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Particulate matter air pollution in Europe in a +2 °C warming world

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HIGHLIGHTS

- We evaluated the impacts of meteorological forcings upon chemistry transport models.
- 30-years simulations for present and future scenarios were performed.
- Impacts of emissions and +2 °C climate change upon particulate matter were studied.
- Reductions of PM can be mostly attributed to emission reduction policies.
- Changes due to a +2 °C warming alone are robustly predicted despite a weak signal.

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ABSTRACT

In the framework of the IMPACT2C project, we have evaluated the future European particulate matter concentrations under the influence of climate change and anthropogenic emission reductions. To do so, 30-year simulations for present and future scenarios were performed with an ensemble of four regional Chemical Transport Models. +2 °C scenarios were issued from different regional climate simulations belonging to the CORDEX experiment (RCP4.5 scenario). Comparing present day simulations to observations shows that these simulations meet the requested quality criteria even if some biases do exist. Also, we showed that using regional climate models instead of meteorological reanalysis was not critical for the quality of our simulations. Present day as well as future scenarios show the large variability between models associated with different meteorology and process parameterizations. Future projections of PM concentrations show a large reduction of PM10 and PM2.5 concentrations in a +2 °C climate over the European continent (especially over Benelux), which can be mostly attributed to emission reduction policies. Under a current legislation scenario, annual PM10 could be reduced by between 1.8 and 2.9 μg m^{-3} (14.1–20.4%). If maximum technologically feasible emission reductions were implemented, further reductions of 1.4–1.9 μg m^{-3} (18.6–20.9%) are highlighted. Changes due to a +2 °C warming, in isolation from emission changes, are in general much weaker (−1.1 to +0.4 μg m^{-3},-0.3 to +5.1% for annual PM10 averaged over the European domain). Even if large differences exist between models, we have determined that the decrease of PM over Europe associated with emission reduction is a robust result. The patterns of PM changes resulting from climate change (for example the increase of PM over Spain and southern France and the decrease of PM10 over eastern Europe) are also robustly predicted even if its amplitude remains weak compared to changes associated with emission reductions.

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1. Introduction

Among the atmospheric pollutants, particulate matter (PM) is of great concern, inducing degraded visibility (Seinfeld and Pandis, 2006), and affecting human health and ecosystems (Anderson et al., 2012; Aan de Brugh et al., 2011). The classification of PM is based on the size of the particles: PM10 and PM2.5 refer to particles with an aerodynamic diameter smaller than 10 μm and 2.5 μm, respectively. For these species, threshold values have been defined for the protection of human health (European Commission, 1999, 2001, 2008): a daily mean of 50 μg m^{-3} of PM10 should not be exceeded more than 35 times a year and the PM2.5 annual average should not exceed 25 μg m^{-3}. The composition of PM includes primary components, directly emitted into the atmosphere: elemental carbon, organic carbon, mineral dust, and sea salt) and secondary components: secondary inorganic aerosols (sulfates, nitrates, ammonium (SIA)), and secondary organic aerosols (SOA) resulting from the chemical reactions of precursor species (sulfur dioxide (SO_2), nitrogen oxides (NO_x), ammonia (NH_3) and volatile organic compounds (VOC).

The concentrations of PM are influenced by meteorological conditions (temperature, wind speed, humidity, stability of the atmosphere, precipitation, etc). These parameters may have multiple competing effects on PM concentrations. In fall and winter, stagnant conditions enhance levels of primary pollutants (SO_2 and NO_x) and PM while high temperatures and low precipitation during summer favor the production of secondary pollutants. Changes in temperature affect the formation of SIA (Jacob and Winner, 2009; Barmpadimos et al., 2012): increasing temperatures can lead to elevated sulfate concentrations as the rate of SO_2 oxidation increases. In contrast, inorganic and semi-volatile organic aerosols evaporate when temperature increases (Jiménez-Guerrero et al., 2012; Megaritis et al., 2014). Different studies (Sheehan and Bowman, 2001; Clark et al., 2016) provide evidence for a temperature effect, both on biogenic SOA precursor emission, gas-particle partitioning and on the chemical mechanism of SOA formation. Among the meteorological variables, wind speed, mixing height and relative humidity have the largest influence on PM concentrations (Gebhart et al., 2001). Increases in humidity favor nitric acid partitioning to the aerosol phase and can lead to nitrate concentration increases (Galindo et al., 2011; Lecoeur and Seigneur, 2013). However, high PM10 concentrations can also be related to dry conditions in certain areas due to increased potential for suspension of dust and soil (Wise and Comrie, 2005). High wind speed allows for greater ventilation, but can be correlated with high PM10 concentrations in specific regions and under certain conditions due to the resuspension of particles from the ground as well as the long-range transport of particulates (Gustafson and Leung, 2007). Changes in wind speed also lead to changes in marine and desert aerosol production (Aksoyoglu et al., 2011). Stagnant conditions are also correlated with high PM10 concentrations, as they allow particulates and their precursors to accumulate in the boundary layer (Triantafyllou et al., 2002). Changes in mixing height affect the dilution of primary pollutants and the formation of secondary pollutants (Jiménez-Guerrero et al., 2012; Pay et al., 2012). Megaritis et al. (2014) analyzed individual impacts of meteorological parameters on the concentration and composition of PM2.5 over Europe. They concluded that fine particulate matter (PM2.5) appears to be more sensitive to temperature changes than to the other meteorological parameters.

Due to the sensitivity of pollutant concentrations to weather conditions, it is expected that climate change will have an effect on air pollution, and particularly PM. The magnitude of climate change over the next century will depend on the mitigation strategies that will take place in the coming years. A major commitment of nations following the recent Paris agreement is to limit global warming below 2 °C above pre-industrial conditions. However, even a +2 °C global temperature change has been shown to induce significant changes on regional European climate (Vautard et al., 2014). Here we set out to examine what the implications of such a climate scenario would be for particulate matter pollution in Europe, in the framework of the EU-FP7 IMPACT2C project.

In the last decades, several studies have calculated the effects of climate change on air quality with a focus on PM at a European scale (Carvalho et al., 2010; Huszar et al., 2011; Juda-Rezler et al., 2012; Jiménez-Guerrero et al., 2012). These studies have isolated the effects of climate change by keeping similar anthropogenic emissions for the present time and future periods using a single model approach. Juda-Rezler et al. (2012) show a decrease in PM10 concentrations over central and Eastern Europe explained by an increase in precipitation under the IPCC SRES A1B scenario. Colette et al. (2013) show the results of sensitivity simulations, with climate and emission changes, under different scenarios. They point out those changes in air pollutant emissions are the main factor driving future air quality projections.

Both air chemistry and climate change are modeled with uncertainties. In order to estimate uncertainties, particular future climate projections can be studied with an ensemble of models. Here we estimate future concentrations of PM, in a climate that is 2 °C warmer than during pre-industrial times, and their uncertainties using an ensemble of 4 chains of models that simulate future air pollution with different global climate models (GCMs), regional climate models (RCMs) and chemical transport models (CTMs). Future concentrations are compared with historical ones, as simulated by the same modeling chain, and we examine where models agree and disagree. Since regional air pollutant emissions are likely to evolve in future decades due to legislation, we also account for emission changes and evaluate whether a +2 °C climate change is likely to alter the effects of reductions in emissions of air pollutants.

Initially, we examine whether the use of GCM-generated large-scale meteorology, instead of meteorological re-analyses, deteriorates the simulations of air quality. Previous studies have discussed the performance of long-term air quality hindcasts when driven by forcings from climate models (Katragkou et al., 2011; Zanis et al., 2011; Lacressonnière et al., 2012; Manders et al., 2012; Colette et al., 2013). However, statistics were obtained from relatively short periods (5–10 years) compared to the climate time scale due to computational constraints. Here we use 20- and 30-year periods to assess model performance. Also, the previous studies did not compare the performance of an ensemble of different models. Here, four different CTMs are used in coordinated experiments to provide an insight into the robustness of the results and the uncertainty given by the spread among the models. This study is unprecedented given the number of long (20 and 30 years) simulations and the resolution of the models (0.5° × 0.5°) over a European domain.

The description of the modeling systems, the simulations performed, and the databases used to evaluate the models, are given in Section 2. In Section 3, we discuss the performances of the models by comparing the simulations driven by reanalysis meteorological forcings with observations of chemical PM10 components and by comparing GCM-driven simulations with re-analyses-driven simulations. In Section 4, we compare future and historical simulated concentrations. Conclusions and perspectives are presented in Section 5.

2. Models and experiments

2.1. Participating models

The participating models in the project are the CTMs CHIMERE (Bessagnet et al., 2004; Menut et al., 2013), EMEP MSC-W (version 4.4, hereafter referred to as "EMEP") (Simpson et al., 2012), MATCH (Robertson et al., 1999; Andersson et al., 2007) and MOCAGE (Josse et al., 2004; Nho-Kim et al., 2005; Martet et al., 2009). The four models, in their IMPACT2C configurations, have been evaluated against observations for ozone concentrations using real and simulated historical climate (Watson et al., 2015). The key characteristics of the CTMs are described in Watson et al. (2015) and summarized in Table 1, along with their meteorological drivers. Except for MOCAGE, which uses climate projections from the ARPEGE-Climat GCM in its native global grid with a resolution of 0.5° over Europe, all CTMs use GCM data, which are downscaled using a RCM. The Weather Research and Forecasting (WRF) meteorological model was used to downscale meteorological input data for the CHIMERE and EMEP simulations (WRF has a different configuration in each case, see Katragkou et al. (2015)). The RCA4 and ARPEGE-Climat models are used to provide high-resolution meteorological input for the runs of MATCH and MOCAGE, respectively.

The model set-up is described in Lacressonnière et al. (2016). As reported in Table 1, different chemical, physical and dynamical approaches are used by the models to calculate PM concentrations. The number of bins used to represent the distribution of PM10 and PM2.5 varies between the models, from 2 (for EMEP) to 9 (for CHIMERE).

2.2. Air pollutant emissions

For the purposes of IMPACT2C project, the Representative Concentration Pathway (RCP) 4.5 climate scenario was used for the greenhouse gas emissions in the GCM runs simulating a change of 2 °C above pre-industrial times specific to each model. The ECLIPSE v4a anthropogenic emissions (http://eclipse.nilu.no/) were used in the CTMs in this work as these emissions are considered to be more appropriate for air quality modelling than the RCP emissions. They are based on an energy scenario close to RCP6.0 and RCP4.5, that have very similar radiative forcing for the year 2050 and are therefore coherent with the GCM runs. ECLIPSE was an EU collaborative project, in which IIASA developed future emissions data sets using the GAINS (Greenhouse Gas and Air Pollution Interactions and Synergies) model (Amann et al., 2011). These data comprise the annual sums of the emissions of black carbon, CO, NH_3, non-methane hydrocarbons, NOx, organic carbon and SOx. Ten sectors of anthropogenic activity are distinguished (i.e. SNAP sectors). In all CTMs, the time disaggregation was done on the basis of GENEMIS data (Ebel et al., 1994) using monthly, weekly and hourly coefficients depending on the activity sector (Society, 1994). This inventory has a spatial resolution compatible to our models (0.5° × 0.5°), and was available for both 2005 and 2050 (representative of the present-day and the period of +2 °C warming). Two different emission scenarios for the future are provided by ECLIPSE and used in this study: the Current Legislation Emission (CLE) and the Maximum Feasible Reduction (MFR) with the assumption of stringent air quality legislation (Supplementary Material, table A). Even though air pollutant emissions vary across years, we chose to keep constant emissions for all years at the levels of 2005 and 2050 representing the present-day and future respectively to remove inter-annual emission variations.

2.3. Experimental design

All experiments are conceived to be consistent with the EURO-CORDEX regional climate simulations. In order to evaluate the four chains of model simulations, we use a strategy similar to that of Watson et al. (2015). Two simulations are performed for the current climate (see Table 2):

- HINDCAST simulations cover a 20-year period (1989–2008) and are forced by meteorological fields from the CORDEX evaluation runs (ERA-Interim reanalysis forcing, see eg. Vautard et al., 2013; Kotlarski et al., 2014 for further details.)
- HISTORICAL simulations cover a 30-year period (1971–2000) and are forced by climate model outputs.

The difference between HINDCAST and observations will evaluate the quality of the simulation in the experimental framework. The difference between HINDCAST and HISTORICAL will allow for further diagnosis of the potential alteration of simulations when shifting from meteorological forcing by climate-models to forcing by re-analysis.

In order to assess future concentrations, a simulation (S1) using the 2050 CLE emissions and the meteorology for the future +2 °C period was carried out. The simulated +2 °C periods differ for each model (Table 2). By comparing results of S1 with those of HISTORICAL, we evaluate the effect of changes in climate variables superimposed with changes due to air pollutant emissions (hereafter referred to "climate and emission changes"). In order to estimate how climate change acts on future concentrations we conducted an additional simulation (S2) using the 2050 CLE emissions but the meteorology forcing provided by the EURO-CORDEX HISTORICAL simulation. The difference between S1 and S2 concentrations (hereafter refereed to "climate change alone") thus provides the contribution of climate change to simulated future concentrations in the absence of changes in air pollutant emissions. Finally the effect of the MFR scenario was investigated in the S3 simulation, similar to S1 but replacing CLE by MFR 2050 emissions (hereafter referred to "Emission scenario MFR"). Table 2 summarizes the ensemble of 5 simulations that were performed with each model.

2.4. Observational data

We evaluated the PM10 and PM2.5 concentrations and their chemical components with daily observations from the EMEP (European Monitoring and Evaluation Program, http://www.emep.int) and AirBase (http://dataservice.eea.europa.eu/dataservice) databases. Given the spatial resolution of the CTMs in the present study (about 0.5° × 0.5°), not all the reporting sites are fully representative. We used an objective classification of the AirBase sites based on past measurements proposed by Joly and Peuch (2012) in order to overcome issues related to the lack of homogeneity or erroneous information in the metadata. This pollutant-specific classification based on an extensive series of past measurement data defines 10 classes of measuring stations, from the least polluted sites (class 1) to the most polluted ones (class 10). This classification is performed independently for each type of pollutant and takes into account its chemistry, lifetime and transport. As in Lacressonnière et al. (2012), we chose to use classes 1–5 to evaluate the performances of PM10. Due to the lack of sufficient data of PM2.5 within the AirBase data set, there is no classification established for PM2.5 in Joly and Peuch (2012). We chose to use classes 1–5, considering that PM2.5 is a relatively long-lived species like PM10. Considering the stations that provide data for at least 5 years during the period 1998–2008, the number of stations

Table 1
Description of the four CTMs.

Model driving	GCM	RCM	Chemical mechanism	Chemical boundary conditions	Vertical resolution	Aerosol size	Aerosol speciation	Other
CHIMERE [CNRS-IPSL]	IPSL-CM5A-MR	WRF	MELCHIOR2 Lattuati (1997), Derognat et al. (2003): 44 species, 120 reactions	LMDz-INCA (Szopa et al., 2012)	First layer thickness: 20 m 8 levels from surface to 500 hPa	9 bins	Sea salt, desert dust, BC, primary OM (POM), SOA, SIA	Biogenic emissions from MEGAN v2.4 model (Guenther et al., 2006) Version CHIMERE 2001, latest developments in Menut et al. (2013) Dust parametrization (Menut et al., 2013)
EMEP [MET.NO]	NorESM	WRF	Andersson-Sköld and Simpson (1999) Simpson et al. (2012)	LMDz-INCA (Szopa et al., 2012)	First layer thickness: 90 m 20 levels from 90 m to 100 hPa	2 bins	Sea Salt, desert dust, EC, POM, SOA, SIA	Biogenic emissions build upon maps of 115 forest species (Koble and Seufert, 2001). Emission factors for each forest species and for other land-classes are based upon Simpson et al. (1999, 2012), and driven by hourly temperature and light using algorithms from Guenther et al., 1995)
MATCH [SMHI]	EC-EARTH	RCA4	Simpson et al, (1993); Langner and Pleijel (1998); SIA formations as in Andersson et al. (2007) SOA formation by oxidation of anthropogenic and biogenic compounds based on the volatility basis set scheme in the EMEP MSC-W model (Bergström et al., 2012, 2014)	LMDz-INCA (Szopa et al., 2012)	First layer thickness: 60 m Lowest 5 km divided in 20 layers	4 bins	Sea Salt, desert dust, EC, primary OM, SOA, SIA	Isoprene emissions calculated online using the E−94 isoprene emission methodology by Simpson et al. (1995). Terpene emissions calculated online as in Simpson et al. (2012) Sea salt emissions based on Monahan et al. (1986) for larger and Martensson et al. (2003) for finer particles, but with temperature corrections following Sofiev et al. (2011) as described in Soares et al. (2016) No internal desert dust emissions; this compound is purely transported from the boundary. Primary particles (EC, OM, sea salt, desert dust) treated with a 4-bin bulk model without growth processes between the size bins as in Andersson et al. (2007). Secondary particles (SIA and SOA) compounds are treated as trace species.
MOCAGE [Météo-France]	ARPEGE	ARPEGE	RACM (Stockwell et al., 1997) and REPROBUS (Lefèvre et al., 1994): 108 species	MOCAGE	First layer thickness: 40 m 47 levels from 40 m to 5 hPa	6 bins	Sea salt, desert dust, BC, POM	Global and regional domains two way nested Biogenic emissions come from the Global Emissions Inventory Activity (GEIA), modified to match the European emission totals generated with the MEGAN v2.4 model (Guenther et al., 2006)

Table 2
Description of the simulations performed for the current (HINDCAST, HISTORICAL) and future periods (S1, S2 and S3). The future +2 °C period is different for each model. The IPSL GCM reaches this threshold during 2027–2056 in the RCP4.5 scenario while NorESM reaches this threshold for 2056–2085. EC-EARTH reaches it during 2041–2070 and ARPEGE for the 2038–2067 period. * Current legislation emissions. ** Maximum feasible reductions.

Simulations	Climate	Boundary conditions	Emissions
HINDCAST	1989–2008	2005	V4a 2005
HISTORICAL	1971–2000	2005	V4a 2005
S1	+2 °C RCP4.5	2050	V4a 2050 CLE*
S2	1971–2000	2050	V4a 2050 CLE*
S3	+2 °C RCP4.5	2050	V4a 2050 MFR**

reaches 358 for PM10 and 26 for PM2.5.

SIA measurements were obtained through the EMEP database. A total of 16 stations, which provide daily observations for at least 5 years over 1998–2008, were selected (see Supplementary material). The locations of the selected AirBase and EMEP stations are displayed in Fig. 1. For the evaluation of particulate matter concentrations, three statistical metrics were selected for the present study: (i) the mean bias (MB) and (ii) the sigma ratio (σ, i.e. modeled standard deviation divided by observed standard deviation). These statistics were calculated for the daily mean values at each selected station and averaged over annual and seasonal periods. As suggested by Boylan and Russell (2006), we also considered (iii) the mean fractional bias (MFB). These authors proposed that the model performance criteria would be met when |MFB| ≤ 60%. The model performance goal would be met when |MFB| ≤30%.

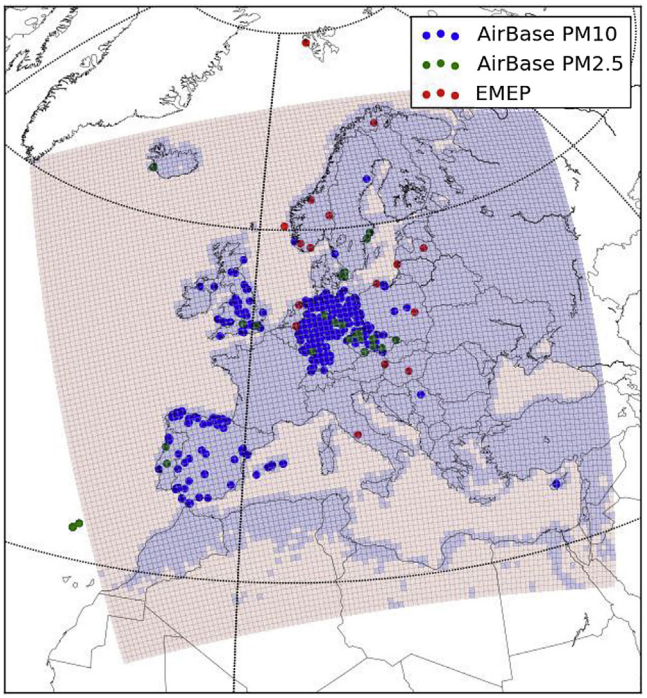

Fig. 1. Map of AirBase stations providing PM10 (circles in blue) and PM2.5 (circles in green) data. EMEP stations (circles in red) provide data of sulfate, nitrate and ammonium aerosols. The blue area shows the EURO-CORDEX domain used for calculations of aerosols average, while the red area (oceans and northern Africa) illustrates the domain excluded. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3. Evaluation of simulations for PM10, PM2.5 and SIA

3.1. HINDCAST simulations vs. observations

We first briefly discuss the overall ability of the HINDCAST simulations to reproduce climatological PM concentrations. Time series of monthly mean PM10 concentrations are presented in Fig. 2. A common underestimation of PM10 is observed for the MOCAGE, EMEP and MATCH models, as found in previous studies (see eg. Stern et al., 2008; Solazzo et al., 2012b).

For CHIMERE, PM10 levels are slightly overestimated from October to December and underestimated for the rest of the year. The seasonal cycles are well reproduced by the CHIMERE, EMEP and MATCH models. The underestimation of PM10 in MOCAGE (Fig. 2) is exacerbated by the lack of secondary organic and inorganic aerosols in this model. Table 3 summarizes the statistics of the daily mean PM10 and PM2.5 levels, averaged for the annual and seasonal (JJA and DJF) periods of 1998–2008 over all the European stations considered. Negative mean biases are found for all the models and seasons. The sigma ratios calculated for both seasons indicate that the models globally well estimate the range of variability (Table 3.), with the exception of MOCAGE during the winter season. The annual sigma ratio for CHIMERE and EMEP agree well with the observed variability. The model performance criteria, suggested by Boylan and Russell (2006), are met for the CHIMERE, EMEP and MATCH models for JJA and DJF periods.

Results for the MB, standard deviation and mean are presented in Table 4 for the SIA simulated with CHIMERE, EMEP and MATCH. MOCAGE is not included due to the fact that it does not simulate secondary inorganic aerosols. The models reproduce the mean sulfate concentrations fairly well, with small mean biases (MB = −0.2–0.8 μg m^{-3}). The variability of the observations (Std Dev = 1.8 μg m^{-3}) is overestimated by CHIMERE and EMEP, and underestimated by MATCH. The mean concentrations of nitrate are slightly overestimated by CHIMERE (MB = 0.4 μg m^{-3}) and EMEP (MB = 0.5 μg m^{-3}) as well as the variability (Std Dev = 2.9 μg m^{-3} for CHIMERE and Std Dev = 2.0 μg m^{-3} for EMEP as compared to 1.6 μg m^{-3} for observations). MATCH agrees well with the observed variability (Std dev = 1.4 μg m^{-3}) and the mean levels of nitrate (Mean = 1.7 μg m^{-3}). For ammonium, the levels simulated by EMEP and MATCH agree well with the observations as suggested by the lower biases.

The statistical scores of PM10 and PM2.5 display equivalent or higher performances compared to previous studies. Significant negative biases for particulate matter concentrations are commonly seen in many air quality models (Lacressonnière et al., 2012; Solazzo et al., 2012a; Colette et al., 2013). In the present study, the use of non-nudged meteorology inside the European domain can also be responsible for degrading the performances of the models, since regional climate models, even driven at the boundaries by re-analyses, can develop their own biases within the model domain. However, the performance criteria set by Boylan and Russell (2006) are met with only few exceptions.

3.2. Cross model variability and alteration of simulations due to global climate model meteorological forcing

In this and the following section, the average concentrations of PM and its components have been calculated using a masked EURO-CORDEX domain, that represents the European land masses and excludes the oceans and parts of northern Africa (Fig. 1).

An analysis of the annual cycle of mean concentrations for primary and secondary components over Europe gives complementary information about the ensemble variability and the impact of the climatic forcing. Fig. 3 shows that for primary components differences between models are weak during winter and spring (about 2.5 μg m^{-3}) but larger in summer/fall due to higher dust

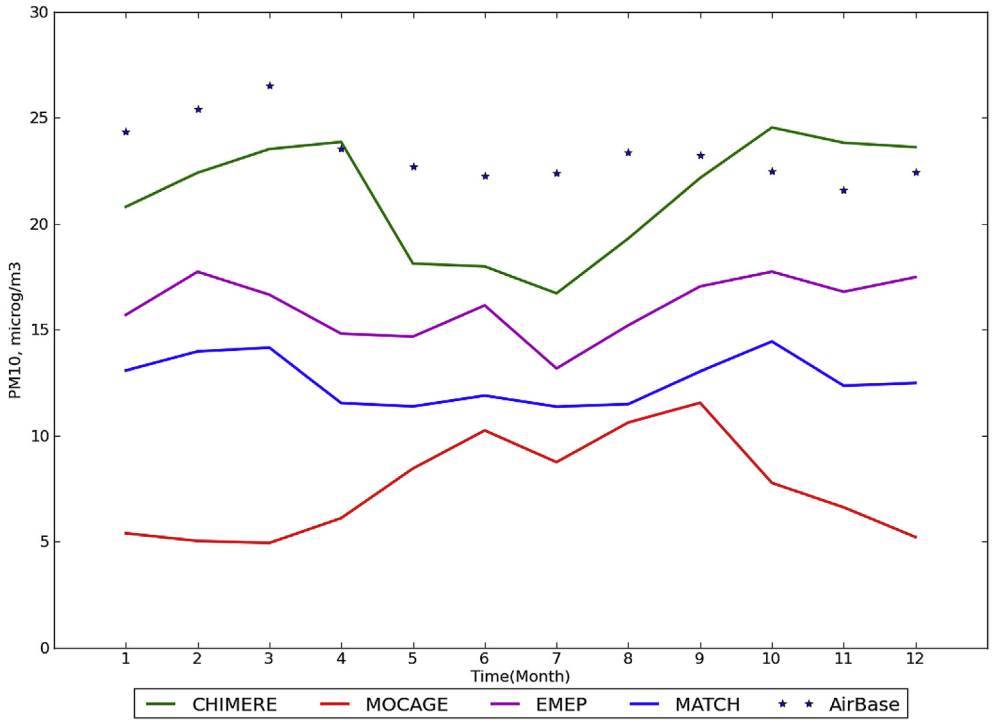

Fig. 2. Time series of monthly average daily PM10 levels (μg m^{-3}) over the period 1998–2008, simulated by CHIMERE (in green), MOCAGE (in red), EMEP (in purple) and MATCH (in blue); and measured by AirBase (blue stars). The time series are averaged over the stations. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 3
Annual and seasonal (JJA: June, July and August, DJF: December, January and February) statistics over Europe at AirBase stations. Statistics are averaged for the 10-yr period of 1998–2008. The calculated statistics are mean bias (MB, μg m^{-3}), sigma ratio (σ), and mean fractional bias (MFB, %). MFB = $\frac{2}{N}\sum_{i=1}^{n}\frac{(Mi-Oi)}{(Mi+Oi)}$ (M refers to the model, O refers to the observations). Statistics are computed for the PM10 and PM2.5 daily mean values.

		PM10 daily mean			PM2.5 daily mean		
		MB	σ	MFB	MB	σ	MFB
Annual	CHIMERE	−0.9	0.9	1.1	−0.5	1.0	0.9
	MOCAGE	−15.7	0.4	−94.6	−8.9	0.6	−72.1
	EMEP	−6.6	0.8	−28.4	−5.2	0.7	−34.4
	MATCH	−10.0	0.6	−50.9	−3.6	0.6	−12.7
JJA	CHIMERE	−3.5	0.8	−14.8	−0.9	1.0	−10.5
	MOCAGE	−12.6	0.6	−78.0	−4.2	1.0	−38.7
	EMEP	−7.3	1.0	−36.9	−3.4	1.1	−32.3
	MATCH	−10.3	0.7	−58.7	−3.5	0.7	−24.9
DJF	CHIMERE	0.1	0.9	8.3	−1.6	1.1	−2.2
	MOCAGE	−18.7	0.2	−97.9	−14.1	0.2	−95.6
	EMEP	−5.8	0.9	−18.9	−6.8	0.7	−36.4
	MATCH	−10.1	0.8	−45.2	−3.6	0.8	0.4

concentrations in CHIMERE and much larger OC concentrations in MOCAGE (not shown). We also note that differences between HISTORICAL and HINDCAST runs rarely exceed 1 μg m^{-3} except for MOCAGE in the summer/fall period. Contrary to primary species, for secondary components differences between models are weak for the summer/fall period but larger in winter/spring. Here again the differences between HISTORICAL and HINDCAST generally remain below 1 μg m^{-3}.

The chemical compositions of PM10 aerosols in HINDCAST and HISTORICAL simulations for the CHIMERE, EMEP and MATCH models (Fig. 4) are also fairly similar. MOCAGE is not included in this analysis due to its lack of secondary organic and inorganic aerosols. All three models represent the sulfate, nitrate, ammonium, sea salt, primary organic matter (POM), black carbon (BC), SOA and desert dust. Among the secondary inorganic aerosols, sulfate makes up the largest proportion in MATCH (up to 23.3%) and CHIMERE (up to 20.2%) by comparison to nitrate and ammonium while the fractions in sulfate and nitrate have a similar importance (up to 17.7% for nitrate and 19.2% for sulfate) in EMEP. The proportions of SOA range between 4.9 and 7.9%. The share of dust in CHIMERE (23.4–25.6%) is higher than in EMEP (12.8–16.1%) and MATCH (16.8–17.6%) due to a larger influx of desert dust at the southern boundary and higher levels over continental Europe in CHIMERE. The absolute concentrations of dust are similar in MATCH and EMEP. Keeping in mind that MATCH only simulates dust as a component forced by model lateral boundaries, this highlights the importance of boundary influx. Putaud et al. (2010) showed that the contributions of dust to PM10 for different types of sites (rural, urban) reach 4–12% over northwestern Europe, 15–28% over southern Europe and 9–15% over central Europe, which is in agreement with model outputs. The fraction of sea salt is lower in CHIMERE and MATCH than in EMEP. The absolute concentration is around 2 μg m^{-3} in both CHIMERE and MATCH, while it reaches 3.5 μg m^{-3} in EMEP. Finally, the proportion of primary organic matter (POM) is lower in EMEP (3.7%) and MATCH (6.6–6.8%) than in CHIMERE (10.3–10.8%).

To summarize, the use of meteorological forcing from a regional climate model driven by global climate model data on its boundaries, as compared to a three-dimensional re-analysis does not affect the chemical composition of PM. Differences in relative fractions averaged over Europe between both scenarios are small, in general less than about 1% (i.e. less than differences between models). We also found only small changes between the simulated primary and secondary PM10 levels in HISTORICAL and HINDCAST simulations for both summer and winter seasons except for summer/fall for MOCAGE, again these differences are in general smaller than those between models. This evaluation gives confidence in the

Table 4
Annual statistics obtained with CHIMERE, EMEP and MATCH over Europe at the EMEP stations. Statistics are averaged for 1998–2008 period. The computed statistics are mean bias (MB, μg m^{-3}), and simulated standard deviation (Std Dev, μg m^{-3}). Statistics are computed for the sulfate, nitrate and ammonium daily mean value. Daily mean values of sulfate, nitrate and ammonium simulated and observed, as well as observed standard deviations are computed.

	Sulfate			Nitrate			Ammonium		
	Mean	MB	Std Dev	Mean	MB	Std Dev	Mean	MB	Std Dev
CHIMERE	3.1	0.8	2.4	2.1	0.4	2.9	1.6	0.6	1.4
EMEP	2.1	−0.2	1.9	2.2	0.5	2.0	1.0	0.0	0.9
MATCH	1.9	−0.4	1.4	1.7	0.0	1.4	0.9	−0.1	0.6
Observation	2.3		1.8	1.7		1.6	1.0		0.8

use of climatic simulation to force the CTMs and derive future air quality conditions.

4. Future changes in PM concentrations

In this section, the robustness of future changes in PM10 and PM2.5 concentrations are discussed. A specific study has been conducted to evaluate the robustness of regional climate change (S1–S2) in Lacressonnière et al. (2016). Fig. 5 shows the PM10 ensemble average changes of the CHIMERE, EMEP and MATCH models due to climate and emission changes (a, S1-HISTORICAL), emission scenario MFR (b, S3–S1) and climate change alone (c, S1–S2). We chose not to include MOCAGE, because of the lack of inorganic and organic aerosols.

The impact of a global +2 °C warming relative to preindustrial times and of estimated emission changes from the present time to the equivalent time period is highlighted in Fig. 5a. A general reduction in PM10 is simulated over continental Europe, with the largest changes of PM10 over Western Europe. This decrease of PM10 is robust in almost all areas (see black bullets in Fig. 5a). Similar results are simulated for PM2.5 (Supplementary Material, Fig. A). On average over the European land domain (Table 5), the concentrations of PM10 in the future CLE scenario are between 1.8 and 2.9 μg m^{-3} (14.1–20.4%) lower than HISTORICAL. The decreases of PM2.5 range from 1.6 μg m^{-3} to 3.1 μg m^{-3} (16.7–31.7%). These changes are mainly due to projected decreases in emissions of primary PM and PM precursor gases (SO_2 and NOx). The largest changes are exhibited over North Africa in CHIMERE, MOCAGE and EMEP (Supplementary Material, Fig. B), and can mainly be attributed to the changes in dust emissions, due to changes in precipitation and winds. Weaker changes of dust concentrations are simulated in MATCH, because dust emissions are not accounted for within the simulation domain.

Compared to the CLE scenario, the mitigation scenario (use of MFR emissions scenario) enhances the decrease of PM10 compared to the current legislation over Europe in all the models (Fig. 5b), with the largest decrease of up to 5 μg m^{-3} over eastern Europe. When averaged over the masked domain (Table 5), the results show further reductions of PM10 that range from 1.4 μg m^{-3} to 1.9 μg m^{-3} (18.6–20.9%). The reductions in PM2.5 range from 24.8% to 32.9%.

The projected changes of PM10 concentrations due to climate change alone (S1–S2) is robust in many areas (Fig. 5c), but the amplitude of the changes remains small (−0.1–0.4 μg m^{-3}, -0.3–5.1%), as compared to the variability of concentrations between the models (Supplementary Material, Fig. A), and to changes that would be obtained from air pollutant emission reduction policies. These findings are in agreement with the conclusions of Lacressonnière et al. (2016) for PM2.5 changes. Changes due to a global +2 °C warming therefore remain uncertain. However, relatively good agreement is found regarding the increase of desert dust concentration over the Iberian Peninsula and southern France (see Lacressonnière et al., 2016), and the decrease of PM10 over eastern Europe (Russia). According to the analysis of PM components, the changes in PM10 are mainly due to changes in natural emissions, such as desert dust, sea salt and biogenic emissions, affected by changes in meteorology. The stronger meridional wind component (Supplementary material, Fig. C) over north Africa and Spain in the future climate favors the transport of desert dust into the domain. Over the northern North Atlantic and the North Sea, westerlies become stronger in the future scenario (Supplementary material, Fig. C) and support the transport of sea salt aerosol to Europe. The decrease of precipitation (Supplementary material, Fig. D) over central Europe partly explains the decrease of PM.

The average aerosol composition across the masked domain is presented in Fig. 6 for the future simulations and HISTORICAL for the CHIMERE, EMEP and MATCH models.

All the secondary aerosol species decrease in the future scenarios as the emissions of anthropogenic precursors decrease. The small increase in NH_3 emissions between 2005 and 2050 CLE scenario is not reflected in the formation of ammonium. The ammonium concentrations decrease from HISTORICAL to S2 in all the models. This feature is likely related to decreased formation of ammonium sulfate and nitrate following the large reductions in available SO_2 and NOx in all future scenarios (Engardt and Langner, 2013). This projection of aerosol compositions shows increasing relative contribution of sea salt and dust aerosols in the future.

5. Conclusions

Within the context of the FP7 IMPACT2C project, we have in this work used four regional CTMs, each driven by a different climate model, to assess the impact of a +2 °C global warming on future European particulate matter concentrations and composition. We first analyzed the ability of the models, forced by re-analysis data at the lateral boundaries and without nudging (ERA-Interim, HINDCAST simulation), to reproduce the observed levels and variability of PM for a past period (1989–2008). The statistical results are similar to previous studies with the participating models. All models exhibited negative biases for PM10 and PM2.5, while the simulated concentrations of secondary inorganic aerosols (mainly contributing to PM2.5) are in good agreement with the observations. Common performance criteria are mostly met by the models. The larger biases observed in MOCAGE are caused by the fact that this model does not take all major species into account. We have also verified the impact of using GCM models as lateral boundary forcings (HISTORICAL simulation) instead of re-analysis (HINDCAST) on the simulated PM concentrations. Over the domain selected for this study, the spatial pattern and amplitude of the differences in PM10 mass concentrations between the two simulations vary greatly among the models. However, seasonal cycles are similar in both simulations when averaged over the continental European domain. The chemical composition of PM10 remains largely unchanged. Furthermore, the contributions of the individual aerosol components appear similar between the models. Finally, the comparisons of the two experiments, constrained by

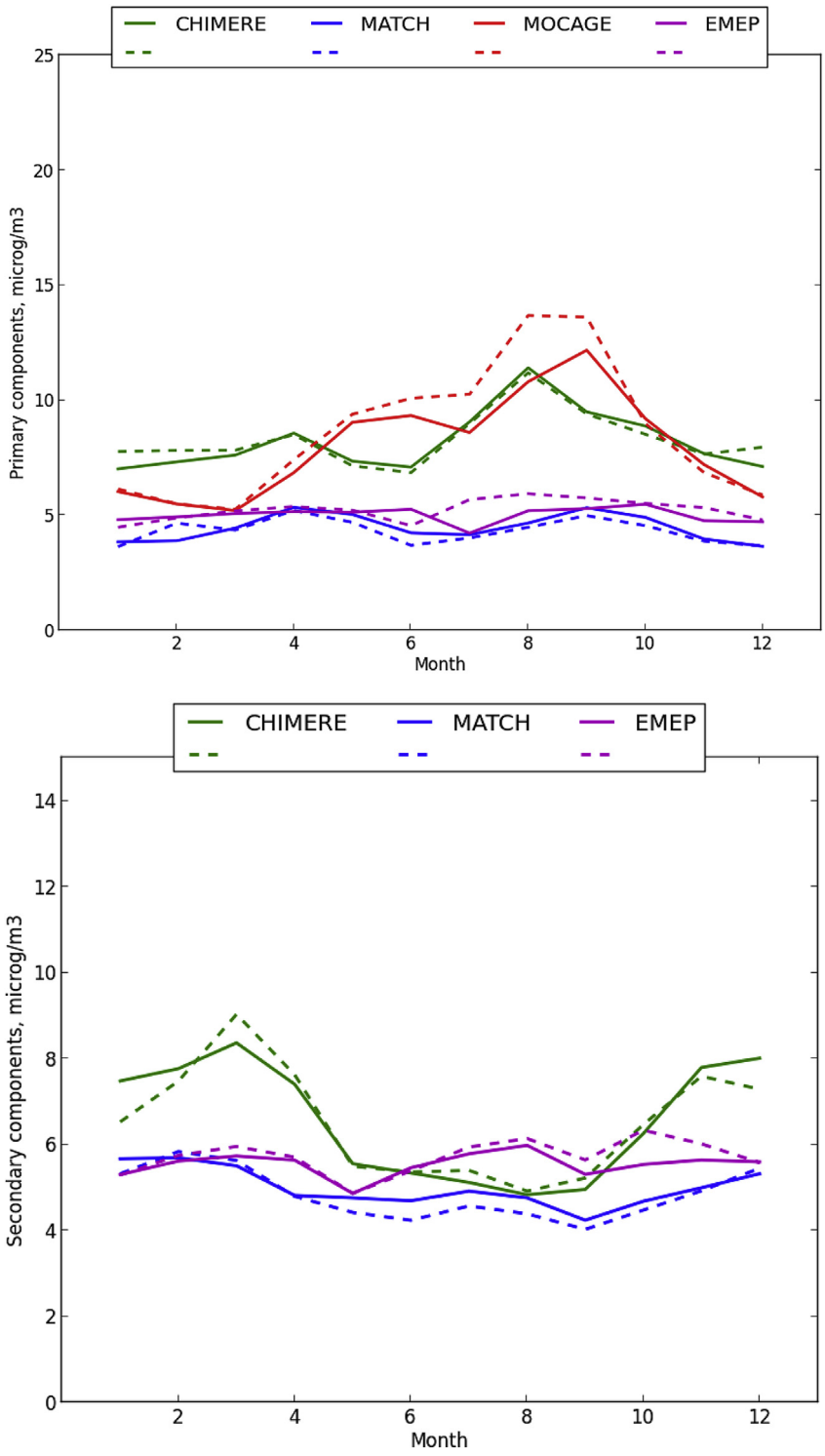

Fig. 3. Time series of monthly mean primary (Top) and secondary (bottom) components of PM10, averaged over the period simulated for HINDCAST (-) and HISTORICAL (- -), across the masked EURO-CORDEX land domain (see Fig. 1).

analyses or climate model forcings, suggest that the induced differences in particulate matter and its chemical components are small compared to the inter-model variability. Finally, we evaluated future change in PM concentrations. Several scenarios were constructed, S1 used +2 °C climate (based on RCP4.5) and future emissions inventories taking into account current legislation reductions, S2 with historical climate, S3 with future climate but more stringent emission reduction scenario. Comparing S1 and HISTORICAL simulations shows large reductions in PM10 over continental Europe (more specifically over Benelux) and even stronger increases and decreases over northern Africa. The decrease of PM10 range from 1.8 to 2.9 μg m^{-3} (14–20.4%) when averaged over the land domain. The S1 minus S2 comparisons indicate that the PM reductions over continental Europe are largely associated to anthropogenic emission reductions, and that the impact of climate change is comparatively small in our modelling

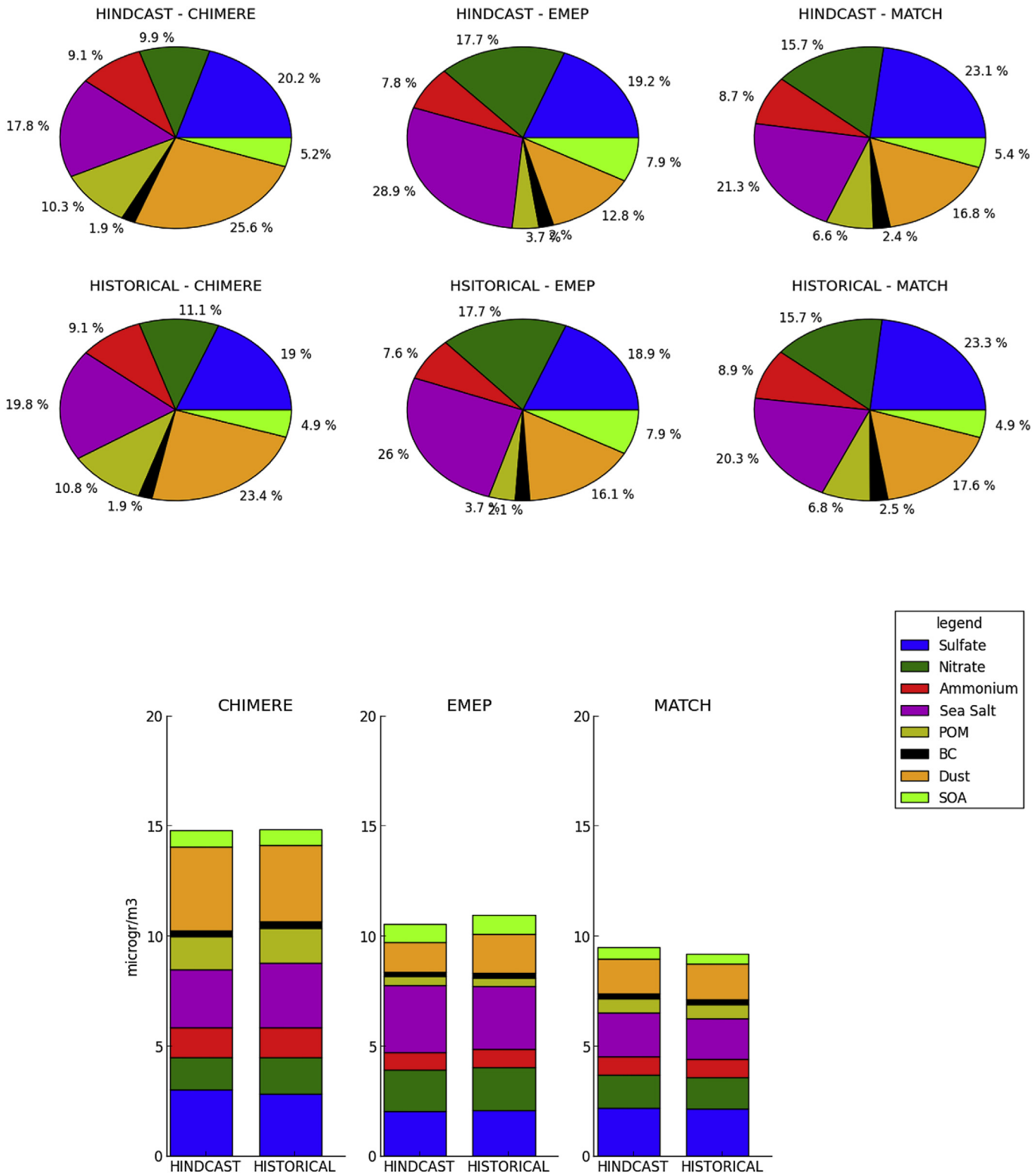

Fig. 4. Contribution (top) and absolute concentrations (bottom) of aerosol species: sulfate, nitrate, ammonium, sea salt, primary organic matter (POM), black carbon, dust and secondary organic matter (SOA) in PM10, simulated by HINDCAST and HISTORICAL averaged over the masked domain.

chains, both for PM10 and individual compounds. Reductions in PM are more important considering MFR scenarios that take into account more stringent emissions reductions than CLE scenarios. In average over the masked domain, further reductions of PM10 that range from 1.4 to 1.9 μg m^{-3} (18.6–20.9%) are obtained with the MFR scenario when comparing to CLE. All secondary components (mainly associated with anthropogenic emissions) are efficiently reduced by emission control, while the impact of a changing climate is much weaker over continental Europe. Using the ensemble average, we have tested the robustness of PM changes.

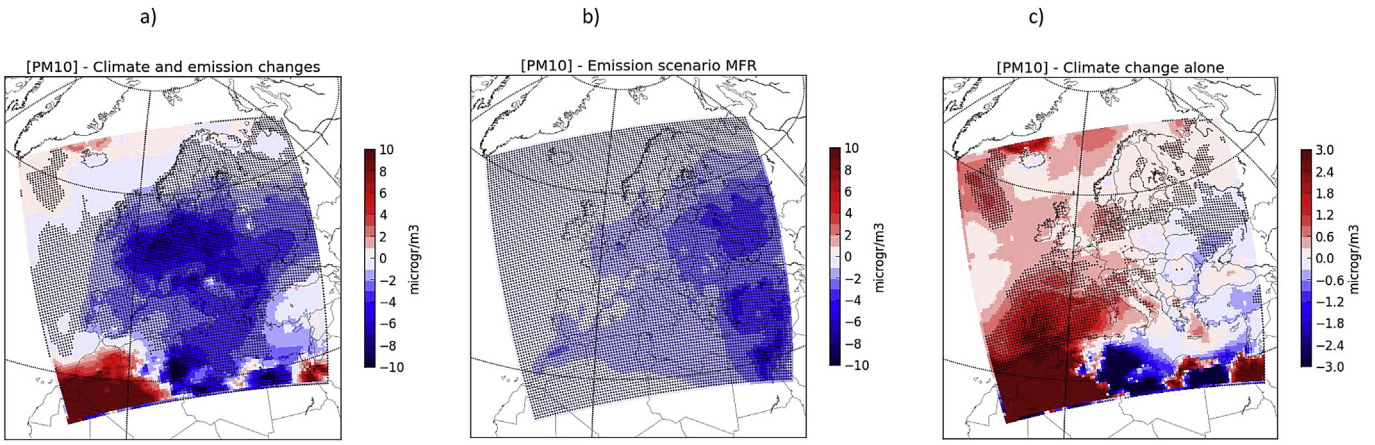

Fig. 5. PM10 average predicted changes for scenarios: (a) CLE scenarios under 2 °C warming with 2050 emissions (S1-HIST), (b) 2050 MFR emissions (S3−S1) and (c) effects of 2 °C climate change only (S1−S2). Black bullets are shown when the three models agree on the sign of PM10 changes.

Table 5
Annual average changes of PM10 and PM2.5 concentrations over the masked EURO-CORDEX land domain (see blue area in Fig. 1) due to climate and emissions changes, emission scenario MFR and climate change alone, simulated by the 4 CTMs (CHIMERE, EMEP and MATCH). In μg m^{-3} and % between brackets.

	PM10			PM2.5		
	Climate and emission changes (S1−HISTORICAL)	Emission scenario MFR (S3 −S1)	Climate changealone (S1−S2)	Climate and emission changes (S1−HISTORICAL)	Emission scenario MFR (S3−S1)	Climate change alone (S1−S2)
CHIMERE	−2.9 (−17.1%)	−1.9 (−18.6%)	−0.1 (−0.3%)	−3.1 (−26.3%)	−1.8 (−26.4%)	−0.3 (−3.5%)
EMEP	−1.8 (−14.1%)	−1.8 (−19.1%)	0.4 (2.7%)	−1.6 (−16.7%)	−1.6 (−24.8%)	0.2 (2.2%)
MATCH	−2.0 (−20.4%)	−1.4 (−20.9%)	0.3 (5.1%)	−2.2 (−31.7%)	−1.4 (−32.9%)	0.1 (2.0%)

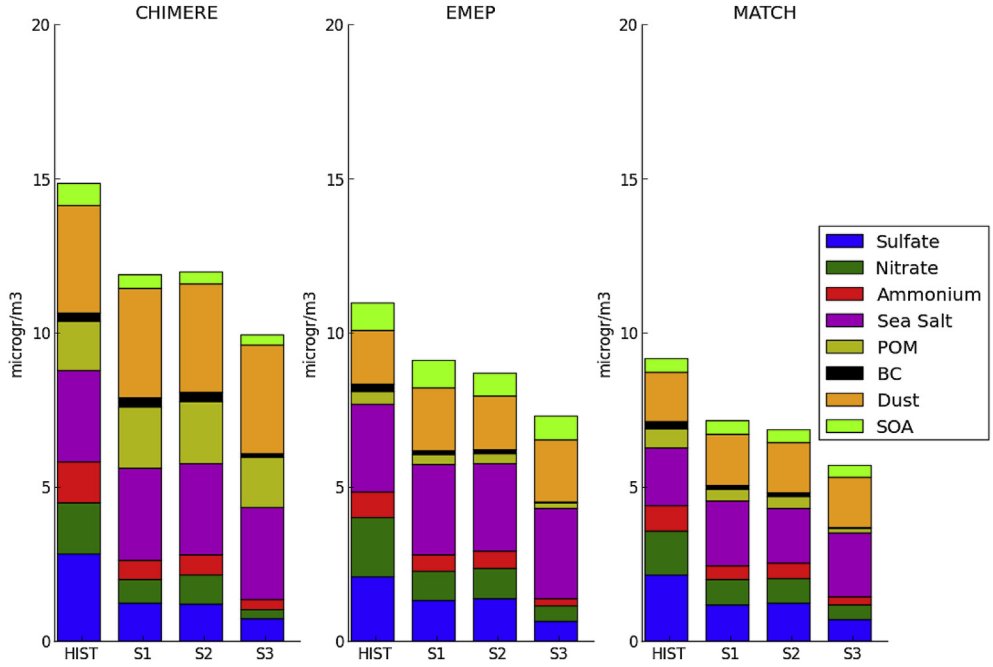

Fig. 6. PM10 aerosol composition over the masked land domain in HISTORICAL, and the future scenarios S1, S2 and S3.

The decrease associated to emission reductions exhibits a robust pattern all over Europe and is in range of 4–6 μg m^{-3} with CLE and up to 9 μg m^{-3} with MFR over continental Europe. Concerning the impact of a +2 °C climate, the model results are robust over large part of Europe even if the signal is weak.

To conclude, future European air quality levels could be largely improved if all feasible mitigation measures were implemented to reduce anthropogenic emissions, while the effects of a 2° global warming are comparatively small.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2017.01.037.

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