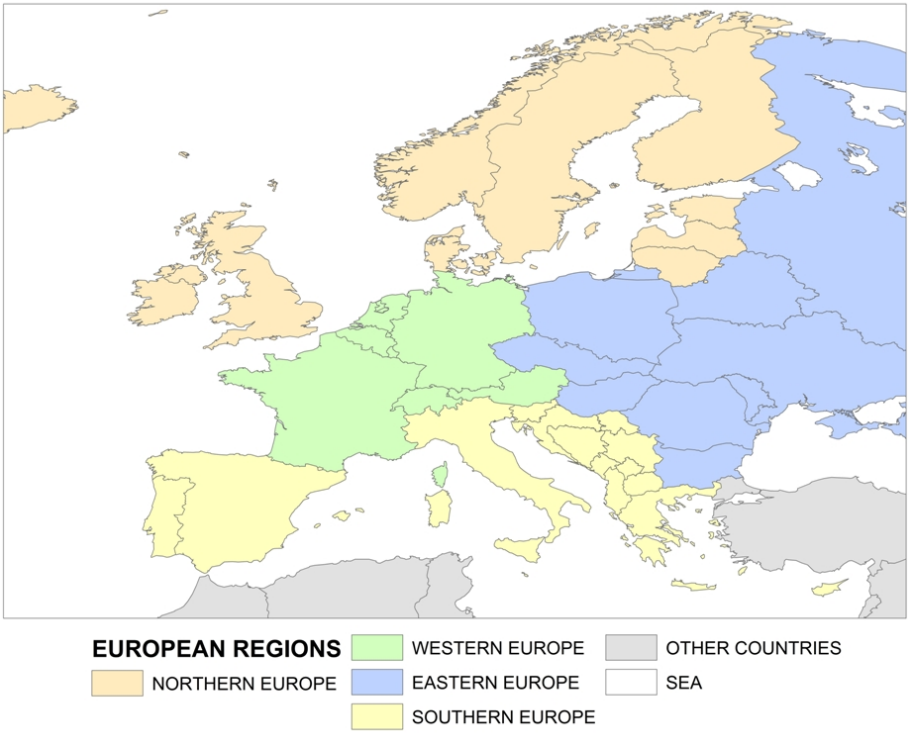
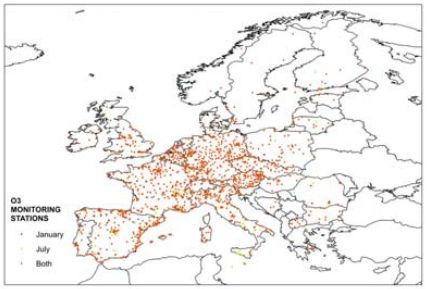
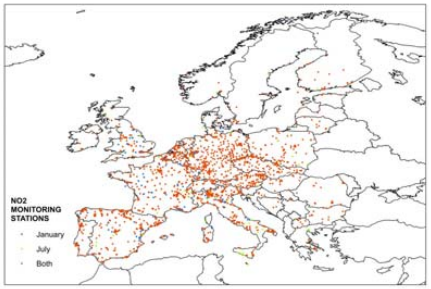


Fig. 1. Modeling domain and the regional European grouping used by the United Nations Statistics Department.

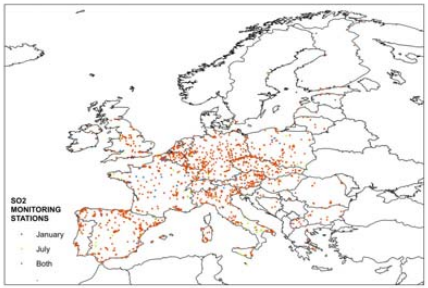
ACPD
13, 6681–6705, 2013



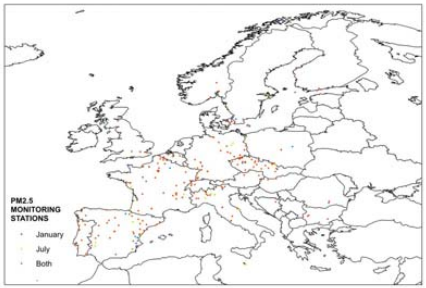
O_3 monitoring stations



NO_2 monitoring stations



SO_2 monitoring stations



$PM_{2.5}$ monitoring stations

Fig. 2. Location of O_3, NO_2, SO_2 and $PM_{2.5}$ monitoring stations (red color for monitoring stations active in both months, blue color for monitoring stations active only in January, green color for monitoring stations active only in July).

ACPD
13, 6681–6705, 2013

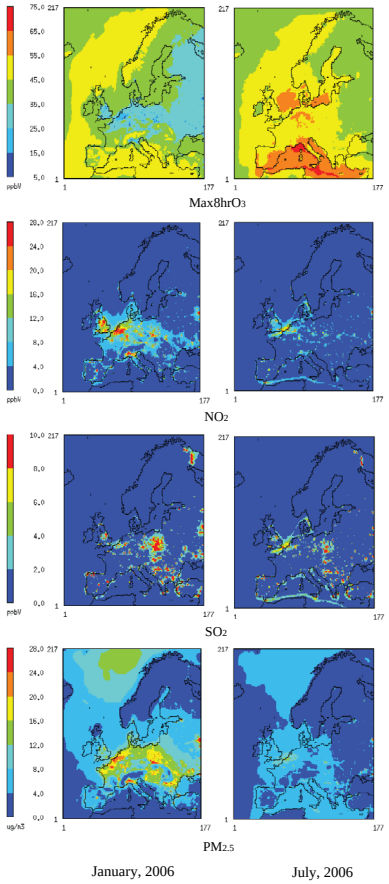


Fig. 3. Simulated daily (Max8hrO_3, $PM_{2.5}$) and hourly (NO_2, SO_2) average concentrations for January (left column) and July (right column) 2006.

ACPD
13, 6681–6705, 2013

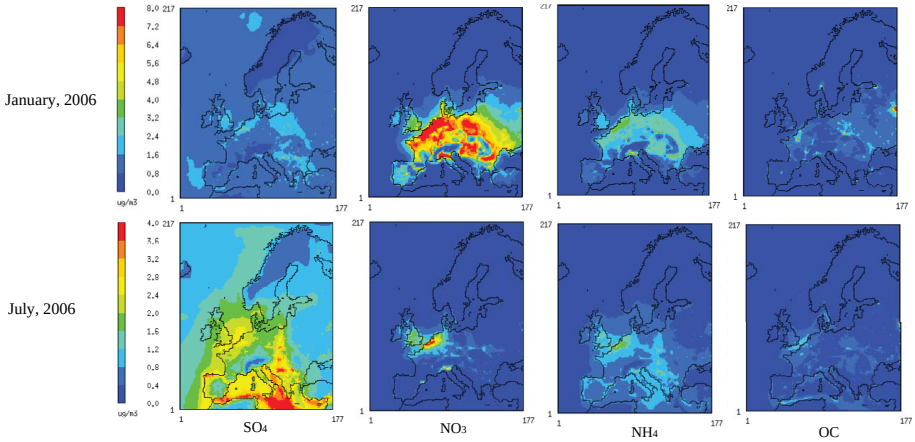


Fig. 4. Simulated $PM_{2.5}$ component daily average concentrations for January and July, 2006.

ACPD
13, 6681–6705, 2013