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Modeling Europe with CAMx for Phase I of the Air Quality Model Evaluation International Initiative (AQMEII)

Final Report

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Foreword

Under CRC Project A-75-1 "Modeling Europe with CAMx for Phase I of the Air Quality Model Evaluation International Initiative (AQMEII)," ENVIRON International Corporation collaborated with researchers from Europe (from INERIS in France and the University of Athens in Greece) to model ozone and particulate matter (PM) for Europe in 2006. The modeling was performed as part of the AQMEII study organized by the European Commission's Joint Research Centre (JRC) and the United States Environmental Protection Agency (EPA) to promote research and collaboration between scientists in Europe and North America on regional air quality model evaluation. The results from A-75-1 are presented in the format of a draft journal manuscript. This manuscript will be expanded under CRC Project A-75-2 by including results of sensitivity tests to model inputs and assumptions and analysis of ozone sensitivity to precursor emissions. The expanded manuscript will be submitted to a special issue of Atmospheric Environment devoted to the AQMEII study.

The draft manuscript is included in this report along with an Executive Summary and supporting materials for the journal article.

Draft Final Report

CRC PROJECT A-75-1

Modeling Europe with CAMx for Phase I of the Air Quality Model Evaluation International Initiative (AQMEII)

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Executive Summary

The Comprehensive Air Quality Model with Extensions (CAMx) photochemical grid model was used to model ozone (O_3) and particulate matter (PM) for most of Europe in the framework of the Air Quality Model Evaluation International Initiative (AQMEII) phase 1. The AQMEII study is organized by the European Commission's Joint Research Centre (JRC) and the United States Environmental Protection Agency (EPA). The aims are to promote research and collaboration between scientists in Europe and North America on regional air quality model evaluation. AQMEII provided input data for 2006 emissions, meteorology and boundary conditions (BCs) and requested that participants simulate one year of air quality for Europe and/or North America using AQMEII input data where possible. The AQMEII project is evaluating the resulting ensembles of model results for each continent. For this purpose, ENVIRON interpolated the CAMx results onto a specified analysis grid and transferred them to JRC for statistical and graphical evaluation using a software system called ENSEMBLE The objective is to provide consistent analysis methods and enable evaluation of the ensemble predictions of all models. ENVIRON also performed an independent evaluation of the CAMx modeling results.

This report for CRC project A 75-1 discusses ENVIRON's application of CAMx to Europe using the input data provided by AQMEII. A future report for project CRC A 75-2 will discuss model sensitivity analyses including use of alternate input data and assumptions. Model performance for January and July 2006 exhibited under-estimation trends for all pollutants both in winter and summer, except for SO_2. SO_2 generally had little bias although some over-estimation occurred at coastal locations and this was attributed to incorrect vertical distribution of emissions from marine vessels. Performance for NOx and NO_2 was better in winter than summer. For O_3, the spatial distribution was modelled well in summer but with some tendency to underestimate daily maximum 8-hour O_3. The tendency to under-predict daytime NOx and O_3 in summer may result from insufficient NOx emissions or overstated daytime dilution (e.g., too deep planetary boundary layer). Winter $O_3$was biased low and this was attributed to a low-bias in the O_3 boundary conditions. PM_{10} was widely under-predicted in both winter and summer. The poor PM_{10} was influenced by under-estimation of coarse PM emissions. It is clear from these results that the underlying emission inventory and metrological input data play a crucial role in the air quality model performance. Model performance was better in some parts of Europe and the eastern European countries may have higher uncertainty of emissions and in the Po Valley of northern Italy the meteorological simulation at 35 km resolution may not have captured the

strong influence of topography on pollutant concentration within the Po Valley which is bounded to the north by the Alps.

It is worth noting that observed concentrations may sometimes exhibit a "country bias," meaning that the behaviour of the stations is quite homogeneous inside each country, but rather different country by country. This is particularly true for PM observations and to a lesser extent also for SO_2, NO_2 and CO. This behaviour could descend from differences in anthropogenic activities within each country (e.g. different fuels, different abatement technologies), but it could be also related to different measurement methodologies.

The AQMEII approach to applying many models was to promote use of consistent data sources (e.g., emissions, BCs) and minimize uncertainties associated with use of differing inputs by each model. However, most models are using different meteorological data, several are using different BCs and a few are using different emissions. AQMEII is evaluating the ensemble of predictions from all models applied for Europe and may not be able to untangle the consequences of differing input data and assumptions. In follow-on work under CRC Project A75-2, ENVIRON is investigating the influence of input data, assumptions and uncertainties on model performance using the CAMx application described here for the EU domain. Multiple simulations will be conducted first to identify the role played by different input data and some of the uncertainties discussed here. For example, alternate BCs will be used and the impact on winter O_3 evaluated; ship emissions will be divided between the first two model layers; alternate meteorological input data will be utilized. Several revised model configurations will be developed for model performance evaluation and the sensitivity of O_3 to precursor emissions (anthropogenic NOx and VOC, biogenic VOC) will be evaluated.

Draft Journal Manuscript

Modeling Europe with CAMx for Phase I of the Air Quality Model Evaluation International Initiative (AQMEII)

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1. Introduction

The Comprehensive Air Quality Model with Extensions (CAMx) photochemical grid model (ENVIRON, 2010) was used to model ozone (O_3) and particulate matter (PM) for most of Europe in the framework of the Air Quality Model Evaluation International Initiative (AQMEII) phase 1. The AQMEII study is organized by the European Commission's Joint Research Centre (JRC) and the United States Environmental Protection Agency (EPA). The aims are to promote research and collaboration between scientists in Europe and North America on regional air quality model evaluation. Participants in phase I simulated one year of air quality in Europe and/or North America and submitted results in a standardized format for shared analysis and interpretation. The AQMEII approach to model performance evaluation is to interpolate all results from all models to a common grid where they can be evaluated using a software system called ENSEMBLE. The objective is to provide consistent analysis methods and enable evaluation of the ensemble predictions of all models. To promote consistent model applications the AQMEII organizers made available key model input data such as emissions, boundary conditions and meteorology. This report for CRC project A 75-1 discusses ENVIRON's application of CAMx to Europe using the input data provided by AQMEII. A future report for project CRC A 75-2 will discuss model sensitivity analyses including use of alternate input data.

2. Methodology

2.1. CAMx Photochemical Grid Model

Air quality modeling for the European (EU) domain and calendar year 2006 used CAMx version 5.21 to simulate physical and chemical processes governing the formation and transport of ozone and PM (ENVIRON, 2010) with Carbon Bond 05 (CB05) gas phase chemistry (Yarwood et al., 2005). Model inputs were prepared from data provided by AQMEII supplemented by other data sources as described below. The CAMx modeling domain was defined in latitude and longitude with 207 by 287 grid cells and 23 vertical layers. The modeling domain covered most of Europe, from 15.875°W to 35.875 °E

and 34.5625°N to 70.4375°N, with a grid resolution of 0.125° latitude by 0.25° longitude (equivalent to about 15 to 20 km). The grid resolution of the CAMx domain was aligned to the emission inventory in order to avoid spatial interpolation of gridded emissions data. The extent of the CAMx domain encompasses the common grid for analysis of model results, from 15°W to 35°E and 35°N to 70°N at 0.25° resolution.

2.2. Meteorology

Meteorological data for calendar year 2006 were developed for AQMEII using the MM5 model (Duhdia, 1993) with 35 km resolution by the Laboratoire des Sciences du Climat et de l'Environnement (CEA) in Paris, France (Vautard, 2010). The MM5 domain was defined in Mercator projection with 180 by 220 grid cells and 32 vertical layers with a 30 meter deep surface layer. The MM5CAMx preprocessor for CAMx was modified to interpolate from the Mercator projection employed by MM5 to the more finely resolved latitude-longitude coordinate system used by CAMx. CAMx employed fewer vertical layers (23) than MM5 (32) to reduce the computational burden of the air quality simulations. The CAMx vertical layers exactly matched those used in MM5 for the lowest 14 layers (up to ~1,800 m) and above this altitude were aggregates of several MM5 layers.

2.3. Emission inventory

2.2.1 Anthropogenic Emissions

Anthropogenic emissions for 2006 were developed by TNO Environment and Geosciences (Denier van der Gon, 2010). The data consisted of annual average emissions for 10 SNAP (Selected Nomenclature for sources of Air Pollution) sectors (Visschedijk et al., 2007) on a 1/16 by 1/8 degree latitude-longitude grid. Major point sources were gridded, which combined sources of the same SNAP sector in each grid cell, and plume rise was accounted using layer-fractions which were constant spatially and temporally for each SNAP sector. Chemical constituents included methane (CH_4), carbon monoxide (CO), nitrogen oxides (NOx), sulfur oxides (SOx), non-methane volatile organic compounds (NMVOC), ammonia (NH_3) and particulate matter of 10 and 2.5 micrometers or less (PM_{10} and $PM_{2.5}$).

The Emissions Processing System version 3 (EPS3) was used to prepare emissions data for input to CAMx using temporal allocation and vertical layer distribution profiles provided by TNO for each SNAP sector. Speciation profiles for NMVOC to the CB05 chemical mechanism (Yarwood et al., 2005) were developed based on data from Passant (2002). TNO provided PM speciation profiles to allocate PM10 to sulfate (PSO4), elemental carbon (EC), primary organic carbon (POC), Sodium (Na),

other PM fine, and other PM coarse. CAMx models the total mass of organic aerosol (i.e., POA for primary organic aerosols) rather than carbon mass (i.e., POC) and factors of 1.45-1.8 were applied to the POC mass to calculate POA and subtracting the mass difference from "other PM fine" to conserve total PM mass.

The 2006 anthropogenic emissions for the CAMx modeling domain are summarized by SNAP sector in Table 1 and by country or sea area in Table S1. NOx emissions are primarily from on-road and off-road mobile sources (63%) which includes marine vessels. The largest contributor to SO_2 emissions (56%) is the power generation sector. Solvent use contributes 37% of NMVOC emissions on-road mobile sources (22%). Agricultural sources dominate NH_3 emissions (93%). Emissions in sea areas are dominated by commercial shipping.

2.2.2 Biogenic Emissions

Biogenic emissions depend strongly on meteorology and landcover and were estimated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN; Guenther, et al., 2006; Sakulyanontvittaya, et al., 2008) at each hour for each grid cell. MEGAN has a global database of landcover derived from satellite data at 1 km resolution. Meteorological input data for MEGAN were taken from the MM5 predictions. MEGAN estimates emissions of isoprene, methylbutenol, terpenes, sesquiterpenes, other VOCs (OVOCs) and soil NOx.

2.2.3 Biomass Burning Emissions

Biomass burning emissions were estimated by the Finnish Meteorological Institute (FMI; Sofief, 2010) using the fire radiative power (FRP) data product from MODIS equipped satellites. The dataset consisted of daily PM emissions for each fire gridded at 0.1° resolution. Scaling factors were provided to calculate gaseous components (CO, HCHO, NOx, NH_3, and SO_2) as ratios to PM. FMI suggested distributing emissions vertically by placing 50% of emissions below 200 m and 50% between 200 m and 1 km (Sofiev, 2010) but US modeling studies have used higher plume rise (ASI, 2005). Plume rise is related to the spatial extent of fires, and other factors, which are likely to differ for the conditions analyzed by FMI and the US studies. For this study, fire plume rise was modeled by analyzing the emission inventory data to categorize the area burned by each fire and then using plume rise equations specific for fires of differing spatial extent (ASI, 2005).

2.4.4 Sea-salt Emissions

Emissions of sea-salt particles, including sodium, chloride, and sulfate (SO_4), were estimated from the MM5 hourly, gridded meteorology using flux equations for open ocean (Smith and Harrison 1998; Gong, 2003) and breaking waves in the surf zone (de Leeuw et al., 2000).

Average daily emissions in January and July 2006 for each source category are summarized in Table S2.

2.5. Boundary/Initial Conditions (BCs/ICs)

Boundary conditions (BCs) for CAMx were from data provided by the the European Centre for Medium-Range Weather Forecasts (ECMWF) GEMS project (http://gems.ecmwf.int). The GEMS data were a composite of two models, namely MOZART for gases and IFS for particles. EPA evaluated the GEMS BCs by comparison with climatological values and GEOS-Chem model results for North America (Schere, 2010) and concluded generally that differences between the three data sources were within the uncertainty ranges. However, EPA recommended not using sea-salt from GEMS because concentrations were high. The SO_2 and SO_4 data from GEMs also were not recommended as they were based on simple assumptions for emissions and removal rather than a complete atmospheric transformation mechanism. Neglecting sulfur from the boundaries should not greatly affect the simulations, since SO_2/SO_4 should be strongly forced by emissions within the domain. The GEMS data did not provide PM nitrate or ammonium. In this study, BCs were extracted from GEMS data and formatted for CAMx. Background concentrations were assumed for nitrate, ammonium, sulfate and other aerosol species missing from the GEMS data. The 2006 annual simulation was initialized on December 17, 2005, to limit the influence of the ICs on results for 2006.

3. Performance Evaluation

The AQMEII project is evaluating the ensemble of model results for Europe as submitted by project participants. For this purpose, ENVIRON and other AQMEII participants interpolated their model results onto a common grid and transferred them to JRC for statistical and graphical evaluation using a software system called ENSEMBLE (Galmarini et al., 2004). The objective is to provide consistent analysis methods and enable evaluation of the ensemble predictions of all models. However, the ENSEMBLE system is being extended to meet the needs of the AQMEII project and only limited performance evaluation results were available for use in this report. Therefore, this report uses

performance evaluation results prepared using the Atmospheric Model Evaluation Tool (AMET; Appel et al., 2010).

Ambient air quality measurements from the AirBase database for Europe (EEA, 2010) were used with AMET to compute statistical metrics of model performance. Background monitors (i.e., reported as being removed from traffic and industrial sources) below 700 m elevation and with data availability exceeding 75% were included in this analysis (~ 1,400 sites). The AirBase system classifies monitors according to location type with most of the selected stations classified as urban background, 379 as suburban background and 360 as rural background. Statistical metrics for PM constituents were computed using data from the European Monitoring and Evaluation Program (EMEP) database (EMEP, 2010). Monthly normalized mean bias (NMB), normalized mean error (NME), fractional bias (FB) and fractional error (FE) statistics (Table 2) were calculated for January and July using paired predictions and observations. Concentration thresholds were applied to the observed data (i.e., NOx $\geq$ 0.5 ppb, $NO_2 \geq$ 0.5 ppb, $O_3 \geq$ 5 ppb, $SO_2 \geq$ 0.2 ppb, CO $\geq$ 10 ppb, $PM_{10} \geq$ 1.0 µg/m3) to focus on conditions that exceed measurement thresholds. Table 3 reports the statistical performance metrics at the selected AirBase stations for January and July 2006. The spatial distributions of monthly average concentration at individual monitors and FB in the monthly average concentration are displayed in Figures 1, 2, S1 and S2.

Overall, all species except SO_2 are under-predicted in both January and July (Table 2). SO_2 has less than 10% bias (NMB and FB) in both months but greater than 60% error (NME and FE) indicating that the average concentrations are predicted correctly but with substantial scatter. For O_3 and CO, model performance improves in July compared to January. NOx, NO_2 and PM_{10} are substantially underestimated and performance is poor for both months with similar magnitude bias and error statistics indicating that the underestimation trends are consistent both spatially and temporally. Analyzing the FB and FE statistics for January by monitor location type (Figure S4) shows less under-prediction tendency at rural monitors than at suburban/urban monitors for most species except O_3, as discussed below.

Maps of FB (Figures 1, 2, S1 and S1) can reveal whether spatial patterns exist in model bias. The spatial display of model bias for O_3 shows widespread good performance in July (generally within 10% FB) with some larger under-predictions in the Po Valley region of Northern Italy (Figure 2a). A similar analysis for the July average daily maximum 8-hour O_3 (Figure S3) shows greater under-estimation of high O_3 than average O_3 although the FB values are still within 30%. The diurnal cycle of July O_3 (Figure S5) shows that the model reproduces well the daily modulation in O_3. In contrast to

July, January O_3 performance is poor showing consistent under-predictions across the domain (Figure 1a). Analyzing the FB and FE statistics for January by monitor location type (Figure S4) shows that January O_3 is under-predicted for all monitor types (rural, suburban and urban). The diurnal cycle of January O_3 (Figure S5) shows that the model reproduces the daily modulation in O_3 but with an offset due to a consistent low bias. Since O_3 production by atmospheric chemistry is generally suppressed in winter, O_3 transport from the model boundaries (i.e., BCs) is expected to be the dominant factor in causing the low bias for O_3 in January. The effect of BCs on January O_3 should be studied further through a sensitivity test employing BCs from a different source.

In contrast to O_3, NOx performance is fairly good at rural background stations in January (Figures 1b and S4) with the model over-estimating NOx in parts of the UK, Belgium, Netherlands and Germany, but under-estimating NOx elsewhere, particularly in Northern Italy and Eastern Europe. The NOx performance is poorer in July (Figure 2b) with under-estimation across Europe. In July, CAMx predicted much lower day-time NOx (both NO and NO_2) than observed (Figure S5) which may have contributed to the tendency to under-predict daily maximum 8-hour O_3 (Figure S3), discussed above. These problems may stem from insufficient NOx emissions or overstated daytime dilution (e.g., too deep planetary boundary layer) of NOx emissions and O_3 in July.

SO_2 performance is highly variable, showing a fractional bias varying between ± 100% in both January and July (Figures S1 and S2). The bias is positive at most coastal stations while the modeled and observed concentrations are in a fairly good agreement inland suggesting that contributions from ship emissions to surface SO_2 might be over-estimated. A likely reason for SO_2 overestimation at coastal locations is that all ship emissions that were placed in the first model layer. In reality, many large vessels have sufficient stack height to release emissions into the second model layer. It will be useful to conduct a sensitivity test dividing ship emissions between the first two model layers, although a difficulty in performing this test will be the fact that ship emissions are combined with other off-road mobile sources in the available emission files.

PM_{10} is underestimated in all countries in January (Figure 1c), except France where the model and observations are in a fair agreement with a fractional bias within 30%. PM_{10} performance has greater under-estimation in July, although time series (Figure S5) show that the model prediction and observation correlate better in July than January. Figure S5 also shows that $PM_{2.5}$ performance is fairly good, especially in July, suggesting that the poor PM_{10} performance is primarily due to under-estimation of coarse material mass which suggests emission inventory problems. Analysis of PM_{10} and speciated components of PM using EMEP data (Figure S6) confirms that CAMx could not

reproduce PM_{10} episodes, showing a mean low bias of 13.0 $\mu g/m^3$ in January at rural EMEP stations. The combined inorganic PM species (i.e. PSO4, PNO3, PNH4) measured are generally less than 5 $\mu g/m3$ (compared to 20-40 $\mu g/m^3$ of PM_{10}) and the model could reproduce most of the mass, especially for PNO3. The analysis suggests that emissions of coarse PM were underestimated.

4. Conclusions and Future work

CAMx modelling for the EU domain was completed for 2006 using input data for emissions, meteorology and BCs developed by AQMEII. Model performance for January and July exhibited under-estimation trends for all pollutants both in winter and summer, except for SO_2. SO_2 generally had little bias although some over-estimation occurred at coastal locations and this was attributed to incorrect vertical distribution of emissions from marine vessels. Performance for NO_X and NO_2 was better in winter than summer. For O_3, the spatial distribution was modelled well in summer but with some tendency to underestimate daily maximum 8-hour O_3. The tendency to under-predict daytime NOx and O_3 in summer may result from insufficient NOx emissions or overstated daytime dilution (e.g., too deep planetary boundary layer). Winter $O_3$was biased low and this was attributed to a low-bias in the O_3 boundary conditions. PM_{10} was widely under-predicted in both winter and summer. The poor PM_{10} was influenced by under-estimation of coarse PM emissions. It is clear from these results that the underlying emission inventory and metrological input data play a crucial role in the air quality model performance. Model performance was better in some parts of Europe and the eastern European countries may have higher uncertainty of emissions and in the Po Valley of northern Italy the meteorological simulation at 35 km resolution may not have captured the strong influence of topography on pollutant concentration within the Po Valley which is bounded to the north by the Alps.

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The AQMEII approach to applying many models was to promote use of consistent data sources (e.g., emissions, BCs) and minimize uncertainties associated with use of differing inputs by each model. However, most models are using different meteorological data, several are using different BCs and a few are using different emissions. AQMEII is evaluating the ensemble of predictions from all models

applied for Europe and may not be able to untangle the consequences of differing input data and assumptions. In follow-on work under CRC Project A75-2, ENVIRON is investigating the influence of input data, assumptions and uncertainties on model performance using the CAMx application described here for the EU domain. Multiple simulations will be conducted first to identify the role played by different input data and some of the uncertainties discussed here. For example, alternate BCs will be used and the impact on winter O_3 evaluated; ship emissions will be divided between the first two model layers; alternate meteorological input data will be utilized. Several revised model configurations will be developed for model performance evaluation and the sensitivity of O_3 to precursor emissions (anthropogenic NOx and VOC, biogenic VOC) will be evaluated.

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Table 1. Anthropogenic emissions by SNAP sector for 2006 (metric tons/year)

	SNAP Sector	CO	NOx	NMVOC	CH4	NH3	SO2	PM10
1	Combustion in energy industries	762,912	2,903,396	120,552	774,388	5,984	7,781,377	431,632
2	Non-industrial combustion	11,340,097	833,530	1,137,160	677,509	10,978	791,519	866,201
3	Combustion in manufacturing Industry	4,003,572	1,849,805	177,135	278,575	5,854	1,900,364	313,757
4	Production processes	3,282,061	378,349	1,082,172	61,159	120,157	492,550	535,376
5	Energy extraction and distribution	149,083	41,399	941,238	5,595,385	930	239,703	66,655
6	Solvent use	27,422	184	4,495,530	0	9,760	6,766	59,816
7	Road transport	14,262,267	5,085,578	2,635,363	113,785	81,671	90,220	402,004
8	Other mobile sources	3,288,189	5,408,350	756,676	6,159	3,048	2,563,899	496,021
9	Waste treatment and disposal	1,582,985	30,175	118,913	8,609,183	121,147	7,753	102,764
10	Agriculture	190,261	193,548	538,112	12,749,030	4,889,872	3,173	412,733
	Total	38,888,849	16,724,314	12,002,851	28,865,173	5,249,401	13,877,324	3,686,959

Table 2. Definitions of statistical metrics of model performance

Metric (potential range)	Definition
Normalized Mean Bias (-100% to +∞)	$NMB = \frac{\sum_{i=1}^{N}(C_m - C_o)}{\sum_{i=1}^{N} C_o}$
Normalized Mean Error (0% to +∞)	$NME = \frac{\sum_{i=1}^{N}\lvert C_m - C_o\rvert}{\sum_{i=1}^{N} C_o}$
Fractional Bias (-200% to +200%)	$FB = \frac{1}{N}\sum_{i=1}^{N}\frac{(C_m - C_o)}{\left(\frac{C_o + C_m}{2}\right)}$
Fractional Error (0% to +200%)	$FE = \frac{1}{N}\sum_{i=1}^{N}\frac{\lvert C_m - C_o\rvert}{\left(\frac{C_o + C_m}{2}\right)}$

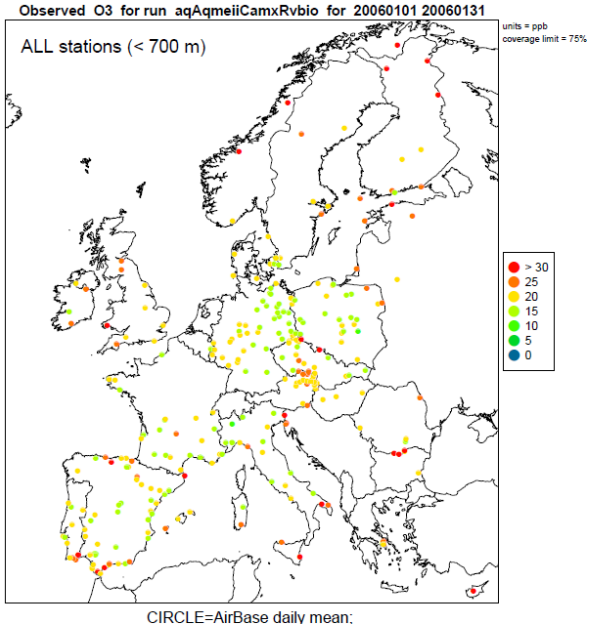
C_o = observation
C_m = model prediction
N = number of data pairs (C_o, C_m)

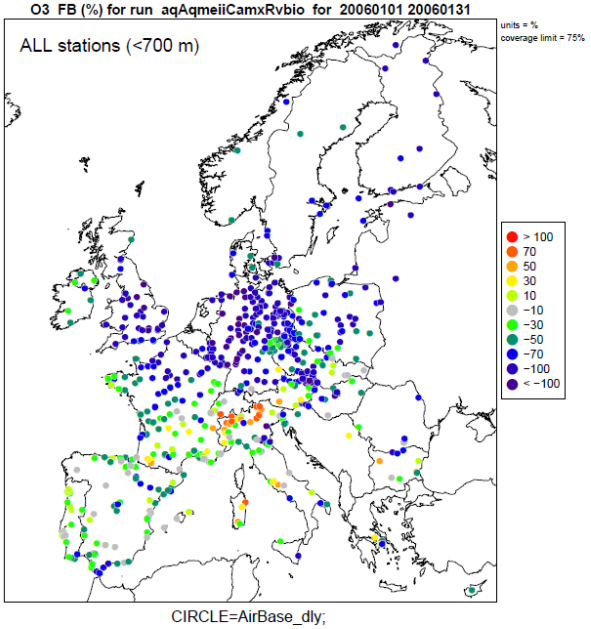
Table 3. Statistical metrics of model performance for January and July 2006

	N	NMB	NME	FB	FE
January					
O_3	25278	-40.4	50.9	-48.1	69.0
NOx	14840	-36.5	46.7	-51.3	74.1
NO_2	35728	-38.1	39.9	-48.0	61.1
CO	11370	-36.8	42.2	-45.4	62.9
SO_2	24651	1.1	60.9	0.4	68.2
PM_{10}	27251	-38.0	50.8	-46.8	73.1
July					
O_3	32069	-4.1	16.2	-1.9	20.5
NOx	14624	-51.1	53.6	-75.2	83.4
NO_2	35668	-45.6	49.4	-61.8	73.0
CO	10769	-12.5	33.0	-10.8	44.4
SO_2	22762	9.1	60.0	6.9	63.8
PM_{10}	27990	-44.2	45.9	-58.8	64.4

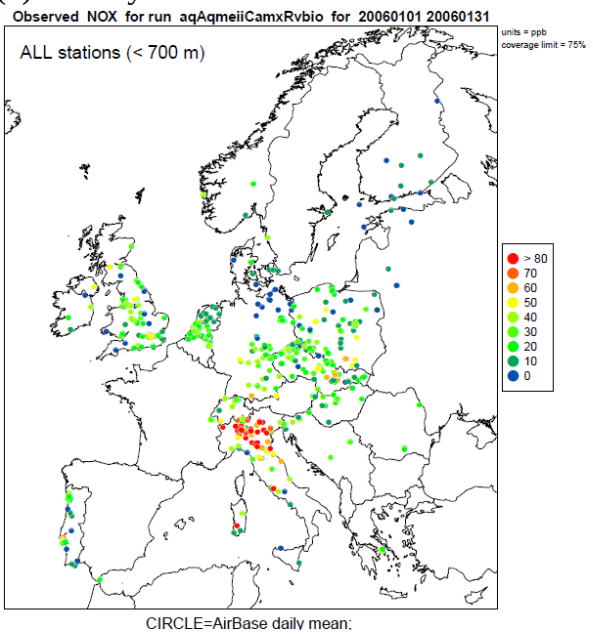
See Table 2 for definitions of the statistical metrics

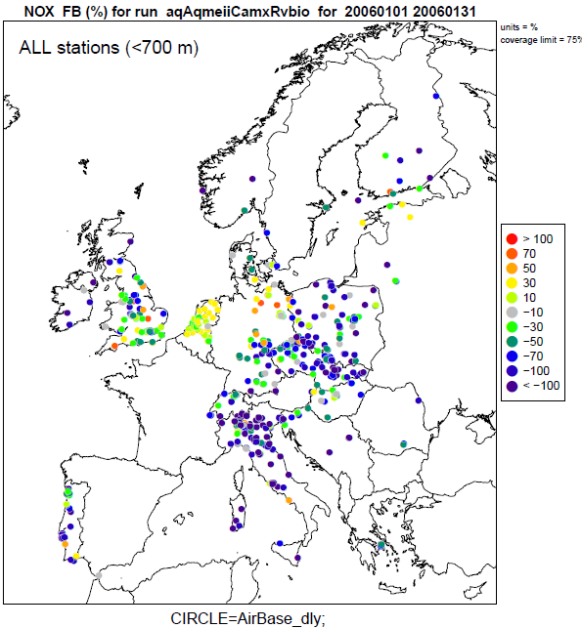
(a) January O_3





(b) January NOx





(c) January PM_{10}

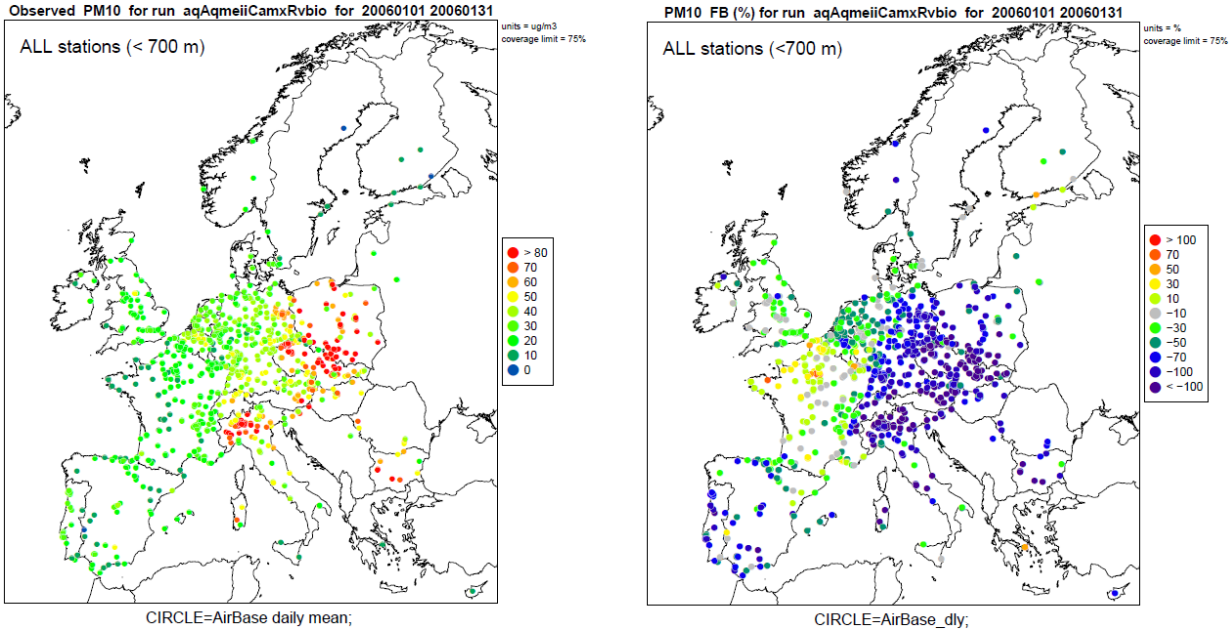
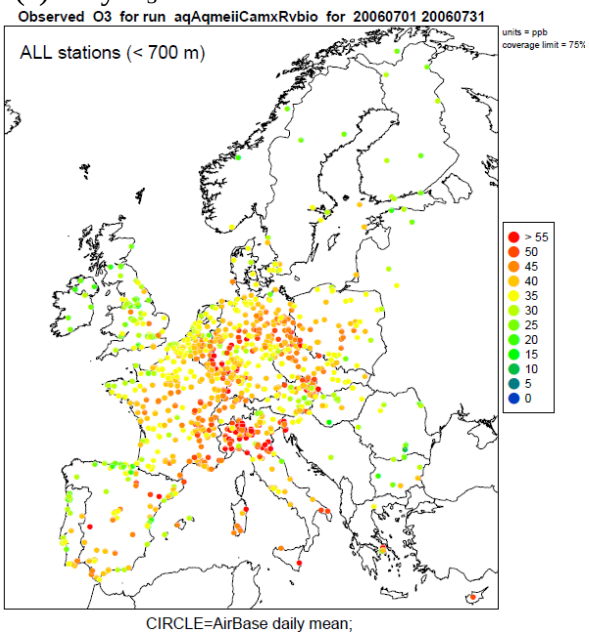
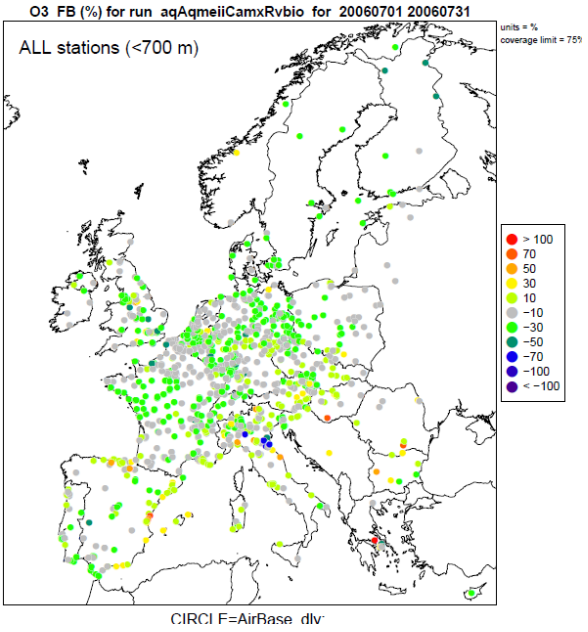


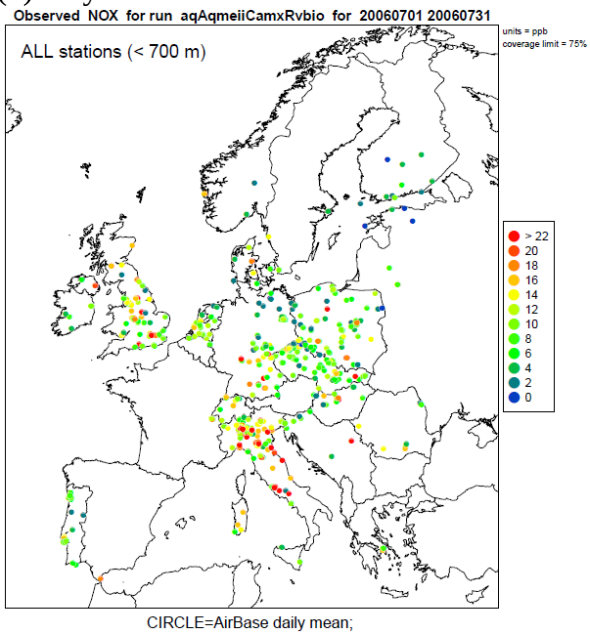
Figure 1. Observed daily mean concentrations (left) and fractional bias (right) of (a) O_3, (b) NOx, and (c) PM_{10} for January, 2006. (Figure S1 shows NO_2, SO_2 and CO).

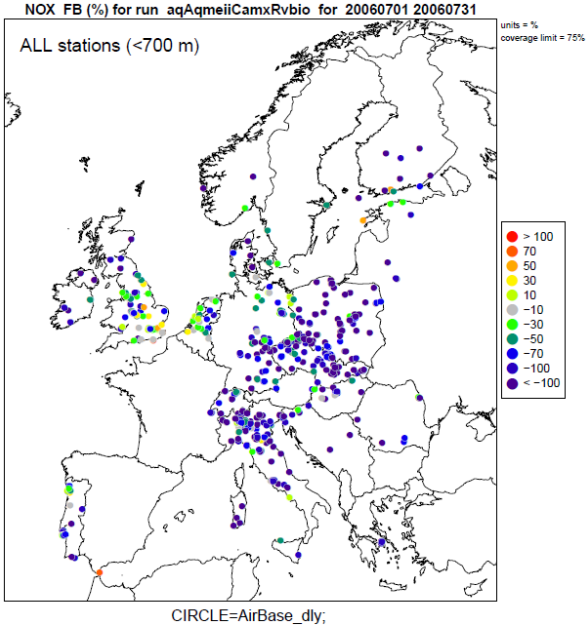
(a) July O_3





(b) July NOx





(c) July PM_{10}

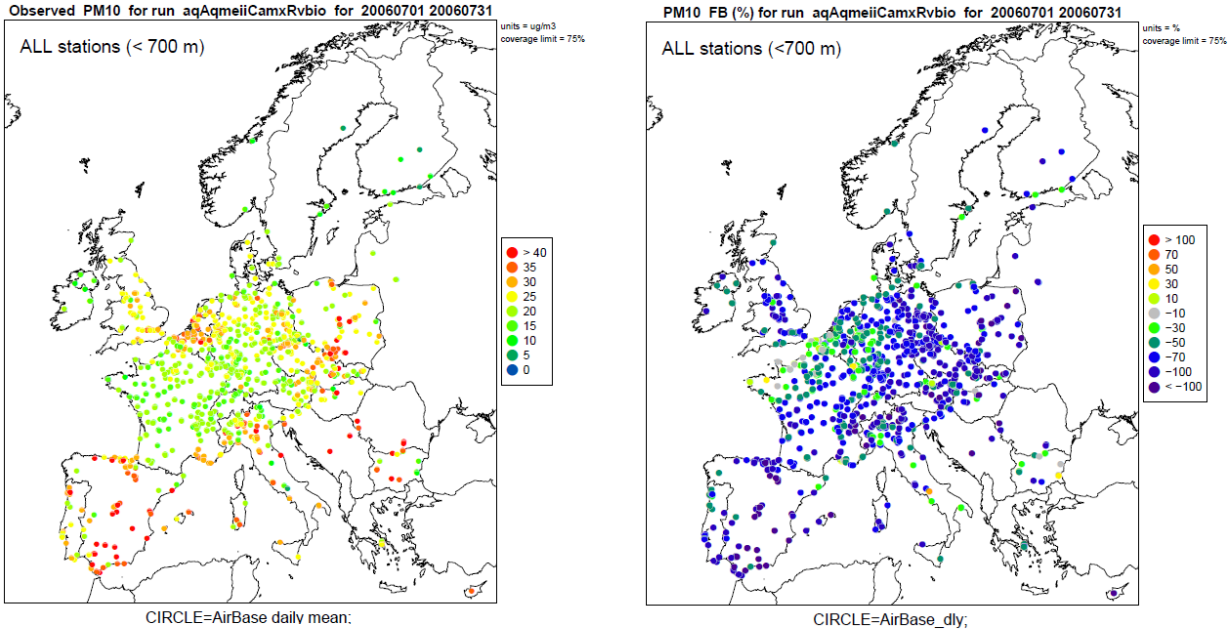


Figure 2. Observed daily mean concentrations (left) and fractional bias (right) of (a) O_3, (b) NOx, and (c) PM_{10} for July, 2006. (Figure S2 shows NO_2, SO_2 and CO).

Supplementary Information

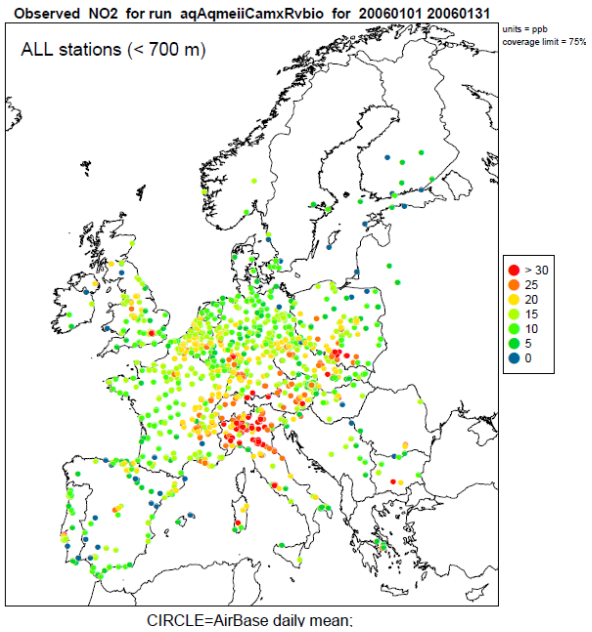
Table S1. Anthropogenic emissions by country or sea area for 2006 (metric tons/year)

Country	CO	NOx	NMVOC	CH4	NH3	SO2	PM10
Albania	112,326	25,535	32,309	177,786	24,398	31,255	9,085
Atlantic Ocean	51,968	517,315	17,888	0	0	359,895	43,423
Austria	705,024	215,984	164,276	331,057	63,927	28,083	46,382
Baltic Sea	36,123	346,739	12,166	0	0	224,834	26,126
Belarus	530,830	160,124	189,010	748,068	138,689	77,009	35,701
Belgium	851,588	276,772	198,578	364,228	73,206	136,104	42,188
Bosnia and Herzegovina	181,765	52,278	47,835	157,609	17,266	423,640	43,167
Black Sea	7,829	74,664	2,690	0	0	53,663	6,235
Bulgaria	759,381	222,748	153,547	473,961	57,186	850,382	82,997
Croatia	311,951	66,388	90,236	151,310	44,861	57,534	23,450
Cyprus	40,862	17,309	13,960	47,266	5,245	12,108	2,811
Czech Republic	479,382	266,888	177,971	496,595	67,524	201,389	33,910
Denmark	611,562	186,621	113,448	265,148	88,460	24,857	40,752
Estonia	152,419	30,496	33,671	89,287	9,251	69,561	25,191
Finland	512,576	192,753	132,647	217,140	36,819	86,065	49,467
France	5,169,230	1,151,452	1,349,980	2,634,445	727,597	432,793	492,543
Germany	4,038,311	1,426,299	1,209,808	2,161,306	616,090	567,324	194,266
Greece	605,382	297,184	341,096	403,967	71,982	528,801	68,922
Hungary	579,742	205,224	177,069	365,692	80,164	429,133	53,681
Ireland	211,893	110,097	58,976	621,653	111,965	60,162	21,318
Italy	3,973,855	1,133,175	1,227,009	1,826,637	420,590	471,378	162,170
Latvia	329,923	42,812	65,278	81,543	14,082	11,139	15,290
Lithuania	195,033	60,016	80,253	162,992	34,854	40,426	21,253
Luxembourg	41,313	14,142	12,804	16,540	5,254	2,759	3,506
Malta	0	11,396	8,846	17,976	892	8,026	644
Mediterranean Sea	158,543	1,546,054	53,020	0	0	1,083,591	126,537
Moldavia	140,142	65,556	38,265	217,119	27,101	122,205	43,879
Moldova	103,700	39,368	25,814	90,380	7,340	103,569	18,668
Netherlands	587,992	325,026	169,808	773,771	131,972	51,193	38,851
North Sea	77,421	746,737	26,495	0	0	483,699	58,468
Norway	418,560	190,528	191,419	210,359	22,485	21,328	52,776
Poland	3,524,572	675,397	945,791	1,814,791	290,750	1,308,061	284,969
Protugal	601,561	250,065	285,711	519,955	69,912	192,286	45,949
Russia	1,442,338	347,498	319,504	2,170,613	88,484	427,290	187,464
Serbia	1,368,525	290,248	391,122	1,216,704	195,507	571,585	144,961
Slovakia	290,949	86,596	74,815	199,527	26,586	87,708	24,350
Slovenia	77,066	58,019	41,911	99,100	18,703	35,681	9,405
Spain	2,191,555	1,445,946	1,051,691	1,751,802	442,340	1,235,055	203,821
Sweden	573,083	201,452	191,128	262,253	51,646	39,180	52,663
Switzerland	318,258	83,363	104,143	166,429	55,130	16,276	19,331
Turkey	1,924,232	662,163	523,038	1,531,449	257,292	1,349,459	269,119
Ukraine	2,014,849	845,175	569,494	3,172,935	465,971	530,874	325,959
United Kingdom	2,289,536	1,601,490	943,601	2,321,972	316,584	688,957	151,998
Yugoslavia	313,851	165,492	147,141	529,754	67,740	358,676	84,075
Total	**38,907,001**	**16,730,584**	**12,005,262**	**28,861,119**	**5,245,845**	**13,894,993**	**3,687,721**

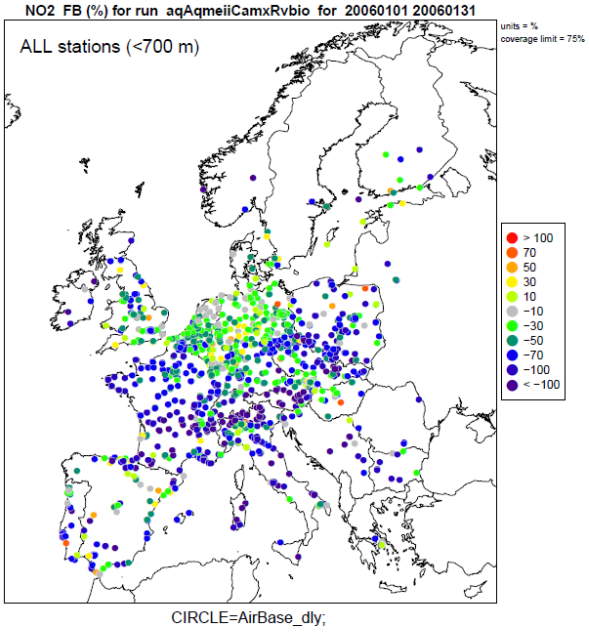
Table S2. Average daily emissions by source category in January and July 2006 (metric tons/day)

Source Category	CO	NOx	NMVOC	CH4	NH3	SO2	PM10
				January			
Anthropogenic	124,122	45,790	31,554	64,742	7,034	43,647	11,243
Biogenic	1,438	155	5,295	119	-	-	-
Fire	4,977	154	69	-	74	21	671
Sea Salt	-	-	-	-	-	-	65,148
Total	**130,537**	**46,099**	**36,918**	**64,861**	**7,108**	**43,668**	**77,062**
				July			
Anthropogenic	80,309	41,990	27,873	68,870	12,373	31,458	7,708
Biogenic	22,979	2,295	171,372	1,289	-	-	-
Fire	196,991	6,078	2,719	-	2,934	838	26,594
Sea Salt	-	-	-	-	-	-	15,917
Total	**300,279**	**50,363**	**201,964**	**70,159**	**15,307**	**32,296**	**50,219**

(a) NO_2



CIRCLE=AirBase daily mean;



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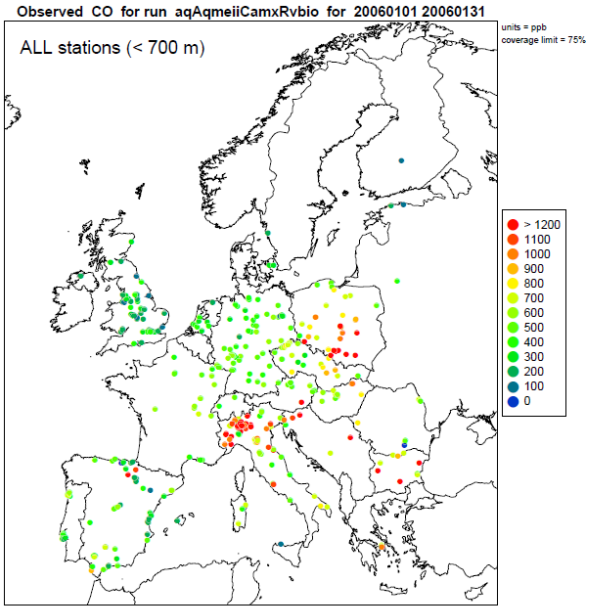
(b) CO



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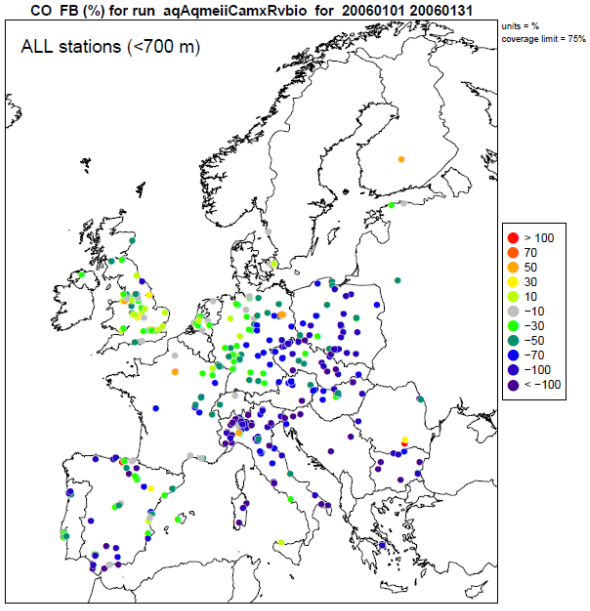


CIRCLE=AirBase_dly;

(c) SO_2

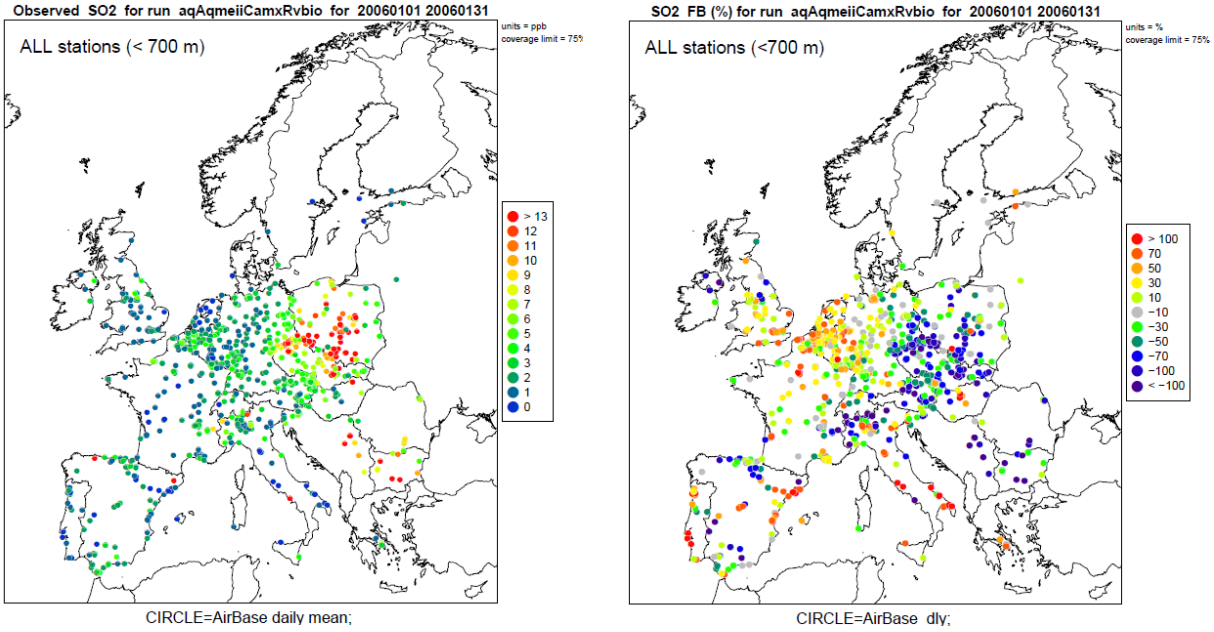
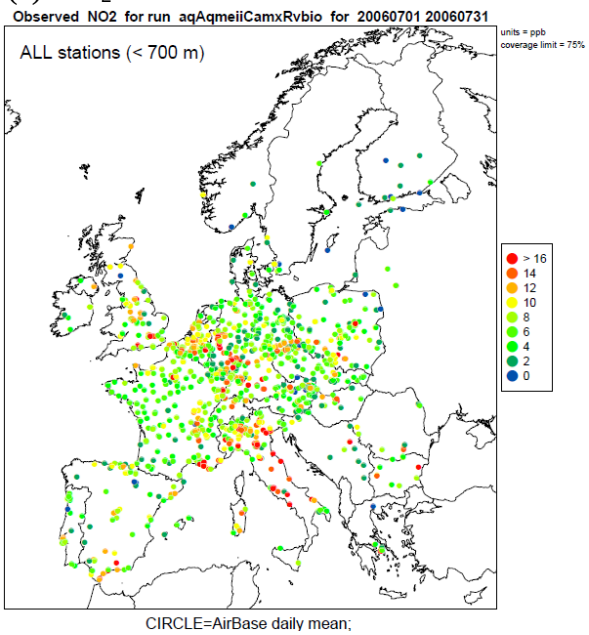
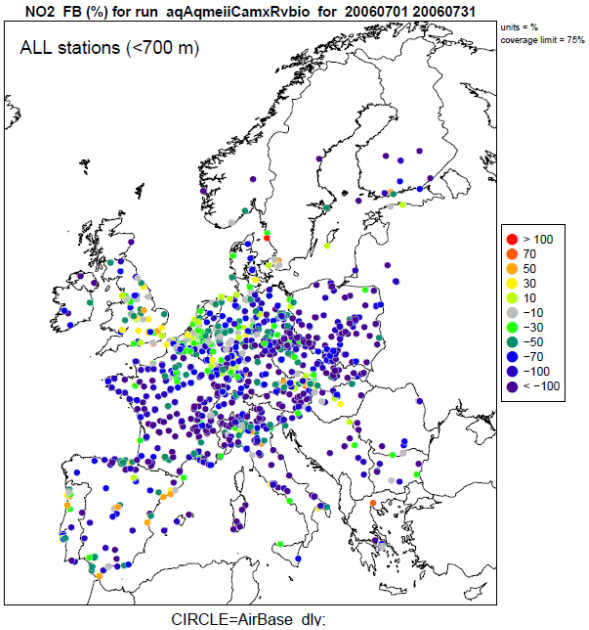


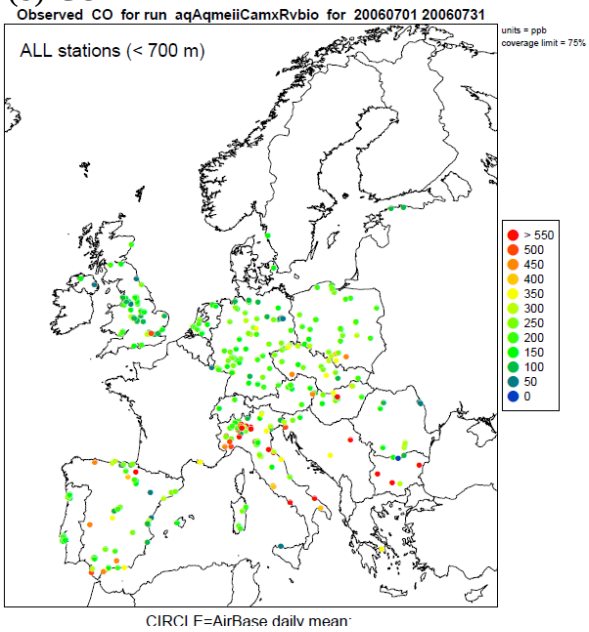
Figure S1. Observed daily mean concentrations (left) and fractional bias in daily mean concentrations (right) of (a) NO_2, (b) CO, and (c) SO_2 for January 2006.

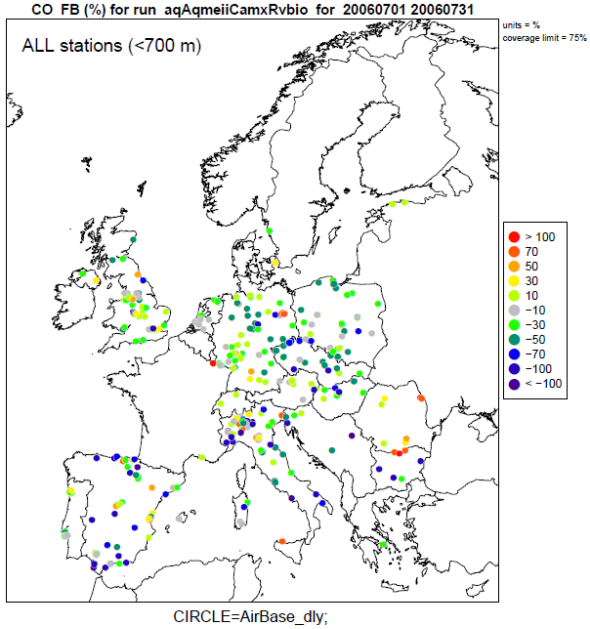
(a) NO_2





(b) CO





(c) SO_2

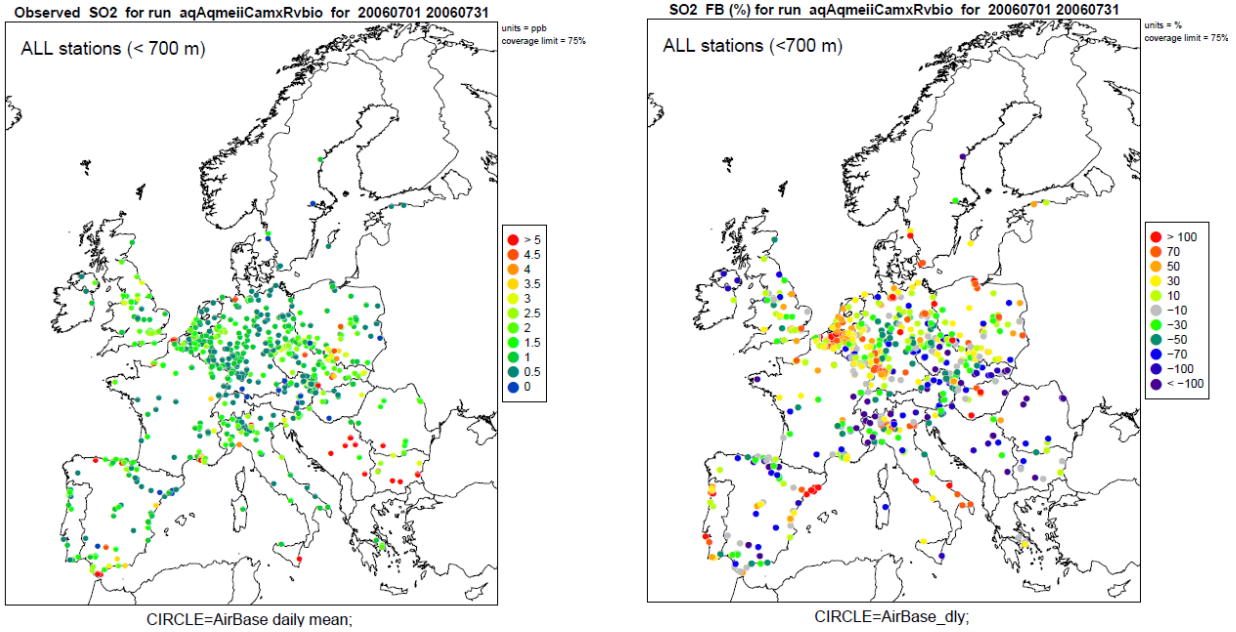


Figure S2. Observed daily mean concentrations (left) and fractional bias (right) of (a) NO_2, (b) CO, and (c) SO_2 for July 2006.

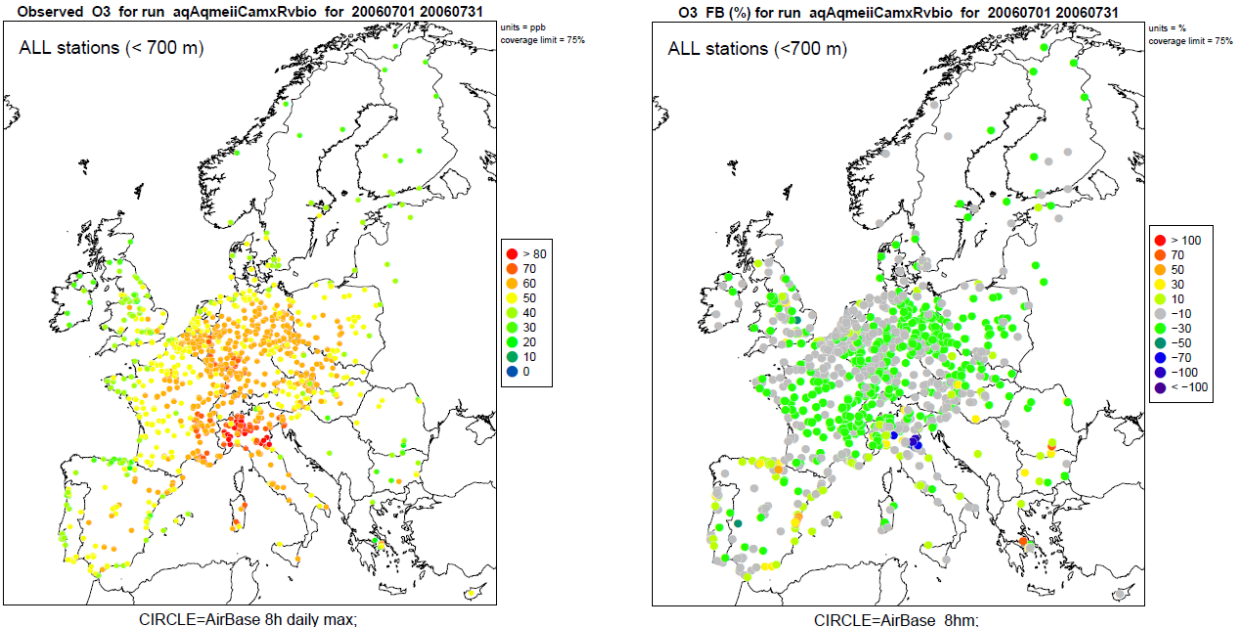


Figure S3. Observed 8-hour maximum O_3 concentrations (left) and fractional bias (right) in July, 2006.

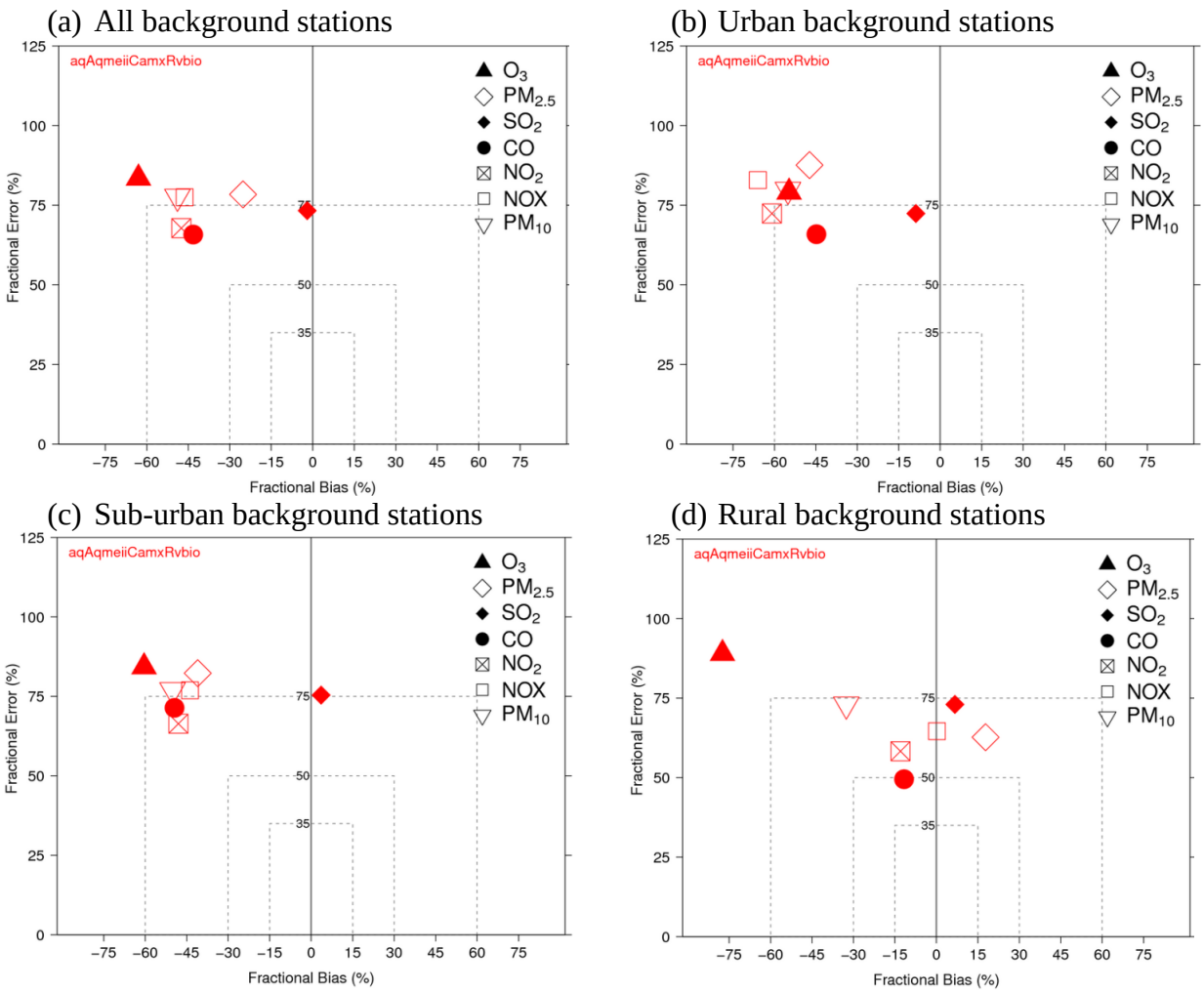


Figure S4. Fractional bias and error in hourly concentrations (daily for PM) in January, 2006 at (a) all, (b) urban, (c) sub-urban, and (d) rural background AirBase monitors.

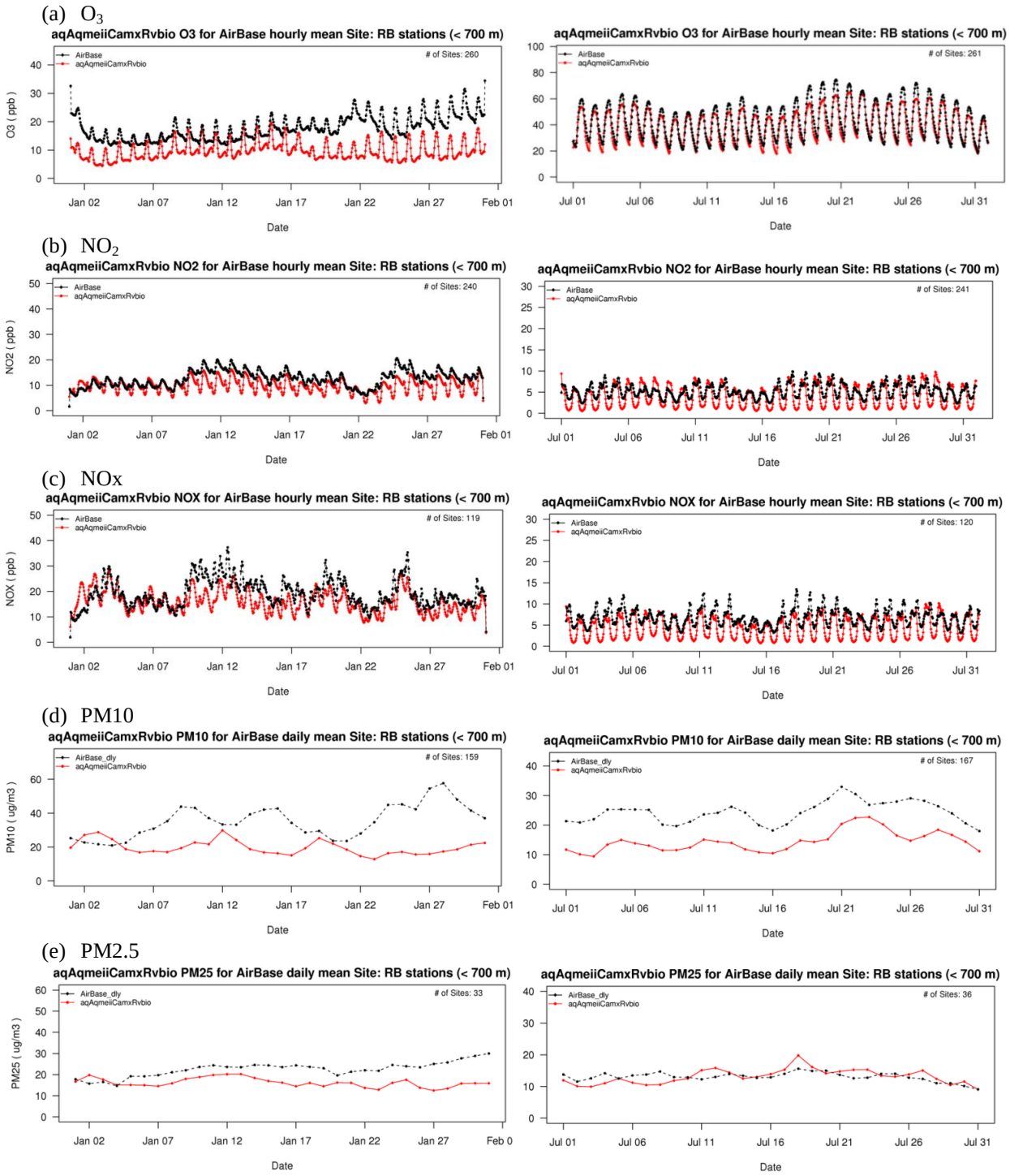


Figure S5. Modeled and observed hourly concentrations of (a) O_3, (b) NO_2, (c) NOx and daily concentrations of (d) PM10 and (e) PM2.5 at AirBase rural background stations for January (left) and July (right), 2006.

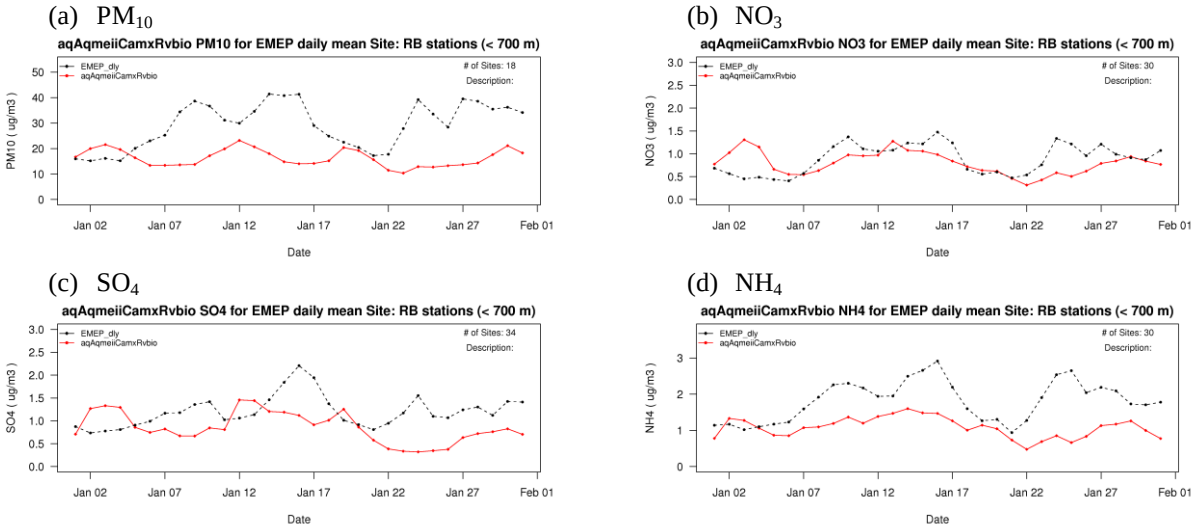


Figure S6. Modeled and observed daily mean concentrations of (a) PM_{10}, (b) PNO3, (c) PSO4, and (d) PNH4 averaged over all selected EMEP stations for January, 2006.