



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

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Abstract. A 9 yr 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 ($36 \mu\text{g m}^{-3}$) 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 criterion. 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 1 yr 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), which 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 tem-

perature. 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.

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, although uncertainties remain concerning semivolatile $PM_{2.5}$ components.

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 $2.5 \mu\text{m}$ ($PM_{2.5}$) are regulated in North America and Europe as a consequence of their effects on human health (respiratory and cardiovascular diseases). $PM_{2.5}$ 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 traffic, 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 nonvolatile and semivolatile 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., 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., 2011), 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 semivolatile 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. To date, there have only been a few studies about the effect of climate on PM_{2.5} over Europe, as a consequence of the limited amount of daily PM_{2.5} observations. Nevertheless, some observations provide also PM_{2.5} chemical composition, which is important to understand the effect of meteorology on PM_{2.5}, thus the present study focuses on PM_{2.5}.

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 in-

formation 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 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 of ozone (O₃) precursors over the United States (e.g., Gilliland et al., 2008; Yarwood et al., 2003) and to the response of O₃, nitrogen dioxide (NO₂), and PM₁₀ over Europe (Colette et al., 2011). 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. Such an assessment of model performance appears needed since air quality models are increasingly being used to investigate the effect of climate change on future PM concentrations.

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 1 yr model performance evaluations for PM₁₀, 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 9 yr (2000–2008) of concentrations of gaseous and particulate pollutants over Europe. Polyphemus is an air quality modeling platform that has been used for many applications at different scales (from local to continental). Polair3D is the chemical-transport model (CTM) of Polyphemus.

The modeling domain covers a geographical area that 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 reanalyses of the European Centre for Medium-Range Weather Forecasts (ECMWF). The horizontal resolution (both longitude and latitude) for specific years 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 Programme¹ (EMEP) inventory for 2000 to 2008 for all sectors. Surface emissions were directly injected into the model surface layer and elevated point source emissions were injected into the model layers following the EMEP table for vertical distribution. No horizontal disaggregation was needed because the model and EMEP inventory have similar grid spacings. Temporal profiles by source sectors provided by EMEP were used. Biogenic emissions were computed with the Model of Emissions of Gases and Aerosols from Nature (MEGAN, Guenther et al., 2006). NO_x from soils was excluded from SNAP10 of the EMEP inventory to avoid double counting. Sea salt emissions are parameterized following Monahan et al. (1986). Forest fire emissions were not considered as they were not available. Moreover, modeling the impact of forest fires is still an area of research due to uncertainties in emission rates and smoke plume heights.

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).

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 6 h 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 year 2004 alone could have been considered for those boundary conditions since emission control policies led to decreases in emissions in Europe and North America over the period of interest. However, boundary conditions depend also on meteorology, and a mean over 5 yr provides more robustness. The MOZART-4 chemical mechanism includes 85 gas-phase species, 12 bulk aerosol compounds, 39 photolysis and 157 thermal gas-phase reactions. Dust and sea salt aerosol data are distributed in

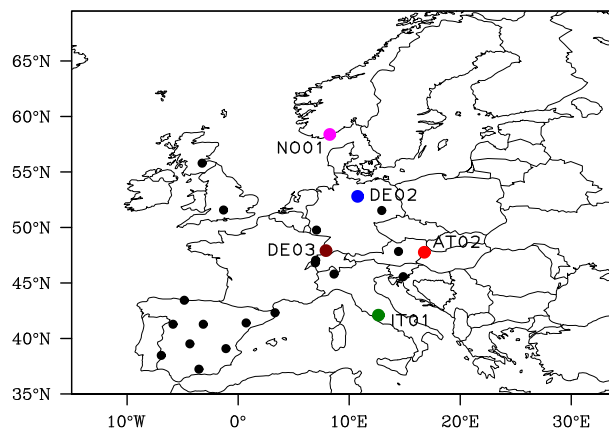


Fig. 1. Geographical domain of the simulation and EMEP stations used for the dynamic evaluation (Sect. 4). The stations with a colored dot provide data for PM_{2.5}, sulfate, nitrate, and ammonium, while those with a black dot only provide PM_{2.5} data.

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 (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. We used 60 species from MOZART-4 in the boundary conditions. The MOZART-4 VOC species were matched to CB05 following information available in Emmons et al. (2010). For example, BIGALK is considered as ALK3 and thus matched to 3.0 PAR, BIGENE to 1.0 PAR + 1.0 OLE, and TOLUENE to TOL (instead of a combination of TOL and XYL). The aerosol species were also matched to Polair3D species. Most of the species were directly matched to Polair3D, except OC1 and OC2, which were converted to 2.25 PSOAI_P + 2.88 PSOAm_P + 3.87 PSOAh_P. These coefficients, which depend on the SVOC/POA and OM/OC ratios are taken from Couvidat et al. (2013).

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). Aerosol dynamics (coagulation, nucleation, condensation, and evaporation) is treated. Inorganic PM is simulated with the ISORROPIA thermodynamic model (Nenes et al., 1998) and organic PM is modeled with a hydrophilic/hydrophobic organic (H²O) model using a molecular surrogate approach (Couvidat et al., 2012). Nitrate aerosol in the coarse fraction is treated. The dry deposition velocities for gases are preprocessed with the parameterization of Zhang et al. (2003). Vertical diffusion is

¹<http://emep.int/>

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 $\mu\text{g m}^{-3}$), the Netherlands (25 $\mu\text{g m}^{-3}$), northeastern Spain and France, and Eastern European countries (around 22 $\mu\text{g m}^{-3}$), while concentrations are lower in the northern and easternmost parts of Europe and in Spain (between 4 and 14 $\mu\text{g m}^{-3}$). Figure 2b depicts higher concentrations of sulfate over Eastern Europe, with two peaks in Romania and Bulgaria (5.5 $\mu\text{g m}^{-3}$) and a general south–north gradient and a land–ocean gradient. Figure 2c shows high concentrations of nitrate at the center of the domain (up to 12 $\mu\text{g m}^{-3}$ over northern Italy), while low concentrations are less than 4 $\mu\text{g m}^{-3}$ over the rest of the continent. High concentrations of ammonium are localized in northern Italy, Benelux, and southern Poland (Fig. 2d), with a maximum of 5 $\mu\text{g m}^{-3}$ in the Milan area. As expected, sea salt concentrations are highest over the Atlantic Ocean (between 4 and 7 $\mu\text{g m}^{-3}$), the Mediterranean Sea and the Baltic Sea (between 2.5 and 4 $\mu\text{g m}^{-3}$), and along the coasts of the countries bordering the sea and the ocean (between 2 and 3 $\mu\text{g m}^{-3}$). Sea salt concentrations are near zero on the continent (Fig. 2e). There is a sea salt gradient in the northwestern part of the domain, which is a consequence of two factors: high sea salt emissions in this part of the domain, and different parameterizations used in the model computation of sea salt emissions (Monahan et al., 1986) and in the boundary conditions (Mahowald et al., 2006). PM_{2.5} organic matter concentrations are high in northwestern Portugal, eastern France, and over northern Italy (between 8.5 and 13 $\mu\text{g m}^{-3}$). Slovenia, Poland, and Romania also show high concentrations of organic matter (around 8 $\mu\text{g m}^{-3}$) (Fig. 2f). As expected, there is an ocean–land gradient since both anthropogenic and biogenic emissions are higher on land than over water due to the presence of vegetation and human activities. Black carbon concentrations are below 1 $\mu\text{g m}^{-3}$ over all of Europe, except for the northeastern part of France (3 $\mu\text{g m}^{-3}$) and in some localized areas in France, Italy, and Romania (1.25 $\mu\text{g m}^{-3}$) (Fig. 2g). The concentrations of mineral dust vary from 1.5 to 5 $\mu\text{g m}^{-3}$ below 52° N, while they vary from 0.75 to 1.5 $\mu\text{g m}^{-3}$ 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} con-

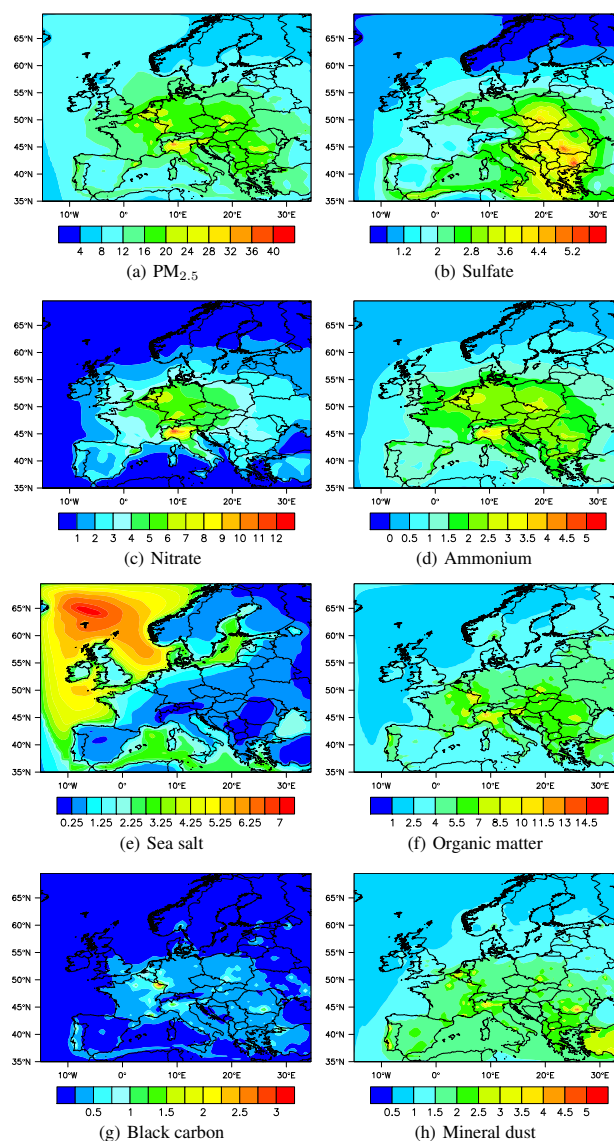


Fig. 2. Nine-year (2000–2008) averaged surface concentrations of PM_{2.5} and its components, expressed in $\mu\text{g m}^{-3}$.

sists mainly of mineral dust (from 14 % to 20 %), nitrate for the southernmost (and most industrial) part of Scandinavia (12 to 30 %), and organic matter (from 40 % to 60 %). The MEGAN emissions show high monoterpene emissions over Scandinavia, which, combined with the low pollution in this region, explain the high fraction of organic matter in PM_{2.5}. Nitrate, ammonium, and organic matter account for around 60 % of PM_{2.5} in Germany, Switzerland, Austria, and northern Italy, while sulfate and mineral dust represent together around 20 %.

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 around 20 % sea salt. Organic matter

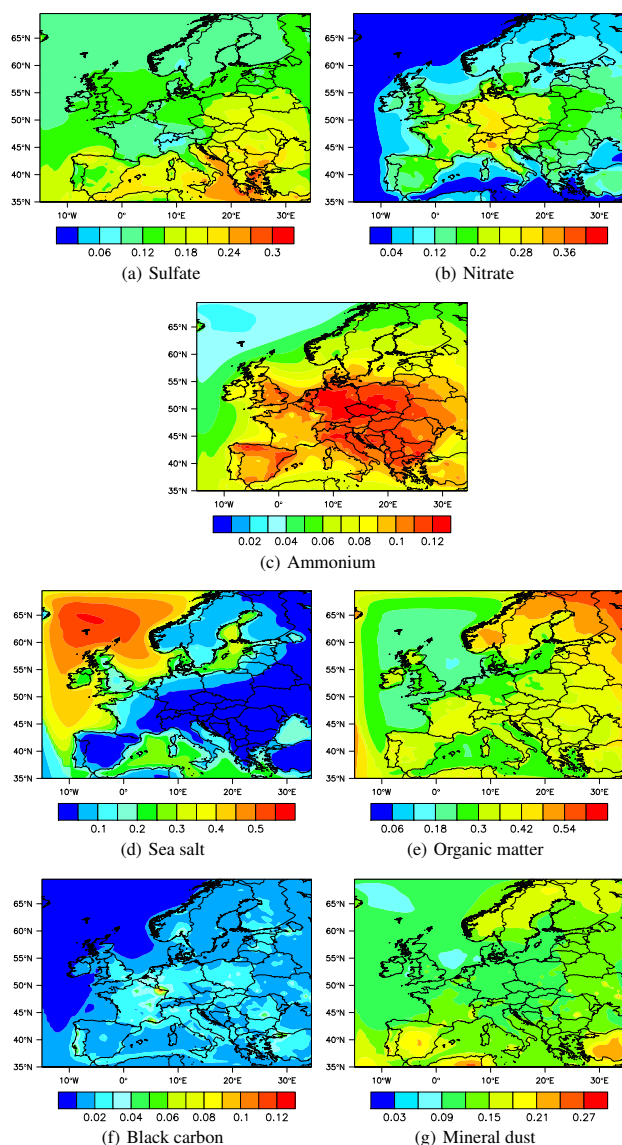


Fig. 3. Nine-year (2000–2008) averaged surface fractions of sulfate, nitrate, ammonium, sea salt, organic matter, black carbon, and mineral dust in PM_{2.5}.

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 of Europe, except for some localized areas in Portugal, France, Romania, and Turkey (4 % to 8 %) as well as a peak of 10 % that is observed at the border of France with Luxembourg and Germany.

3 Operational evaluation

3.1 Statistical scores

The operational evaluation was performed for each year for ozone (O₃) and PM. Available PM measurements include PM₁₀ and PM_{2.5} mass concentrations and PM_{2.5} sulfate (SO₄²⁻), nitrate (NO₃⁻), and ammonium (NH₄⁺) concentrations. Although organic carbon and elemental carbon data are available at the EMEP stations for 1 yr (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) (μg m⁻³) are presented in Table 1 to provide a common overview of model performance for O₃ 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 U.S. 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 μg m⁻³ to evaluate hourly O₃. A threshold of 80 μg m⁻³ was used here. The suggested performance criteria are |MNB| ≤ 15 % and MNE ≤ 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 ≤ 75 % and |MFB| ≤ 60 %.

The total number of stations that provide data for at least 1 yr in the whole period is 91 for O₃, 77 for SO₄²⁻, 34 for NH₄⁺, 33 for NO₃⁻ and PM₁₀, and 22 for PM_{2.5}. If the number of stations for which data are available each year is about constant for O₃ (around 70 stations), SO₄²⁻ (around 20 stations), and PM₁₀ (around 10 stations), that number is more variable for the other species. For example, 20 stations 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 remote rural background stations, 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, artifacts associated with NO₃⁻ and NH₄⁺ measurements occur due to evaporation (or condensation) of semivolatile ammonium nitrate (NH₄NO₃) from the particles collected on the filter due to fluctuations in temperature and relative humidity and/or pressure drop across the filter, which perturb the gas-particle equilibrium. In California these uncertainties have been estimated to be up to 30 % (Hering and Cass, 1999); however, they could be less

Table 1. Temporal 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 $\mu\text{g m}^{-3}$) for O₃ (hourly basis), PM₁₀, PM_{2.5}, SO₄[−], NO₃[−], and NH₄⁺ (daily basis).

Year	O ₃				PM ₁₀				PM _{2.5}			
	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	13.0	16.1	24.7	51.1	13.0	13.6	22.3
2001	64.0	28.6	56.0	74.0	61.0	9.9	18.0	18.0	62.0	8.5	14.0	16.5
2002	62.8	28.6	60.5	75.3	56.0	11.0	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	75.0	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	15.0	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	9.0	13.3	15.9
Year	SO ₄ [−]				NO ₃ [−]				NH ₄ ⁺			
	ρ	RMSE	μ_{obs}	μ_{sim}	ρ	RMSE	μ_{obs}	μ_{sim}	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	2.0	3.6	67.0	0.6	0.5	0.7
2005	50.4	1.6	2.2	2.0	39.4	2.9	2.0	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	2.0	3.6	58	1.1	0.9	1.4

Table 2. Operational evaluation of the model using the criteria* of Russell and Dennis (2000) for O₃ and of Boylan and Russell (2006) for PM and its components (expressed in %).

Years	O ₃		PM ₁₀		PM _{2.5}		SO ₄ [−]		NO ₃ [−]		NH ₄ ⁺	
	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

* The performance criteria are |MNE| < 35 % and MNB < 15 % for O₃ modeling; a threshold of 80 $\mu\text{g m}^{-3}$ was used here. The performance goal (resp. criterion) is met when |MFB| < 30 % (60 %) and MFE < 50 % (75 %) for PM modeling.

in Europe, where most NH₄NO₃ formation occurs during the cold season. An unbiased uncertainty of 15 % has been reported for NO₃[−] measurements (Schaap et al., 2004). 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 $\mu\text{g m}^{-3}$. On average, hourly O₃ is overestimated by about 23 %, the correlation coefficient is 62.9 %, and the RMSE is 28.2 $\mu\text{g m}^{-3}$ (the observed mean concentration is 60.6 $\mu\text{g m}^{-3}$). The criteria of Russell and Dennis (2000) are met with an MNE of 18.4 % and an MNB of 5.3 % on

average. Those criteria are met for all years except 2000, when MNB is 17 % (instead of ≤ 15 %, see Table 2).

PM₁₀ is estimated with a correlation coefficient of 56.6 % and a RMSE of $10.7 \mu\text{g m}^{-3}$ on average (the observed mean concentration is $17.3 \mu\text{g m}^{-3}$). 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 an RMSE of $9 \mu\text{g m}^{-3}$ (the observed mean concentration is $13.3 \mu\text{g m}^{-3}$) and a correlation coefficient of 59 %. On average, MFB (29.8 %) and MFE (47.4 %) also meet the performance goals. These performance goals are met for 5 yr (2001, 2003, 2004, 2006, and 2008); for the other years, PM_{2.5} meets the performance criteria. SO₄⁻ provides the best results: the correlation coefficient is 56.5 % and the RMSE is $1.7 \mu\text{g m}^{-3}$ on average (the observed mean concentration is $2.3 \mu\text{g m}^{-3}$). Simulated concentrations are on average 4 % lower than the observations. Both MFB (-2.1 %) and MFE (43.2 %) meet the model performance goals for every single year. Model performance is lower for NO₃⁻ and NH₄⁺. Simulated concentrations are overestimated compared to the observed concentrations (80 % for NO₃⁻ and 55 % for NH₄⁺ on average). NH₄⁺ has a better correlation coefficient and RMSE (58 % and $1.1 \mu\text{g m}^{-3}$, the observed mean concentration is $0.9 \mu\text{g m}^{-3}$) than NO₃⁻ (42 % and $3.3 \mu\text{g m}^{-3}$, the observed mean concentration is $2 \mu\text{g m}^{-3}$) because a fraction of NH₄⁺ is associated with SO₄⁻. NO₃⁻ does not meet the performance criteria (MFB=20.5 % and MFE=83.2 % on average), but NH₄⁺ does (MFB=20.8 % and MFE=56 %). For NO₃⁻ the performance criteria are met for 4 yr (2003, 2006, 2007, and 2008). For NH₄⁺ the performance goals are met for the last 3 yr (2006–2008), and the performance criteria for all years except 2000.

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 the Polyphe-mus/Polair3D model and nine other models, applied 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 PM₁₀ and PM_{2.5}. We compare here our model performance evaluation to that of the other 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. It is interesting to ensure that our model performance did not degrade when simulating a longer time period. The observational data for the AQMEII model evaluation include stations

from the AirBase and EMEP databases, while we only used the latter (AirBase does not provide data for the components of PM, while EMEP does). Table A1 summarizes the statistics obtained for PM₁₀ and PM_{2.5}.

Daily PM₁₀ is slightly overestimated by Polair3D over 2000–2008 ($15.9 \mu\text{g m}^{-3}$ measured against $16.6 \mu\text{g m}^{-3}$ simulated), while the AQMEII model concentrations are on average $10 \mu\text{g m}^{-3}$ 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 average 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 PM₁₀ for 10 model simulations in the context of AQMEII for the whole of year 2006. The RMSE ranges from 7.3 to $15.2 \mu\text{g m}^{-3}$ for the different models, which is consistent with the RMSE obtained here, which is $10.7 \mu\text{g m}^{-3}$ on average (see Table 1). The MFB for PM₁₀ 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 ($11.5 \mu\text{g m}^{-3}$ measured against $16 \mu\text{g m}^{-3}$ simulated on average). The AQMEII models show both under and overestimations ($13.3 \mu\text{g m}^{-3}$ measured against a range of 5 to $21.4 \mu\text{g m}^{-3}$ simulated). The Polair3D average RMSE ($7.2 \mu\text{g m}^{-3}$) 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.

3.3 Comparison with the performance evaluation of 1 yr simulations

We also compared our model performance evaluation to that of four other chemical-transport models that have been used for 1 yr simulations 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.

All models simulated different years, with different models and configuration. Péré et al. (2010) used respectively EMEP and MEGAN to generate anthropogenic and biogenic emissions. PM and trace gases (CO, VOC, NO, NO₂) released by the important fires in 2003 were also taken into account. Pay et al. (2010) used the Advanced Research Weather Research and Forecasting (WRF-ARW) Model to

provide meteorology, the EMEP database for anthropogenic emissions and the Dust Regional Atmospheric Model (BSC-DREAM8b) to simulate the atmospheric cycle of mineral dust. Boundary conditions were provided by LMDz-INCA2. Appel et al. (2012) forced WRF with boundary conditions from the North American Model from the National Centers for Environmental Prediction to generate meteorological data. Anthropogenic and biogenic emissions were respectively provided by MEGAN and TNO (<http://www.tno.nl/>). Fire emissions from the Moderate Resolution Imaging Spectroradiometer (MODIS) fire radiative power product were taken into account. Boundary conditions were generated via the Global and Regional Earth System Monitoring using Satellite and in situ data (GEMS) product with the help of data provided by GEOS-Chem for chemical boundary conditions. The AERO05 aerosol module and the CB05 mechanism were used. Tuccella et al. (2012) forced WRF/Chem with meteorological initial and boundary conditions provided by ECMWF. The chemical boundary conditions of trace gases are based upon the results from the NOAA Aeronomy Lab Regional Oxidant Model (NALROM). The gas-phase chemistry model used is the Regional Acid Deposition Model, version 2 (RADM2), and the aerosol module includes the Modal Aerosol Dynamics Model for Europe (MADE) for the inorganic fraction, and the Secondary Organic Aerosol Model (SORGAM) for the carbonaceous secondary fraction.

The performance of PM_{2.5} obtained 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 SO₄²⁻, Polair3D and CHIMERE show good agreement with the observations on average (within 0.1 μg m⁻³), whereas WRF/Chem underestimates significantly. All models overestimate NO₃⁻, but the bias is lower for CHIMERE than for WRF/Chem and Polair3D. Performance results for NH₄⁺ are similar for Polair3D, CHIMERE, and WRF/Chem.

3.4 Discussion

The operational model evaluation showed that NO₃⁻ and NH₄⁺ are overestimated. This overestimation results from a combination of various factors. First, artifacts in the measurements methods, due to the volatilization of NH₄NO₃ from filters, can contribute to the model overestimation, although an evaluation of nitrate measurement methods in Europe did not show any significant bias (Schaap et al., 2004). In addition, the overestimation of nitrate could be due to the slight underestimation of sulfate by the model (2.2 μg m⁻³ simulated against 2.3 μg m⁻³ in the measurements). Thus, not enough ammonia is consumed by SO₄²⁻, favoring the formation of additional NH₄NO₃. Also, there is still a significant uncertainty about ammonia emissions, including their magnitude and temporal variability. Finally, taking the mean

over 5 yr to generate pseudo-climatological boundary conditions for aerosols and gases is also a source of uncertainties.

The overestimation of PM₁₀ is slight (18.4 μg m⁻³ simulated against 17.3 μg m⁻³ in the measurements), but Polyphemus tends to simulate greater PM₁₀ concentrations than other models over Europe (Sartelet et al., 2012). The comprehensive treatment of organic aerosols in Polyphemus, which leads to reasonable agreement with observations of carbonaceous aerosols (Couvidat et al., 2012), can explain in part this slight overestimation because the NO₃⁻ overestimation is not compensated by an organic underestimation as it was the case in some earlier modeling studies). The overestimation of daily PM_{2.5} can be explained by the overestimation of NO₃⁻, which is a greater fraction of PM_{2.5} than of PM₁₀.

The evolution of model performance over the years shows a clear improvement for O₃, NH₄⁺, and NO₃⁻ 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 and is similar to that of other models.

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}, SO₄²⁻, NO₃⁻ and NH₄⁺ 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 SO₄²⁻, NO₃⁻, and NH₄⁺ observations on a daily basis. The AT02 station is located near Lake Neusiedl in Austria at 117 m above mean sea level (a.m.s.l.). The closest city is Illmitz (2416 inhabitants). The DE02 station is located at 74 a.m.s.l., in a forest with agriculture and meadows at a distance of 1 km and is surrounded by small

stationary SO₂ 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 approximately 3 cars per day within 3 km around the station. The DE03 station is situated on a mountain in the Black Forest, at 1205 a.m.s.l. and surrounded by forests and meadows, where there is minor agricultural activity for some parts of the year. Freiburg (206 000 inhabitants) is 12 km from the station and there are approximately 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 a.m.s.l. and is 30 km from Rome. The station has the particularity to be inside the Research Area of the National Council of Italy; therefore, the site could be influenced by the vehicles of the research personnel. There is no relevant industry 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., SO₄⁻, NO₃⁻, and NH₄⁺), but those should be taken into account by the model. The NO01 station is surrounded mainly by forest, meadows, freshwater lakes, and low-intensity agricultural areas; it is located at 190 a.m.s.l. There are some local known emissions, 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 ENSEMBLES² observations for temperature and precipitation (horizontal resolution of 0.5° × 0.5°), and the ERA Interim³ 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 associ-

ation 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 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 4 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 in-depth analysis on the 5 stations that provide PM_{2.5}, SO₄⁻, NO₃⁻, and NH₄⁺ 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} measurements (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 linear 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). In this evaluation, the day lag ranges from 0 to 2 for two reasons. First, the general evolution of the correlation as a function of the day lag does not show linear relationships over 10 days but does over the first 3 days. Moreover, the best correlation coefficients for precipitation and wind speed suggest that they have little impact on PM beyond 4 days. If both the regression coefficients and the best correlations are

²<http://eca.knmi.nl/download/ensembles/ensembles.php>

³<http://www.ecmwf.int/research/era/do/get/era-interim>

Table 3. Dynamic evaluation for PM_{2.5} (without speciation): linear 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 (ranging from 0 to 2) for the observed (obs) and simulated (sim) concentrations. 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	<i>r</i>	reg	<i>r</i>	reg	<i>r</i>	reg	<i>r</i>	reg	<i>r</i>	reg	<i>r</i>
AT02	-0.02	-0.46	-0.01	-0.43	-0.02	-0.21	-0.01	-0.19	0.03	-0.11	0.1	-0.46
AT48	-0.03	-0.09	-0.03	0.14	0.02	-0.22	0.02	-0.19	0.02	-0.16	0.09	-0.45
CH02	-0.03	-0.41	-0.02	0.11	-0.03	-0.34	0.05	-0.3	0.02	-0.22	0.05	-0.41
CH04	-0.03	0.18	-0.02	0.11	0	-0.37	0.06	-0.32	0.02	-0.32	0.06	-0.38
DE02	-0.03	-0.3	-0.02	-0.11	-0.04	-0.23	-0.02	-0.18	0.02	-0.25	0.05	-0.41
DE03	-0.03	0.32	-0.03	0.21	-0.01	-0.27	0.06	-0.35	0.02	-0.3	0.06	-0.53
DE04	-0.03	-0.12	-0.04	0.18	-0.01	-0.39	0.06	-0.3	0.08	-0.38	0.08	-0.54
DE44	-0.03	-0.31	-0.02	-0.14	-0.03	-0.25	0	-0.22	0.05	-0.31	0.06	-0.47
ES07	-0.04	0.48	-0.01	0.26	-0.02	-0.19	0.08	-0.32	0.03	-0.18	0.05	-0.31
ES08	-0.05	0.23	-0.02	0.1	0.01	-0.27	0.04	-0.22	0.07	-0.31	0.05	-0.24
ES09	-0.03	0.55	-0.03	0.06	0	-0.16	0.03	-0.2	0.03	-0.39	0.06	-0.53
ES10	-0.02	0.12	-0.03	0.16	-0.03	-0.12	0.07	-0.28	-0.03	-0.24	0.06	-0.5
ES11	-0.03	0.44	-0.01	-0.05	0	-0.23	0.05	-0.22	0.03	-0.35	0.08	-0.48
ES12	-0.03	0.38	-0.01	0.2	0.03	-0.17	0.05	-0.2	0.06	-0.41	0.07	-0.57
ES13	-0.04	0.37	-0.02	-0.05	0.01	-0.21	0.05	-0.27	0.08	-0.37	0.08	-0.54
ES14	-0.02	-0.05	-0.02	-0.1	-0.01	-0.14	0	-0.09	0.11	-0.42	0.1	-0.53
ES15	-0.03	0.51	-0.02	-0.15	-0.01	-0.16	0.05	-0.27	-0.01	-0.25	0.06	-0.48
GB36	-0.02	-0.13	-0.02	-0.1	-0.04	-0.2	-0.01	-0.18	0.04	-0.31	0.06	-0.4
GB48	-0.01	-0.19	0.04	-0.27	-0.02	-0.17	0.03	-0.16	-0.01	-0.19	0.03	-0.19
IT01	-0.03	-0.18	-0.02	-0.18	0.02	-0.27	0.04	-0.24	0.08	-0.24	0.12	-0.53
IT04	0	-0.55	-0.01	0.12	-0.01	-0.22	-0.04	-0.29	0.03	-0.23	0.09	-0.27
NO01	-0.03	0.14	-0.01	-0.16	-0.04	-0.17	-0.06	0.08	-0.01	-0.2	0.03	-0.1
SI08	-0.02	-0.09	-0.02	-0.11	-0.07	-0.29	0.1	-0.32	0.02	0.09	0.07	-0.26

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 best results are AT02, CH04, ES08, ES12, ES14, GB36, and IT01 (values of the regression coefficients and correlations given in this section are the arithmetic means of the observational and simulation data, unless noted otherwise; the individual values are given in Table 3). At CH04 the regression coefficients for temperature are -0.02 and -0.03, respectively, in simulation and 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. At ES08 the regression coefficients for temperature, precipitation, and wind speed are close to -0.03, 0.02, and 0.06, respectively, and the maximum correlations for temperature, precipitation, and wind speed are close to 0.15, 0.25, and -0.27, respectively, in both simulation and observation. At GB36 the regression coefficients for temperature, precipitation, and wind speed are respectively close to -0.02, -0.02, and 0.05 for both simulation and observation. The maximum correlations are also close for each variable in both simula-

tion and observation. AT02, ES12, ES14, and IT01 present similar results.

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, AT48, DE02, DE44, ES08, ES09, ES11, ES12, ES13, ES14, GB36, GB48, 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 -0.04 to 0.05 among the stations, with close values between observation and simulation (with a difference lower than 0.03 except at ES11 and ES13). 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 -0.23), 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 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, ES14, GB36, IT01, and SI08 stations, where the curves are nearly identical. At these stations the regression coefficients are either -0.02 or -0.03 and are nearly the same in simulation and observation. The best observed correlation is around -0.45 at AT02, $+0.15$ at ES10, -0.07 at ES14, -0.12 at GB36, -0.18 at IT01, and around -0.10 at SI08 for both simulation and observation. Other stations present similar regressions, but with a constant difference between the values of the correlation between the observed and the simulated data (AT48, DE02, DE03, DE44, ES07, ES08, ES09, ES12, and GB48). For some other stations, the correlation is not well reproduced by the model (CH02, DE04, ES11, ES13, ES15, IT04, and NO01). For these 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 GB36 station provides the best results with the two correlation curves nearly identical (regression coefficients of 0.05, and best correlation of around -0.35 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., SO₄⁻, NO₃⁻, NH₄⁺, organics, ...), unlike precipitation and wind speed, which impact PM_{2.5} components in the same way. Exceptions are sea salt and mineral 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 SO₄⁻, NO₃⁻, and NH₄⁺.

Such an analysis is conducted below for the 5 stations that have joint observations for PM_{2.5}, SO₄⁻, NO₃⁻, and NH₄⁺.

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., SO₄⁻, NO₃⁻, and NH₄⁺, 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 SO₄⁻, NO₃⁻, and NH₄⁺ concentrations as a function of changes in precipitation and temperature are well represented. The evolution of SO₄⁻ as a function of changes in temperature is well represented by the model, while the model gives a slightly larger correlation in absolute values for NO₃⁻ (~ -0.4 vs. ~ -0.3) and NH₄⁺ (~ -0.3 vs. ~ -0.25). 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. Simulation data show that the negative correlation between temperature and PM_{2.5} is driven by all the PM components (around -0.15 for SO₄⁻, -0.2 for sea salt, -0.4 for NO₃⁻, NH₄⁺, and organic matter, and -0.6 for black carbon), except mineral dust (near 0). Lower temperatures in winter are associated with greater emissions of NO_x, SO₂, elemental carbon and primary organic carbon (residential heating and fossil-fueled power plants), which may explain the greater concentrations of SO₄⁻, carbonaceous PM, and nitric acid. Lower temperatures also favor the formation of semivolatile NH₄NO₃. 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 some 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 SO₄⁻ as a function of changes in

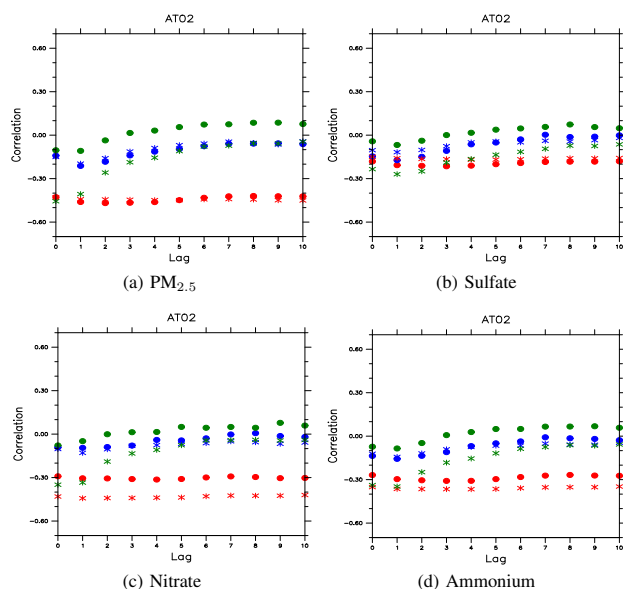


Fig. 4. Correlations between meteorology (temperature in red, precipitation in blue, wind speed in green) on a given day and pollutant concentrations (PM_{2.5}, sulfate, nitrate, and ammonium), ranging up to 10 days after at AT02 (2003–2008). Simulated data are represented with a star, while observational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

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 NO₃[−] and NH₄⁺ (correlations of −0.30 to −0.40 for the observations and of −0.05 to −0.10 for the model). NO₃[−] modeled concentrations are similar in winter (3.7 μg m^{−3}) and in summer (3 μg m^{−3}), whereas they are significantly different in the observations (4.7 μg m^{−3} in winter against 1.1 μg m^{−3} in summer) (see Table 4). The differences between modeled and observed concentrations could be due to artifacts in the NO₃[−] measurements in summer due to NH₄NO₃ 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, overestimate NO₃[−]. Simulation data show that the negative correlation between temperature and PM_{2.5} is driven by all the PM components (around −0.10 for SO₄[−], NO₃[−], NH₄⁺, 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.

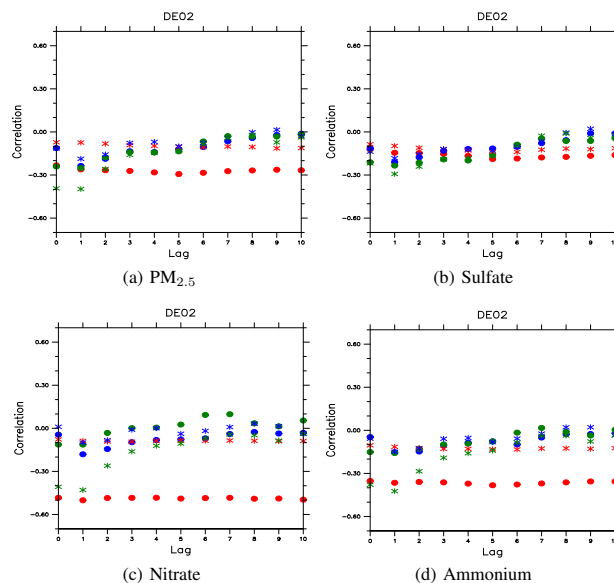


Fig. 5. Correlations between meteorology (temperature in red, precipitation in blue, wind speed in green) on a given day and pollutant concentrations (PM_{2.5}, sulfate, nitrate, and ammonium), ranging up to 10 days after at DE02 (2006–2008). Simulated data are represented with a star, while observational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

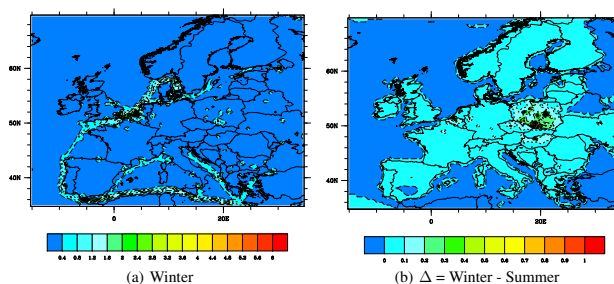
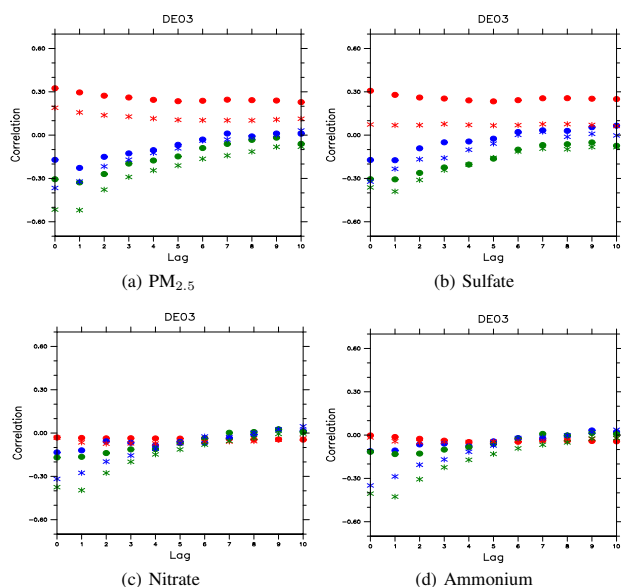


Fig. 6. Average sulfur dioxide emissions over Europe in winter, and difference (Δ) between winter and summer, expressed in $\mu\text{g m}^{-2} \text{h}^{-1}$.

The AT02 and DE02 stations show negative correlations between temperature and SO₄[−]. SO₄[−] concentrations depend on SO₂ emissions (shown in Fig. 6), which may vary by season, and on the conversion rate of SO₂ to SO₄[−], which is greater in summer when oxidant concentrations are greater and kinetics faster. SO₂ emissions are greater over Poland in winter than in summer (i.e., when temperatures are low, which suggests that SO₄[−] concentrations at these two stations are governed more by SO₂ emissions than by the kinetics of SO₂ to SO₄[−] conversion). A first reason comforting this hypothesis is that the SO₂ emission impact is more visible at the AT02 station (correlation of around −0.3), which is closer to the SO₂ emission sources, than at the DE02 station (correlation of around −0.15). Furthermore, daily mean

Table 4. Mean simulated (sim) and observed (obs) concentrations of sulfate, nitrate, and ammonium in winter (December–January–February, DJF) and in summer (June–July–August, JJA) at the five stations, expressed in $\mu\text{g m}^{-3}$.

	SO ₄ [−]		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	2.4	1.3	1.5	0.8
DE02	2.2	1.6	3.5	2.7	3.7	3.0	4.7	1.1	1.8	1.4	2.0	0.7
DE03	1.4	1.5	1.0	2.1	3.7	3.2	0.8	0.9	1.5	1.4	0.4	0.5
IT01	1.2	2.0	1.8	3.5	4.1	1.8	4.4	2.25	1.6	1.2	1.7	1.4
NO01	0.9	1.1	1.0	1.5	1.3	0.7	0.7	0.8	0.7	0.5	0.4	0.4

**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.

concentrations of SO₄[−] are higher at these stations in winter ($3.1 \mu\text{g m}^{-3}$ at AT02 and $2.2 \mu\text{g m}^{-3}$ at DE02) than in summer ($2.2 \mu\text{g m}^{-3}$ at AT02 and $1.6 \mu\text{g m}^{-3}$ at DE02; see Table 4); these seasonal differences demonstrate that SO₄[−] concentrations at AT02 and DE02 are more affected by SO₂ emissions than by the kinetics of SO₄[−] 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 SO₄[−] 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 temperature and precipitation show better agreement for PM_{2.5} and SO₄[−] than for NO₃[−] and NH₄⁺. NO₃[−] modeled concentrations are four times greater than the observations, which may be the result of the overestimation of NO₃[−] by the model and/or negative artifacts in the measurements. The model overestimates the correlation between wind speed and NO₃[−] and NH₄⁺, but it correctly reproduces the low correlation between temperature and both NO₃[−] and NH₄⁺. Simulation data show that the positive correlation between temperature and PM_{2.5} is the result of low positive correlations with SO₄[−] (0.05), mineral dust (0.20), and organic matter (0.25) and of low negative correlations with NH₄⁺ (-0.05), NO₃[−] (-0.05), black carbon (-0.25), and sea salt (-0.15). The correlation between SO₄[−] 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 SO₂ emission sources that show strong seasonal variability and that it is impacted by SO₂ sources with low seasonal variability (e.g., maritime traffic). The fact that the model correctly reproduces this opposite response suggests that it represents the relationship between meteorology and SO₄[−] 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 anticorrelation. 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 NO₃[−] and NH₄⁺ and is well represented by

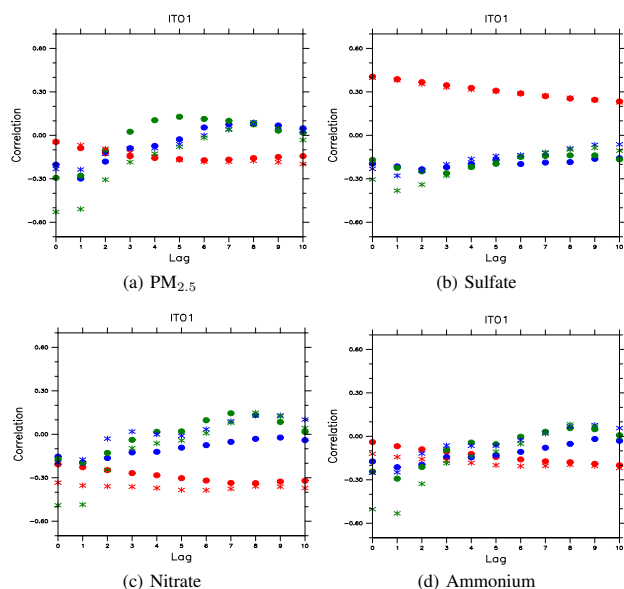


Fig. 8. Correlations between meteorology (temperature in red, precipitation in blue, wind speed in green) on a given day and pollutant concentrations (PM_{2.5}, sulfate, nitrate, and ammonium), ranging up to 10 days after at IT01 (2007–2008). Simulated data are represented with a star, while observational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

the model. The model correctly reproduces the evolution of SO₄[−], NO₃[−], and NH₄⁺ as a function of changes in temperature and precipitation (although the strength of the correlation between precipitation and NO₃[−] is slightly underestimated by the model). The correlation between SO₄[−] 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 the anticorrelation between wind speed and NH₄⁺ or NO₃[−]. Simulation data show that the negative correlation between temperature and PM_{2.5} is driven by NH₄⁺ (−0.2), NO₃[−] (−0.35), black carbon (−0.5), sea salt (−0.15), and organic matter (−0.2).

Correlations between NO₃[−]/NH₄⁺ and wind speed present large differences between the observations and the simulation at the AT02, DE02, DE03, and IT01 stations. Simulated NO₃[−] is overestimated, which is a recurring issue in PM modeling over Europe. Moreover, there is less NO₃[−] in summer than in winter in the simulation, and 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 NO₃[−] 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 temper-

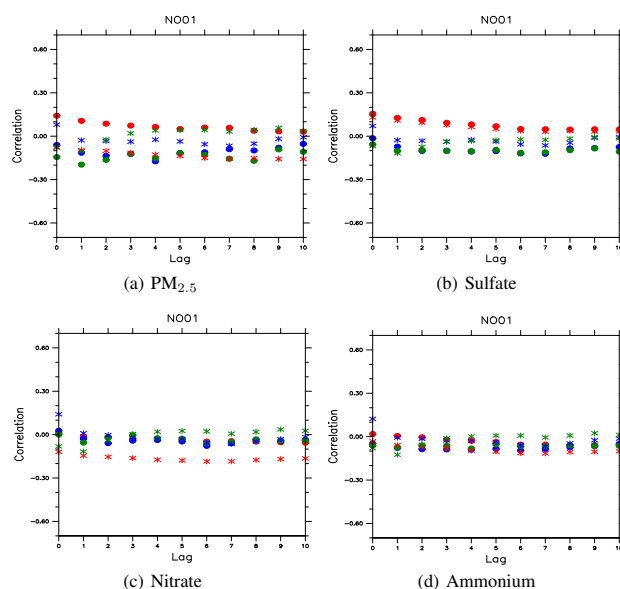


Fig. 9. Correlations between meteorology (temperature in red, precipitation in blue, wind speed in green) on a given day and pollutant concentrations (PM_{2.5}, sulfate, nitrate, and ammonium), ranging up to 10 days after at NO01 (2002–2008). Simulated data are represented with a star, while observational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

ature is not well represented (0.1 for the observations versus −0.1 for the simulation as a consequence of the performance for NO₃[−]). In the observations, the dependence of PM_{2.5} on temperature is driven by that of SO₄[−] and NH₄⁺ because NO₃[−] 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 SO₄[−] 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 NO₃[−] and NH₄⁺. This can be explained by the fact that PM_{2.5} observed mean concentrations at NO01 (4.4 μg m^{−3}) are significantly lower than those at most of the other stations (19.9 μg m^{−3} at AT02, 12.9 μg m^{−3} at DE02, 5.9 μg m^{−3} at DE03, and 22 μg m^{−3} at IT01), added to the fact that there are significantly fewer PM_{2.5} data at the NO01 station compared to the four other sites.

The positive correlations between temperature and SO₄[−] at DE03, IT01, and NO01 suggest that atmospheric oxidation of SO₂ (favored by faster kinetics at higher temperatures and greater oxidant concentrations in summer) dominates over greater SO₂ 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 SO₄²⁻ are higher in summer (1.5 μg m⁻³ at DE03, 2.0 μg m⁻³ at IT01, and 1.1 μg m⁻³ at NO01) than in winter (1.4 μg m⁻³ at DE03, 1.2 μg m⁻³ at IT01, and 0.9 μg m⁻³ at NO01) comforts this hypothesis.

This dynamic evaluation needed to be performed on stations that provide daily data of PM_{2.5} and its components for a common period of over at least a year. These requirements were meant to increase the significance of the statistical indicators. Since PM_{2.5} measurements are relatively recent in Europe, the number of stations meeting these requirements is small, which is a limitation of this study. Therefore, one should be cautious not to extrapolate the model dynamic performance to regions far remote from these stations. Nevertheless, the ability of the model to reproduce the effect of meteorology on PM_{2.5} and its inorganic components is satisfactorily characterized at several stations, which are located in various parts of Europe and which provide different types of responses.

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 measurements 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 μg m⁻³ over northern Italy to low concentrations of 6 μg m⁻³ over Scandinavia). PM_{2.5} composition varies also significantly. For example, the PM_{2.5} SO₄²⁻ fraction is highest in Eastern Europe, the NO₃⁻ 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 O₃ meets the model performance criteria and that PM_{2.5}, PM₁₀, and SO₄²⁻ meet the performance goals. NO₃⁻ and NH₄⁺ are overestimated by the model; NH₄⁺ meets the performance criteria, but NO₃⁻ does not. The correlation coefficients between simulated and observed data are 63 % for O₃, 57 % for PM₁₀, 59 % for PM_{2.5}, 56.5 % for SO₄²⁻, 58 % for NH₄⁺, and 42 % for NO₃⁻. The comparison with other recent 1 yr model simulations over Europe shows that all models overestimate NO₃⁻ and that the bias for Polair3D is comparable to the bias for WRF/Chem, but greater to that for CHIMERE. The performance for PM_{2.5}, SO₄²⁻, and NH₄⁺ 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 SO₄²⁻ to changes in temperature varies significantly among stations and can be opposite depending on the distance of the station from certain SO₂ 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. For example, the lack of PM_{2.5} chemical composition data in the western part of the domain (i.e., the countries near the Atlantic Ocean: Ireland, France, the United Kingdom, Spain, and Portugal) is a limitation of this study. The results of studies on the impact of meteorology/climate change on PM_{2.5} compositions on that westernmost part of Europe should therefore be treated with caution. Analyses of correlation between observed temperature and NO₃⁻ concentrations in the US have shown results that differed from modeled responses of NO₃⁻ to temperature (Tai et al., 2010). Such analyses in Europe would require conducting 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 extend this analysis to carbonaceous PM_{2.5}, particularly 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 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. Nevertheless, the results of this study provide the first dynamic evaluation on an PM_{2.5} air quality model with respect to meteorology.

Appendix A

Comparison with other modeling studies in the context of AQMEII

Table A1. Comparisons to observations for surface PM₁₀ and PM_{2.5} over Europe (concentrations and RMSE are in $\mu\text{g m}^{-3}$) from 7 July to 31 August for this simulation (2000–2008) and the AQMEII models (2006) (Sartelet et al., 2012).

	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 %

Appendix B

Comparison with model performance evaluations of 1 yr simulations over Europe

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). The data from this work are averaged over 2000–2008.

References Models Year(s) simulated	Péré et al. CHIMERE (2003)	Pay et al. CALIOPE-EU (2004)	Tuccella et al. WRF/Chem (2007)	Appel et al. CMAQ (2006)	This work Polair3D (averaged 2000–2008)
Number of stations	11	16	NA	160	22
Obs ($\mu\text{g m}^{-3}$)	12.2	13.0	12.6		13.3
Sim ($\mu\text{g m}^{-3}$)	15.1	6.3	8.6		15.9
Correlation (%)	73	45	41		59
RMSE ($\mu\text{g m}^{-3}$)		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

Table B2. Comparison for sulfate with Péré et al. (2010) and Tuccella et al. (2012). The data from this work are averaged over 2000–2008.

References Models Year(s) simulated	Péré et al. CHIMERE (2003)	Tuccella et al. WRF/Chem (2007)	This work Polair3D (averaged 2000–2008)
Number of stations	11	NA	77
Obs ($\mu\text{g m}^{-3}$)	3.9	2.4	2.3
Sim ($\mu\text{g m}^{-3}$)	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

Table B3. Comparison for nitrate with Péré et al. (2010) and Tuccella et al. (2012). The data from this work are averaged over 2000–2008.

References Models Year(s) simulated	Péré et al. CHIMERE (2003)	Tuccella et al. WRF/Chem (2007)	This work Polair3D (averaged 2000–2008)
Number of stations	11	NA	33
Obs ($\mu\text{g m}^{-3}$)	3.1	2.9	2
Sim ($\mu\text{g m}^{-3}$)	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

Table B4. Comparison for ammonium with Péré et al. (2010) and Tuccella et al. (2012). The data from this work are averaged over 2000–2008.

References Models Year(s) simulated	Péré et al. CHIMERE (2003)	Tucella et al. WRF/Chem (2007)	This work Polair3D (averaged 2000–2008)
Number of stations	11	NA	34
Obs ($\mu\text{g m}^{-3}$)	2.1	1.8	0.9
Sim ($\mu\text{g m}^{-3}$)	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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