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Dynamic evaluation of a multi-year model simulation of particulate matter concentrations over Europe

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Abstract

A nine-year air quality simulation is conducted from 2000 to 2008 over Europe using the Polyphemus/Polair3D chemical-transport model (CTM) and then evaluated against the measurements of the European Monitoring and Evaluation Programme (EMEP).

The spatial distribution of $PM_{2.5}$ over Europe shows high concentrations over northern Italy (36 $\mu g\,m^{-3}$) and some areas of eastern Europe, France, and Benelux, and low concentrations over Scandinavia, Spain, and the easternmost part of Europe. $PM_{2.5}$ composition differs among regions.

The operational evaluation shows satisfactory model performance for ozone (O_3). $PM_{2.5}$, PM_{10}, and sulfate (SO_4^{2-}) meet the performance goal of Boylan and Russell (2006). Nitrate (NO_3^-) and ammonium (NH_4^+) are overestimated, although NH_4^+ meets the performance criteria. The correlation coefficients between simulated and observed data are 63 % for O_3, 57 % for PM_{10}, 59 % for $PM_{2.5}$, 57 % for SO_4^{2-}, 42 % for NO_3^-, and 58 % for NH_4^+. The comparison with other recent one-year model simulations shows that all models overestimate nitrate. The performance of $PM_{2.5}$, sulfate, and ammonium is comparable to that of the other models.

The dynamic evaluation shows that the response of $PM_{2.5}$ to changes in meteorology differs depending on location and the meteorological variable considered. Wind speed and precipitation show a strong negative day-to-day correlation with $PM_{2.5}$ and its components (except for sea salt, which shows a positive correlation), that tends towards 0 as the day lag increases. On the other hand, the correlation coefficient is near constant for temperature, for any day lag and $PM_{2.5}$ species, but it may be positive or negative depending on the species and, for sulfate, depending on the location. The effects of precipitation and wind speed on $PM_{2.5}$ and its components are better reproduced by the model than the effects of temperature. This is mainly due to the fact that temperature has different effects on the $PM_{2.5}$ components, unlike precipitation and wind speed which impact most of the $PM_{2.5}$ components in the same way.

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These results suggest that state-of-the-science air quality models reproduce satisfactorily the effect of meteorology on $PM_{2.5}$ and, therefore, are suitable to investigate the effects of climate change on particulate air quality.

1 Introduction

Atmospheric particulate matter (PM) pollution has become a field of great interest because of its impacts on human health, climate change, and atmospheric visibility. Therefore, air quality regulations have been implemented for PM concentrations. In particular, fine particles with an aerodynamic diameter less than or equal to 2.5 µm ($PM_{2.5}$) are regulated in North America and Europe. $PM_{2.5}$ is a complex mixture of particles of different sizes and chemical compositions. These chemical compositions include primary PM, which is directly emitted in the atmosphere from various sources (e.g. road trafic, construction sites, soil dust, fires), and secondary PM, which is formed in the atmosphere via chemical reactions in the gas and aqueous phases, leading to the oxidation of precursors such as sulfur dioxide (SO_2), nitrogen oxides (NO_x), and volatile organic compounds (VOC) to non-volatile and semi-volatile species. The processes that govern the secondary particle concentrations are various and complex. In particular, they depend strongly on meteorology (temperature, solar radiation, humidity, presence of clouds and fog). Emissions of primary particles and precursors of secondary PM are also strongly affected by meteorology (wind speed, temperature, solar radiation). Furthermore, precipitation removes PM from the atmosphere. Therefore, climate change is expected to affect PM concentrations via the effect of meteorological variables on the emissions, formation, and removal of PM.

Studies of the effect of climate change on air quality have focused initially on ozone (e.g. Meleux et al., 2007; Loon et al., 2007; Mahmud et al., 2008; Wu et al., 2008; Chen et al., 2009; Katragkou et al., 2011), and the study of its effect on PM concentrations is more recent. So far, most of the PM studies have focused on the United States and, to a lesser extent, Europe (e.g. Racherla and Adams, 2006; Dawson et al.,

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2007, 2009; Zhang et al., 2008; Avise et al., 2009; Pye et al., 2009; Tagaris et al., 2009; Mahmud et al., 2010; Singh and Palazoglu, 2012; Tai et al., 2010, 2012; Kelly et al., 2012), but simulations have typically been limited to a year or several months. The individual effects of various meteorological variables have been examined for the United States by perturbing each meteorological variable separately. The results suggest that the strongest effects of changes in meteorology on $PM_{2.5}$ concentrations are the effects of temperature, wind speed, absolute humidity, mixing height, and precipitation. According to these studies (Dawson et al., 2007; Mahmud et al., 2010; Galindo et al., 2010), temperature tends to increase average sulfate concentrations and decrease average nitrate and organic concentrations, leading to an overall decrease in $PM_{2.5}$ concentrations. Increasing absolute humidity increases nitrate aerosol, which leads to increased $PM_{2.5}$ concentrations. Changes in mixing height lead to mixing and dilution effects, with $PM_{2.5}$ concentrations generally decreasing as mixing height increases. $PM_{2.5}$ concentrations decrease with increased precipitation rate and the extent of the precipitation area. Increases in wind speed lead to changes in advection and transport resulting in decreases in $PM_{2.5}$ concentrations. Because meteorology may affect $PM_{2.5}$ components in opposite ways (e.g. an increase in temperature favors the emissions of biogenic VOC and their oxidation to semi-volatile organic compounds (SVOC) but increases SVOC volatility), no strong consensus has yet been reached on the effects of the overall present and future climate on $PM_{2.5}$ concentrations.

Before one investigates the effects of climate change on PM concentrations, it is primordial to ensure that our current understanding of the relationships between meteorology and PM concentrations is correct. Typically, the evaluation of model performance is limited to the ability of the model to reproduce $PM_{2.5}$ and its components and provides no information on the ability of a model to predict the response of $PM_{2.5}$ components to changes in meteorology. Four levels of model performance evaluation may be considered: operational, diagnostic, dynamic, and probabilistic (Seigneur et al., 2000; Dennis et al., 2010). The operational evaluation tests the ability of the model to correctly estimate PM concentrations, while the diagnostic evaluation focuses on the estimation of

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the components of PM and precursors (Dennis et al., 2010 included PM components in the operational evaluation and we follow their categorization here for simplicity). The dynamic evaluation tests the ability of the model to predict the response of PM concentrations to changes in meteorology and emissions. Finally, the probabilistic evaluation takes also into account the uncertainties associated with the model predictions and observations of PM. To date, dynamic model performance evaluations have been limited to emission changes over the United States (Gilliland et al., 2008; Yarwood et al., 2003). To our knowledge, there has been no comprehensive dynamic evaluation conducted with respect to meteorology. Therefore, the goal of this study is to conduct such a dynamic evaluation using a multi-year simulation of $PM_{2.5}$ over Europe.

A brief description of the Polyphemus/Polair3D modeling system used here is given in Sect. 2, along with the characteristics of the model simulation and the spatial distribution and composition of modeled $PM_{2.5}$. The Polyphemus system is used for simulating concentrations over Europe for years 2000 to 2008. An evaluation is then made for each year for both gases and aerosols. An operational model performance evaluation using available data is presented in Sect. 3. Those results are compared with those obtained recently in the AQMEII project and in other one-year model performance evaluations for PM_{10}, $PM_{2.5}$ and its components. A dynamic evaluation performed with respect to meteorology is presented in Sect. 4. Conclusions and future prospects are presented in Sect. 5.

2 Model simulation

2.1 Input data and model configuration

We used the Polyphemus/Polair3D model (Mallet et al., 2007; Debry et al., 2007; Sartelet et al., 2007; Couvidat et al., 2012) to simulate nine years (2000–2008) of concentrations of gaseous and particulate pollutants over Europe. Polyphemus is an air quality modeling platform, which has been used for many applications at different

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scales (from local to continental). Polair3D is the chemical-transport model (CTM) of Polyphemus.

The modeling domain covers a geographical area, which spreads from 15° W to 34.5° E in longitude and from 35° N to 69.5° N in latitude. Therefore, the domain covers an area of 100° × 70° with a step of 0.5° along both longitude and latitude as shown in Fig. 1. Fourteen levels are considered from the ground up to 12 000 m. The boundary heights of the different model layers are 30, 60, 100, 150, 200, 300, 500, 750, 1000, 1500, 2400, 3500, 6000, and 12 000 m.

Meteorological data were obtained from the European Centre for Medium-Range Weather Forecasts (ECMWF). The horizontal resolution (both longitude and latitude) of these meteorological fields is 1.125° for 2000, 0.36° for 2001–2005, and 0.25° for 2006–2008. The vertical resolution includes 36 levels for 2000–2002, 2005 and January 2006, 31 levels for 2003–2004, and 54 levels for the remainder of 2006 and 2007–2008.

Anthropogenic emissions for gases and particles were generated with the Environmental Monitoring and Evaluation Programme[1] (EMEP) inventory for 2000 to 2008 for all sectors. Biogenic emissions were computed with the Model of Emissions of Gases and Aerosols from Nature (MEGAN, Guenther et al., 2006). Sea-salt emissions are parameterized following Monahan et al. (1986).

Boundary conditions are obtained from the outputs of the Model of OZone And Related Tracers (MOZART-4, Emmons et al., 2010) for the years 2004 to 2008, with six-hour resolution. No MOZART output is available for years 2000 to 2002, and the year 2003 is incomplete; we thus computed the mean of years 2004 to 2008 to create climatological boundary conditions for years 2000–2003. The MOZART-4 chemical mechanism includes 85 gas-phase species, 12 bulk aerosol compounds, 39 photolysis and 157 gas-phase reactions. Dust and sea salt aerosol data are distributed in MOZART4 among four size sections (0.05–0.5, 0.5–1.25, 1.25–2.5, and 2.5–5.0 µm for dust and 0.1–0.5, 0.5–1.5, 1.5–5.0, and 5.0–10.0 µm for sea salt). The concentrations from the input data are proportionally redistributed among the five sections of Polair3D

[1] http://emep.int/

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(0.01–0.04, 0.04–0.16, 0.16–0.63, 0.63–2.5, and 2.5–10.0 µm). Black carbon, organic carbon, nitrate, ammonium, and sulfate aerosol input data follow a normal size distribution, which is distributed over the five sections of Polair3D.

The chemical mechanism chosen for the simulation is CB05 (Yarwood et al., 2005). It has been shown to perform satisfactorily in previous applications to Europe (Kim et al., 2009). Photolysis rates are computed offline, using the photolysis preprocessor Fast-J, which calculates photolysis rates in the presence of an arbitrary mix of cloud and aerosol layers (Wild et al., 2000). The dynamics of the PM size distribution is simulated according to a sectional representation of the PM mass distribution (Debry et al., 2007). Inorganic PM is simulated with the ISORROPIA thermodynamic model (Nenes et al., 1998) and organic PM is modeled with a hydrophilic/hydrophobic organic (H^2O) model using a molecular surrogate approach (Couvidat et al., 2012). The dry deposition velocities for gases are preprocessed with the parameterization of Zhang et al. (2003). Vertical diffusion is computed using the Troen and Mahrt (1986) parameterization within the planetary boundary layer. For land-use coverage, the United States Geological Survey (USGS) cover map is used. For each year of simulation, the initial conditions are computed by using a spin-up period of 15 days from 15 to 31 December of the previous year.

2.2 $PM_{2.5}$ spatial distribution and chemical composition over Europe

The spatial distribution of particulate species over Europe is shown in Fig. 2 for the 9-yr averaged surface concentrations of $PM_{2.5}$, sulfate, nitrate, ammonium, sea salt, organic matter, black carbon, and mineral dust. Figure 2a shows high concentrations of $PM_{2.5}$ over northern Italy (36 $\mu g\,m^{-3}$), the Netherlands (25 $\mu g\,m^{-3}$), northeastern Spain and France and eastern European countries (around 22 $\mu g\,m^{-3}$), while concentrations are lower in the northern and easternmost parts of Europe and in Spain (between 4 and 14 $\mu g\,m^{-3}$). Figure 2b depicts higher concentrations of sulfate over eastern Europe, with two peaks in Romania and Bulgaria (5 $\mu g\,m^{-3}$). The spatial distribution for nitrate (Fig. 2c) is similar to the distribution for $PM_{2.5}$, while high concentrations of ammonium

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are localized in eastern Europe (Fig. 2d), with a maximum of 5 $\mu g\,m^{-3}$ in Bulgaria. As expected, sea salt concentrations are the highest over the Atlantic Ocean (between 4 and 7 $\mu g\,m^{-3}$), the Mediterranean and the Baltic seas (between 2.5 and 4 $\mu g\,m^{-3}$), and along the coasts of the countries bordering the sea and the ocean (between 2 and 3 $\mu g\,m^{-3}$). Sea salt concentrations are near zero on the continent (Fig. 2e). Organic matter is high in northwestern Portugal, eastern France and over northern Italy (between 8.5 and 13 $\mu g\,m^{-3}$). Slovenia, Poland and Romania also show high concentrations of organic matter (around 8 $\mu g\,m^{-3}$) (Fig. 2f). Black carbon concentrations are below 1 $\mu g\,m^{-3}$ over all Europe, except for the northeastern part of France (3 $\mu g\,m^{-3}$), and in some localized areas in France, Italy and Romania (1.25 $\mu g\,m^{-3}$) (Fig. 2g). The concentrations of mineral dust vary from 1.5 to 5 $\mu g\,m^{-3}$ below 52° N, while they vary from 0.75 to 1.5 $\mu g\,m^{-3}$ above 52° N.

Figure 3 presents the 9-yr averaged surface fractions of sulfate, nitrate, ammonium, sea salt, organic matter, black carbon, and mineral dust in $PM_{2.5}$. In Scandinavia, $PM_{2.5}$ consists mainly of organic matter (from 40 % to 60 %), mineral dust (from 14 % to 20 %), and nitrate for the southernmost (and most industrial) part of Scandinavia (12 to 30 %). Nitrate and organic matter account for around 60 % of $PM_{2.5}$ in Germany, Switzerland, Austria and northern Italy, while ammonium, sulfate, and mineral dust represent together around 36 %. As expected, $PM_{2.5}$ is mainly sea salt over the ocean and the sea (from 22 % to 60 % of its composition); $PM_{2.5}$ along the west coast of France, Ireland, Great Britain, the Netherlands and Denmark is made of around 20 % of sea salt. Organic matter represents around 20 % in Germany, Belgium, southeastern England and northwestern France, and from 30 % to 50 % in the rest of Europe. Black carbon accounts for less than 4 % in all Europe, except for some localized areas in Portugal, France, Romania and Turkey (4 % to 8 %) and a peak of 10 %, which is observed at the border of France with Luxembourg and Germany.

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3 Operational evaluation

3.1 Statistical scores

The operational evaluation was performed for each year for ozone (O_3) and PM. Available PM measurements include PM_{10} and $PM_{2.5}$ mass concentrations and $PM_{2.5}$ sulfate, nitrate, and ammonium concentrations. Although organic carbon and elemental carbon data are available at the EMEP stations for one year (Yttri et al., 2007), no operational evaluation was performed for these species, as Polair3D has recently been evaluated for carbonaceous species (Couvidat et al., 2012). The correlation coefficient (%) and the root mean square error (RMSE) ($\mu g\,m^{-3}$) are presented in Table 1 to provide a common overview of model performance for O_3 and PM. Other statistical metrics that are used routinely to evaluate model performance are presented in Table 2. The model fits best to the observations when the RMSE is small compared to the observed mean and the correlation coefficient is large. The US EPA (EPA, 1991; Russell and Dennis, 2000) recommends using the mean normalized bias (MNB) (%) and the mean normalized error (MNE) (%) with an observation-based minimum threshold of about 80–120 $\mu g\,m^{-3}$ to evaluate hourly ozone. A threshold of 80 $\mu g\,m^{-3}$ was used here. The suggested performance criteria are $|MNB| \leq 15\,\%$ and $MNE \leq 35\,\%$. Bias indicates whether the model tends to under or overpredict the observations, and error and RMSE indicate how much it deviates from the observations. The mean fractional bias (MFB) (%) and the mean fractional error (MFE) (%) are recommended to evaluate PM (Boylan and Russell, 2006). The model performance goal is met when both MFE and |MFB| are less than or equal to 50 % and 30 %, respectively, and the model performance criterion is met when both $MFE \leq 75\,\%$ and $|MFB| \leq 60\,\%$.

The total number of stations that provide data for at least one year in the whole period is 91 for ozone, 77 for sulfate, 34 for ammonium, 33 for nitrate and PM_{10}, and 22 for $PM_{2.5}$. If the number of stations for which data are available each year is about constant for ozone (around 70 stations), sulfate (around 20 stations), and PM_{10} (around 10 stations), that number is more variable for the other species. For example, 20 stations

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provide data for $PM_{2.5}$ in 2005, while there are only 2 in 2000. There are some uncertainties in the observational data. Although most of the sites are background sites, they could nevertheless be impacted by some proximate source or be affected by local meteorological conditions that are not resolved by the model (representativeness issue). Moreover, the observational error may be non-negligible due to artifacts in the measurement methods, particularly for semi-volatile species (i.e. nitrate and ammonium). Also, the aerosol water content is not taken into account in model-to-data comparisons but a small amount of water may remain in the PM mass measurements.

Table 1 shows the annual mean correlations between the simulated and the observed concentrations (ρ in %), the RMSE, and the mean concentrations of the observed data (μ_{obs}) and of the simulated data (μ_{sim}), expressed in $\mu g\,m^{-3}$. On average, hourly ozone is overestimated by about 23 %, but the correlation coefficient is 62.9 %, and the RMSE is 28.2 $\mu g\,m^{-3}$ (the observed mean concentration is 60.6 $\mu g\,m^{-3}$). The criteria of Russell and Dennis (2000) are met with a MNE of 18.4 % and a MNB of 5.3 % on average. Those criteria are met for all years except 2000 when MNB is 17 % (instead of $\leq$ 15 %, see Table 2).

PM_{10} is well estimated with a correlation coefficient of 56.6 % and a RMSE of 10.7 $\mu g\,m^{-3}$ on average (the observed mean concentration is 17.3 $\mu g\,m^{-3}$). On average, MFB and MFE are 15.2 % and 42.9 %, respectively, meeting the performance goal of Boylan and Russell (2006) (see Table 2). The performance goal is met every year except in 2000 when both MFB and MFE exceed the goals but meet the criteria. $PM_{2.5}$ is overestimated by 20 % on average with a RMSE of 9 $\mu g\,m^{-3}$ (the observed mean concentration is 13.3 $\mu g\,m^{-3}$) and a correlation coefficient of 59 %. On average, MFB (29.8 %) and MFE (47.4 %) also meet the performance goal. These performance goals are met for five years (2001, 2003, 2004, 2006, and 2008); for the other years, $PM_{2.5}$ meets the performance criteria. Sulfate provides the best results: the correlation coefficient is 56.5 % and the RMSE is 1.7 $\mu g\,m^{-3}$ on average (the observed mean concentration is 2.3 $\mu g\,m^{-3}$). Simulated concentrations are on average 4 % lower than the observations. Both MFB (−2.1 %) and MFE (43.2 %) meet the model performance goal

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for every single year. Model performance is lower for nitrate and ammonium. Simulated concentrations are overestimated compared to the observed concentrations (80 % for nitrate ad 55 % for ammonium on average). Ammonium has a better correlation coefficient and RMSE (58 % and 1.1 $\mu g\,m^{-3}$, the observed mean concentration is 0.9 $\mu g\,m^{-3}$) than nitrate (42 % and 3.3 $\mu g\,m^{-3}$, the observed mean concentration is 2 $\mu g\,m^{-3}$) because a fraction of ammonium is associated with sulfate. Nitrate does not meet the performance criteria (MFB = 20.5 % and MFE = 83.2 % on average), but ammonium does (MFB = 20.8 % and MFE = 56 %). For nitrate, the performance criterion is met for four years (2003, 2006, 2007, and 2008). For ammonium, the performance goal is met for the last three years (2006–2008), and the performance criterion for all years except 2000.

The evolution of model performance over the years shows a clear improvement for ozone, ammonium, and nitrate from 2000 to 2008. This improvement could be due to improvement in the measurements, the emission inventory, the meteorology (better spatial resolution for the more recent years), the boundary conditions (year-specific values starting in 2004) or a combination thereof. It is not possible at this point to identify conclusively the driving source of this evolution, nevertheless, model performance is satisfactory on average for $PM_{2.5}$ and its components.

3.2 Comparison with other model evaluations in the context of AQMEII

Sartelet et al. (2012) summarized the results of an operational model performance evaluation conducted for nine models, excluding the Polyphemus/Polair3D model, in the context of the Air Quality Model Evaluation International Initiative (AQMEII, Solazzo et al., 2012). The mean of the statistics of the nine AQMEII models was computed, along with the minimum and maximum values, for PM_{10} and $PM_{2.5}$. We compare here our model performance evaluation to that of the AQMEII models. The AQMEII model evaluation was performed for a 2-month period (from 7 July to 31 August 2006). We thus computed the performance statistics for this study from 7 July to 31 August for each year and averaged the results over 2000–2008 for the comparison. The

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observational data for the AQMEII model evaluation include stations from the Airbase and EMEP databases, while we only used the latter (Airbase doest not provide data for the components of PM, while EMEP does). Table A1 summarizes the statistics obtained for PM_{10} and $PM_{2.5}$.

Daily PM_{10} is well-estimated by Polair3D over 2000–2008 (15.9 $\mu g\,m^{-3}$ measured against 16.6 $\mu g\,m^{-3}$ simulated), while the AQMEII model concentrations are on average 10 $\mu g\,m^{-3}$ lower than the observations. The Polair3D average RMSE is about half the mean observed value, whereas the mean RMSE for the AQMEII models is commensurate with the observed value. The Polair3D averaged correlation coefficient is significantly higher than the best correlation coefficient for the AQMEII models. The MFE and MFB of Polair3D meet the performance goal criteria, while the AQMEII models do not. Solazzo et al. (2012) performed statistical analysis for PM_{10} for 10 model simulations in the context of AQMEII for the whole 2006 year. The RMSE ranges from 7.3 to 15.2 $\mu g\,m^{-3}$ for the different models, which is consistent with the RMSE obtained here, which is 10.7 $\mu g\,m^{-3}$ on average (see Table 1). The MFB for PM_{10} at rural stations ranges between −70 % and +10 % for the different AQMEII models, while, in this work, it is 15.2 % on average. The MFE for the AQMEII models spreads from 25 % to 75 % for the different models, while it is 42.9 % on average in this work.

Daily $PM_{2.5}$ is overestimated compared to the observations (11.5 $\mu g\,m^{-3}$ measured against 16 $\mu g\,m^{-3}$ simulated on average). The AQMEII models show both under and overestimations (13.3 $\mu g\,m^{-3}$ measured against a range of 5 to 21.4 $\mu g\,m^{-3}$ simulated). The Polair3D average RMSE (7.2 $\mu g\,m^{-3}$) is smaller than those of the AQMEII models, with similar mean observed values. The correlation coefficient is significantly better than those of the AQMEII models (68 % against 3 to 21 %). Compared to the AQMEII models, the MFE and MFB of this simulation show better results and meet the model performance criteria of Boylan and Russell (2006), while the AQMEII models do not.

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3.3 Comparison with the performance evaluation of one-year simulations

We also compared our model performance evaluation to that of four other chemical-transport models that have been used for a one-year simulation over Europe (see Appendix B): CHIMERE (Péré et al., 2010), CALIOPE-EU (Pay et al., 2010), WRF/Chem (Tuccella et al., 2012), and CMAQ (Appel et al., 2012), which respectively simulated 2003, 2004, 2007, and 2006. The Polair3D results are averaged over 2000–2008 for this comparison. The performance of $PM_{2.5}$ is comparable to that of the other models; CHIMERE shows better correlations but similar normalized mean bias (NMB), the correlation obtained with Polair3D is better than those of CALIOPE-EU and WRF/Chem, but WRF/Chem shows lower MNB and MNE than Polair3D. For sulfate, Polair3D and CHIMERE show good agreement with the observations on average (within 0.1 $\mu g\,m^{-3}$), whereas WRF/Chem underestimates significantly. For nitrate, all models overestimate the observations but the bias is lower for CHIMERE than for WRF/Chem and Polair3D. Performance results for ammonium are similar for Polair3D, CHIMERE, and WRF/Chem.

4 Dynamic evaluation

4.1 Data sets and method

A dynamic evaluation of an air quality model with respect to meteorology requires a long period to provide sufficient meteorological variability to evaluate the response of $PM_{2.5}$ concentrations to variations in meteorology. The long duration used here (2000–2008) allows one to perform such a dynamic evaluation and test the ability of the model to correctly reproduce the variability of the concentrations of $PM_{2.5}$ and of its components in response to meteorology (e.g. temperature, wind speed, precipitation). Available $PM_{2.5}$ EMEP measurements provide 23 stations, which give daily observations for at least a year, but only 5 stations have EMEP joint observations for $PM_{2.5}$, sulfate,

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nitrate and ammonium for the same period and for which a reasonable percentage of the data is available. The station locations are shown in Fig. 1. The five stations, which include joint observations of $PM_{2.5}$ and inorganic components, are AT02 in Austria (2003–2008), DE02 and DE03 in Germany (2006–2008), IT01 in Italy (2007–2008), and NO01 in Norway (2002–2008). NO01 does not provide daily $PM_{2.5}$ observations (only 2 to 3 times per week); however, it provides sulfate, nitrate, and ammonium observations on a daily basis. The AT02 station is located near Lake Neusiedl in Austria at 117 m above mean sea level (msl). The closest city is Illmitz (2416 inhabitants). The DE02 station is located at 74 m msl, in a forest with agriculture and meadows at a distance of 1 km and is surrounded by small stationary SO_2 and NO_x sources at distances greater than 1 km. The two largest and closest cities are Ützen (35 600 inhabitants, 22 km from the station) and Salzwedel (21 000 inhabitants, 27 km from the station). Local emissions from cars should not affect the measurements, as there are approximatively 3 cars per day within 3 km around the station. The DE03 station is situated on a mountain in the Black Forest, at 1205 m msl and surrounded by forests and meadows, where there is a minor agricultural activity for some parts of the year. Freiburg (206 000 inhabitants) is 12 km from the station and there are approximatively 5 vehicles per day within 10 km around the station. The high altitude of the DE03 station compared to the other sites should not impact the results of the dynamic evaluation, as both measured and simulated concentrations are surface data (Polair3D uses terrain-following coordinates). The IT01 station is located at 48 m msl and is 30 km from Rome. The station has the particularity to be inside the Reasearch Area of the National Council of Italy, therefore the site could be influenced by the vehicles of the research personel. There is no relevant industries near the area but there is a highway situated 1.7 km from the station. The proximity to a large city and a highway could lead to greater concentrations of primary PM, as well as some lesser influence on concentrations of secondary PM (i.e. sulfate, nitrate, and ammonium), but those should be taken into account by the model. The NO01 station is mainly surrounded by forest, meadow, freshwater lakes, low intensity agricultural areas; it is located at 190 m msl. There are some local known

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esmissions which have minor or negligible influence on the air quality of the site. Some local agricultural activities occasionally yield elevated ammonia concentrations.

For the observation data set, we used the EMEP observations for the pollutants, the ENSEMBLES[2] observations for temperature and precipitation (horizontal resolution of 0.5° × 0.5°), and the ERA Interim[3] data for wind speed (horizontal resolution of 1.5° × 1.5°). The Polyphemus/Polair3D simulation results were used for both $PM_{2.5}$ concentrations and meteorology of the simulation data set.

We computed for each station the correlations between the meteorology on a given day and the PM daily concentrations ranging from 0 to 10 days after for both observed and simulated values. We refer to the differences between those days as the lag (i.e. ranging from 0 to +10). For example, at lag = 0, the computed correlation corresponds to a day-to-day correlation. For lag = +10, the correlation is computed with the meteorology on a given day and the PM concentrations 10 days later, therefore, it may reflect the impact of meteorology on the PM concentrations 10 days later. We assume in our analysis that the association between a meteorological variable and a PM concentration reflects the impact of meteorology on PM levels. If the correlation is highest when the lag is 0, then the correlation represents an association between a meteorological variable and a PM concentration for the same day. It is the case at several stations (see station specific discussions below) for wind speed and precipitation. A negative correlation between precipitation and PM concentrations can be interpreted as wet scavenging of PM: greater precipitation leads to lower PM concentrations. Similarly, a negative correlation between wind speed and PM concentrations can be interpreted as greater dispersion of primary PM emissions (and/or emissions of precursors): greater wind speed leads to lower PM concentrations. Because the PM concentration is a 24-h average value, a meteorological event (e.g. precipitation) may impact the PM concentrations more the following day than the same day if it occurs near the end of that same day at a large spatial scale. The concentrations measured the following day will thus represent

[2]http://eca.knmi.nl/download/ensembles/ensembles.php

[3]http://www.ecmwf.int/research/era/do/get/era-interim

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an air mass that has been affected by the meteorological event. If the spatial scale of the meteorological event is significant and/or if there is stagnation, the "memory" of the meteorological event may last for several days and the correlation may remain significant for a few days. The correlation between PM and wind speed/precipitation reaches its maximum value (absolute value) for a lag equal to 0 or +1 and then tends towards 0 for the lag equal to 10. This suggests that wind speed/precipitation have little impact on PM beyond four days. The correlations between temperature and pollutants are nearly constant for a given station and a given pollutant. This suggests that temperature impacts PM over a much longer period compared to wind speed and precipitation. This behavior reflects the fact that temperature differences are significant among seasons and synoptic systems but show little day-to-day variation except for frontal passages.

We divide our dynamic evaluation into two parts. We first focus on the 23 stations that provide $PM_{2.5}$ measurements in Sect. 4.2. We conduct a greater depth analysis on the 5 stations that provide $PM_{2.5}$, sulfate, nitrate, and ammonium in Sect. 4.3.

4.2 Dynamic evaluation of $PM_{2.5}$ at EMEP stations

The correlations computed above may be represented with curves depicting the evolution of the correlations as a function of the day-lag (one graph per station). The large number of stations providing $PM_{2.5}$ data (23) prevents us from using this approach, which is used for a detailed analysis at the five stations that include $PM_{2.5}$ components. Instead, we choose to perform here a regression analysis to describe the evolution of these correlations as a function of the day lag, in both simulation and observation, allowing us a more compact presentation of these results (see Table 3). If both the regression coefficients and the best correlations are close in the observations and in the simulation for a given station and a given meteorological variable, then we assume that the evolution of the relationship between this meteorological variable and the $PM_{2.5}$ concentrations as a function of the day lag is well reproduced by the model.

The stations that give the better results are CH04, ES08, GB36, and GB48. At CH04, the regression coefficients for temperature are −0.01 in both simulation and

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observation, and the maximum correlation (0.18 in the observation, 0.11 in the simulation) occurs at lag = 0 in both cases. The regression coefficients for both simulation and observation are close to 0.04 for wind speed and precipitation, and with a maximum correlation of around −0.35 at lag = 1. ES08 presents similar results. At GB36, the regression coefficients for temperature, precipitation, and wind speed are respectively close to −0.01, 0.01, and 0.03 for both simulation and observation. The maximum correlations are also close for each variable in both simulation and observation. These results are even better at GB48.

The evolution of the correlation between the $PM_{2.5}$ concentrations and the precipitation is typically what is best reproduced by the model at most of the stations (AT02, CH02, DE02, DE03, DE04, DE44, ES07, ES09, ES10, ES12, ES14, IT01, and IT04). The correlation curves at most of these stations are nearly identical, which can be seen on the values of the regression coefficients, ranging from 0.01 to 0.04 among the stations, with close values between observation and simulation (with a difference of 0.01 except at ES15). The lag for which the best correlation is observed is lag = 1 for most of the cases and for both simulation and observation. The values of the best correlation coefficients are also close in both simulation and observation within 30 % at 17 out of 23 stations, for example, at CH02 (around −0.33), DE44 (around −0.22), ES11 (around −0.23), and IT01 (around −0.25). The differences between the values of the regression coefficients in simulated and observed data mainly come from a difference between the values of the correlation for lag = 0 and 1 (at ES10 for example). At other stations, the profile is the same between observation and simulation but with a slight constant difference (at ES11 and NO01 for example).

The evolution of the correlation between temperature and $PM_{2.5}$ concentrations is not as well reproduced by the model. The results are best at the AT02, ES10, IT01, and SI08 stations, where the curves are nearly identical. At these stations, the regression coefficients are either 0 or −0.01 and are the same in simulation and observation. The best observed correlation is around −0.45 at AT02, +0.15 at ES10, −0.18 at IT01, and around −0.10 at SI08 for both simulation and observation. Other stations present

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similar regressions, but with a constant difference between the values of the correlation between the observed and the simulated data (DE02, DE03, DE44, ES07, and ES12). For some other stations, the correlation is not well reproduced by the model (AT48, CH02, DE04, ES09, ES11, ES13, ES15, IT04, and NO01). For example, at the CH02 and IT04 stations, the best correlation is of opposite sign in the simulation compared to the observation.

The evolution of the correlation between wind speed and $PM_{2.5}$ concentrations is also difficult to correctly reproduce, mainly because of the resolution of the ERA Interim data (150 km instead of 50 km for Polyphemus and the ENSEMBLES data). The Italian station IT04 provides the best results with the two correlation curves nearly identical (regression coefficients of 0.02, and best correlation of around −0.25 observed at lag = 0 in both simulation and observation). However, at most of the stations, the profile of the evolution is correctly reproduced by the model but with a tendency of the model to overestimate the values of the correlations (at 21 out of 23 stations).

The ability of the model to predict changes in $PM_{2.5}$ in response to changes in meteorology may be summarized as follows. Correlations with precipitation are always negative as expected, as it removes PM from the atmosphere. The best correlation is observed for lag = 0 or lag = 1, and the regression coefficient is positive for both observation and simulation. We conclude that the model reproduces the effect of precipitation on PM correctly. Correlations with wind speed have the same profile as for precipitation, suggesting that greater wind speed disperses the polluted air mass more efficiently. This effect is reproduced by the model; however, it tends to be overestimated. Correlations with temperature are more difficult to reproduce. The best correlation coefficient can be either positive or negative, depending on the stations. The regression coefficients are almost always 0, suggesting that temperature impacts $PM_{2.5}$ in the same way for several consecutive days. The difficulty to correctly reproduce these correlations is mainly due to the fact that temperature has different effects on the $PM_{2.5}$ components (i.e. sulfate, nitrate, ammonium, organics, ...), unlike precipitation and wind speed, which impact $PM_{2.5}$ components in the same way. Exceptions are sea salt and

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soil dust, for which emissions increase with wind speed; however, these components are mostly present in coarse PM. A more detailed analysis could not be performed here, because most of these stations do not provide daily observations of sulfate, nitrate, and ammmonium. Such an analysis is conducted below for the 5 stations that have joint observations for $PM_{2.5}$, sulfate, nitrate, and ammonium.

4.3 Dynamic evaluation of $PM_{2.5}$ and its main components at EMEP stations

We analyze here the ability of the model to reproduce the effects of meteorology on $PM_{2.5}$ inorganic components, i.e. sulfate, nitrate, and ammonium, by comparing the correlations between concentrations of $PM_{2.5}$ components and meteorological variables obtained in the observations and simulation.

Figure 4 presents these correlations for the AT02 station in Austria. Figure 4a shows that the model represents well the variation of $PM_{2.5}$ as a function of changes in meteorology, particularly for temperature and precipitation. Correlations for temperature and precipitation versus $PM_{2.5}$ are nearly identical for both simulated and observed data, while correlations for wind speed versus $PM_{2.5}$ have the same profile, but not the same intensity (-0.38 for simulated data against -0.11 for observational data with a lag taken at 0). The evolution of sulfate, nitrate, and ammonium concentrations as a function of changes in precipitation and wind speed are well represented. For wind speed, the simulated data give a larger correlation in absolute value than the observations for all three species, which is consistent with the $PM_{2.5}$ results. The evolution of sulfate as a function of changes in temperature is well represented by the model, while the model gives a slightly larger correlation in absolute value for nitrate (~ -0.4 vs ~ -0.3) and ammonium (~ -0.25 vs ~ -0.3). Simulation data show that the negative correlation between temperature and $PM_{2.5}$ is driven by all the PM components (around -0.15 for sulfate, -0.2 for sea salt, -0.4 for nitrate, ammonium, and organic matter, and -0.6 for black carbon), except mineral dust (near 0). Lower temperatures in winter are associated with greater emissions of nitrogen oxides, sulfur dioxide, elemental carbon and primary organic carbon (residential heating and fossil-fuel fired

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power plants), which may explain the greater concentrations of sulfate, carbonaceous PM and nitric acid. Lower temperatures also favor the formation of semi-volatile ammonium nitrate. Higher temperatures favor emissions of biogenic precursors of secondary organic aerosol (SOA); however, the results suggest that the variability of primary organic aerosol (POA) dominates over that of SOA. Profiles for wind and precipitation are identical for all species with variation on the intensity of the correlation for a lag taken at 0 (the correlation then increases from lag = 0 to lag = +10), except for sea salt, for which the maximum value of the correlation is positive. Over the sea, increases in wind speed are often linked to increases in precipitation rate and lead to the emissions and subsequent transport of suspended sea salt particles. The negative correlation of sea salt with temperature may result from the association of low temperature with high wind speeds (winter storms).

Figure 5 presents correlations for the DE02 station in Germany. The evolution of sulfate as a function of changes in meteorology is well represented. As for AT02, the model reproduces well the evolution of PM concentrations as a function of changes in precipitation. Similarly, the observations show correlations between PM and wind speed that have similar profiles, but are greater than those of the simulation. The evolution of $PM_{2.5}$ as a function of changes in temperature is not as well represented by the model (−0.10 for the simulation against −0.25 for the observations) as for AT02, because of the difficulty of the model to correctly represent changes in nitrate and ammonium (correlations of −0.30 to −0.40 for the observations and of −0.05 to −0.10 for the model). Nitrate modeled concentrations are similar in winter ($3.7\,\mu g\,m^{-3}$) and in summer ($3\,\mu g\,m^{-3}$), whereas they are significantly different in the observations ($4.7\,\mu g\,m^{-3}$ in winter against $1.1\,\mu g\,m^{-3}$ in summer) (see Table 4). The differences between modeled and observed concentrations could be due to artifacts in the nitrate measurements in summer due to ammonium nitrate volatility (Hering and Cass, 1999), which would then explain the differences between the modeled and simulated correlations, but it could also result from uncertainties in the simulation, which, as shown in Sect. 3, overestimates nitrate. Simulation data show that the negative correlation between temperature and $PM_{2.5}$ is

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driven by all the PM components (around −0.10 for sulfate, nitrate, ammonium, and organic matter, −0.20 for sea salt, and −0.30 for black carbon), except mineral dust (around 0.15). As for AT02, profiles for wind speed and precipitation are identical for all species, except for sea salt.

The AT02 and DE02 stations show negative correlations between temperature and sulfate. Sulfate concentrations depend on SO_2 emissions (shown in Fig. 6), which may vary by season, and on the conversion rate of SO_2 to sulfate, which is greater in summer when oxidant concentrations are greater and kinetics faster. SO_2 emissions are greater over Poland in winter (i.e. when temperatures are low, which suggests that sulfate concentrations at these two stations are governed more by SO_2 emissions than by the kinetics of SO_2 to sulfate conversion). A first reason comforting this hypothesis is that the SO_2 emission impact is more visible at the AT02 station (correlation of around −0.3), which is closer to the SO_2 emission sources, than at the DE02 station (correlation of around −0.15). Furthermore, daily mean concentrations of sulfate are higher at these stations in winter (3.1 $\mu g\,m^{-3}$ at AT02 and 2.2 $\mu g\,m^{-3}$ at DE02) than in summer (2.2 $\mu g\,m^{-3}$ at AT02 and 1.6 $\mu g\,m^{-3}$ at DE02, see Table 4); these seasonal differences demonstrate that sulfate concentrations at AT02 and DE02 are more affected by SO_2 emissions than by the kinetics of sulfate formation.

Figure 7 shows the results for the DE03 station in Germany. The general evolution of $PM_{2.5}$ as a function of changes in meteorology is well represented by the model: the correlation coefficients for the simulation and observations have similar profiles, but they differ by about 0.10. The model correctly reproduces the evolution of sulfate as a function of changes in precipitation and wind speed. The modeled correlations for $PM_{2.5}$ and its components versus precipitation are between −0.2 and −0.35 at lag = 0 or +1 compared to observed correlations between −0.1 and −0.2 also at lag = 0 or +1. The modeled correlations for $PM_{2.5}$ and its components versus wind speed are between −0.4 and −0.5 at lag = 0 or +1 compared to observed correlations between −0.15 and −0.3 also at lag = 0 or +1. The correlations with wind speed and precipitation show better agreement for $PM_{2.5}$ and sulfate than for nitrate and ammonium. Nitrate

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modeled concentrations are four times greater than the observations, which may be the result of the overestimation of nitrate by the model and/or negative artifacts in the measurements. The model underestimates the correlation between temperature and sulfate (+0.1 vs +0.3), but it correctly reproduces the low correlation between temperature and both nitrate and ammonium. Simulation data show that the positive correlation between temperature and $PM_{2.5}$ is the result of low positive correlations with sulfate (0.05), mineral dust (0.20), and organic matter (0.25) and of low negative correlations with ammonium (−0.05), nitrate (−0.05), black carbon (−0.25), and sea salt (−0.15). The correlation between sulfate and temperature is positive (+0.3). This result is opposite to those at AT02 and DE02. It reflects the fact that this station is remote from large SO_2 emission sources that show strong seasonal variability and that it is impacted by SO_2 sources with low seasonal variability (e.g. maritime trafic). The fact that the model correctly reproduces this opposite response suggests that it represents the relationship between meteorology and sulfate formation during long-range transport correctly. The effect of wind speed and precipitation on concentrations of $PM_{2.5}$ and its components is similar to those at the previous stations.

Figure 8 presents correlations for the IT01 station in Italy. The evolution of $PM_{2.5}$ as a function of changes in temperature and precipitation is well represented by the model; for wind speed, the correlation coefficient has the same profile for the simulation and observations, but differs by around 0.30 with the model overestimating the strength of the anti-correlation. For a lag equal to or greater than 3, the correlation between wind speed and $PM_{2.5}$ is positive (up to 0.15 for a lag equal to 5). This profile, which differs from the other stations, is driven by nitrate and ammonium and is well represented by the model. The model correctly reproduces the evolution of sulfate, nitrate, and ammonium as a function of changes in temperature and precipitation (although the strength of the correlation between precipitation and nitrate is underestimated by the model). The correlation between sulfate and temperature is positive as for DE03 and, therefore, shows a relationship opposite to that obtained at AT02 and DE02. The model reproduces this correlation perfectly. The model overestimates the strength of

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the anti-correlation between wind speed and ammonium or nitrate. Simulation data show that the negative correlation between temperature and $PM_{2.5}$ is driven by ammonium (−0.2), nitrate (−0.35), black carbon (−0.5), sea salt (−0.15), and organic matter (−0.2).

Correlations between nitrate/ammonium and wind speed present large differences between the observations and the simulation at the AT02, DE02, DE03, and IT01 stations. Simulated nitrate is overestimated, which is a recurring issue in PM modeling over Europe. Moreover, there is less nitrate in summer than in winter in the simulation, significantly less at some stations (−63 % AT02, −20 % at DE02, −14 % at DE03, and −56 % at IT01, see Table 4). The correlations obtained with the model are strong and may reflect the availability of daily and gridded data. These strong correlations may not be seen in the observations because of the lower concentrations of nitrate in the measurements, especially in summer, and the fact that observations are not always available everyday.

Figure 9 presents results for the NO01 station in Norway. The evolution of $PM_{2.5}$ as a function of changes in temperature is not well represented (0.1 for the observations versus −0.1 for the simulation). In the observations, the dependence of $PM_{2.5}$ on temperature is driven by that of sulfate and ammonium, because nitrate shows very low correlations. The correlations between $PM_{2.5}$ and precipitation or wind speed are very low in the observations, which is a major difference with the four other sites. The model correctly reproduces this behavior with negative but low (between −0.15 and 0) correlations compared to those obtained at the other stations. The evolution of sulfate as a function of changes in meteorology is well represented, although the strength of the correlations is slightly overestimated for precipitation and wind speed. NO01 correlations are smaller in absolute value than the correlations at the other stations in both observations and simulation, especially for nitrate and ammonium. This can be explained by the fact that $PM_{2.5}$ observed mean concentrations at NO01 (4.4 $\mu g\,m^{-3}$) are significantly lower than those at most of the other stations (19.9 $\mu g\,m^{-3}$ at AT02, 12.9 $\mu g\,m^{-3}$ at DE02, 5.9 $\mu g\,m^{-3}$ at DE03, and 22 $\mu g\,m^{-3}$ at IT01), added to the fact

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that there is significantly fewer $PM_{2.5}$ data at the NO01 station, compared to the four other sites.

The positive correlations between temperature and sulfate at DE03, IT01, and NO01 suggest that atmospheric oxidation of SO_2 (favored by faster kinetics at higher temperatures and greater oxidant concentrations in summer) dominates over greater SO_2 emissions (expected with low temperatures in eastern Europe, see Fig. 6). The fact that there are no large sources of emissions around these stations (Fig. 6) and that daily mean concentrations of sulfate are higher in summer (1.5 $\mu g\,m^{-3}$ at DE03, 2.0 $\mu g\,m^{-3}$ at IT01, and 1.1 $\mu g\,m^{-3}$ at NO01) than in winter (1.4 $\mu g\,m^{-3}$ at DE03, 1.2 $\mu g\,m^{-3}$ at IT01, and 0.9 $\mu g\,m^{-3}$ at NO01) comforts this hypothesis.

5 Conclusions

A 9-yr air quality simulation has been conducted over Europe with the Polyphemus/Polair3D CTM. The results of the simulation were compared with available EMEP data and both an operational/diagnostic evaluation and a dynamic evaluation (with respect to meteorology) were conducted.

Modeled $PM_{2.5}$ concentrations vary over Europe by a factor of 6, from high concentrations of 36 $\mu g\,m^{-3}$ over northern Italy to low concentrations of 6 $\mu g\,m^{-3}$ over Scandinavia). $PM_{2.5}$ composition varies also significantly. For example, the $PM_{2.5}$ sulfate fraction is highest in eastern Europe, the nitrate fraction is highest in central Europe, and the organic fraction is highest in Scandinavia, Portugal, eastern France, and eastern Europe.

The operational/diagnostic evaluation shows that O_3 meets the model performance criteria and that $PM_{2.5}$, PM_{10}, and SO_4^{2-} meet the performance goal. NO_3^- and NH_4^+ are overestimated by the model; NH_4^+ meets the performance criteria, but NO_3^- does not. The correlation coefficients between simulated and observed data are 63 % for O_3, 57 % for PM_{10}, 59 % for $PM_{2.5}$, 56.5 % for SO_4^{2-}, 58 % for NH_4^+, and 42 % for NO_3^-. The

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comparison with other recent one-year model simulations shows that all models overestimate nitrate and that the bias for Polair3D is comparable to the bias for WRF/Chem, but greater to that for CHIMERE. The performance of $PM_{2.5}$, sulfate, and ammonium, which is comparable to that of the other models.

The dynamic evaluation shows that the evolution of $PM_{2.5}$ as a function of changes in meteorology is well represented for precipitation and wind speed overall, although the model tends to overestimate the $PM_{2.5}$ response to wind speed. Results are mixed for temperature because of the complex relationships between $PM_{2.5}$ components and temperature, but the model shows good agreement for half of the $PM_{2.5}$ stations. The correlations show that the response of $PM_{2.5}$ to changes in meteorology differs according to the location of the station and the meteorological variable considered. Wind and precipitation show mostly a strong negative correlation with pollutants (except for sea salt, for which a positive correlation is modeled) for lags of 0 or 1 day, and a correlation near 0 with a larger day lag. The correlation coefficient is nearly constant for temperature, for any lag and pollutant species. The response of $PM_{2.5}$ and sulfate to changes in temperature varies significantly among stations and can be opposite depending on the distance of the station from certain SO_2 emission regions with strong seasonality. These different responses are correctly reproduced by the model. The correlation profiles for observed data at the NO01 station differ from those at the other stations; the model also reproduces these differences correctly.

This dynamic evaluation is limited by the amount of data on $PM_{2.5}$ and its composition over large periods at European stations. Analyses of correlation between observed temperature and nitrate concentrations in the US have shown results that differed from modeled responses of nitrate to temperature (Tai et al., 2010). Such analysis in Europe would require to conduct the dynamic evaluation by season; however the observational data set is not sufficient to conduct a meaningful statistical analysis by season. Furthermore, it would be interesting to extent this analysis to carbonaceous $PM_{2.5}$, particulary since particulate organic matter displays a complex relationship to temperature. As the monitoring of $PM_{2.5}$ with chemical speciation increases over Europe, further dynamic

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evaluations should be conducted to test the ability of air quality models to reproduce the effect of meteorology on $PM_{2.5}$ concentrations and composition.

Appendix A

Comparison with other model studies in the context of AQMEII

See Table A1.

Appendix B

Comparison with model performance evaluations of one-year simulations over Europe

See Tables B1–B4.

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Table 1. Correlations between the simulated and observed data (ρ, expressed in %), RMSE, mean of the observations (μ_{obs}) and of the simulation (μ_{sim})(expressed in $\mu g\,m^{-3}$) for O_3, PM_{10}, $PM_{2.5}$, SO_4^{2-}, NO_3^-, and NH_4^+.

	Ozone				PM_{10}				$PM_{2.5}$			
Year	ρ	RMSE	μ_{obs}	μ_{sim}	ρ	RMSE	μ_{obs}	μ_{sim}	ρ	RMSE	μ_{obs}	μ_{sim}
2000	61.6	34.1	58.8	80.2	59.5	13	16.1	24.7	51.1	13	13.6	22.3
2001	64.0	28.6	56	74	61.0	9.9	18	18	62.0	8.5	14	16.5
2002	62.8	28.6	60.5	75.3	56.0	11	17.9	19.2	60.2	9.3	13.3	17.4
2003	65.0	28.4	64.2	75.6	69.3	11.8	20.4	19.5	61.6	10.4	15.3	17.5
2004	60.1	28.1	60.5	75	54.5	9.3	16.8	17.7	60.5	8.0	13.4	14.8
2005	59.9	27.7	63.7.	74.9	53.3	11.3	17.6	16.8	60.4	8.0	11.8	14.6
2006	65.3	27.1	61.5	74.2	48.9	11.4	18.7	17.3	52.9	9.8	15	14.3
2007	62.8	26.0	61.9	72.6	56.6	9.4	15.3	16.8	69.1	7.6	10.8	14.1
2008	65.1	25.2	58.3	72.1	49.4	9.3	15.4	16.1	51.3	7.0	10.5	11.8
Average 2000–2008	62.9	28.2	60.6	74.9	56.6	10.7	17.3	18.4	59.1	9	13.3	15.9

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Table 1. Continued.

Year	SO_4^{2-} ρ	RMSE	μ_{obs}	μ_{sim}	NO_3^- ρ	RMSE	μ_{obs}	μ_{sim}	NH_4^+ ρ	RMSE	μ_{obs}	μ_{sim}
2000	58.8	2.2	2.1	3.0	27.1	6.3	1.8	6.4	47.7	2.4	0.9	2.7
2001	53.3	1.7	2.2	2.3	28.6	3.1	1.8	3.4	47.5	1.1	1.1	1.4
2002	59.6	1.8	2.7	2.5	43.2	3.3	2.2	3.6	51.4	1.0	0.7	0.9
2003	60.5	2.0	2.7	2.5	52.2	2.9	2.1	3.3	70.8	1.0	0.6	1.1
2004	57.3	1.5	2.2	2.2	39.0	3.0	2	3.6	67.0	0.6	0.5	0.7
2005	50.4	1.6	2.2	2	39.4	2.9	2	3.3	55.0	1.0	1.0	1.5
2006	56.2	1.9	2.6	2.2	51.1	2.7	2.8	3.4	56.4	1.2	1.3	1.8
2007	57.0	1.3	1.9	1.8	44.3	2.7	1.9	2.9	60.9	1.0	1.0	1.5
2008	55.6	1.2	1.8	1.6	49.5	2.4	1.7	2.4	65.4	0.9	0.9	1.3
Average 2000–2008	56.5	1.7	2.3	2.2	41.6	3.3	2	3.6	58	1.1	0.9	1.4

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Table 2. Operational evaluation of the model using the criteria (a) of Russell and Dennis (2000) for ozone and of Boylan and Russell (2006) for PM and its components.

	Ozone		PM_{10}		$PM_{2.5}$		SO_4^{2-}		NO_3^-		NH_4^+	
Years	MNB	MNE	MFB	MFE	MFB	MFE	MFB	MFE	MFB	MFE	MFB	MFE
2000	17	23.8	47	54	55	62	28	48	89	103	84	89
2001	9.8	19.5	10	41	25	42	0	45	3	93	1	55
2002	7.6	19	17	44	36	50	−6	44	25	84	−7	55
2003	3.4	19.5	7	38	25	45	−9	43	28	76	43	59
2004	5.2	17.6	13	39	22	42	−3	40	29	83	−5	52
2005	1.2	18.1	8	45	33	48	−7	45	6	86	7	57
2006	2.4	17.3	3	40	10	40	−12	44	5	71	23	50
2007	0.9	17	21	45	37	53	−3	41	8	78	24	47
2008	0.2	14.1	11	40	25	47	−7	39	−8	75	17	40
Average 2000–2008	5.3	18.4	15.2	42.9	29.8	47.4	−2.1	43.2	20.5	83.2	20.8	56

(a) The performance criteria are |MNE| < 35 % and MNB < 15 % for ozone modeling; a threshold of 80 $\mu g\,m^{-3}$ was used here. The performance goal (resp. criterion) is met when |MFB| < 30 % (60 %) and MFE < 50 % (75 %) for PM modeling.

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Table 3. Dynamic evaluation for $PM_{2.5}$ (without speciation): regression analysis of the correlation coefficients between the $PM_{2.5}$ and temperature (T° C), precipitation (PR), and wind speed (WS) as a function of the day-lag. reg is the regression coefficient obtained from the regression analysis and r is the best correlation coefficient.

Station	correlation of $PM_{2.5}$ vs. T° C				correlation of $PM_{2.5}$ vs. PR				correlation of $PM_{2.5}$ vs. WS			
	obs		sim		obs		sim		obs		sim	
	reg	r	reg	r	reg	r	reg	r	reg	r	reg	r
AT02	0	−0.46	0	−0.43	0.02	−0.21	0.01	−0.19	0.02	−0.11	0.04	−0.46
AT48	0	−0.09	−0.01	0.14	0.03	−0.22	0.02	−0.19	0.02	−0.16	0.05	−0.45
CH02	0	−0.41	−0.01	0.11	0.03	−0.34	0.03	−0.30	0.03	−0.22	0.03	−0.41
CH04	−0.01	0.18	−0.01	0.11	0.03	−0.37	0.04	−0.32	0.04	−0.32	0.04	−0.38
DE02	0	−0.30	−0.01	−0.11	0.02	−0.23	0.02	−0.18	0.03	−0.25	0.04	−0.41
DE03	−0.01	0.32	−0.01	0.21	0.03	−0.27	0.03	−0.35	0.03	−0.30	0.04	−0.53
DE04	0	−0.12	−0.01	0.18	0.04	−0.39	0.03	−0.30	0.04	−0.38	0.05	−0.54
DE44	0	−0.31	−0.01	−0.14	0.03	−0.25	0.03	−0.22	0.04	−0.31	0.05	−0.47
ES07	−0.02	0.48	−0.01	0.26	0.01	−0.19	0.02	−0.32	0.01	−0.18	0.03	−0.31
ES08	−0.02	0.23	−0.01	0.10	0.02	−0.27	0.02	−0.22	0.03	−0.31	0.02	−0.24
ES09	−0.01	0.55	−0.01	0.06	0.01	−0.16	0.02	−0.20	0.03	−0.39	0.05	−0.53
ES10	−0.01	0.12	−0.01	0.16	0.01	−0.12	0.02	−0.28	0.02	−0.24	0.04	−0.50
ES11	−0.02	0.44	−0.01	−0.05	0.01	−0.23	0.02	−0.22	0.03	−0.35	0.04	−0.48
ES12	−0.01	0.38	0	0.20	0.01	−0.17	0.02	−0.20	0.03	−0.41	0.04	−0.57
ES13	−0.01	0.37	−0.01	−0.05	0.01	−0.21	0.02	−0.27	0.03	−0.37	0.05	−0.54
ES14	−0.01	−0.05	0	−0.10	0.01	−0.14	0.01	−0.09	0.04	−0.42	0.04	−0.53
ES15	−0.01	0.51	−0.01	−0.15	0.01	−0.16	0.03	−0.27	0.02	−0.25	0.04	−0.48
GB36	0	−0.13	−0.01	−0.10	0.01	−0.20	0.01	−0.18	0.03	−0.31	0.04	−0.40
GB48	−0.01	−0.19	0.01	−0.27	0.01	−0.17	0.01	−0.16	0.03	−0.19	0.04	−0.19
IT01	0	−0.18	−0.01	−0.18	0.03	−0.27	0.03	−0.24	0.03	−0.24	0.06	−0.53
IT04	0	−0.55	−0.01	0.12	0.01	−0.22	0.02	−0.29	0.02	−0.23	0.02	−0.27
NO01	−0.01	0.14	−0.01	−0.16	0	−0.17	0	−0.08	0	−0.20	0.01	−0.1
SI08	−0.01	−0.09	−0.01	−0.11	0.03	−0.29	0.03	−0.32	0	0.09	0.03	−0.26

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Table 4. Mean simulated and observed concentrations of sulfate, nitrate, and ammonium in winter (DJF) and in summer (JJA) at the five stations, expressed in $\mu g\,m^{-3}$.

	SO_4^{2-}				NO_3^-				NH_4^+			
	sim		obs		sim		obs		sim		obs	
	DJF	JJA	DJF	JJA	DJF	JJA	DJF	JJA	DJF	JJA	DJF	JJA
AT02	3.1	2.2	3.5	2.5	4.9	1.8	2.0	0.70	3.1	2.2	1.5	0.8
DE02	2.2	1.6	3.5	2.7	3.7	3.0	4.7	1.1	2.2	1.6	2.0	0.7
DE03	1.4	1.5	1.0	2.1	3.7	3.2	0.8	0.9	1.4	1.5	0.4	0.5
IT01	1.2	2.0	1.8	3.5	4.1	1.8	4.4	2.25	1.2	2.0	1.7	1.4
NO01	0.9	1.1	1.0	1.5	1.3	0.7	0.7	0.8	0.9	1.1	0.4	0.4

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Table A1. Comparisons to observations for surface PM_{10} and $PM_{2.5}$ over Europe (concentrations and RMSE are in $\mu g\,m^{-3}$) from 7 July to 31 August for this simulation (2000–2008) and the AQMEII models (2006).

	PM_{10}				$PM_{2.5}$			
	This work	AQMEII models			This work	AQMEII models		
		Min	Mean	Max		Min	Mean	Max
Number of stations	12	235	235	235	7	39	39	39
Mean observed	15.9	23.2	23.2	23.2	11.5	13.3	13.3	13.3
Mean simulated	16.6	6.2	12.9	23.4	16	5	12.3	21.4
RMSE	7.7	16.2	23.2	24.6	7.2	11.4	24.1	69.2
Correlation	57.3 %	8.2 %	17.3 %	25 %	67.6 %	3.2 %	11.8 %	21.1 %
MFB	7.6 %	−111.0 %	−64.3 %	3.9 %	32.7 %	−85.7 %	−30.5 %	44.9 %
MFE	35.4 %	44.5 %	80.8 %	113 %	43.4 %	55.1 %	72.3 %	94.2 %

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Table B1. Comparison for $PM_{2.5}$ with Péré et al. (2010), Pay et al. (2010), Tuccella et al. (2012), and Appel et al. (2012).

	Péré et al.	Pay et al.	Tucella et al.	Appel et al.	This work
Obs ($\mu g\,m^{-3}$)	12.2	13.0	12.6		13.3
Sim ($\mu g\,m^{-3}$)	15.1	6.3	8.6		15.9
Correlation (%)	73	45	41		59
RMSE ($\mu g\,m^{-3}$)		11.6			9
MFB (%)		−74			29.8
MFE (%)		81			47.7
MNB (%)			−7.3		62
MNE (%)			59.6		76
NMB (%)	24.2			−46.6	26.4
NME (%)				55.2	53.1

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Table B2. Comparison for sulfate with Péré et al. (2010) and Tuccella et al. (2012).

	Péré et al.	Tucella et al.	This work
Obs ($\mu g\,m^{-3}$)	3.9	2.4	2.3
Sim ($\mu g\,m^{-3}$)	4.0	0.9	2.2
Correlation (%)	50	50	56
MNB (%)		−46.9	16.6
MNE (%)		64.9	51.3
NMB (%)	4.25		4.5

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Table B3. Comparison for nitrate with Péré et al. (2010) and Tuccella et al. (2012).

	Péré et al.	Tucella et al.	This work
Obs ($\mu g\,m^{-3}$)	3.1	2.9	2
Sim ($\mu g\,m^{-3}$)	4.6	4.4	3.6
Correlation (%)	59	48	42
MNB (%)		115.2	123.8
MNE (%)		169.3	163.7
NMB (%)	36.7		112.2

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Table B4. Comparison for ammonium with Péré et al. (2010) and Tuccella et al. (2012).

	Péré et al.	Tucella et al.	This work
Obs ($\mu g\,m^{-3}$)	2.1	1.8	0.9
Sim ($\mu g\,m^{-3}$)	3.1	1.7	1.4
Correlation (%)	60	57	58
MNB (%)		96.4	59.9
MNE (%)		139	86.2
NMB (%)	48.5		47.7

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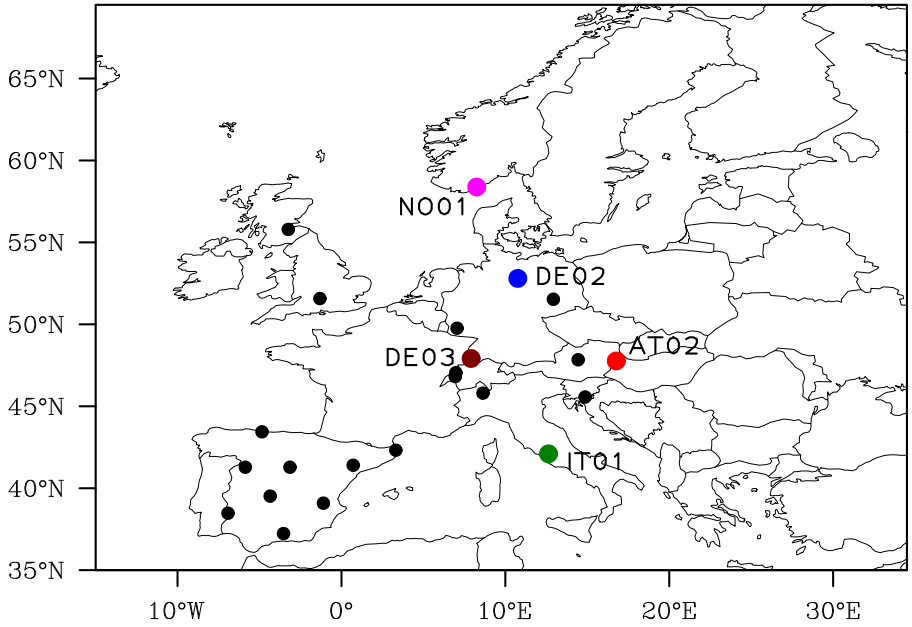

Fig. 1. Geographical domain of the simulation and EMEP stations used for the dynamic evaluation (Sect. 4). The stations with a colored dot provide data for $PM_{2.5}$, sulfate, nitrate, and ammonium, while those with a black dot only provide $PM_{2.5}$ data.

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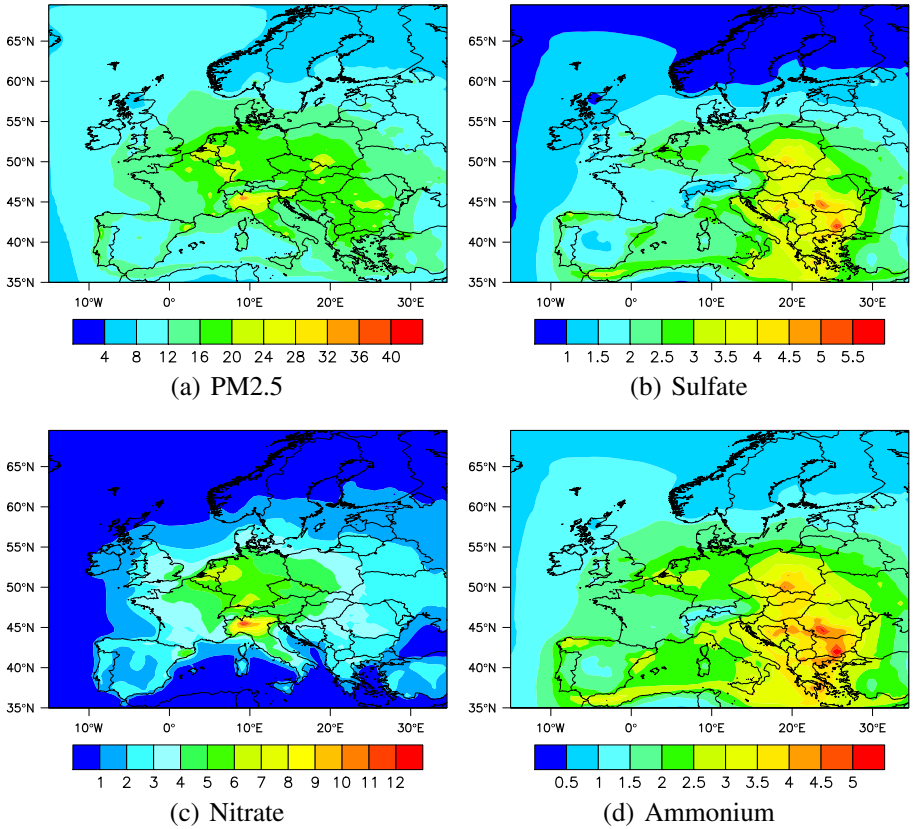

Fig. 2. Nine-year averaged surface concentrations of $PM_{2.5}$ and its components, expressed in $\mu g\,m^{-3}$.

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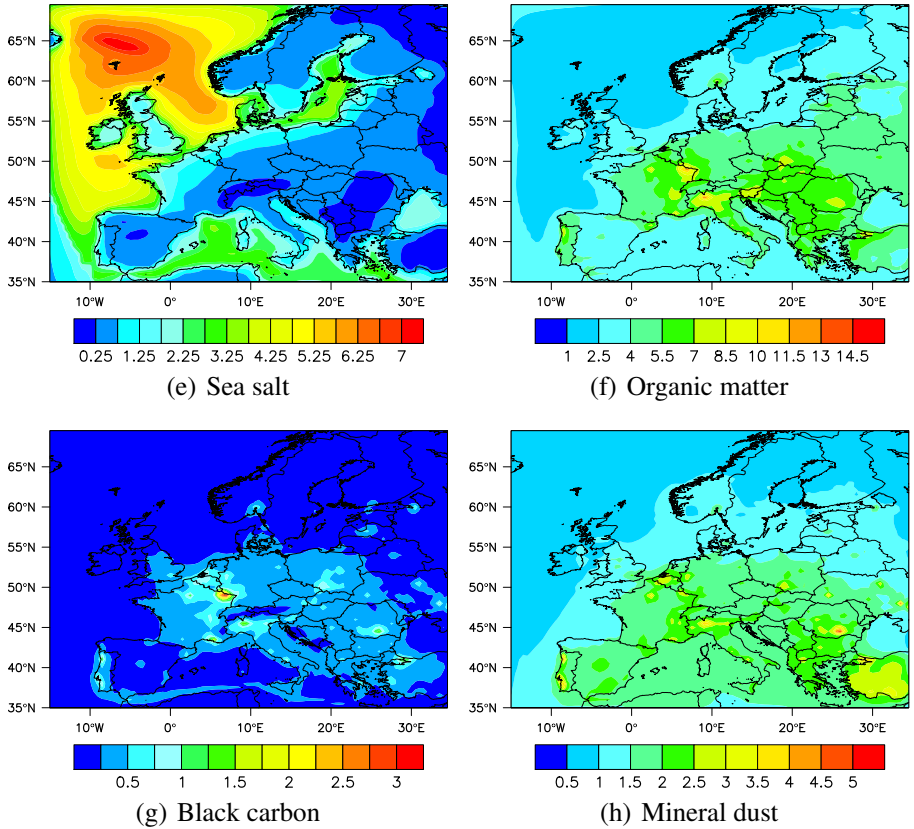

(e) Sea salt (f) Organic matter

(g) Black carbon (h) Mineral dust

Fig. 2. Continued.

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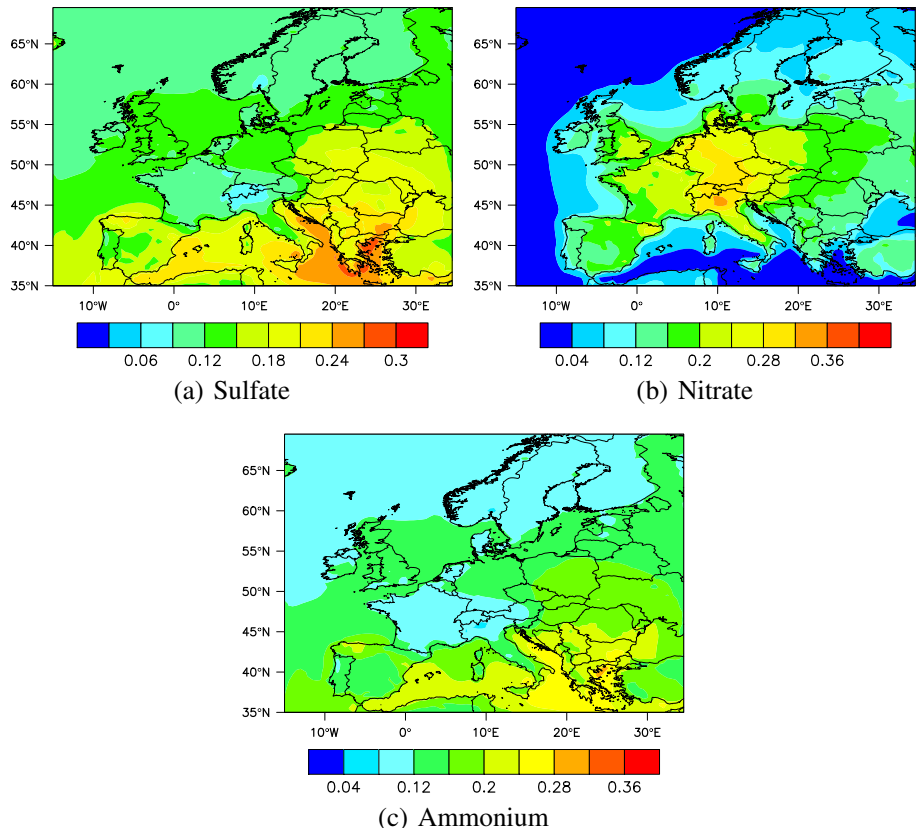

Fig. 3. Nine-year averaged surface fractions of sulfate, nitrate, ammonium, sea salt, organic matter, black carbon and mineral dust in $PM_{2.5}$.

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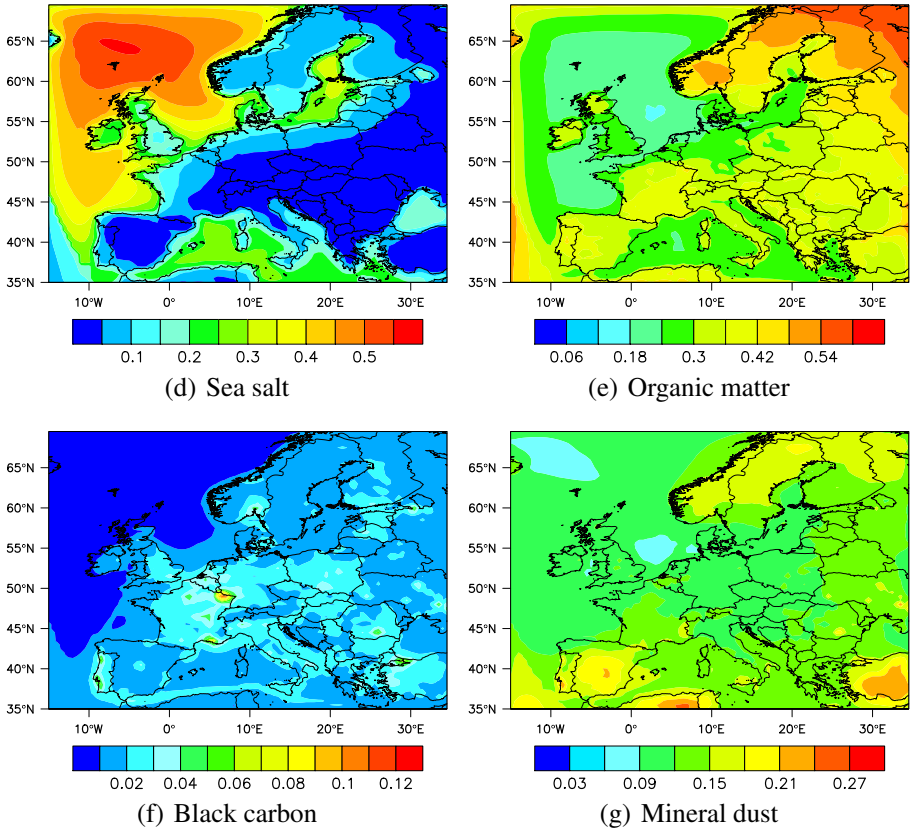

(d) Sea salt (e) Organic matter

(f) Black carbon (g) Mineral dust

Fig. 3. Continued.

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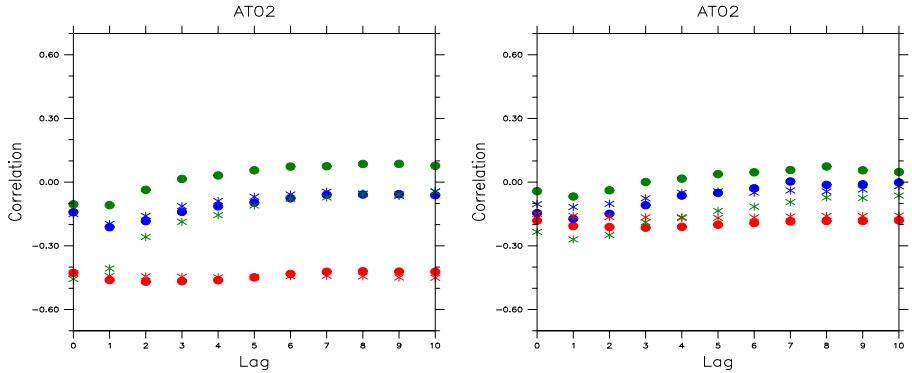
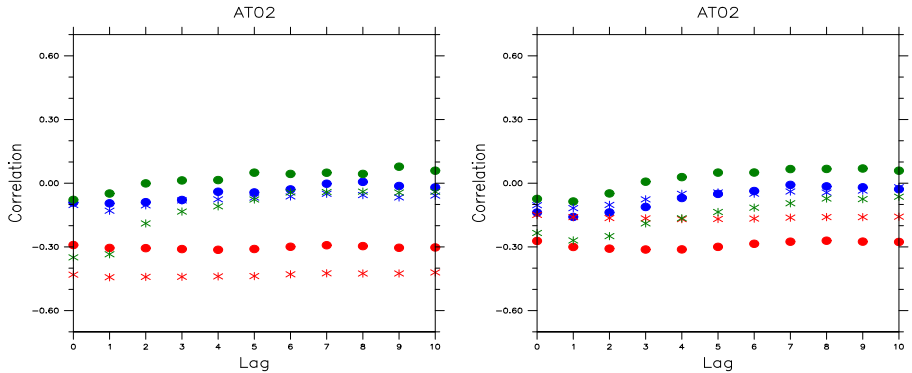


(a) PM2.5

(b) Sulfate



(c) Nitrate

(d) Ammonium

Fig. 4. Correlations between meteorology (temperature in red, precipitation in blue, wind speed in green) on a given day and pollutant concentrations ($PM_{2.5}$, sulfate, nitrate and ammonium), ranging up to 10 days after at AT02 (2003–2008). Simulated data are represented with a star, while observational data are represented with a dot.

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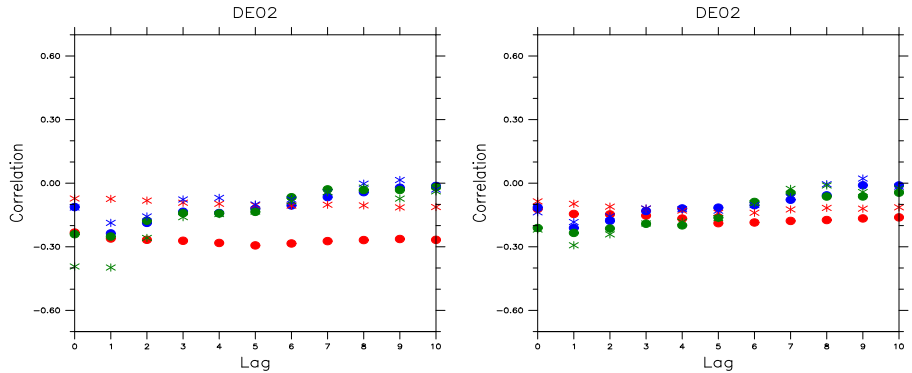
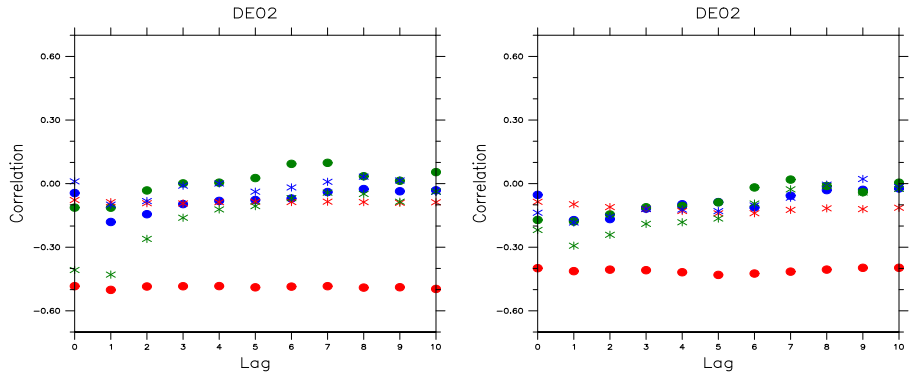


(a) PM2.5

(b) Sulfate



(c) Nitrate

(d) Ammonium

Fig. 5. Correlations between meteorology (temperature in red, precipitation in blue, wind speed in green) on a given day and pollutant concentrations ($PM_{2.5}$, sulfate, nitrate and ammonium), ranging up to 10 days after at DE02 (2006–2008). Simulated data are represented with a star, while observational data are represented with a dot.

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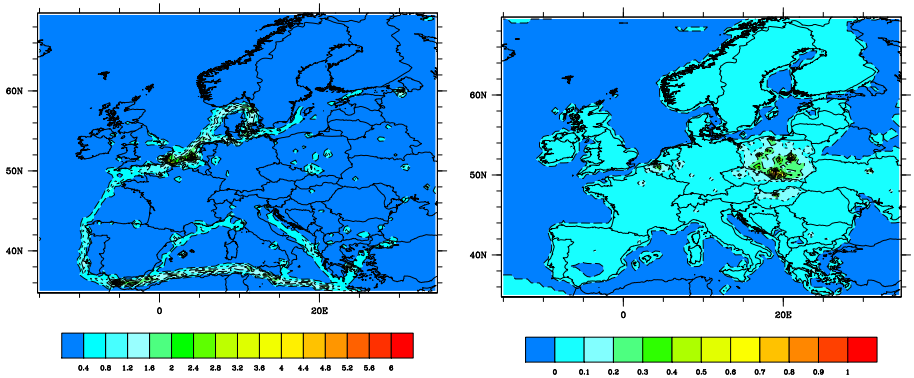

(a) Winter (b) Δ = Winter - Summer

Fig. 6. Average SO_2 emissions over Europe in winter, and difference (Δ) between winter and summer, expressed in $\mu g\,m^{-3}$.

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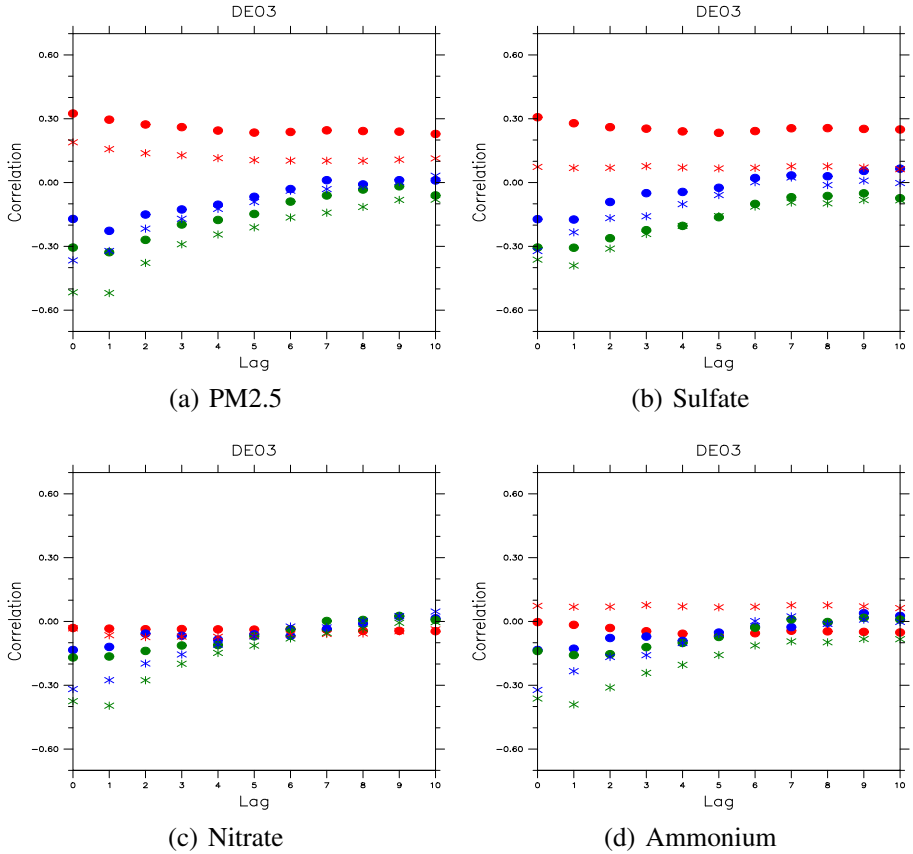

Fig. 7. Correlations between meteorology (temperature in red, precipitation in blue, wind speed in green) on a given day and pollutant concentrations ($PM_{2.5}$, sulfate, nitrate and ammonium), ranging up to 10 days after at DE03 (2006–2008). Simulated data are represented with a star, while observational data are represented with a dot.

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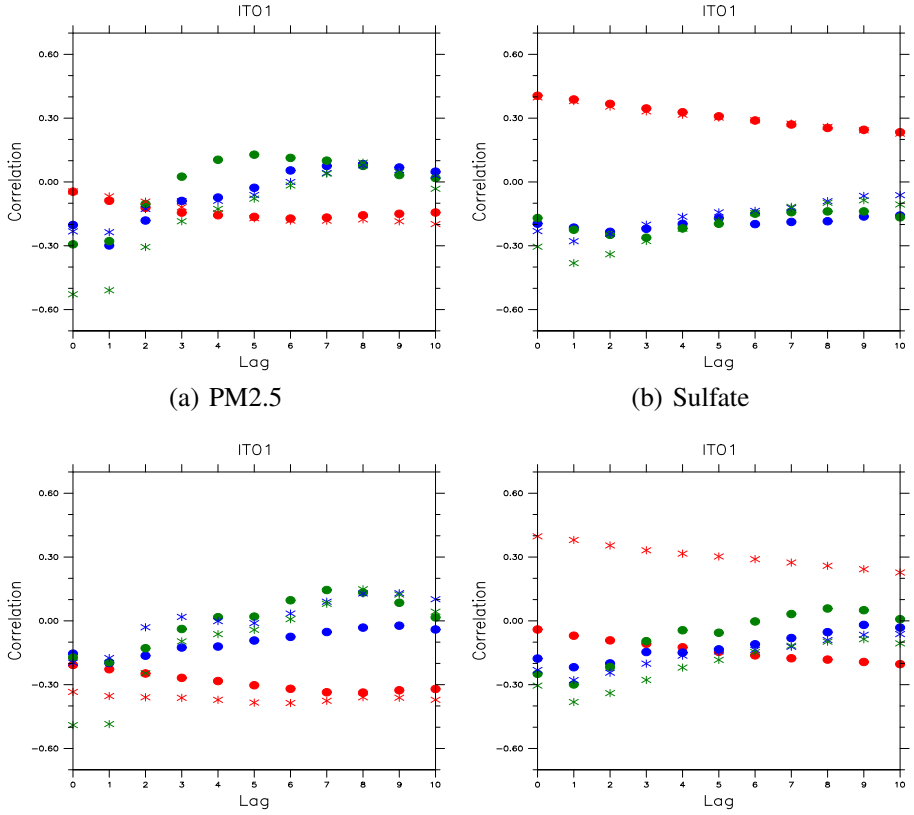

Fig. 8. Correlations between meteorology (temperature in red, precipitation in blue, wind speed in green) on a given day and pollutant concentrations ($PM_{2.5}$, sulfate, nitrate and ammonium), ranging up to 10 days after at IT01 (2007–2008). Simulated data are represented with a star, while observational data are represented with a dot.

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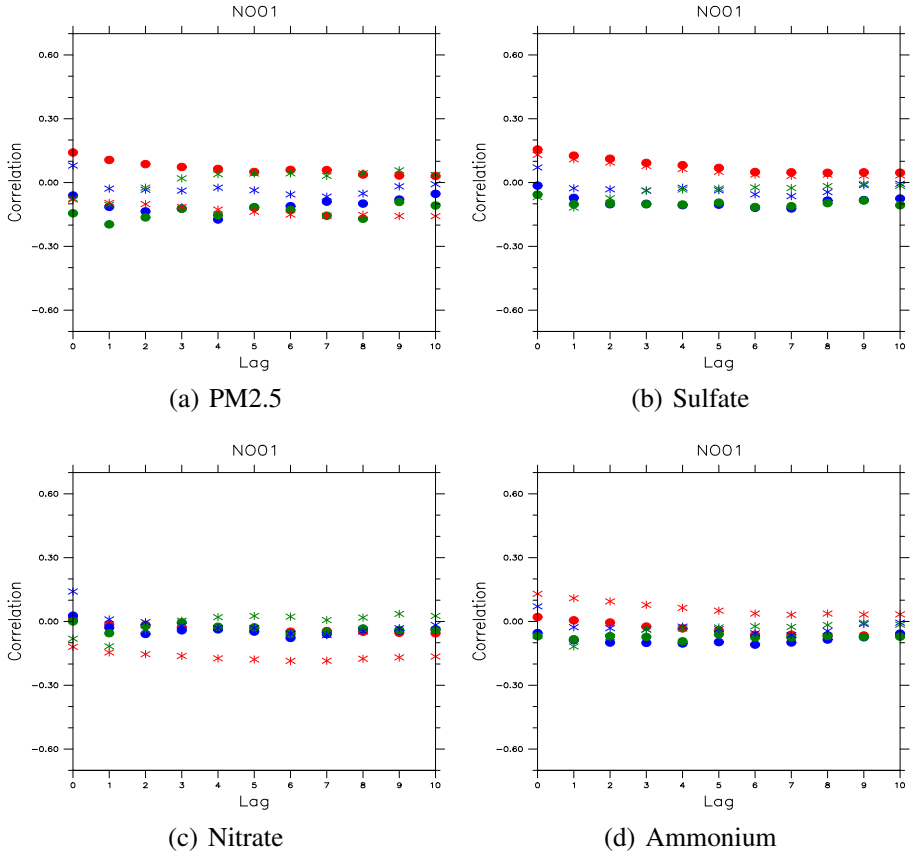

Fig. 9. Correlations between meteorology (temperature in red, precipitation in blue, wind speed in green) on a given day and pollutant concentrations ($PM_{2.5}$, sulfate, nitrate and ammonium), ranging up to 10 days after at NO01 (2002–2008). Simulated data are represented with a star, while observational data are represented with a dot.

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