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Linking climate and air quality over Europe: effects of meteorology on $PM_{2.5}$ concentrations

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Abstract

The effects of various meteorological parameters such as temperature, wind speed, absolute humidity, precipitation and mixing height on $PM_{2.5}$ concentrations over Europe were examined using a three-dimensional chemical transport model, PMCAMx-2008. Our simulations covered three periods, representative of different seasons (summer, winter, and fall). $PM_{2.5}$ appears to be more sensitive to temperature changes compared to the other meteorological parameters in all seasons.

$PM_{2.5}$ generally decreases as temperature increases, although the predicted changes vary significantly in space and time, ranging from $-700\,\mathrm{ng\,m^{-3}\,K^{-1}}$ ($-8\,\%\,\mathrm{K^{-1}}$) to $300\,\mathrm{ng\,m^{-3}\,K^{-1}}$ ($7\,\%\,\mathrm{K^{-1}}$). The predicted decreases of $PM_{2.5}$ are mainly due to evaporation of ammonium nitrate, while the higher biogenic emissions and the accelerated gas-phase reaction rates increase the production of organic aerosol (OA) and sulfate, having the opposite effect on $PM_{2.5}$. The predicted responses of $PM_{2.5}$ to absolute humidity are also quite variable, ranging from $-130\,\mathrm{ng\,m^{-3}}\,\%^{-1}$ ($-1.6\,\%\,\%^{-1}$) to $160\,\mathrm{ng\,m^{-3}}\,\%^{-1}$ ($1.6\,\%\,\%^{-1}$) dominated mainly by changes in inorganic $PM_{2.5}$ species. An increase in absolute humidity favors the partitioning of nitrate to the aerosol phase and increases the average $PM_{2.5}$ during summer and fall. Decreases in sulfate and sea salt levels govern the average $PM_{2.5}$ response to humidity during winter. A decrease of wind speed (keeping constant the emissions) increases all $PM_{2.5}$ species (on average $40\,\mathrm{ng\,m^{-3}}\,\%^{-1}$) due to changes in dispersion and dry deposition. The wind speed effects on sea salt emissions are significant for $PM_{2.5}$ concentrations over water and in coastal areas. Increases in precipitation have a negative effect on $PM_{2.5}$ (decreases up to $110\,\mathrm{ng\,m^{-3}}\,\%^{-1}$) in all periods due to increases in wet deposition of $PM_{2.5}$ species and their gas precursors. Changes in mixing height have the smallest effects (up to $35\,\mathrm{ng\,m^{-3}}\,\%^{-1}$) on $PM_{2.5}$.

Regarding the relative importance of each of the meteorological parameters in a changed future climate, the projected changes in precipitation are expected to have the largest impact on $PM_{2.5}$ levels during all periods (changes up to $2\,\mathrm{\mu g\,m^{-3}}$ in the

fall). The expected effects in future $PM_{2.5}$ levels due to wind speed changes are similar in all seasons and quite close to those resulting from future precipitation changes (up to $1.4\,\mu g\,m^{-3}$). The expected increases in absolute humidity in the future can lead to large changes in $PM_{2.5}$ levels (increases up to $2\,\mu g\,m^{-3}$) mainly in the fall due to changes in particulate nitrate levels. Despite the high sensitivity of $PM_{2.5}$ levels to temperature, the small expected increases of temperature in the future will lead to modest $PM_{2.5}$ changes and will not dominate the overall change.

1 Introduction

Over the past decades, increased levels of atmospheric particulate matter (PM) have received considerable attention due to their impact on human health, climate change, and visibility. In particular, fine particulate matter with an aerodynamic diameter less than 2.5 µm ($PM_{2.5}$), has detrimental effects on human health as it is associated with increases in mortality, as well as respiratory and cardiovascular diseases (Schwartz et al., 1996; Bernard et al., 2001; Pope et al., 2009). $PM_{2.5}$ has also been implicated in various air quality problems such as changes of the energy balance of the planet (IPCC, 2007), visibility reduction (Seinfeld and Pandis, 2006), and the formation of acid rain and acid fogs (Burtraw et al., 2007).

Concentrations of PM are strongly influenced by meteorology. For example, increasing temperature can lead to elevated sulfate concentrations due to increased rate of SO_2 oxidation (Aw and Kleeman, 2003; Dawson et al., 2007; Jacob and Winner, 2009; Lecoeur and Seigneur, 2013). In contrast, semi-volatile organic and inorganic aerosols evaporate as temperature increases (Sheehan and Bowman, 2001; Dawson et al., 2007; Tsigaridis and Kanakidou, 2007; Jimenez-Guerrero et al., 2012). Temperature has also a significant indirect effect on secondary organic aerosol (SOA) concentrations. In a warmer climate, secondary organic aerosol can increase due to higher biogenic VOC emissions (Heald et al., 2008; Jacob and Winner, 2009). Changes in absolute humidity also affect $PM_{2.5}$ levels. Increases in humidity favor nitric acid partitioning to the aerosol phase and therefore can lead to nitrate concentration increases (Dawson et al., 2007; Galindo et al., 2011; Lecoeur and Seigneur, 2013). Wet deposition is in most areas the major removal process for $PM_{2.5}$, hence changes in precipitation rate or the area extent of precipitation have a significant impact on aerosol concentrations (Dawson et al., 2007; Lecoeur and Seigneur, 2013). Changes in wind speed lead to changes in dispersion and transport as well as to changes in marine and desert aerosol production (Jacob and Winner, 2009; Aksoyoglu et al., 2011). Finally mixing height determines to a large extend the dilution of primary and the formation of secondary pollutants (Jimenez-Guerrero et al., 2012; Pay et al., 2012).

Over the next decades, climate is expected to change and this change will influence $PM_{2.5}$ concentrations. Based on IPCC projections for Europe (IPCC, 2007), temperature is expected to rise from 1 to 5.5 K over the next century. Emissions of biogenic VOCs are also expected to increase as temperature increases. Forkel and Knoche (2007) predicted a 30 % increase (locally up to 50 %) of biogenic VOC emissions in Europe due to a predicted 1.7–2.4 °C temperature increase, under the IPCC IS92a scenario within the next 30 years. Higher temperatures in a future climate, will also lead to increases in absolute humidity (IPCC, 2007). Precipitation is also expected to change over Europe in the future, having large spatial and seasonal variations. Based on the IPCC A2 emission scenario, Räisänen et al. (2004) predicted an increase in mean winter precipitation in northern and central Europe (up to 50 %) and a substantial decrease in southern Europe in the next century. During summer, precipitation was projected to decrease throughout central and southern Europe. Similar projections for precipitation were also reported by other modeling studies (Giorgi and Meleux, 2007; Hedegaard et al., 2008; Kjellström et al., 2010). In addition, general circulation models (GCMs) and regional climate models (RCMs) predict changes in both rainfall intensity and frequency (Christensen and Christensen, 2004; Frei et al., 2006; Buonomo et al., 2007; Boe et al., 2009; Argüeso et al., 2012). Jacob and Winner (2009) suggested that the critical variable that affects PM concentrations is precipitation frequency rather than precipitation rate. Wind speed is also predicted to change in a future climate. Andersson and Engardt (2010) predicted increases in wind speed over northern Europe, and decreases in the southern regions. Similar projections for wind speed were reported by other model studies (Räisänen et al., 2004; Kjellström et al., 2010; Katragkou et al., 2011). Hedegaard et al. (2013) reported increasing mixing height in most of Europe under a future climate (above 100 m in southeastern Europe), but Jimenez-Guerrero et al. (2011) predicted an average decrease for most continental Europe.

The impact of various climate scenarios on air quality over Europe as well as the correlation between meteorology and PM concentrations have been the subject of several studies (Koch et al., 2003; Heald et al., 2008; Hedegaard et al., 2008, 2013; Jacob and Winner, 2009; Redington et al., 2009; Roustan et al., 2010; Galindo et al., 2011; Im et al., 2012; Manders et al., 2012; Pay et al., 2012; Megaritis et al., 2013). Carvalho et al. (2010) applied a regional CTM, CHIMERE, over Europe with downscaled meteorology generated by a global GCM to study the impact of climate change on ozone and PM_{10} levels, using the IPCC A2 scenario, which describes a very heterogeneous world, with continuously increasing population, self-reliance and preservation of local identities. Their predicted PM_{10} concentration changes showed a strong spatial and temporal variability with increases over the continental regions and decreases over water. They concluded that the PM_{10} response was mainly driven by changes in the boundary layer height and wind speed. Jimenez-Guerrero et al. (2012) used a regional modeling system, MM5-CHIMERE, over southwestern Europe in order to study how concentrations of air pollutants respond to a changing climate for 2100 under the IPCC A2 scenario. Their findings suggest that aerosol species are strongly influenced by the higher future temperatures. They predicted an increase of sulfate and secondary organic aerosols (SOA) due to faster reactions and higher emissions of biogenic VOCs, and a decrease of particulate nitrate. In a multi-year simulation (2000–2008), Lecoeur and Seigneur (2013) used a three-dimensional CTM, Polyphemus/Polair3D, to investigate the response of $PM_{2.5}$ species to changes in meteorology. Their results suggest that wind speed and precipitation have a strong negative effect on $PM_{2.5}$ and its components (with sea salt being the only exception, for which a positive correlation with wind speed was predicted), while the response of $PM_{2.5}$ to temperature changes varied significantly among the $PM_{2.5}$ species considered. The negative response of $PM_{2.5}$ to wind speed changes and the variable effects caused by changes in temperature were also reported by Aksoyoglu et al. (2011). Additional work has been conducted in several areas over the world with the majority focused on the United States (Hogrefe et al., 2004; Racherla and Adams, 2006; Dawson et al., 2007, 2009; Tagaris et al., 2007, 2008; Zhang et al., 2008; Avise et al., 2009; Pye et al., 2009; Mahmud et al., 2010; Day and Pandis, 2011; Singh and Palazoglu, 2012; Tai et al., 2012; Jeong and Park, 2013). The predicted $PM_{2.5}$ changes due to climate are quite variable in space and time, and there are often conflicting conclusions about the meteorological variables driving these changes.

Most of the earlier modeling studies have focused on the overall effect of future climate on $PM_{2.5}$ concentrations. While this has provided valuable insights, it has often been difficult to quantify the effects of changes of individual meteorological parameters and processes. One of the few available studies has focused on the United States, studying the sensitivity of $PM_{2.5}$ to different meteorological perturbations (Dawson et al., 2007). However, this study covered a relatively short simulation period, and it did not assess how important these meteorological changes are for individual processes that are related to the formation, transport and removal of $PM_{2.5}$ components. In addition, to our knowledge, there has been little work trying to quantify how these individual processes (such as the partitioning of semi-volatile PM components, the marine aerosol production, etc) can be affected by changes in meteorology and eventually, how sensitive $PM_{2.5}$ is to these changes. The goal of this study is to conduct a detailed sensitivity analysis quantifying how changes in temperature, wind speed, absolute humidity, precipitation, and mixing height, and their subsequent effects on different processes, can influence fine particulate matter ($PM_{2.5}$) concentrations over Europe. Each of these parameters is studied separately so that the relative importance of each as well as the subsequent response of $PM_{2.5}$ can be quantified. For this purpose we use a three-dimensional CTM, PMCAMx-2008 over Europe. PMCAMx-2008 implements a state-of-the-art organic module for organic aerosol (OA) modeling based on the volatility basis set framework (VBS) (Donahue et al., 2006), which has not been used in earlier versions of the model, as well as in earlier climate-air quality interactions studies. The model uses also updated inorganic aerosol modules for the simulation of inorganic PM species. In addition, we covered three month-long simulation periods, in order to obtain more representative results regarding the seasonal dependence of the $PM_{2.5}$ response to changes in meteorology.

A brief description of the PMCAMx-2008 along with the characteristics of its application in the European domain is given in Sect. 2. The PMCAMx-2008 base-case predictions for $PM_{2.5}$ concentrations and some information regarding the model evaluation are given in Sect. 3. The description of each sensitivity simulation conducted in this study as well as the predicted response of $PM_{2.5}$ to these meteorological perturbations are presented in the next sections. Finally the relative importance of the various meteorological parameters and the main conclusions are presented.

2 The PMCAMx-2008 CTM

2.1 Model description

PMCAMx-2008 (Fountoukis et al., 2011; Megaritis et al., 2013) uses the framework of the CAMx air quality model (Environ, 2003). The chemical mechanism used in this study to describe the gas-phase chemistry is based on the SAPRC99 mechanism (Environ, 2003; Carter, 2010) and includes 211 reactions of 56 gases and 18 free radicals. For the simulation of the aerosol species, the model uses three detailed modules: inorganic aerosol growth (Gaydos et al., 2003; Koo et al., 2003), aqueous phase chemistry (Fahey and Pandis, 2001) and SOA formation and growth (Murphy and Pandis, 2009). These modules employ a sectional approach using ten aerosol size sections, spanning the diameter range from 40 nm to 40 µm. In this study inorganic aerosol formation was simulated using the "bulk equilibrium approach" where the bulk inorganic aerosol and gas phase are assumed to be always in equilibrium. The organic aerosol treatment in PMCAMx-2008 is based on the volatility basis set framework (Donahue et al., 2006; Stanier et al., 2008). Primary organic aerosol is assumed to be semivolatile in PMCAMx-2008, while the model treats all organic species (primary and secondary) as chemically reactive. Further details regarding the simulation of inorganic and organic aerosol species in PMCAMx-2008 can be found in Fountoukis et al. (2011).

For the simulation of wet scavenging the model assumes that the scavenging rate within or below a cloud due to precipitation is equal to the product of the concentration of a pollutant and the respective scavenging coefficient (Seinfeld and Pandis, 2006). Dry deposition, for the gas-phase species, is simulated using the resistance model of Wesely (1989), while for aerosol species the PMCAMx-2008 uses the resistance approach of Slinn and Slinn (1980) as implemented in UAM-AERO (Kumar et al., 1996). More information about the simulation of removal processes can be found in Fountoukis et al. (2011) and Megaritis et al. (2013).

2.2 Model application

PMCAMx-2008 was set to simulate the atmosphere over Europe covering a 5400 km × 5832 km region with a 36 km × 36 km resolution grid and 14 vertical layers extending up to approximately 6 km altitude. Three month-long periods, representative of different seasons (summer, winter, and fall) were simulated. The summer simulations were based on a hot late spring period (1–29 May 2008), the fall modeled period was from 15 September to 17 October 2008, while the winter simulation covered a cool late winter period (25 February–23 March 2009). The first two days from each simulation were used as model initialization days and were excluded from the analysis. All three periods showed a variety of meteorological conditions and pollution levels. The summer period was characterized by a blocking anticyclone (especially in the first half of May) leading to stable meteorological conditions and enhanced pollution over Central Europe. In addition, high temperatures were observed in most of Europe (Pikridas et al., 2010; Hamburger et al., 2011; Poulain et al., 2011; Mensah et al., 2012), typical for summer conditions. Fall represented the transition from summer to winter with a moderate temperature (which was decreasing during this period), less stable atmospheric pressure and frequent precipitation events (EMEP, 2010; Poulain et al., 2011) while the winter period was characterized by low temperatures in most of Europe (Hildebrandt et al., 2010b; Freney et al., 2011; Poulain et al., 2011; Mensah et al., 2012).

The necessary inputs to the model included emissions, meteorological conditions, land use data and initial and boundary conditions of the simulated PM species. Anthropogenic gas emissions included land as well as international shipping emissions and were developed by the TNO team as a continuation of the work in GEMS and MACC (Visschedijk et al., 2007; Denier van der Gon et al., 2010). Anthropogenic particulate organic and elemental carbon emissions were based on the EUCAARI Pan-European Carbonaceous Aerosol Inventory (Kulmala et al., 2011). Biogenic emissions were produced by utilizing the MEGAN (Model of Emissions of Gases and Aerosols from Nature) model (Guenther et al., 2006). Sea salt emissions (O'Dowd et al., 2008) as well as wildfire emissions (Sofiev et al., 2009) were also included. Further details about the emissions data used in this study can be found in Fountoukis et al. (2011). The meteorological input into the model included hourly data of temperature, pressure, water vapor, clouds, rainfall, horizontal wind components and vertical diffusivity generated using the meteorological model WRF (Weather Research and Forecasting) (Skamarock et al., 2008). For the boundary conditions of the major PM species we used the same concentrations as Fountoukis et al. (2011). The boundary conditions were chosen on the basis of measurements taken in sites close to the boundaries (e.g., Seinfeld and Pandis, 2006; Zhang et al., 2007).

3 Base case simulations and model evaluation

The predicted concentrations of total $PM_{2.5}$ during the three modeled base case periods are presented in Fig. 1. During the summer period, the domain-average ground-level concentration of total $PM_{2.5}$ is 7.7 $\mu g\,m^{-3}$. Elevated $PM_{2.5}$ concentrations are predicted in most of Western Europe (up to 25 µg m^{-3}), due mainly to high ammonium nitrate levels in this area (maximum concentration of 11 µg m^{-3}). In Central and Northern Europe fine organic matter dominates with biogenic secondary OA and oxidized primary OA (including OA from IVOCs) contributing the most (approximately 60 % and 25 % respectively). $PM_{2.5}$ concentrations in these areas may exceed 20 µg m^{-3}. Sulfate is predicted to be the dominant $PM_{2.5}$ species over the Eastern Mediterranean. The strong photochemical activity in this area favors the conversion of sulfur dioxide to sulfate and can partly explain the relatively high $PM_{2.5}$ levels in this area (up to 15 µg m^{-3}).

During the winter period the mean predicted ground-level concentration of total $PM_{2.5}$ over the domain is 7.1 µg m^{-3}. Sulfate and organics are predicted to be the major components contributing approximately 25 % and 28 % of total $PM_{2.5}$ mass. Peak period-average concentrations of total $PM_{2.5}$ (mostly over Central and Northern Europe) exceed 20 µg m^{-3} especially in areas with large industrial activity or large urban emissions. OA accounts for up to half of total $PM_{2.5}$ in these areas with fresh primary OA being the dominant OA component.

In the fall period the model predicts an average total $PM_{2.5}$ concentration of 8.3 µg m^{-3} over the domain. The elevated $PM_{2.5}$ levels are due to a combination of high ammonium nitrate, sulfate, and organic aerosol. On a domain-average basis organic aerosol and sulfate are predicted to account for 28 % of total $PM_{2.5}$ mass each, followed by ammonium (12 %), and nitrate (10 %). High levels of $PM_{2.5}$ over the Balkans and the Mediterranean (up to 22 µg m^{-3}), are mainly due to high sulfate concentrations while ammonium nitrate dominates over the western parts of the domain. The largest OA concentrations, with a peak of 6.8 µg m^{-3}, are predicted in the Po Valley area. High OA levels are also predicted over the Balkans.

Fountoukis et al. (2011) have evaluated the PMCAMx-2008 predictions against aerosol mass spectrometer (AMS) measurements during the summer EUCCARI intensive period (May 2008) used here. The observed data included ground measurements taken at four sites in Europe (Cabauw, Finokalia, Mace Head, Melptiz) as well as measurements aloft from an aircraft campaign (Morgan et al., 2010). PMCAMx-2008 had the most accurate performance for PM_1 organic aerosol (monthly average measured concentration: 3.3 $\mu g\,m^{-3}$, PMCAMx-2008: 3 $\mu g\,m^{-3}$), reproducing more than 87 % of the hourly averaged data within a factor of 2, showing a normalized mean bias (NMB) of −11 % and a normalized mean error (NME) of 30 %. The model reproduced more than 94 % of the observed daily averaged organic aerosol data within a factor of 2. The monthly average concentration of PM_1 sulfate predicted by the model in the four sites was 2.9 $\mu g\,m^{-3}$ compared to the measured value of 2.8 $\mu g\,m^{-3}$. PMCAMx-2008 was able to reproduce more than 70 % of the hourly averaged data and more than 82 % of the daily averaged points within a factor of 2. The normalized mean error (NME) for PM_1 sulfate concentration was 47 % and the corresponding mean error and fractional error, 1.3 $\mu g\,m^{-3}$ and 0.4 respectively. The model had also a reasonable performance for PM_1 nitrate and PM_1 ammonium in Cabauw, Finokalia and Melpitz (the corresponding mean bias and NMB were 0.4 $\mu g\,m^{-3}$ and 29 % for PM_1 nitrate, and 0.03 $\mu g\,m^{-3}$ and 2 % for PM_1 ammonium respectively). However in Mace Head, the model significantly overpredicted both fine nitrate and ammonium concentrations. These errors as suggested by Fountoukis et al. (2011) could be to some extent, attributed to the assumption of bulk equilibrium that PMCAMx-2008 uses for the inorganic aerosol simulation. In Mace Head a significant fraction of nitrate is associated with sea salt and shifts to the coarse mode, an effect that is not well captured by the model (Capaldo et al., 2000).

The comparison of PMCAMx-2008 predictions against the airborne measurements showed also an encouraging agreement. The model predicted average concentrations for PM_1 OA, sulfate, nitrate, and ammonium were 2.6, 1.6, 1.6 and 1.2 $\mu g\,m^{-3}$ respectively, while the corresponding measured average values were 2.2, 1.6, 1.4 and 1.3 $\mu g\,m^{-3}$. PMCAMx-2008 was able to capture more than 75 % and 77 % of the measured OA and sulfate data with concentrations higher than 1 $\mu g\,m^{-3}$ within a factor of 2. For PM_1 nitrate and ammonium, the model showed small normalized mean bias

(NMB) (1 % and −14 % respectively) and a mean absolute gross error (MAGE) of 1.1 and 0.7 µg m^{-3} respectively.

The comparison of PMCAx-2008 predictions against AMS hourly ground measurements during the EUCAARI winter intensive period also indicated a reasonable agreement for PM$_1$ OA and sulfate (see Supplement of Megaritis et al., 2013). On average the monthly measured concentrations for PM$_1$ OA, nitrate, sulfate and ammonium were 2.3, 2.1, 1.0 and 0.9 µg m^{-3} respectively compared to the predicted average values of 1.1, 1.8, 0.9 and 1.2 µg m^{-3}. The underprediction of OA was attributed to underestimation of wood burning emissions. The model reproduced 44 % of the hourly averaged PM$_1$ OA data and 42 % of the hourly averaged PM$_1$ sulfate data within a factor of 2. The model predictions for OA and sulfate were subject to significant scatter (for OA the fractional bias was −0.3 and the fractional error 0.9, while for sulfate the fractional bias was 0.1 and the fractional error 0.9).

During the fall period, hourly AMS measurements performed at several sites over Europe (Hyytiala, k-Puszta, Melpitz, Puy de Dome, Payerne, Puijo, and Vavihill) during an EMEP intensive campaign, were used for the PMCAMx-2008 evaluation. The model reproduced more than 74 % of the hourly averaged OA data within a factor of 2, having a fractional bias of −0.1 and a fractional error of 0.48. Sulfate, ammonium and nitrate were characterized by small to moderate fractional bias (0.1 for sulfate, 0.11 for nitrate and 0.29 for ammonium) and larger fractional errors (up to 0.8 for nitrate). The predicted monthly average concentrations for PM$_1$ OA, sulfate, nitrate, and ammonium were 2.5, 1.5, 1.6, and 1.2 µg m^{-3} respectively, close to measurements of 2.9, 1.4, 1.4, and 1.0 µg m^{-3}.

4 Sensitivity to meteorological variables

For each of the three modeled periods, we performed a suite of individual sensitivity simulations by perturbing various meteorological parameters, one at a time. The sensitivity tests included perturbations in temperature, wind speed, absolute humidity, precipitation rate, precipitating area and mixing height (Table 1).

Sensitivity to temperature was tested by performing four different simulations. The impact of temperature on biogenic emissions and $PM_{2.5}$ levels was examined, using temperature sensitive biogenic emissions, produced by the MEGAN model, based on an increase of 2 K. In this simulation the only change was on the biogenic emissions inventory. The temperatures used by the model (to simulate chemistry, thermodynamics, etc) were those of the base case scenario. The effect of temperature on aerosol thermodynamics was tested in another simulation where we increased temperature by 2 K only for the modules of PMCAMx-2008 that simulate the partitioning of semi-volatile inorganic and organic $PM_{2.5}$ species. Similar to the first simulation, temperatures for the other processes in PMCAMx and all the other meteorological parameters were the same as in the base case simulation. The third test studied the sensitivity of $PM_{2.5}$ to the temperature dependence of the gas-phase reaction rates. The overall temperature effect on $PM_{2.5}$ concentrations (using also temperature-dependent biogenic emissions) was studied in a different simulation where all surface and air temperatures were increased uniformly over the domain by 2 K, keeping all the other meteorological inputs constant.

The effect of wind speed on $PM_{2.5}$ concentrations was studied by two different simulations. We used first a simplified scenario where horizontal wind speed was decreased uniformly over the entire domain by 10 % keeping all other inputs constant. The vertical wind components were calculated from the perturbed horizontal wind speeds to ensure mass conservation. In this simulation the only changes were on the dispersion coefficients, as well as the transport (vertical velocity, advection, dilution) and removal processes (dry deposition rate), while sea-salt emissions were kept constant as in the base case. In the second test, we examined the effect of wind speed on marine aerosol emissions, recalculating the corresponding emissions inventory for wind speeds decreased by 10 %. This simulation examines only changes in sea salt emissions, therefore wind speed and all other meteorological data used as input by the model were those of the base case scenario.

The effect of absolute humidity was tested based on a uniform increase of 5 % over the entire domain. Precipitation intensity was increased uniformly by 10 % to study its effects. Sensitivity to the spatial extent of precipitation was investigated in a simulation where the area undergoing precipitation was increased by +10 %. This was done by extending the existing precipitating area into non-precipitating but adjacent cells which were chosen randomly. In addition, the sensitivity of $PM_{2.5}$ to mixing height changes was examined in a simulation where the mixing height was increased by one model layer. This was done by changing the vertical diffusivity in only the layer immediately above the base case mixing height. The corresponding average change was an increase in mixing height by approximately 150 m.

Table 1 summarizes the sensitivity simulations imposed in this study and the processes that were perturbed directly in each change. Initial and boundary conditions of the modeled PM species did not change compared to the baseline scenario, in all tests. Emissions of all pollutants were also kept constant as in the base case conditions in all tests, except for the two simulations using temperature sensitive biogenic emissions and new sea salt emissions due to wind speed change.

5 Sensitivity to temperature

5.1 Temperature-dependent biogenic emissions

The predicted changes (sensitivity scenario – base case) in average ground-level concentrations of total $PM_{2.5}$ due to higher biogenic emissions (based on a 2 K temperature increase) are shown in Fig. 2. During the modeled summer period, $PM_{2.5}$ is predicted to increase by 10 $ng\,m^{-3}\,K^{-1}$ (0.13 $\%\,K^{-1}$) on a domain average basis, with a maximum increase of 250 $ng\,m^{-3}\,K^{-1}$ (2 $\%\,K^{-1}$) in France (Fig. 2a). This is mainly due to an OA increase of 1.5 $\%\,K^{-1}$ on average, which reaches up to 5 $\%\,K^{-1}$. Higher biogenic emissions lead to increases in biogenic SOA concentrations which account for almost 90 % of the OA increase. The increased biogenic VOCs, on the other hand result in reductions of OH in several areas. The reduced OH levels, slow down the gas phase formation of sulfate (through SO_2 oxidation), and also lead to decreases of ammonium nitrate. This negative effect of increased biogenic VOCs on OH levels and hence on inorganics was also noted by Zhang et al. (2008). However, the predicted decreases of inorganic $PM_{2.5}$ components are less than the increases of total OA, thus the net impact is an increase of total $PM_{2.5}$ levels.

Biogenic emissions have also a positive effect on total $PM_{2.5}$ concentrations during the modeled winter and fall periods. $PM_{2.5}$ is predicted to increase throughout the domain by $10\,\mathrm{ng\,m^{-3}\,K^{-1}}$ ($0.1\,\%\,\mathrm{K^{-1}}$) and $20.3\,\mathrm{ng\,m^{-3}\,K^{-1}}$ ($0.25\,\%\,\mathrm{K^{-1}}$) on average, during the winter and fall respectively (Fig. 2b and c). The predicted increases during the winter period can reach up to $130\,\mathrm{ng\,m^{-3}\,K^{-1}}$ ($1\,\%\,\mathrm{K^{-1}}$) while during fall are even higher (up to $200\,\mathrm{ng\,m^{-3}\,K^{-1}}$ or $1.5\,\%\,\mathrm{K^{-1}}$). Increases in OA levels dominate the response of total $PM_{2.5}$, while inorganic $PM_{2.5}$ is less sensitive to biogenic emissions during these seasons.

5.2 Temperature effects on gas/aerosol partitioning

Increasing temperature by 2 K only for the partitioning of semi-volatile PM components has a significant effect on total $PM_{2.5}$ levels in all three periods (Fig. 3). The predicted response of $PM_{2.5}$ shows a strong spatial variability, as a result of competing changes in inorganic species concentrations and, to a lesser extent, in organic ones. In the modeled summer period, total $PM_{2.5}$ concentrations decrease by $49\,\mathrm{ng\,m^{-3}\,K^{-1}}$ ($1\,\%\,\mathrm{K^{-1}}$) on average, although the change is quite variable and ranges from $-700\,\mathrm{ng\,m^{-3}}$ ($-5\,\%\,\mathrm{K^{-1}}$) to $50\,\mathrm{ng\,m^{-3}}$ ($1.5\,\%\,\mathrm{K^{-1}}$). The predicted $PM_{2.5}$ decrease is largely due to significant decreases of ammonium nitrate. Rising temperature leads to increased volatilization of ammonium nitrate, which partitions to the gas-phase (Seinfeld and Pandis, 2006). As a result, less ammonium nitrate exists in the particulate phase (approximately 15 % in this simulation), leading to significant decreases of nitrate which reach up to 600 ng m^{-3} K^{-1} (14 % K^{-1}). On the contrary, as particulate nitrate decreases, the cloud pH increases and the aqueous-phase formation of particulate sulfate accelerates. This complex effect of temperature changes on partitioning of semi-volatile inorganic PM$_{2.5}$ is consistent with the results of other studies (e.g. Dawson et al., 2007; Aksoyoglu et al., 2011; Jimenez-Guerrero et al., 2012). OA is also sensitive to temperature mainly due to changes in the levels of secondary OA components and to a lesser extent on primary OA. Higher temperature leads to evaporation of all OA components and subsequently to decreases of their levels. The sensitivity of OA to temperature, as well as the increased gas-phase partitioning as temperature increases, have been also reported by earlier studies (Dawson et al., 2007; Megaritis et al., 2013).

During the modeled winter period, total PM$_{2.5}$ shows also a negative response to temperature, with an average decrease of 25 ng m^{-3} K^{-1} (0.4 % K^{-1}) (Fig. 3b) over the domain. The predicted decrease of PM$_{2.5}$ is significant in Central Europe and reaches up to 500 ng m^{-3} K^{-1} (2 % K^{-1}), due largely to decreases in nitrate (up to 8 % K^{-1}) and to a lesser extent in OA levels.

During the modeled fall period, total PM$_{2.5}$ decreases by 88 ng m^{-3} K^{-1} (1 % K^{-1}) on average over the domain. Significant decreases are predicted mainly over the central and south western areas of the domain, approximately 700 ng m^{-3} K^{-1} (7 % K^{-1}) and 500 ng m^{-3} K^{-1} (3.5 % K^{-1}) respectively (Fig. 3c). Nitrate is significantly reduced (its predicted decreases exceed 10 % K^{-1}), and along with total OA decreases dominate the response of total PM$_{2.5}$, despite the predicted increases in sulfate levels.

5.3 Temperature-dependent gas-phase reaction rates

Changes in gas-phase reaction rates, due to temperature changes, could also affect total PM$_{2.5}$ levels (Dawson et al., 2007). At higher temperatures, gas-phase reactions will accelerate (Dawson et al., 2007; Jacob and Winner, 2009; Day and Pandis, 2011;

Im et al., 2011). In all three modeled periods, $PM_{2.5}$ is predicted to increase due to the combined increases on the individual $PM_{2.5}$ components. In the modeled summer period, $PM_{2.5}$ concentrations are predicted to increase by 26 $ng\,m^{-3}\,K^{-1}$ (0.3 % K^{-1}) on a domain average basis. The effect is stronger over continental Europe, where $PM_{2.5}$ increases by 50 $ng\,m^{-3}\,K^{-1}$ (0.8 % K^{-1}) on average, while in some areas in Western Europe, increases in $PM_{2.5}$ reach up to 400 $ng\,m^{-3}\,K^{-1}$ (2 % K^{-1}) (Fig. 4a). The predicted response of total $PM_{2.5}$ is mainly driven by increases of nitrate levels (approximately 45 % of the $PM_{2.5}$ increase is due to nitrate), followed by increases in OA (largely attributed to secondary OA) and sulfate.

The lower oxidant availability during the winter leads to a lower increase of $PM_{2.5}$ compared to summertime (13.5 $ng\,m^{-3}\,K^{-1}$ or 0.2 % K^{-1} on average) (Fig. 4b). Over continental Europe, the predicted increases are higher, up to 120 $ng\,m^{-3}\,K^{-1}$ (1 % K^{-1}). Changes in organics and nitrate dominate (each of these two components accounts for around 40 % of the $PM_{2.5}$ increase), while increases in sulfate tend to be rather small.

The effects are higher during the modeled fall period (an average increase of 47 $ng\,m^{-3}\,K^{-1}$ or 0.6 % K^{-1} over the domain). The largest changes are in Central and Western Europe where $PM_{2.5}$ increases by approximately 160 $ng\,m^{-3}\,K^{-1}$ (1.4 % K^{-1}) (Fig. 4c). Increases of fine particulate nitrate and organics are driving the $PM_{2.5}$ response, while there are moderate increases in sulfate.

5.4 Overall temperature effects

An increase in temperature by 2 K is predicted to have a negative effect on average $PM_{2.5}$ levels for all three modeled periods. On a domain average basis $PM_{2.5}$ decreases by 25 $ng\,m^{-3}\,K^{-1}$ (0.3 % K^{-1}) in the summer, 7 $ng\,m^{-3}\,K^{-1}$ (0.1 % K^{-1}) in the winter and 33 $ng\,m^{-3}\,K^{-1}$ (0.4 % K^{-1}) in the modeled fall period. However the overall effect of temperature on $PM_{2.5}$ levels is quite variable in space and time (Fig. 5) due to the different effects on the individual processes as well as the competing responses of the $PM_{2.5}$ species. The predicted changes on $PM_{2.5}$ concentrations range from –720 $ng\,m^{-3}\,K^{-1}$

(−8 % K^{-1}) to 280 ng m^{-3} K^{-1} (7 % K^{-1}). Over continental Europe, $PM_{2.5}$ changes are dominated by decreases in nitrate, which reach up to 650 ng m^{-3} K^{-1} (12 % K^{-1}) during the modeled summer period. These decreases are mainly due to the evaporation of ammonium nitrate, leading to a reduction of average nitrate levels by 18 %. On the contrary in several parts of the domain, the higher biogenic VOC emissions and the increased rate of SO_2 oxidation enhance the production of OA and sulfate respectively. These increases can reach up to 225 ng m^{-3} K^{-1} (7 % K^{-1}) for sulfate and up to 190 ng m^{-3} K^{-1} (4 % K^{-1}) for OA. These results support the findings from previous studies that suggest the competing effects of temperature among the different processes and $PM_{2.5}$ species (Dawson et al., 2007; Heald et al., 2008; Jacob and Winner, 2009; Jimenez-Guerrero et al., 2012). Summarizing, the semi-volatile $PM_{2.5}$ evaporation appears to dominate and determine the overall $PM_{2.5}$ response to temperature changes over Europe, during all seasons. The average changes in $PM_{2.5}$ are higher during the fall.

6 Wind speed

Decreasing wind speed by 10 %, without any change on sea-salt emissions (as well as on emissions from other sources), affects all $PM_{2.5}$ components, resulting in increases of their levels in all three modeled periods (Fig. 6). During summer, total $PM_{2.5}$ is predicted to increase by 41 ng m^{-3} $\%^{-1}$ (0.6 % $\%^{-1}$) on average over the entire domain (Fig. 6a). The effects of wind speed were found to be highest in the more polluted and populated areas of the domain. For example, in Western Europe, a high ammonium nitrate area during summer, total $PM_{2.5}$ increases up to 340 ng m^{-3} $\%^{-1}$ (1.5 % $\%^{-1}$), driven mainly by increases of nitrate. Decreases in wind speed affect advection, dispersion and mixing as well as lead to changes in dry deposition. Approximately 7–13 % less $PM_{2.5}$ is dry deposited due to the simulated 10 % reduction in wind speed.

The effects of wind speed on total $PM_{2.5}$ levels are similar during the other two periods. During winter, $PM_{2.5}$ increases by 36 ng m^{-3} %$^{-1}$ (0.5 % %$^{-1}$) on average over the domain. Significant increases are found mainly over North Europe, where $PM_{2.5}$ increases up to 250 ng m^{-3} %$^{-1}$ (0.8 % %$^{-1}$), as well as in Central and Southwestern Europe (Fig. 6b), mainly due to increases of total OA and sulfate. In the modeled fall period $PM_{2.5}$ shows a similar sensitivity. On a domain average basis, $PM_{2.5}$ increases by 38 ng m^{-3} %$^{-1}$ (0.5 % %$^{-1}$). In Central Europe, the predicted increases in $PM_{2.5}$ reach up to 225 ng m^{-3} %$^{-1}$ (0.9 % %$^{-1}$) (Fig. 6c). The predicted $PM_{2.5}$ response is driven mainly by increases in particulate nitrate (it accounts for approximately 40 % of total $PM_{2.5}$ increase) and to a lesser extent in ammonium, sulfate and organics.

Our results, regarding the $PM_{2.5}$ response to wind speed, are consistent with those by Dawson et al. (2007), who found a $PM_{2.5}$ sensitivity to wind equal to 0.77 % %$^{-1}$ during summer and 0.56 % %$^{-1}$ during winter in the Eastern US. This negative effect of wind speed on $PM_{2.5}$ has been also reported in earlier modeling studies over Europe (Carvalho et al., 2010; Aksoyoglu et al., 2011; Lecoeur and Seigneur, 2013).

6.1 Wind effects on sea-salt emissions

The predicted changes (sensitivity scenario – base case) in average ground-level concentrations of $PM_{2.5}$ using a new sea salt emission inventory (based on a 10 % decrease of wind speed) are shown in Fig. 7. As expected, lower sea salt emissions result in lower $PM_{2.5}$ concentrations in all modeled periods, especially over water and in coastal areas. The predicted $PM_{2.5}$ response is as expected not uniform throughout the domain. During the modeled summer period, the predicted $PM_{2.5}$ decrease exceeds 60 ng m^{-3} %$^{-1}$ (or 0.5 % %$^{-1}$), and may reach up to 170 ng m^{-3} %$^{-1}$ (0.9 % %$^{-1}$), mainly due to decreases in particulate sodium and chloride. The predicted decreases are even larger (up to 200 ng m^{-3} %$^{-1}$ or 2.7 % %$^{-1}$) during the winter modeled period, as sea salt emissions and the accompanying concentrations of particulate sodium chloride were higher, while similar results have been obtained for the fall. Over continental

Europe the effects on $PM_{2.5}$ levels due to lower marine aerosol emissions are small. $PM_{2.5}$ is also reduced, however the predicted decrease does not exceed 20 ng m^{-3} %$^{-1}$ (0.1 % %$^{-1}$) in all three periods.

7 Effects of absolute humidity

Changes in absolute humidity affect total $PM_{2.5}$ concentrations, however its predicted response varies significantly in space (Fig. 8) due to the competing changes among $PM_{2.5}$ species. In the modeled summer period, increases of absolute humidity by 5 % result to an average increase of total $PM_{2.5}$ by 8 ng m^{-3} %$^{-1}$ (0.2 % %$^{-1}$) over the entire domain. This is consistent with the Dawson et al. (2007) study for the Eastern US who reported a 20 ng m^{-3} %$^{-1}$ increase in summer $PM_{2.5}$ levels due to increases in absolute humidity by 5–20 %. The highest changes are predicted in Western Europe, with $PM_{2.5}$ increases up to 160 ng m^{-3} %$^{-1}$ (1.6 % %$^{-1}$) (Fig. 8a) as a result of significant increases in nitrate. Increases in relative humidity shift the equilibrium of the ammonia-nitric acid system toward the particles (Seinfeld and Pandis, 2006). As absolute humidity increases by 5 %, approximately 15 % more HNO_3 is predicted to move to the aerosol phase, leading to higher particulate nitrate concentrations. These changes in nitrate, along with increases in ammonium and OA are driving the $PM_{2.5}$ response over land. On the contrary, over the ocean, total $PM_{2.5}$ decreases as humidity increases, due mainly to changes is sulfate and sodium chloride. The negative response of $PM_{2.5}$ in this area (reaching up to a reduction of 140 ng m^{-3} %$^{-1}$ or 1.5 % %$^{-1}$) arises from increases in the size of the particles and accelerated dry deposition (in all modeled periods a 5 % increase in absolute humidity resulted in a 9–15 % increase in dry deposited mass of sulfate, sodium, and chloride).

Absolute humidity has also a positive effect on $PM_{2.5}$ levels during the modeled fall period. Significant increases are predicted in most areas of continental Europe (up to 130 ng m^{-3} %$^{-1}$ or 1 % %$^{-1}$) (Fig. 8c), mainly due to significant increases in particulate nitrate (approximately 65 % of the $PM_{2.5}$ increase). Over the ocean, total $PM_{2.5}$ decreases (its predicted reduction can exceed 50 ng m^{-3} %$^{-1}$ or 0.4 % %$^{-1}$). The predicted increases of nitrate along with the increase in ammonium and total OA exceed the decreases in sulfate and sea salt, thus the net impact on total $PM_{2.5}$ is an average increase of 11.5 ng m^{-3} %$^{-1}$ (0.2 % %$^{-1}$).

In the modeled winter period, the predicted response of total $PM_{2.5}$ to absolute humidity differs. In spite of the increase in nitrate concentrations, the predicted decreases in fine particulate sulfate and sea salt aerosol dominate and determine the response of total $PM_{2.5}$ (Fig. 8b). On a domain average basis, the net effect of absolute humidity on $PM_{2.5}$ is a decrease by 7.5 ng m^{-3} %$^{-1}$ (0.2 % %$^{-1}$), while the predicted concentration changes range from –130 ng m^{-3} %$^{-1}$ (–1.6 % %$^{-1}$) to 44 ng m^{-3} %$^{-1}$ (0.5 % %$^{-1}$).

8 Precipitation

8.1 Precipitation rate

The effect of the precipitation rate on $PM_{2.5}$ concentrations is similar during all the modeled periods. The predicted response of average-ground level $PM_{2.5}$ concentrations after a 10 % increase in precipitation rate (without changing the precipitation area) is shown in Fig. S1 (see Supplement). As it is expected, increases in precipitation rate, accelerate the wet removal of $PM_{2.5}$ species and their gas precursors and consequently result in decreases of their concentrations. In this simulation we predict a 2–4 % increase in $PM_{2.5}$ wet deposited mass as well as a 5–12 % increase in the wet deposition of $PM_{2.5}$ gas precursors due to a 10 % increase in precipitation rate.

During the modeled summer period, total $PM_{2.5}$ is predicted to decrease as precipitation increases, by 13 ng m^{-3} %$^{-1}$ (0.2 % %$^{-1}$) on average. Precipitation affects all the individual $PM_{2.5}$ species leading to reductions of their levels in most areas of the domain (Fig. S1a). Over the western parts of the domain, total $PM_{2.5}$ is reduced up to

$110\,\mathrm{ng\,m^{-3}\,\%^{-1}}$ ($1.8\,\%\,\%^{-1}$). However even in areas with little rainfall during this period (e.g. eastern Mediterranean) (Fig. S2, Supplement), total $PM_{2.5}$ also decreased indicating that changes due to precipitation in upwind areas can affect the levels of $PM_{2.5}$ over downwind areas. Similar effects are predicted during the other two periods. $PM_{2.5}$ is also reduced as precipitation rate increases, having an average decrease of $0.2\,\%\,\%^{-1}$ in both periods. The predicted effects are strongest in areas receiving moderate or little precipitation with a maximum reduction of $65\,\mathrm{ng\,m^{-3}\,\%^{-1}}$ ($1\,\%\,\%^{-1}$) during the modeled winter period and up to $68\,\mathrm{ng\,m^{-3}\,\%^{-1}}$ ($1.2\,\%\,\%^{-1}$) in the modeled fall period. This negative correlation has been also pointed in earlier studies (Hedegaard et al., 2008; Jacob and Winner, 2009; Jimenez-Guerrero et al., 2012; Manders et al., 2012; Lecoeur and Seigneur, 2013). Dawson et al. (2007) predicted quite similar sensitivities for total $PM_{2.5}$ during summer (approximately $0.2\,\%\,\%^{-1}$).

8.2 Precipitation area

The predicted reduction of total $PM_{2.5}$ for a 10 % increase in the spatial extent of precipitation covers a significant portion of Europe, during all periods (Fig. S3, Supplement). During summer the predicted reduction of $PM_{2.5}$ reaches a maximum of $19\,\mathrm{ng\,m^{-3}\,\%^{-1}}$ ($0.3\,\%\,\%^{-1}$) with an average sensitivity of $8\,\mathrm{ng\,m^{-3}\,\%^{-1}}$ ($0.1\,\%\,\%^{-1}$). The predicted reductions arising mainly from the increases in $PM_{2.5}$ wet deposited mass (approximately 2–5 %). The predicted effect is quite similar during the winter period (average reduction of $7.5\,\mathrm{ng\,m^{-3}\,\%^{-1}}$ or $0.1\,\%\,\%^{-1}$), while in the modeled fall period the predicted response of total $PM_{2.5}$ is a little higher, $13\,\mathrm{ng\,m^{-3}\,\%^{-1}}$ ($0.16\,\%\,\%^{-1}$) on average. Our results support the conclusion that not only the precipitation intensity but the area undergoing precipitation as well, can affect total $PM_{2.5}$ concentrations (Lecoeur and Seigneur, 2013).

9 Mixing height

During the base case simulations the average predicted mixing height was 550 m in the summer, 380 m in the winter and 440 m in the fall period respectively. The predicted simulation-averaged changes in $PM_{2.5}$ due to an increase in mixing height (by approximately 150 m) are shown in Fig. S4 (see Supplement).

As expected, increases in mixing height affect all the individual $PM_{2.5}$ components resulting in decreases in their concentrations during all modeled periods. In the summer, the average total $PM_{2.5}$ concentrations decrease by $3.5\,\mathrm{ng\,m^{-3}\,\%^{-1}}$ (or $0.05\,\%\,\%^{-1}$). Similar effects on $PM_{2.5}$ levels are also predicted for the other two periods (an average reduction of $0.03\,\%\,\%^{-1}$). The effect of mixing height is strongest over polluted areas, where the predicted reduction of total $PM_{2.5}$ can exceed $35\,\mathrm{ng\,m^{-3}\,\%^{-1}}$ ($0.8\,\%\,\%^{-1}$) (over Western Europe, during the modeled summer period). Our results are consistent with those by Dawson et al. (2007), who predicted a $PM_{2.5}$ sensitivity to mixing height equal to $0.08\,\%\,\%^{-1}$ during summer and $0.05\,\%\,\%^{-1}$ during winter in the Eastern US.

10 Relative importance of meteorological parameters

In order to evaluate the relative importance of the various meteorological parameters, we estimated the potential effects that each of them may have on total $PM_{2.5}$ concentrations in a future climate. Our estimates were based on the predicted average $PM_{2.5}$ sensitivities to the meteorological perturbations (Fig. 9) and the projected future changes for each parameter. The projected meteorological changes are shown in Table S1 (see Supplement). According to the different IPCC (2007) scenarios, the average temperature in Europe is expected to increase over the next century from 1 to 5.5 K. Projections for wind speed and precipitation in Europe vary significantly in space. Based on the IPCC SRES A2 scenario, wind speed is predicted to change from −10 to 10 %, while precipitation could change from −40 % to 40 %. In our calculations, the changes in precipitation area and intensity were chosen to represent future projections for total precipitation over Europe. For mixing height, a potential range of changes was assumed, based on the estimates of Hedegaard et al. (2013). For this first order estimate, we assumed the same meteorological changes for all seasons.

In all three periods, $PM_{2.5}$ appears to be more sensitive to temperature changes compared to the other meteorological parameters (Fig. 9). On average, $PM_{2.5}$ shows a negative sensitivity to temperature changes, which is higher during fall compared to the other periods (Table S1). However, the predicted $PM_{2.5}$ sensitivities to temperature are spatially and temporally variable as a result of the different effects among the individual processes and the different responses of the $PM_{2.5}$ species. During all seasons, the increased volatilization of ammonium nitrate dominates, causing large decreases in $PM_{2.5}$ with increasing temperature. The negative predicted sensitivities reach up to 440 $\mathrm{ng\,m^{-3}\,\%^{-1}}$ in the fall and 310 $\mathrm{ng\,m^{-3}\,\%^{-1}}$ in the summer period (lower during winter) (Fig. 9). At the same time, the increasing temperatures lead to higher biogenic VOC emissions and accelerate the gas-phase chemical reactions. $PM_{2.5}$ shows also a strong sensitivity to wind speed and its accompanying effects on the marine aerosol production. However the predicted changes are somewhat lower compared to the $PM_{2.5}$ sensitivities to temperature (Fig. 9). The sensitivity is similar in all seasons, and ranges from −115 $\mathrm{ng\,m^{-3}\,\%^{-1}}$ (due to changes in wind speed, without any change in the emissions) to 132 $\mathrm{ng\,m^{-3}\,\%^{-1}}$ (due to the effects of wind speed on sea salt emissions). $PM_{2.5}$ appears to be less sensitive to absolute humidity changes. In all periods, $PM_{2.5}$ concentrations respond differently to absolute humidity, due to the competing effects between the individual $PM_{2.5}$ species (e.g., increases in nitrate, decreases in sulfate), thus the average sensitivity does not exceed 12 $\mathrm{ng\,m^{-3}\,\%^{-1}}$ and the largest $PM_{2.5}$ sensitivities are close to 55 $\mathrm{ng\,m^{-3}\,\%^{-1}}$. Changes in precipitation result in negative sensitivities for $PM_{2.5}$ levels which are comparable to those of absolute humidity, while mixing height seems to have a relatively small effect on average $PM_{2.5}$ levels.

In a future climate, the projected changes in precipitation are expected to have the largest impact on $PM_{2.5}$ levels during all periods (Fig. 10). $PM_{2.5}$ concentrations could potentially change by several $\mathrm{\mu g\,m^{-3}}$ (up to approximately 2 $\mathrm{\mu g\,m^{-3}}$ during the fall period), with changes in precipitation intensity being rather more important than changes in precipitating area. Wind speed and absolute humidity may also lead to appreciable changes in future $PM_{2.5}$ levels. The expected effects on $PM_{2.5}$ due to changes in wind speed as well as its accompanying effects on the marine aerosol production are similar in all three periods and quite close to those resulting from future precipitation changes (up to $1.4\,\mu g\,m^{-3}$). In addition, absolute humidity could potentially lead to large changes in $PM_{2.5}$ mainly during the fall period (increases up to $2\,\mu g\,m^{-3}$). The increased particulate nitrate levels, as higher absolute humidity favors its partitioning, are dominant during this period causing large increases in $PM_{2.5}$. In the other two periods the expected changes in $PM_{2.5}$ are smaller due to the competing responses among the individual $PM_{2.5}$ species. Temperature is expected to have a lower impact on future $PM_{2.5}$ compared to the other meteorological parameters, in all seasons. The expected $PM_{2.5}$ concentration changes range from $-1.1\,\mu g\,m^{-3}$ to $0.5\,\mu g\,m^{-3}$, driven from the offsetting effects of increased nitrate volatilization, higher biogenic VOC emissions and accelerated gas-phase chemistry. Mixing height is expected to have a relatively small impact on $PM_{2.5}$ levels in a future climate.

11 Conclusions

Climate affects air quality through a complex web of interactions starting with changes in the major meteorological variables like temperature, wind speed, absolute humidity, precipitation intensity, precipitation area, mixing height, etc. and progressing through changes in pollutant concentrations, formation and removal rates. In this study, we used a detailed three-dimensional CTM, PMCAMx-2008, to quantify the individual effects of the major meteorological parameters on the concentration and composition of $PM_{2.5}$ over Europe.

Precipitation is expected to have the largest impact on $PM_{2.5}$ concentrations under a changed future climate. In all periods, $PM_{2.5}$ shows a negative sensitivity to precipitation, driven mainly by the accelerated wet deposition of $PM_{2.5}$ species and their gas precursors. The average $PM_{2.5}$ sensitivity is quite similar during all seasons (an approximate decrease of 15 $\mathrm{ng\,m^{-3}\,\%^{-1}}$) and taking also account the significant projected precipitation changes, $PM_{2.5}$ concentrations could potentially change by several $\mu\mathrm{g\,m^{-3}}$ (up to 2 $\mu\mathrm{g\,m^{-3}}$ in the fall) in the future.

Wind speed can also have appreciable effects on future $PM_{2.5}$ levels due to changes in dispersion and transport, dry deposition and marine aerosol production. The projected changes in wind speed over Europe in the future are expected to change $PM_{2.5}$ levels up to 1.4 $\mu\mathrm{g\,m^{-3}}$.

Changes in absolute humidity influence mainly the inorganic $PM_{2.5}$ species, resulting in competing responses. An increase in absolute humidity favors the partitioning of nitrate to the aerosol phase and leads to higher particulate levels. During the fall period, this effect dominates the overall $PM_{2.5}$ response, and as absolute humidity is expected to rise in the future, it could lead to large increases of $PM_{2.5}$ (up to 2 $\mu\mathrm{g\,m^{-3}}$). On the contrary, the increase in absolute humidity could lead to decreases in sulfate, and sea salt levels due to the increase in the size of the particles and the accelerated dry deposition. These negative effects may, to some extent, offset the predicted increases in nitrate, thus during summer and winter the expected changes in future $PM_{2.5}$ due to absolute humidity are smaller.

Temperature is expected to have a lower average impact on future $PM_{2.5}$ levels compared to the rest of the meteorological parameters due to the competing effects among the individual processes and the different responses of the $PM_{2.5}$ species. The evaporation of semi-volatile $PM_{2.5}$ species is found to be the dominant process and determines to a large extent the $PM_{2.5}$ response to temperature changes over Europe, during all seasons. Significant effects are predicted mainly on particulate ammonium nitrate, as the increase in temperature reduces its concentration levels up to 15 $\%\,\mathrm{K^{-1}}$. Especially during fall, the predicted reduction of nitrate drives the overall $PM_{2.5}$ response, and as temperature is expected to rise in a future climate, could potentially lead to decreases in $PM_{2.5}$ levels up to 1.1 $\mu\mathrm{g\,m^{-3}}$. However as temperature increases, biogenic VOC emissions are expected to increase and gas-phase chemical reactions will accelerate, which will offset to some extent the reductions of $PM_{2.5}$, leading to even smaller changes in future $PM_{2.5}$ levels during the summer and winter period. $PM_{2.5}$ concentrations generally decrease as mixing height increases. However the predicted effects are not as significant as those of the other parameters for the average $PM_{2.5}$ levels, due to the importance of secondary $PM_{2.5}$ components that have a strong regional character.

Supplementary material related to this article is available online at http://www.atmos-chem-phys-discuss.net/14/10345/2014/acpd-14-10345-2014-supplement.pdf.

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Table 1. Description of performed sensitivity simulations.

Meteorological parameter	Change examined	Directly affected in simulation
Temperature	+2 K	Biogenic VOC emissions only.
	+2 K	Organic and inorganic aerosol thermodynamics only.
	+2 K	Gas-phase chemistry only.
	+2 K	All temperature-dependent processes (including BVOC emissions).
Wind speed	−10 %	Turbulent dispersion coefficients, advection, dry deposition. Emissions (including marine) were kept constant.
	−10 %	Marine aerosol emissions only.
Absolute humidity	+5 %	Reaction rates with H_2O, aerosol thermodynamics.
Precipitation rate	+10 %	Wet deposition.
Precipitation area	+10 %	Wet deposition.
Mixing height	+1 model layer	Vertical dispersion.

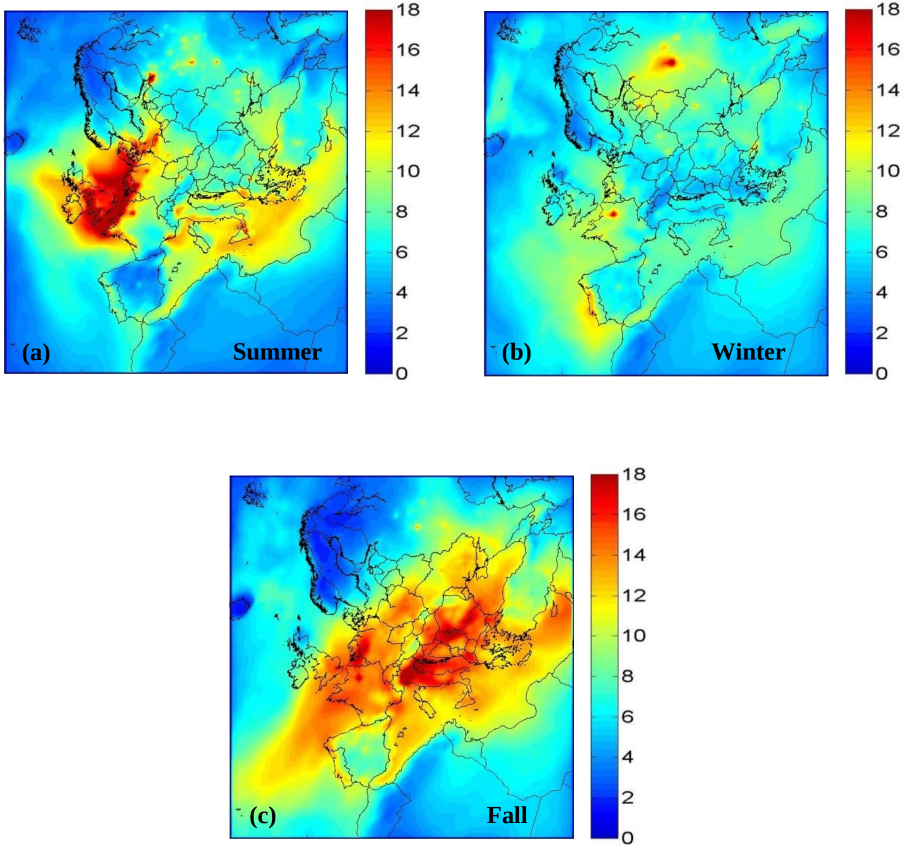

Fig. 1. Predicted average base case $PM_{2.5}$ ground-level concentrations ($\mu g\,m^{-3}$) during the modeled **(a)** summer, **(b)** winter and **(c)** fall periods.

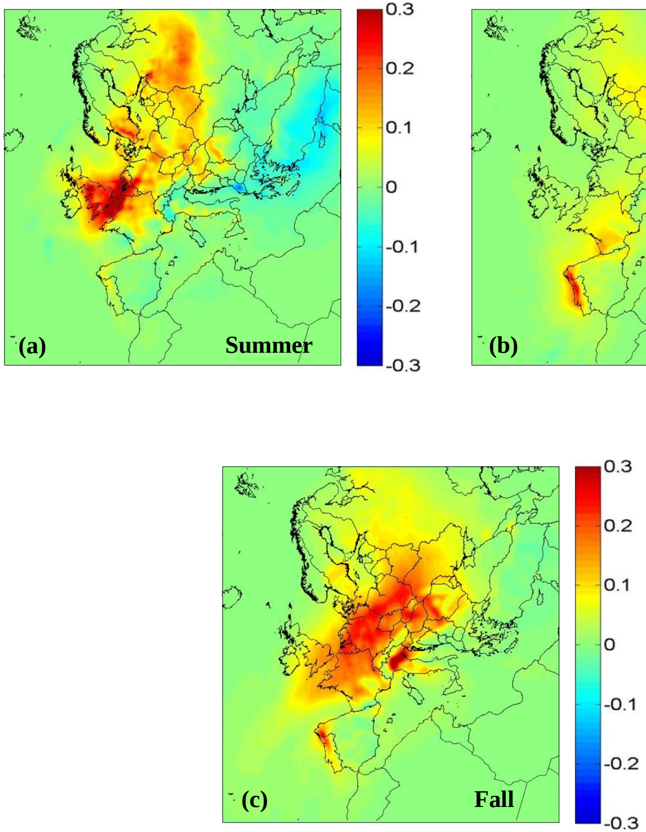

Fig. 2. Predicted average change in ground-level concentrations (µg m^{-3}) of total PM$_{2.5}$ due to changes on biogenic VOC emissions (based on a 2 K temperature increase) during the modeled **(a)** summer, **(b)** winter, and **(c)** fall periods. A positive value corresponds to an increase.

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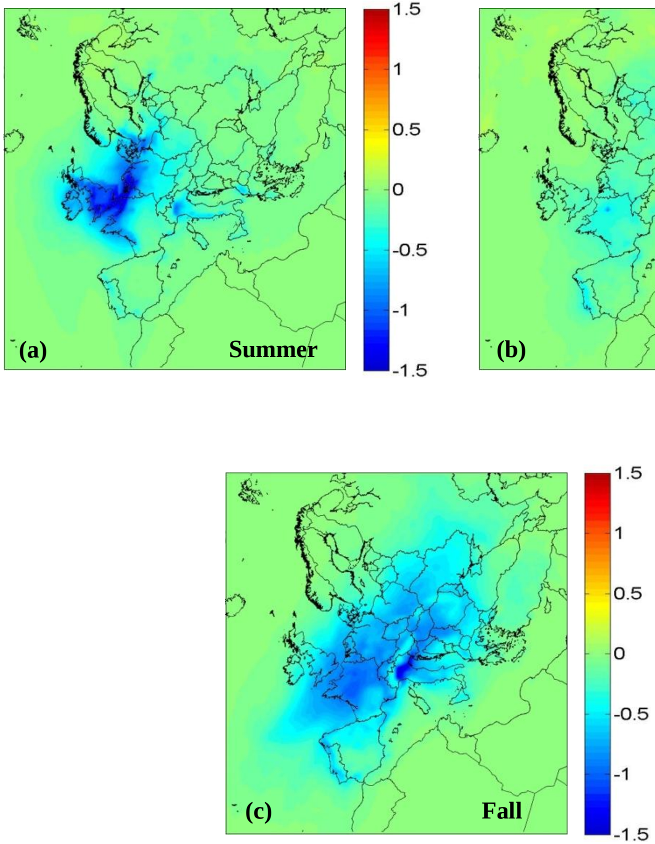

Fig. 3. Predicted average change in ground-level concentrations ($\mu g\,m^{-3}$) of total $PM_{2.5}$ due to changes on gas/aerosol partitioning (based on a 2 K temperature increase) during the modeled **(a)** summer, **(b)** winter, and **(c)** fall periods. A positive value corresponds to an increase.

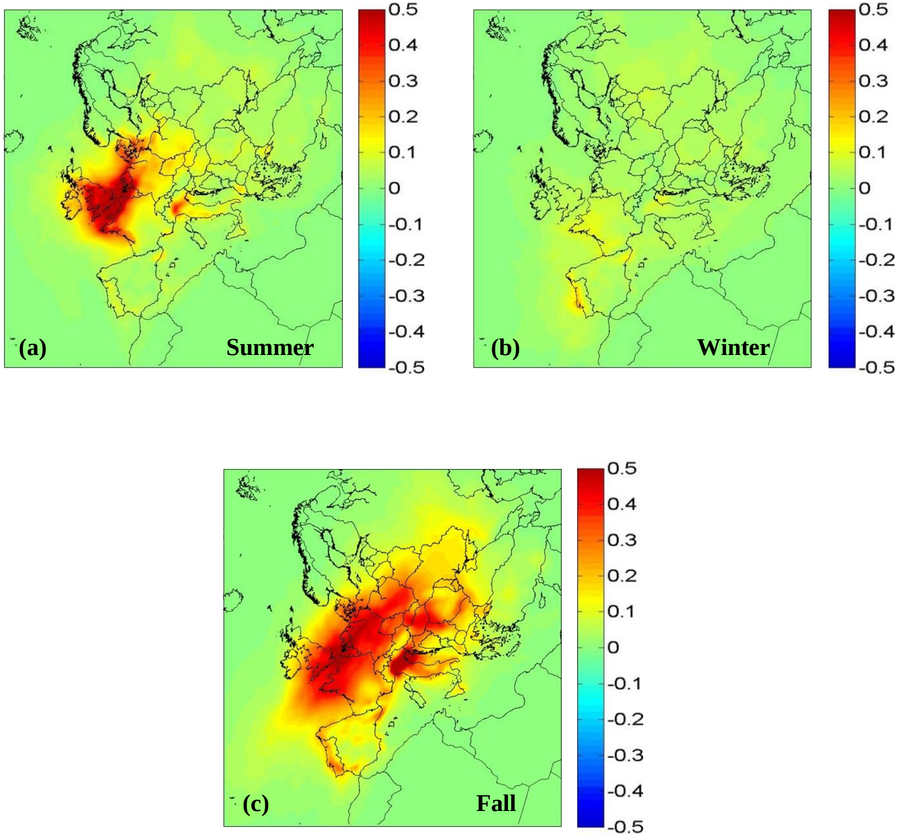

Fig. 4. Predicted average change in ground-level concentrations (µg m^{-3}) of total PM$_{2.5}$ due to changes on gas-phase reaction rates (based on a 2 K temperature increase) during the modeled **(a)** summer, **(b)** winter, and **(c)** fall periods. A positive value corresponds to an increase.

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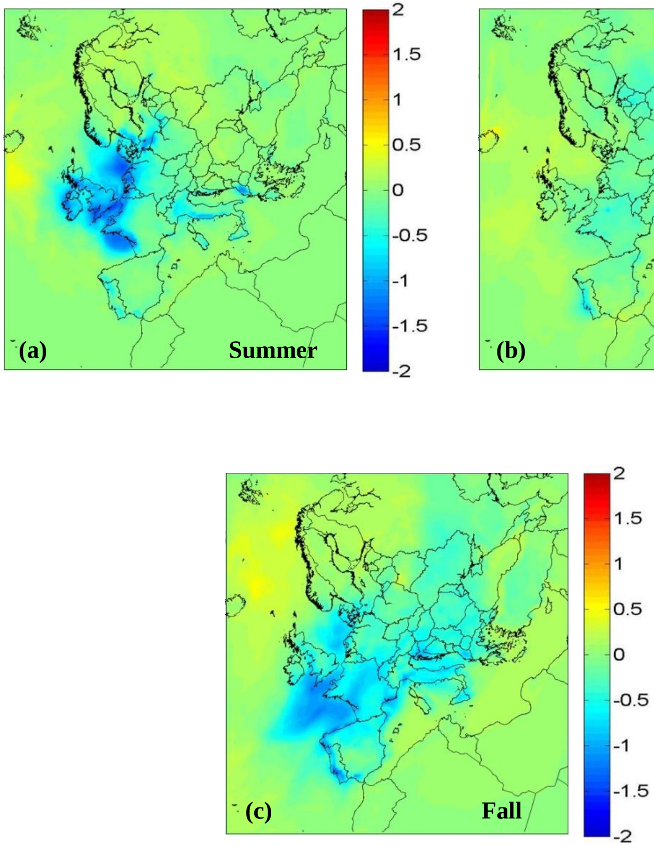

Fig. 5. Predicted average change in ground-level concentrations (µg m^{-3}) of total PM$_{2.5}$ due to an overall temperature increase by 2 K during the modeled **(a)** summer, **(b)** winter, and **(c)** fall periods. A positive value corresponds to an increase.

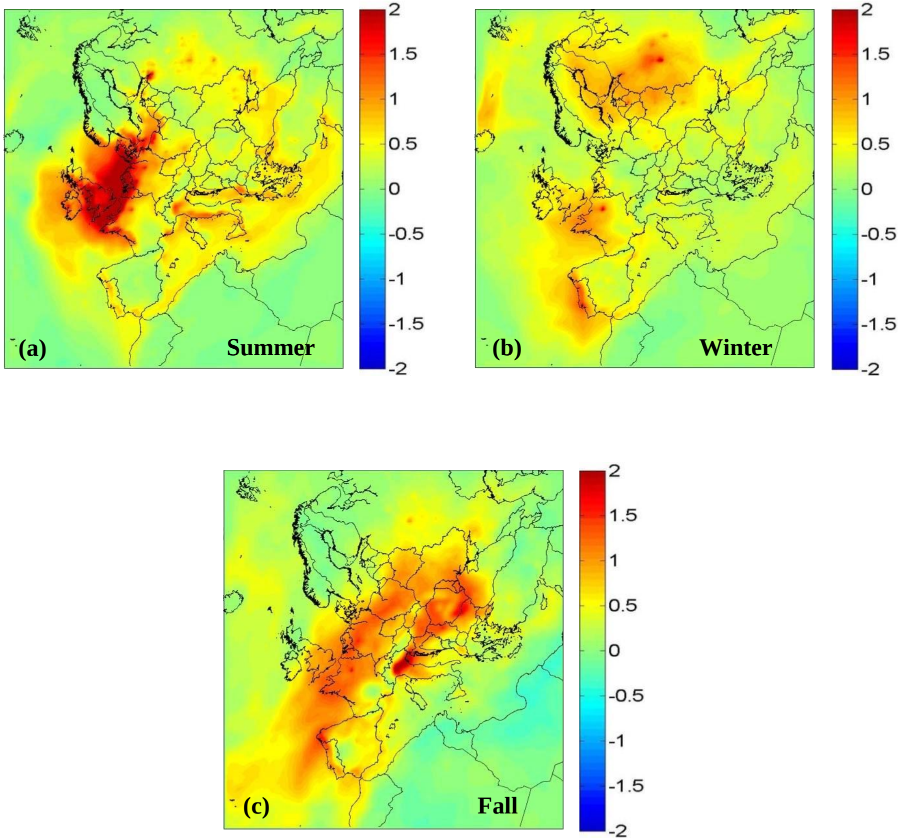

Fig. 6. Predicted average change in ground-level concentrations (µg m^{-3}) of total PM$_{2.5}$ due to a 10 % decrease in wind speed during the modeled **(a)** summer, **(b)** winter, and **(c)** fall periods. A positive value corresponds to an increase.

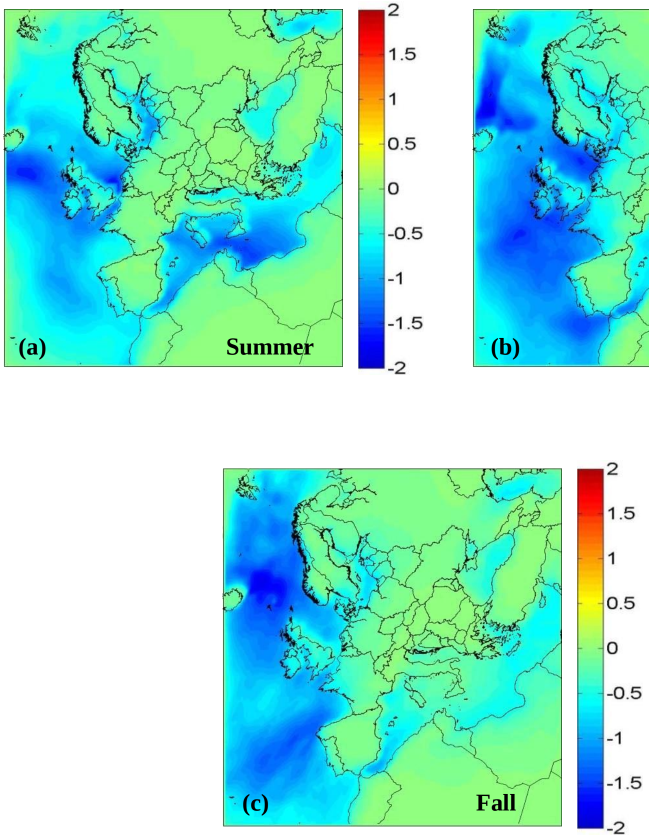

Fig. 7. Predicted average change in ground-level concentrations (µg m^{-3}) of total PM$_{2.5}$ due to changes on sea salt emissions (based on a 10 % decrease in wind speed) during the modeled **(a)** summer, **(b)** winter, and **(c)** fall period. A positive value corresponds to an increase.

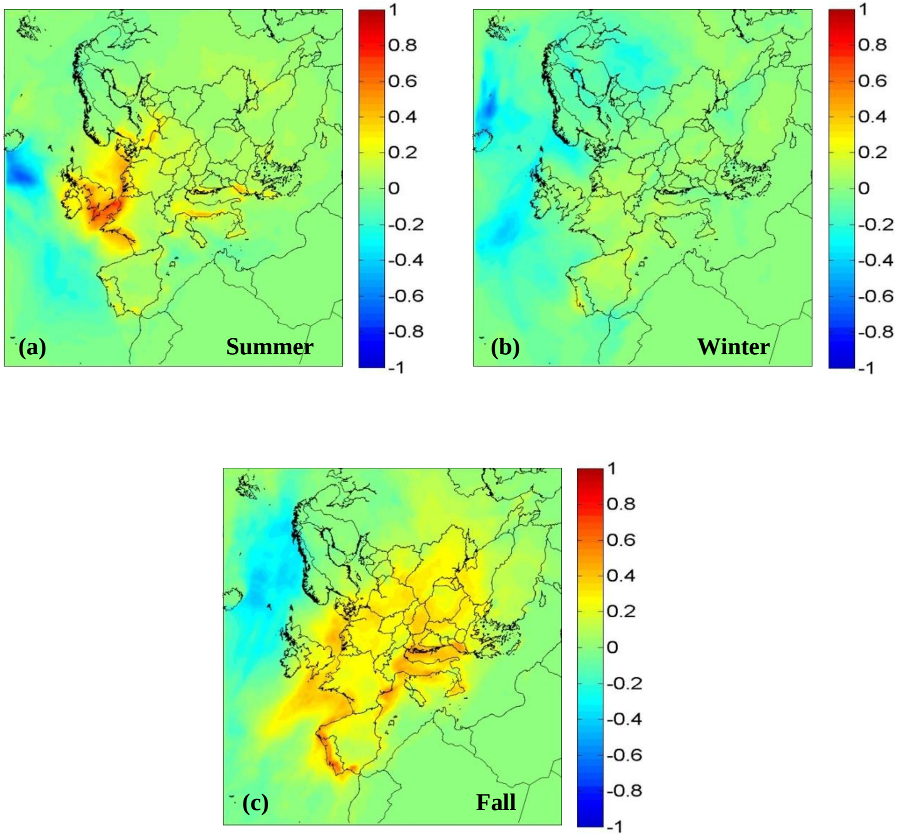

Fig. 8. Predicted average change in ground-level concentrations ($\mu g\, m^{-3}$) of total $PM_{2.5}$ due to a 5 % increase in absolute humidity during the modeled **(a)** summer, **(b)** winter, and **(c)** fall periods. A positive value corresponds to an increase.

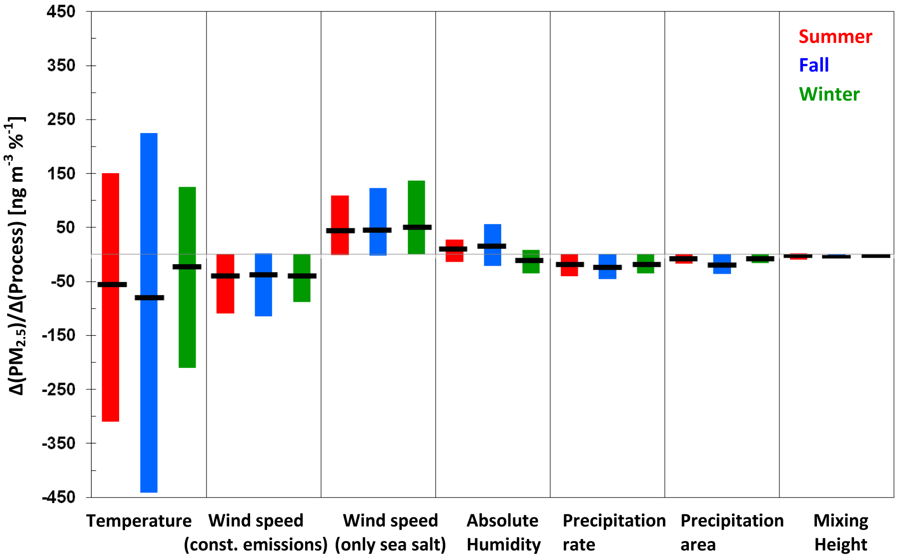

Fig. 9. Predicted simulation-average sensitivities of total $PM_{2.5}$ to changes in temperature, wind speed, sea salt emissions, absolute humidity, precipitation rate, precipitating area, and mixing height, during the three modeled periods. Each bar shows the range between the 10th and 90th percentiles. The black line in each bar shows the mean $PM_{2.5}$ sensitivity over the domain.

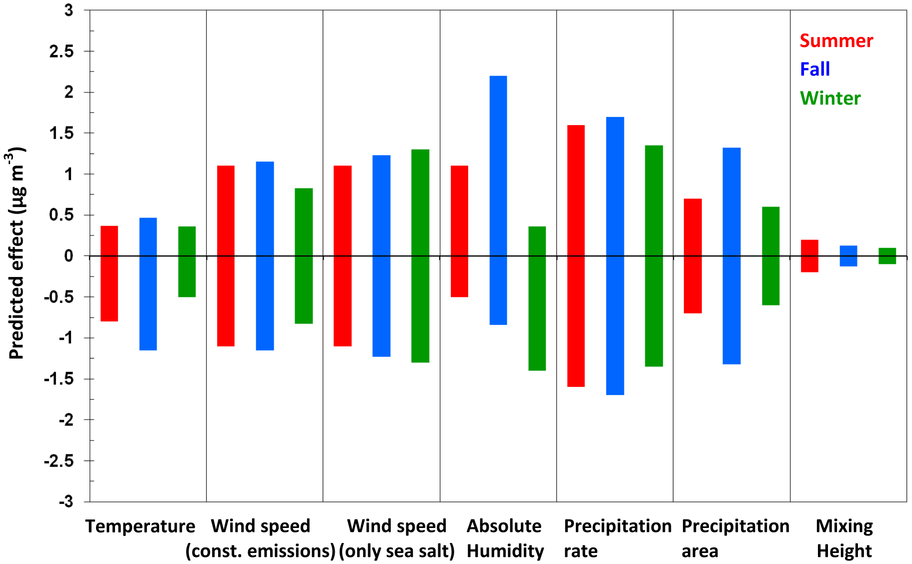

Fig. 10. Expected $PM_{2.5}$ concentration changes due to projected changes in temperature, wind speed, sea salt emissions, absolute humidity, precipitation rate, precipitating area, and mixing height in the future, during the three modeled periods.