



# European atmosphere in 2050, a regional air quality and climate perspective under CMIP5 scenarios

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**Abstract.** To quantify changes in air pollution over Europe at the 2050 horizon, we designed a comprehensive modelling system that captures the external factors considered to be most relevant, and that relies on up-to-date and consistent sets of air pollution and climate policy scenarios. Global and regional climate as well as global chemistry simulations are based on the recent representative concentration pathways (RCP) produced for the Fifth Assessment Report (AR5) of the IPCC (Intergovernmental Panel on Climate Change) whereas regional air quality modelling is based on the updated emissions scenarios produced in the framework of the Global Energy Assessment. We explored two diverse scenarios: a reference scenario where climate policies are absent and a mitigation scenario which limits global temperature rise to within 2 °C by the end of this century.

This first assessment of projected air quality and climate at the regional scale based on CMIP5 (5th Coupled Model Intercomparison Project) climate simulations is in line with the existing literature using CMIP3. The discrepancy between air quality simulations obtained with a climate model or with meteorological reanalyses is pointed out. Sensitivity simulations show that the main factor driving future air quality projections is air pollutant emissions, rather than climate change or intercontinental transport of pollution. Whereas

the well documented “climate penalty” that weights upon ozone (increase of ozone pollution with global warming) over Europe is confirmed, other features appear less robust compared to the literature, such as the impact of climate on PM<sub>2.5</sub>. The quantitative disentangling of external factors shows that, while several published studies focused on the climate penalty bearing upon ozone, the contribution of the global ozone burden is somewhat overlooked in the literature.

## 1 Introduction

Air quality and climate are closely inter-related in their mitigation, their functioning, and their impacts (Jacob and Winner, 2009). Climate policies imply energy efficiency and other technical measures that have an impact on a wide range of human activities and, in turn, on air quality. Reciprocally, air quality mitigation measures may also have an impact on greenhouse gas emissions. In addition, air quality is sensitive to climate change (which affects physical and chemical properties of the atmosphere and therefore drives the frequency of weather events yielding favourable conditions to the build up of pollution). Last, many air pollutants (both gaseous

and particulate) have direct and indirect impacts on climate through the radiative balance of the atmosphere (Forster et al., 2007).

The combined and sometimes competing role of these interlinkages calls for integrated assessment frameworks (EEA, 2004). Integration of technical mitigation measures, their costs and their impact on air quality have been successfully implemented over the past decades to investigate relatively short time periods (Cohan and Napelenok, 2011). In Europe, the GAINS (Greenhouse Gas and Air Pollution Interactions and Synergies) modelling framework (Amann et al., 2011; Amann and Lutz, 2000) is used extensively to support the design of cost-effective emission reduction strategies. The optimisation core of GAINS is based on a number of source–receptor sensitivity simulations with the EMEP chemistry-transport model (Simpson et al., 2012) designed to explore the impact on air quality of incremental changes in European emissions over the next couple of decades. However the robustness of these optimisation tools for longer-term projections is challenged by externalities such as the global burden of pollution and the expected increase of ozone pollution with global warming referred to as “climate penalty” (Wu et al., 2008).

Atmospheric chemistry transport and climate models can contribute to better quantify these externalities. The most established approach to tackle such issues consists in relying on ensembles of models exploring a range of likely futures in order to derive an envelope of projections, as being done in the widely documented IPCC (Intergovernmental Panel on Climate Change) framework (IPCC, 2007). When it comes to atmospheric chemistry there is still a gap between research communities working on global and regional scales. Global chemistry-transport modelling teams are closely aligned with the Climate Model Intercomparison Project (CMIP) (Taylor et al., 2012). A dedicated Atmospheric Composition Change Model Intercomparison Project (ACCMIP) (Lamarque et al., 2013) was recently tailored to produce a consistent envelope of atmospheric composition projections accounting for climate impacts. Such global modelling initiatives often include sophisticated handling of coupling and feedbacks (especially with regard to the radiative impact of short-lived climate forcers; Shindell et al., 2013). However they suffer of a lack of refinement over given regional areas and many of these tools include a simplified formulation of chemical processes, especially with regard to secondary aerosol formation.

Regional air quality (AQ) and climate modelling systems can fill these knowledge gaps. However, robust quantification of regional AQ externalities is still suffering from a lack of coordinated multi-model initiatives that would cover the whole range of processes involved. Over the past few years, there has been a growing body of literature on the ozone climate penalty over Europe (Andersson and Engardt, 2010; Hedegaard et al., 2008, 2013; Katragkou et al., 2011; Langner et al., 2012a, b; Manders et al., 2012; Meleux et al., 2007), amongst which only Langner et al. (2012b) offers an

ensemble perspective. This climate penalty is however rarely compared to other influential factors whereas the latest evidences suggested that reductions in air pollutant (AP) emissions would largely compensate the climate penalty (Hedegaard et al., 2013; Langner et al., 2012a). The role of intercontinental transport of pollution is also somewhat overlooked in the literature apart from the sensitivity studies of Langner et al. (2012a) and the assessment of Szopa et al. (2006). It should also be noted that the vast majority of the regional air quality literature is devoted to ozone with very few studies focusing on particulate matter (Hedegaard et al., 2013; Manders et al., 2012).

We intend to quantify the penalty/benefit brought about by the externalities constituted by climate change and global pollution burden bearing upon ozone and particulate pollution over Europe. Four models are involved: a coupled ocean–atmosphere global circulation model (AOGCM), a global chemistry transport model (GCTM), a regional climate model (RCM) and a regional chemistry transport model (RCTM). Based on this approach, we can cover global and regional scales for both transport and chemistry.

For the global climate and chemistry modelling, we use the representative concentration pathway (RCP) scenarios (van Vuuren et al., 2011) developed for the CMIP5 (Taylor et al., 2012). While the RCPs include estimates of chemically active anthropogenic pollutants and precursors, the scenarios were designed solely to assess the long-term radiative forcing and they were developed with different integrated assessment models. Hence they do not provide consistent scenarios to analyse climate and air pollution policy interactions (see also Butler et al., 2012; and Fiore et al., 2012, for discussion). Given our goal of looking closer at regional air quality issues, for the regional chemistry-transport simulations we selected air pollution scenarios from the more recent Global Energy Assessment (GEA<sup>1</sup>) (Riahi et al., 2012). The GEA scenarios while being consistent with the RCPs – identical long-term radiative forcing levels – also include a detailed representation of air quality policies. To our knowledge, this study is the first to address future air quality over Europe under the hypotheses of the recent RCPs whereas there are a number of global or hemispheric assessments (Hedegaard et al., 2013; Shindell et al., 2013; Young et al., 2013).

This paper is structured as follows. Section 2 describes the chosen emission scenarios for greenhouse gases and chemically active pollutants. The models constituting the regional climate and air quality modelling system are presented in Sect. 3. The modelling results are discussed in Sect. 4.1 for the regional climate projection and Sects. 4.2 and 4.3 for the air quality projection. The sensitivity simulations designed to quantify the respective contribution of climate, intercontinental transport of pollution and regional air pollutant emission changes are discussed in Sect. 4.4.

<sup>1</sup>[http://www.iiasa.ac.at/Research/ENE/GEA/index\\_ga.html](http://www.iiasa.ac.at/Research/ENE/GEA/index_ga.html)

## 2 Emission scenarios

We use the RCP8.5 and RCP2.6 climate scenarios from the CMIP5 set that cover the highest and lowest ranges in terms of radiative forcing explored by the RCP scenarios. The corresponding emissions of short lived species are used in the global chemistry model that will be used to constrain the regional air quality model at its boundaries. The RCP2.6 is designed to keep global warming below 2 °C by the end of the century whereas RCP8.5 does not include any specific climate mitigation policy and thus leads to a high radiative forcing of 8.5 W m<sup>-2</sup> by 2100.

Regarding European air pollutant emissions in the regional air quality simulations, we focused on the two scenarios from the GEA set that include an identical representation of all current air quality legislation in Europe but differ in terms of policies on climate change and energy access. The reference scenario (also called CLE1 in Riahi et al., 2012) assumes no specific climate policy and has a climate response almost identical to the RCP8.5, while the mitigation scenario (CLE2) includes climate policies leading to a stabilisation of global warming (hence resembling the RCP2.6). These scenarios are based on modelling with MESSAGE (Model for Energy Supply Strategy Alternatives and their General Environmental Impact) for the energy system (Messner and Strubegger, 1995; Riahi et al., 2007). MESSAGE distinguishes 11 world regions, including Western Europe and Central & Eastern Europe<sup>2</sup>. The emissions (CH<sub>4</sub>, SO<sub>2</sub>, NO<sub>x</sub> (nitrogen oxides), CO, NMVOC (non-methane volatile organic compounds), black (BC) and organic carbon, PPM (fine primary particulate matter)) are subsequently spatialised on a 0.5° geographical grid using ACCMIP emission data for the year 2000 (Lamarque et al., 2010). Further details of the GEA air pollution modelling framework are available in Rao et al. (2012).

The main strength of the GEA scenarios lies in the use of an explicit representation of all currently legislated air quality policies until 2030 based on detailed information from the GAINS model (Amann et al., 2011). For OECD countries in particular, this includes a wide variety of pollution measures including directives on the sulphur content in liquid fuels, emission controls for vehicles and off-road sources up to the EURO-IV/ EURO-V standards: emission standards for new combustion plants and emission ceilings as well as the revised MARPOL VI legislations for international shipping. The inclusion of detailed AQ policies in the 2005–2030 period has a significant impact on the emissions of pollutants in the GEA scenarios so that, compared to the RCPs, larger co-benefits of climate policies for air pollution can be expected (Colette et al., 2012b). After 2030, the GEA scenarios implicitly assume, through decline in emission factors, contin-

**Table 1.** Total annual anthropogenic emissions (Gg yr<sup>-1</sup>) of NO<sub>x</sub> (in NO<sub>2</sub> equivalent), non-methane VOCs, sulphur dioxide (SO<sub>2</sub>), ammonia (NH<sub>3</sub>), carbon monoxide (CO) and black and organic carbon aggregated over Europe (15° W–40° E, 30–65° N) in the gridded GEA emission projections for 2005 (historical year), and 2050 under the reference (CLE1) and mitigation (CLE2) scenarios.

	GEA 2005	GEA CLE1/2050	GEA CLE2/2050
NO <sub>x</sub>	21 180	9849	4195
NMVOC	18 882	13 003	6115
SO <sub>2</sub>	19 872	4929	1689
NH <sub>3</sub>	7446	9978	9918
CO	63 865	20 019	10 520
BC	780	254	89
OC	1696	397	319

ued air quality legislation given a defined level of economic growth. Further details are provided in (Riahi et al., 2012).

The total emissions of the main anthropogenic pollutants or precursors thereof are given in Table 1. The reference or CLE1 scenario in absence of climate policy already shows a decline by 2050 of about 35–45 % (depending on the constituent) of the current level of emissions, emphasising the efficiency of current legislation with regards to air pollutant emissions in Europe. The decrease is even larger when climate policy is implemented as in the CLE2 scenario. NO<sub>x</sub> and VOC decrease to 14–22 % of current levels, indicating a 50 % co-benefit of climate policy for air quality. For particulate matter, given here as black and organic carbon, the decrease reaches almost a factor 10 in the case of BC in the mitigation scenario.

Using this combination of RCP and GEA scenarios offers the possibility to take into account explicit AQ policies that were not the scope of the RCPs. The only shortcoming of this option is in the inconsistency of chemically active emissions used in the global and regional models, given that the first prescribes boundary conditions of the second. A higher consistency could be achieved either by using GEA data to drive the global models or RCPs to drive the regional AQ model. The first option was ruled out because we preferred to use existing simulations of well established international model intercomparison projects. The second option would have led to ignoring the added value of the consistent representation of air quality policies in the GEA pathways (Fiore et al., 2012; Butler et al., 2012; Colette et al., 2012a, b).

## 3 Modelling framework

The present assessment builds upon a suite of models covering various compartments of the atmospheric system. A global coupled atmosphere–ocean general circulation model and a global chemistry transport model address projected climate change and its impact on global chemistry. The global

<sup>2</sup> <http://www.iiasa.ac.at/web/home/research/researchPrograms/Energy/MESSAGE-model-regions.en.html>

climate and chemistry fields are then downscaled with regional models. The individual tools of this modelling suite are briefly described here.

### 3.1 Global circulation model

The large-scale atmosphere–ocean global circulation model (AOGCM) is the IPSL-CM5A-MR (Institut Pierre Simon Laplace Coupled Model) (Dufresne et al., 2013; Marti et al., 2010). It includes the LMDz meteorological model (Hourdin et al., 2006), the ORCHIDEE land surface model (Krinner et al., 2005), the oceanic NEMO model (Madec et al., 1997) and the LIM sea-ice model (Fichefet and Morales-Maqueda, 1999). The external forcing in terms of anthropogenic radiative forcing is prescribed by the RCPs (Sect. 2). The medium-resolution version of the model is used ( $2.5^\circ \times 1.25^\circ$  in the horizontal and 39 vertical levels).

Switching the meteorological forcing from reanalyses to a climate model is a prerequisite to explore future projections. The difference between the two settings is that the climate model attempts to capture a climate that is representative of present conditions, whereas the reanalysis consists in the actual realisation of the past few years. Considering that significant impact on air quality projections have been reported before (Katragkou et al., 2011; Manders et al., 2012; Zanis et al., 2011; Menut et al., 2012), we decided to investigate here both a GCM-historical scenario (based on the climate model) and a ERA-hindcast (using the ERA-interim reanalyses; Dee et al., 2011).

### 3.2 Regional climate model

The Weather Research and Forecasting (Skamarock et al., 2008) (version 3.3.1) mesoscale model is used as a regional climate model (RCM) for the dynamical downscaling of the IPSL-CM5A-MR global fields. The spatial resolution is 50 km and the domain covers the whole of Europe with  $119 \times 116$  grid points. The set-up of the RCM is the same as that of Menut et al. (2012) which presents a detailed evaluation of the performances of the IPSL-CM5-LR/WRF regional climate modelling suite for air quality modelling purpose. However, we use here an updated, higher resolution version of the AOGCM (IPSL-CM5A-MR) which exhibits a smaller cold bias over the North Atlantic (Hourdin et al., 2012). A somewhat similar set-up is used for the IPSL-INERIS contribution (Vautard et al., 2012) to the forthcoming Coordinated Regional Climate Modelling Experiment (CORDEX; Giorgi et al., 2009). The main differences compared to Vautard et al. (2012) include using 11 yr time slices (of which the first year is used for spin-up and discarded in the following) instead of transient simulations, a slight spectral nudging in the upper layers of the atmosphere, and a lower resolution of 50 km.

### 3.3 Global chemistry-transport model

In order to provide boundary conditions to the regional chemistry transport model and assess the role of global atmospheric chemistry changes on regional air quality we use the LMDz-OR-INCA chemistry–climate model (Folberth et al., 2006; Hauglustaine et al., 2004). The model is run with a horizontal resolution of  $3.75^\circ$  in longitude and  $2.5^\circ$  in latitude and uses 19 vertical levels extending from the surface to 3 hPa. Further details on the model set-up and results can be found in (Szopa et al., 2012) who report a decrease of the tropospheric ozone burden by 2050 compared to 2000 according to the RCP2.6 scenario, while the RCP8.5 lead to an increase, in both cases the magnitude of the change is about 8%. As far as global aerosols are concerned, a decrease is expected by 2050 for all anthropogenic species, while dust and sea salt tend to increase in the future. These global simulations were also used for the ACCMIP experiment and the results of the LMDz-OR-INCA are compared with other global chemistry–climate models in (Shindell et al., 2013; Young et al., 2013).

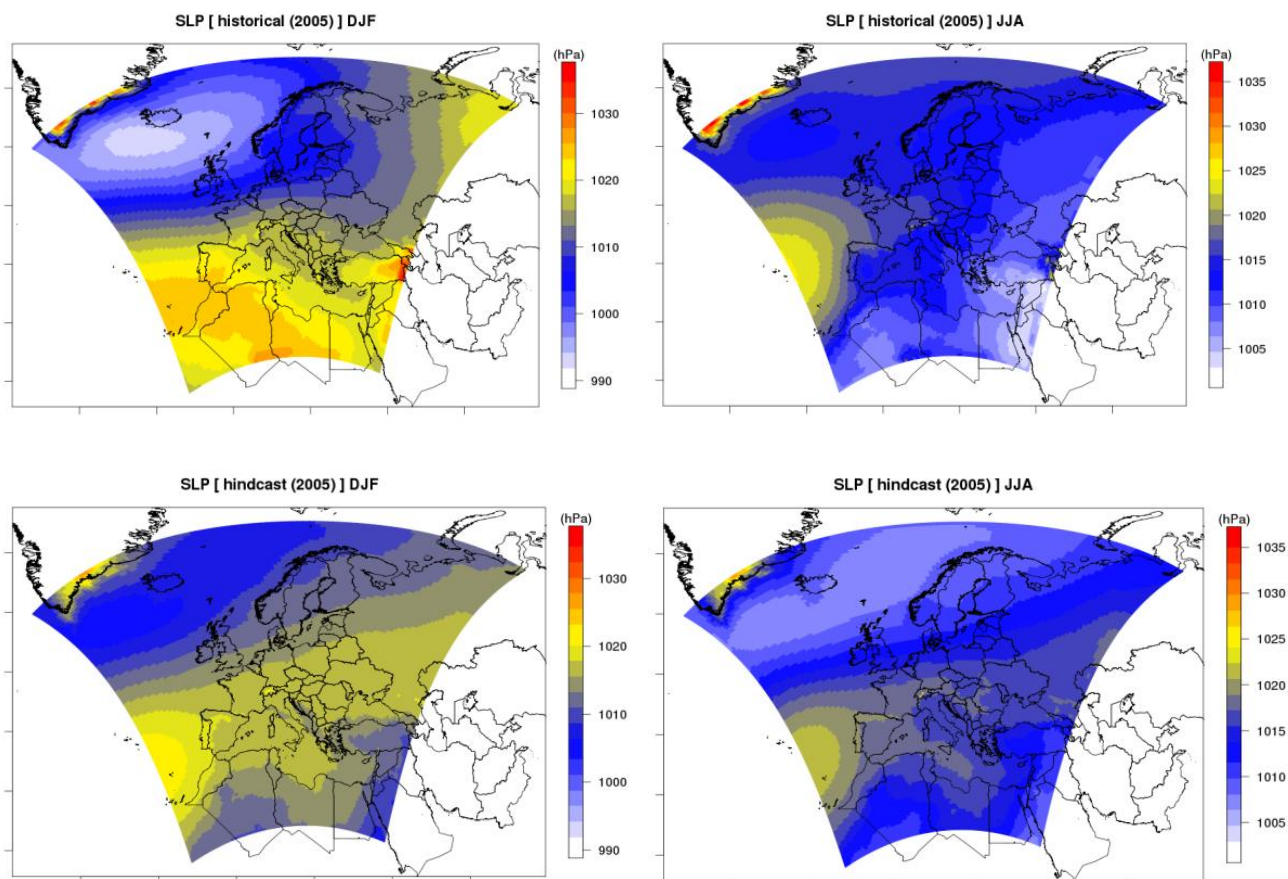
The three dimensional fields of 13 gases (including ozone, methane, carbon monoxide, PAN,  $\text{HNO}_3$ , etc.) as well as various particulate matter compounds (dust, sulphate, black and organic carbon) are then used as boundary conditions for the regional AQ model. Given that we focus on background changes, monthly mean fields averaged over a 10 yr period are used. It should be noted that these background changes combine the impact of distant air pollutant emissions and global climate change since they are based on climate–chemistry simulations.

### 3.4 Regional chemistry-transport model

The CHIMERE<sup>3</sup> model (Bessagnet et al., 2008b; Menut et al., 2013) is used to compute regional air quality. The model is used by a number of institutions in Europe and beyond for event analysis (Vautard et al., 2005), policy scenario studies for the French Ministry of Ecology, the European Commission and the European Environment Agency, operational forecasts (Honoré et al., 2008; Rouil et al., 2009; Zyryanov et al., 2012), model intercomparison exercises (van Loon et al., 2007; Vautard et al., 2007; Solazzo et al., 2012a, b), long-term hindcasts (Colette et al., 2011) and projections (Colette et al., 2012a; Meleux et al., 2007; Szopa et al., 2006).

In the present study, the model is used with 8 vertical levels extending from about 997 to 500 hPa and a horizontal resolution of  $0.5^\circ$ . The relatively coarse horizontal resolution compared to recent air quality model intercomparison initiatives is a trade-off to allow for the long-term simulations presented in Sects. 4.2 and 4.3 but the main computational constrain is carried by the numerous sensitivity experiments discussed in Sect. 4.4.

<sup>3</sup>[www.lmd.polytechnique.fr/chimere](http://www.lmd.polytechnique.fr/chimere)



**Fig. 1.** Mean sea level pressure in the regional climate model for winter (December–January–February, DJF, left) and summer (June–July–August, JJA, right) and for the two representations of current climate: GCM-historical (obtained with the climate simulation) and ERA-hindcast (obtained with the reanalysis). For each panel, the average is over 10 yr.

## 4 Results

The results of the regional air quality and climate simulations are discussed in this section. Four scenarios are investigated in Sects. 4.1 to 4.3 while additional sensitivity experiments will be introduced in Sect. 4.4. The present-day conditions are covered with both ERA-hindcast and GCM-historical simulations and two possible futures are investigated for 2050: one of them (reference) assuming no implementation of any specific climate policy, while the second (mitigation) aims at keeping global warming below 2 °C by the end of the century. The basic experiments that combine all factors (climate, intercontinental transport of pollution and emissions) are discussed in Sects. 4.1 to 4.3 while the sensitivity simulations are the focus of Sect. 4.4.

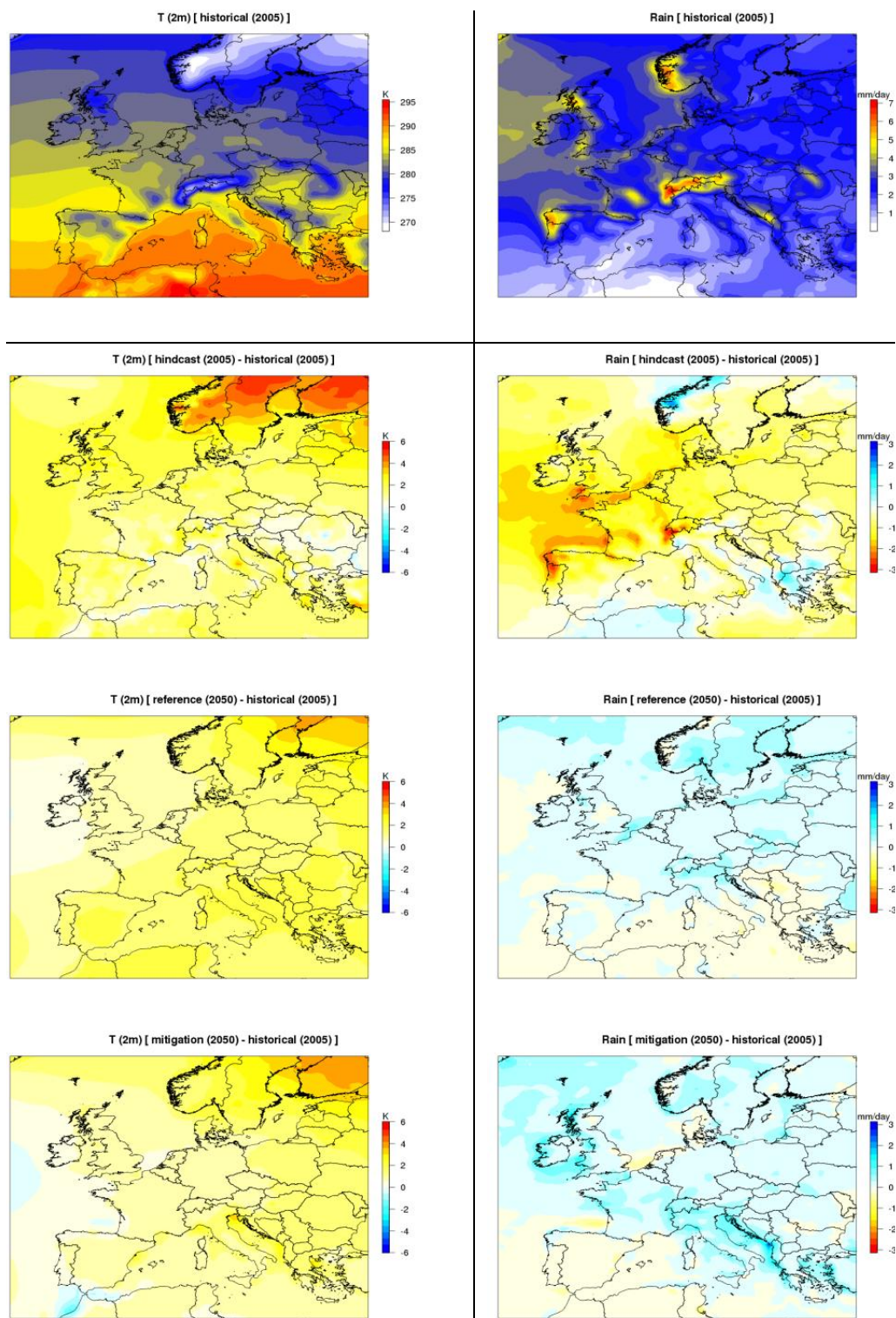
In each case, the simulation covers 10 yr in order to gain statistical significance and minimise the effect of interannual climate variability. This choice of a relatively short time period was made as a compromise between computational time resources and length required to obtain a detectable signal.

### 4.1 Regional climate projections

Even if the focus of the present paper is on impacts for air quality rather than regional climate itself, we describe in this section the main features of the regional climate fields since they will have a critical impact on the modelled air quality.

#### 4.1.1 General circulation

The 10 yr seasonal mean sea level pressure for winter and summer is provided in Fig. 1 for the present day conditions as depicted in the climate model (historical simulation) and the reanalysis (hindcast). In both seasons, important differences are found between the two representations of the climate of the early 21st century. In winter the GCM-historical simulations exhibit a much more zonal flow with a deeper Icelandic low pressure system yielding stronger average westerlies. In summer the differences are not that large. Nevertheless the Azores high pressure system is deeper yet smaller in size, so that average mean sea level pressure over Western Europe is lower than in the reanalysis.



**Fig. 2.** Left column panels: summertime (JJA) high 2 m daily mean temperatures (95th quantile, K) and right column panels: annual mean liquid precipitation ( $\text{mm day}^{-1}$ ). On the first row we display the absolute results of the GCM-historical climate simulation and on the following row the differences compared to the later for the ERA-hindcast, the 2050 reference and the 2050 mitigation projections.

### 4.1.2 Temperature and precipitation

The higher end of the temperature distribution also differs strongly when changing the large-scale forcing. As shown in Fig. 2, the GCM-historical climate model is much colder than the reanalysed ERA-hindcast. This statement is true for both annual mean temperature ( $-0.9$  K in winter and  $-1.5$  K in summer) and the 95th percentile of summer daily mean temperature (left panels in Fig. 2) with an average bias over Western Europe ( $5^{\circ}$  W– $15^{\circ}$  E,  $40$ – $55^{\circ}$  N; see area in Fig. 8) of  $-1.6$  K. This average bias is partly compensated by an opposite bias over sea surfaces. Over continental areas the temperature bias can reach values as high as 5 K. It is noteworthy to emphasise that the 95th percentile of temperature in the 2050 projections is respectively 1.9 K (for the RCP8.5) and 0.5 K (RCP2.6) warmer than the GCM-historical climate, showing that the absolute differences between current and future climate are actually smaller than temperature biases in the present climate.

Differences in daily precipitation are also shown (right panels in Fig. 2). Precipitation is a key variable for air quality because it drives wet scavenging which is an important sink for some trace species. The second row in Fig. 2 shows that the GCM-historical climate simulation is too wet compared to the reanalysis throughout Europe. For precipitation, the difference between the current and projected climate is also clearly lower than the differences of the two realisations of current climate.

### 4.1.3 Summary

Sections 4.1.1 to 4.1.3 have shown important differences between the reanalysed and climate simulations that will be used to drive the air quality modelling performed in the remainder of the study. We anticipate some of these features to have a detrimental impact on the simulation of air pollutant events.

It is not the purpose of this study to assess where such differences come from. Briefly, we can point towards (1) the global climate model, (2) the dynamical regional climate downscaling, or (3) the choice of the time period. The IPSL-CM5-MR model is known to exhibit a cold bias of sea surface temperature over the North Atlantic as a result of a strong underestimation of the Atlantic meridional overturning circulation (Hourdin et al., 2012). The dynamical downscaling has been demonstrated to contribute to an additional cooling (Colette et al., 2012c; Menut et al., 2012). These discrepancies could also be an artefact of the relatively short time period (10 yr) that could be influenced by an unfavourable climate mode (of the North Atlantic Oscillation for instance). The importance of using long time series has been repeatedly emphasized in climate studies and this factor should be taken into account in future air quality and climate assessments when the computing resources are sufficient (Langner et al., 2012a).

Before concluding this section devoted to the climate projection it is important to keep in mind that it is not because the climate model exhibits a bias that its projected changes are not valid. An over-fitted climate would perform ideally for the past, yet being very poor for future projections. That is why, in the vast majority of the climate science literature, model variability is investigated rather than absolute changes. For that respect, climate sensitivity is a more relevant metric than biases over a given period, and the climate sensitivity of the IPSL-CM5-LR model was found to fit in the middle of the ensemble of CMIP5 models (Vial et al., 2013). However, when it comes to climate impact modelling absolute differences and biases do matter, hence raising new challenges. Besides, in the field of climate research, confidence is achieved by making use of model ensembles, which raises a significant computational challenge for air quality projections (that would ideally be based on ensembles of each of the four types of models introduced in Sect. 3, hence multiplying the size of the ensemble). We will discuss in more detail throughout the remainder of the paper how such differences bear upon our confidence in air quality projections.

### 4.2 Evaluation of air quality simulations

Before discussing the modelled changes in air quality at the horizon 2050, we compare the air quality results for the present day to observations. We use ozone (daily max) and PM10 (daily mean) recorded at air quality monitoring stations (1108 for ozone and 688 for PM10) available for the 1998–2007 period in the AIRBASE public air quality database maintained by the European Environmental Agency (<http://air-climate.eionet.europa.eu/databases/AIRBASE/>). Synchronous scores (correlation as  $R^2$  and root mean square error, RMSE) are computed for the air quality simulations driven with the ERA-hindcast meteorological fields, whereas only average biases are provided for GCM-historical that relies on a different meteorology.

The results in Table 2 are in-line with previous implementation of CHIMERE (Solazzo et al., 2012a, b; van Loon et al., 2007) in particular when using this set-up (Colette et al., 2011, 2012a). A positive bias is found for ozone which is expected at such a coarse resolution where primary emissions are smeared out. This bias is partly compensated by a good correlation to achieve a satisfactory RMSE. The climate (GCM-historical) forcing tends to produce much less ozone, hence the small negative bias while a positive ozone bias would be expected at such a coarse resolution. For particulate matter a significant negative bias is found. This range of bias is a common feature in many operational air quality models (Solazzo et al., 2012a), in the present case the bias is even increased because we use only black and organic carbon emissions and ignore other anthropogenic primary particulate matter sources (even though we do account for natural and secondary PM, see Sect. 4.3.2).

**Table 2.** Comparison of the modelled air quality (ozone daily maximum and PM10 daily average) obtained with the reanalysed (ERA-hindcast) or climate (GCM-historical) forcing with respect to the European network of observations at background urban, suburban, and rural sites.

Pollutant	Station type	ERA-hindcast			GCM-historical
		Bias ( $\mu\text{g m}^{-3}$ )	RMSE ( $\mu\text{g m}^{-3}$ )	Corr.	Bias ( $\mu\text{g m}^{-3}$ )
$\text{O}_3^{\text{max}}$	Urban	9.4	1.9	0.75	-1.6
	Suburban	7.6	1.8	0.77	-3.7
	Rural	8.6	1.8	0.74	-2.6
PM <sub>10</sub>	Urban	-10.5	3.6	0.42	-11.5
	Suburban	-9.1	3.3	0.40	-10.3
	Rural	-9.3	3.4	0.41	-10.0

### 4.3 Projected changes in air quality concentration

Figures 3 and 4 display the GCM-historical, ERA-hindcast and projections for ozone and total PM<sub>2.5</sub> (including secondary aerosols) concentration fields. Absolute values are given for the GCM-historical simulation while differences relative to the GCM-historical simulation are provided for the remaining configurations. Such differences are only plotted where found to be statistically significant with a Student *t* test at the 95 % confidence level (the difference being set to zero where insignificant). A qualitative discussion is given in the present subsection. This discussion remains descriptive since the results introduced here combine the impacts of emission projections, climate change and intercontinental transport of pollution. The investigation will be further quantified in Sect. 4.4 regarding the attribution analysis.

#### 4.3.1 Ozone

Projected ozone changes are discussed on the basis of two different metrics in Fig. 3. The average summertime daily maxima (based on 8 h running means) of ozone ( $\text{O}_3^{\text{max}}$ ) is provided since it can be readily compared with the literature. However, in order to perform an exposure-based assessment we also focus on SOMO35, an indicator designed to capture detrimental impacts of ozone on human health and defined as the annual sum of daily maximum over 35 ppbv based on 8 h running means (expressed in  $\mu\text{g m}^{-3}$  day according to EEA, 2009).

The average situation for the GCM-historical (2005) simulation resembles the usual picture (e.g. Colette et al., 2011): a sharp latitudinal gradient with the exception of pollution hotspots over Europe. The difference between the GCM-historical and ERA-hindcast simulations provided on the second row confirms that the climate model is less favourable to ozone build-up than the actual meteorology over the recent past, as anticipated in Sect. 4.1.2.

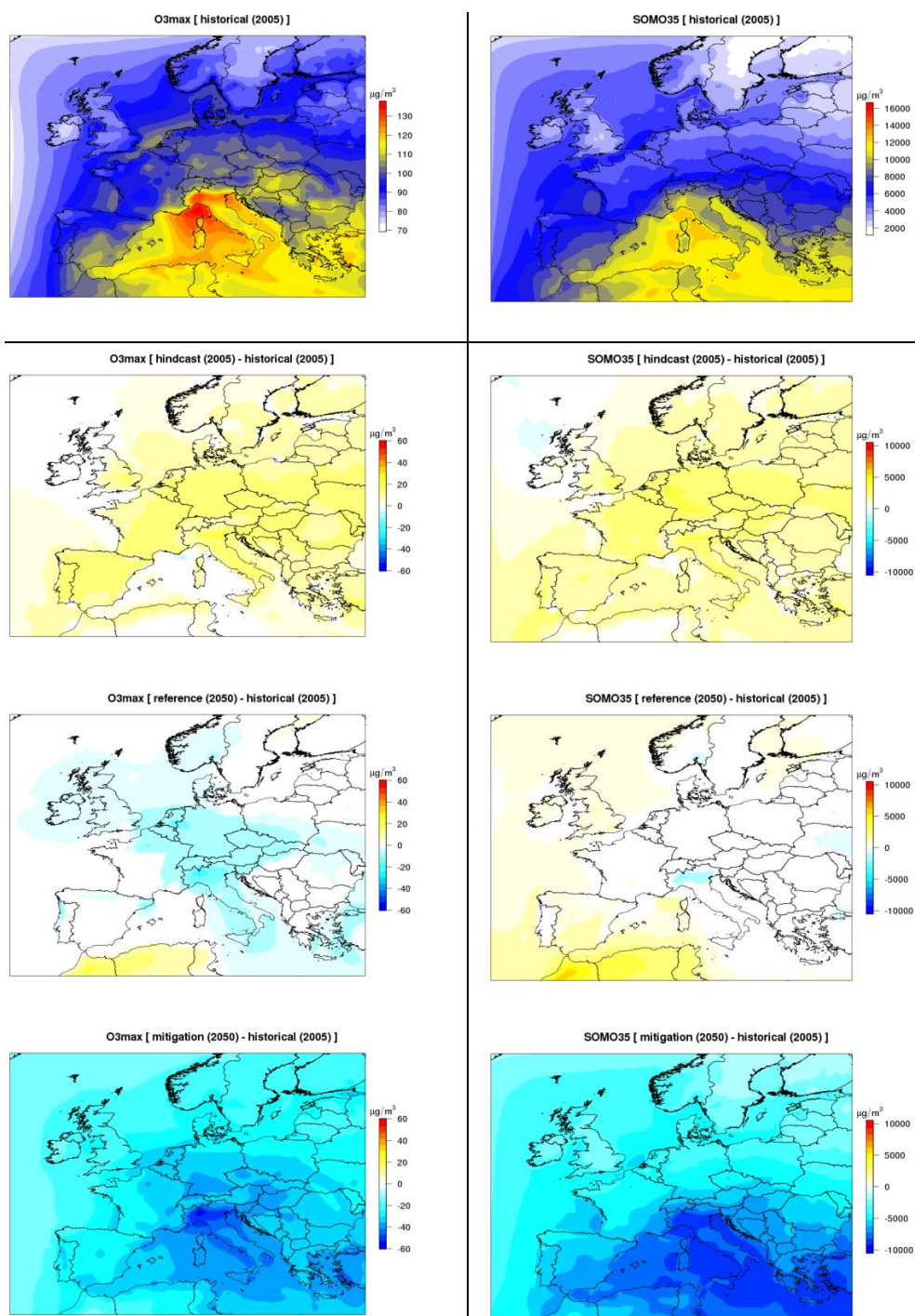
A closer look into the frequency of stagnation episodes rules out a potential responsibility of weather regimes in this underestimation. Manders et al. (2012) reported a decrease of calm spells using a climate model, but in our case the GCM-

historical simulation is actually more conducive to stagnant summertime episodes than the ERA-hindcast with a frequency of calm days (average wind speed below  $3.5 \text{ m s}^{-1}$ ) of 31 and 23 %, respectively, and a mean duration of calm spells of 2.4 and 2.2 days (see the similar findings of Menut et al., 2012; and Vautard et al., 2012).

The differences in summertime temperature and incoming short wave radiation can also lead to a modification of biogenic emissions of ozone precursors. In particular, we found that isoprene emissions were 66 % higher in the ERA-hindcast ( $8797 \text{ Gg yr}^{-1}$ ) than in the GCM-historical ( $5300 \text{ Gg yr}^{-1}$ ) simulation. By looking at the range of correction factors of the biogenic emission model (Curci, 2006; Guenther et al., 2006) for the corresponding range of temperature and incoming short wave radiation, we found that the sensitivity to the radiation was twice as large as the sensitivity to the temperature bias (see the supplementary information for more details on this issue), suggesting that minimising biases in cloud coverage in the climate model would help to reduce the ozone bias. The uncertainty on ozone projections brought about by this feature should be emphasized, especially given that future isoprene emissions reach 6086 and  $7091 \text{ Gg yr}^{-1}$  in the mitigation and reference scenarios, respectively. The projected change is thus smaller than the discrepancy for the current climate. Significant uncertainties related to isoprene projections were reported in the past in multi-model ozone projection experiments. Over Europe, Langner et al. (2012b) found a factor of 5 difference across the model ensemble, while at the global scale Stevenson et al. (2006) reported a factor 3. When compared to the ensemble of Langner et al. (2012b), it appears that CHIMERE is in the mid to upper part of their envelope that ranges from 1592 to  $8018 \text{ Gg yr}^{-1}$  for the present climate. Similarly to all the regional models of that ensemble we did not include the impact of isoprene emission reduction with increasing CO<sub>2</sub> levels (Lathière et al., 2010; Wilkinson et al., 2009).

Lastly, we can mention that temperature, incoming radiation and even specific humidity can also play a direct role in atmospheric chemistry, although these factors are much more difficult to isolate (Menut, 2003).





**Fig. 3.** Top row panels (from left to right): average fields of ozone as summertime average of the daily maxima ( $O_3^{\max}$ ,  $\mu\text{g m}^{-3}$ ), and SOMO35 ( $\mu\text{g m}^{-3}$  day) in the control (2005) simulation (averaged over 10 yr corresponding to the current climate). Following panel rows: difference between the simulations for the reanalysed ERA-hindcast and then for the reference and mitigation 2050 projections taken with respect to the GCM-historical climate simulation (2005). The differences are only displayed where significant given the interannual variability of 10 yr.

Both projections for 2050 indicate a decrease of daily maximum ozone compared to the GCM-historical climate simulation, but the magnitude of this decrease is moderate for the reference scenario. The situation is however more

complex under the reference scenario for the human health exposure to ozone, since the index SOMO35 actually increases over a significant part of Europe. The mitigation scenario achieves a much higher degree of emission reduction.

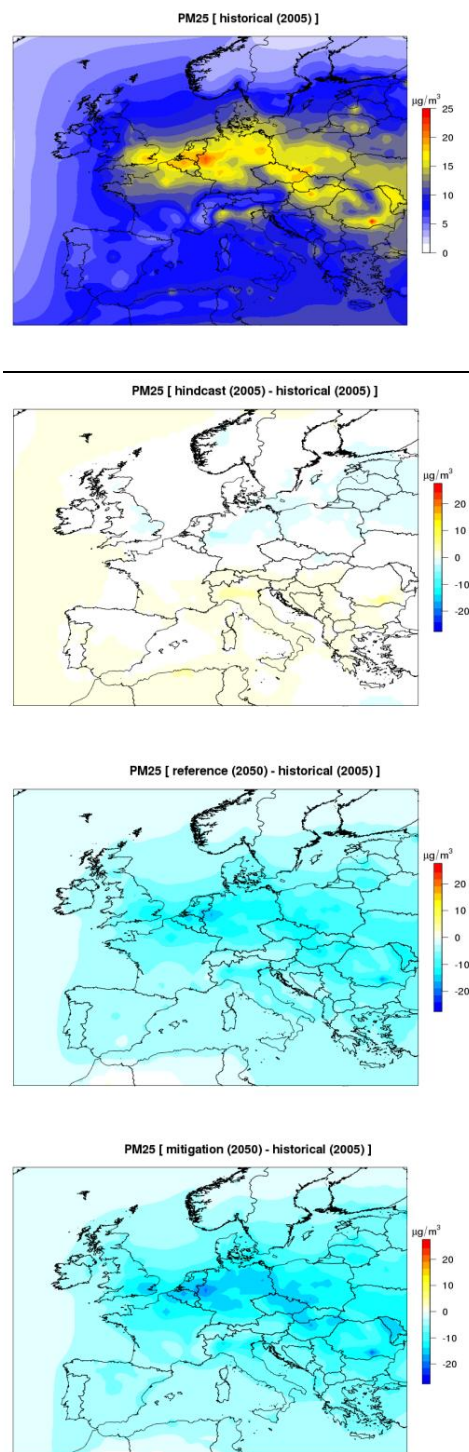
As a result, SOMO35 decreases sharply, especially in the Mediterranean area where the levels were highest. On a more quantitative basis, in order to emphasise the projected changes in high-exposure areas, we apply a weighting function to the SOMO35 fields depending on the population density (obtained from Riahi et al., 2012; and United Nations, 2009, but using the population for 2005 for both the current and prospective scenarios). We find that the population-weighted SOMO35 increases by 7.4 % (standard deviation  $\pm 5.4$  %) in the reference scenario whereas it decreases by 80.4 % ( $\pm 2.1$  %) in the mitigation case.

#### 4.3.2 Particulate matter

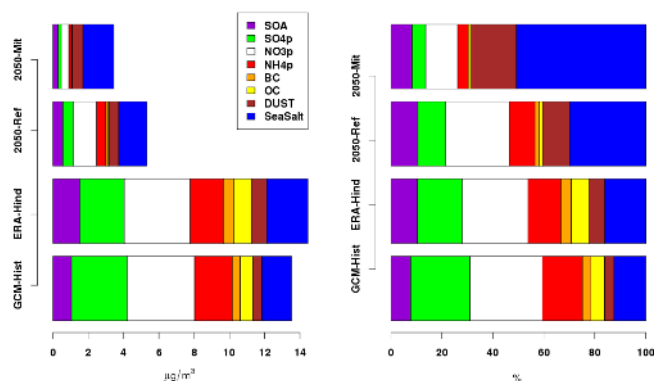
In addition to the primary particulate matter prescribed in the anthropogenic emissions (elemental carbon – EC – and organic carbon – OC) and derived in the natural emissions (dust and sea salt), CHIMERE accounts for the formation of secondary aerosols that undergo a range of microphysical transformations including nucleation, coagulation, and absorption. For inorganic species such as nitrate ( $\text{NO}_3$ ), sulphate ( $\text{SO}_4$ ) and ammonium ( $\text{NH}_4$ ) the thermodynamic equilibrium is diagnosed using the ISORROPIA model (Nenes et al., 1998). For semi-volatile organic species, a partition coefficient is used (Pankow, 1994). Chemical formation of secondary organic aerosols (SOA) is represented with a single step oxidation of the relevant precursors and gas-particle partitioning of the condensable oxidation products (Bessagnet et al., 2008a). Most SOA are issued from the oxidation of monoterpenes computed with the MEGAN model (Guenther et al., 2006). The processes related to aerosol dynamics are described in Bessagnet et al. (2004).

The average fields of fine particles ( $\text{PM}_{2.5}$ ) in Fig. 4 for the GCM-historical (2005) simulation display local maxima over the main air pollution hotspots besides the large influx at the southern boundary of the domain (desert dust). The bias towards too high precipitation (Fig. 2) in the GCM-historical climate simulation has a limited impact on the average load of  $\text{PM}_{2.5}$  over Western Europe, being 12.1 and 12.1  $\mu\text{g m}^{-3}$  in the GCM-historical and reanalysis simulations, respectively. The decrease by 2050 is very large, with  $\text{PM}_{2.5}$  concentrations dropping down to 4.1 and 2.3  $\mu\text{g m}^{-3}$  over areas of Western Europe in the reference and mitigation scenarios, respectively.

A closer look in the average individual aerosol components over Western Europe (as defined in Sect. 4.1.2 and masked to keep only land surface areas and avoid an over-weighting of sea salts) is provided in Fig. 5. Note that individual PM components sum up to  $\text{PM}_{10}$ , instead of the  $\text{PM}_{2.5}$  that are used elsewhere in the paper because of their higher relevance for air quality purposes. Sulphate concentrations are higher in the GCM-historical simulations compared to the ERA-hindcast because of enhanced sulphur aqueous chemistry. On the contrary, nitrate concentrations are similar because of the high availability of free ammonia



**Fig. 4.** Top panel: average fields of annual mean  $\text{PM}_{2.5}$  ( $\mu\text{g m}^{-3}$ ) in the control (2005) simulation (averaged over 10 yr corresponding to the current climate). Following panels: difference between the simulations for the reanalysed ERA-hindcast and then for the reference and mitigation 2050 projections taken with respect to the GCM-historical climate simulation (2005). The differences are only displayed where significant given the interannual variability of 10 yr.



**Fig. 5.** Average aerosol composition over land surfaces of Western Europe for the two simulations corresponding to the present-day conditions (GCM-historical and ERA-hindcast) as well as the two scenarios for 2050 (reference and mitigation). Absolute concentrations are given on the left panel and relative contributions to total  $\text{PM}_{10}$  (expressed in percentages) are given on the right panel.

(defined as the total ammonia minus sulphate, in moles) in the atmosphere.

All the secondary aerosols decrease in the future as a result of decreasing anthropogenic emission of precursors. The most prominent feature in the projection of aerosol composition is the increase of the relative importance of natural aerosols such as dust and sea salts in the future (right panel in Fig. 5), so that in the most stringent scenario the fraction of crustal material becomes dominant in  $\text{PM}_{10}$ . Secondary organic aerosols are the only species that maintain their relative importance due to the contribution of biogenic precursors in their formation process. As far as secondary inorganic aerosols are concerned it is worth mentioning that the small increase of  $\text{NH}_3$  emissions in the GEA projections – Table 1 and Colette et al. (2012b) – is not reflected in the projected formation of particulate  $\text{NH}_4^+$ . Whereas  $\text{NH}_3$  emissions increase by 22 and 21 % for the reference and mitigation scenarios, respectively. Between 2005 and 2050, we find that  $\text{NH}_4^+$  decreases from  $4.0 \mu\text{g m}^{-3}$  in the GCM-historical 2005 simulation to 1.4 and  $0.5 \mu\text{g m}^{-3}$  in the reference and mitigation projections, respectively. This feature emphasises the probable limiting role of  $\text{NO}_x$  emissions through the availability of  $\text{HNO}_3$  in rural areas (Hamaoui-Laguel et al., 2012) that do exhibit a strong decrease in the future. The reason why such behaviour is not reported in coarse global chemistry transport model projections deserves further investigation (Fiore et al., 2012; Shindell et al., 2013). While in the historical simulations nitrate represents about 33 % of non-natural particles, this fraction reaches 39–42 % in 2050.

In terms of exposure, we find that population-weighted  $\text{PM}_{2.5}$  decreases by 61.8 ( $\pm 3.1$ ) and 78.0 % ( $\pm 1.8$ ) in the reference and mitigation scenarios, respectively. It appears that air quality legislation (which is identical in both scenarios) somewhat dominates the relative change in exposure

to  $\text{PM}_{2.5}$ , the impact of the climate policy (which differs in both scenarios) is not as large as observed for the exposure to ozone.

#### 4.4 Disentangling the driving factors

The projected exposure to air pollution discussed in Sect. 4.3 takes into account the whole range of processes playing a role in the future evolution of air quality: global and regional climate, chemical background changes as well as air pollution mitigation measures. This modelling system also offers the opportunity to isolate the contribution of each driving factor to the overall projected change in the basic simulations discussed in Sect. 4.3.

##### 4.4.1 Methodology

We quantify the respective role of each process from sensitivity experiments consisting in replicating the decadal simulations with all things kept equal except one of the driving factors.

The list of simulation experiments is synthesised in Table 3 which includes both the reference and mitigation projections. Only present and future time periods are given in the table, and the future time period refers to either the reference (CLE1 for AP emissions, RCP8.5 for climate and boundary conditions) or the mitigation (CLE2 for AP emissions, RCP2.6 for climate and boundary conditions). Each row of the table refers to the isolation of one driving process, which is achieved by comparing the experiment described in the middle columns to the experiment described in the right columns.

The overall impact of climate, boundary conditions and AP emissions is obtained from the basic simulations (first row from the bottom). The second row from the bottom provides the specification of the simulation used to isolate the impact of switching from reanalyses to a climate model for the present conditions. In order to investigate the regional effects of climate change, the first step consists in using constant present-day AP emission and boundary conditions and then changing the climate forcing (third row from the bottom). However, in order to explore the climate response under gradually changing AP emissions – yet avoiding performing transient simulation – the sensitivity to regional climate change is also tested with future AP emissions (fourth row from the bottom). Symmetrically, the impact of emissions under constant climate and boundary conditions is explored for the present climate and the future climate. Last, the impact of boundary conditions is derived from the experiment on the 7th row from the bottom. We decided to ignore the impact of gradually changing climate and emissions on the role of the boundary conditions, in order to avoid increasing the dimension of the sensitivity matrix to be explored.

Some of the simulations are used several times in the process, so that only the unique sensitivity experiments are bold.

Five unique sensitivity decadal simulations are required for each of the two (reference and mitigation) scenarios, plus two baselines for the present day (ERA-hindcast and GCM-historical) that are common to both scenarios. In total, we have thus twelve decadal regional air quality simulations.

The results are provided in Figs. 6 and 7 as box plots referring to the key provided in Table 3.

Each sensitivity simulation is decadal. Instead of giving the difference of the temporal averages (such as on the maps in the previous figures), we first aggregate spatially by taking the mean over Western Europe ( $5^{\circ}$  W– $15^{\circ}$  E,  $40$ – $55^{\circ}$  N) and then we compute the distribution of differences between the two sets of decadal simulations, which are 55 independent combinations of individual years. These distributions of 55 differences are presented here as box and whisker plots where the boxes provide the three inner quartiles and the whiskers provide the extremes of the distribution. A cross is given where the distribution of difference is statistically significantly different from zero (Student's  $t$  test with a 95 % confidence interval).

Multi-annual sensitivity experiments are common practice in climate studies, but annual simulations are often used in atmospheric chemistry studies to investigate the impact of emission changes or boundary conditions. However, the spread of the distributions obtained here demonstrate the need to use multi-annual sensitivity simulations in order to provide a quantitative perspective of the uncertainty whereas the qualitative conclusions would be unchanged.

The differences between the sensitivity experiments performed under present-day or future conditions (the two shades of orange and blue) also emphasise the need to explore the impact of regional climate change and AP emissions under gradually changing conditions. Whereas the relevance of transient approaches are often pointed out in base-case projections (Langner et al., 2012a), it is not common practice in sensitivity experiments addressing the disentangling of various contributions (Hedegaard et al., 2013; Manders et al., 2012), while Figs. 6 and 7 show that the evolution of the response to climate or emission changes differs under present day and future conditions.

#### 4.4.2 Results

##### Ozone

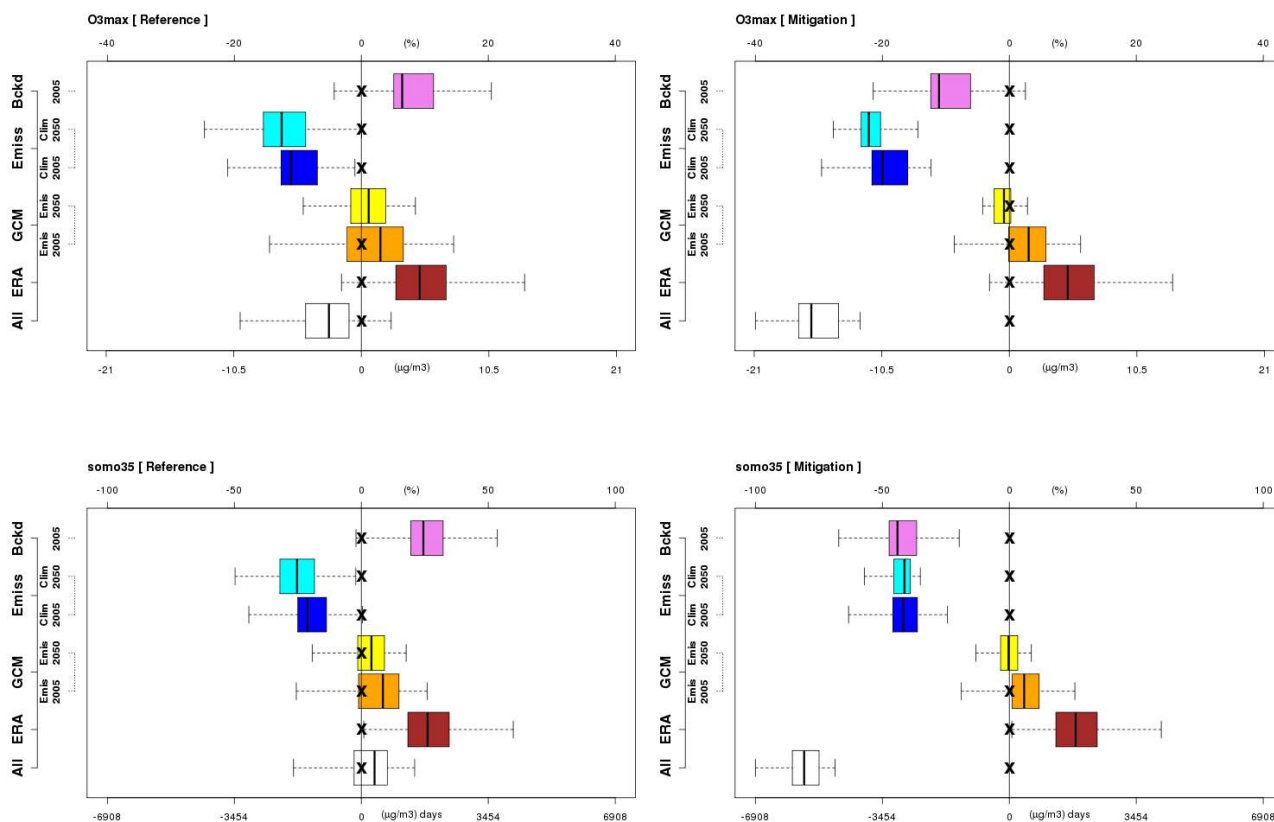
Ozone is presented here (Fig. 6), as in Sect. 4.3, as the average summertime of daily maximum based on 8 h running means as well as SOMO35 and we find again that the overall projection (including all factors: white boxes) consists in a decrease by 2050 for both scenarios for  $O_3^{\max}$  and for the mitigation scenario for SOMO35, while a small increase is found under the reference scenario for SOMO35. In all cases the changes are statistically significant.

We find that the role of AP emissions change (boxes in blue shadings) dominates over the impact of regional climate

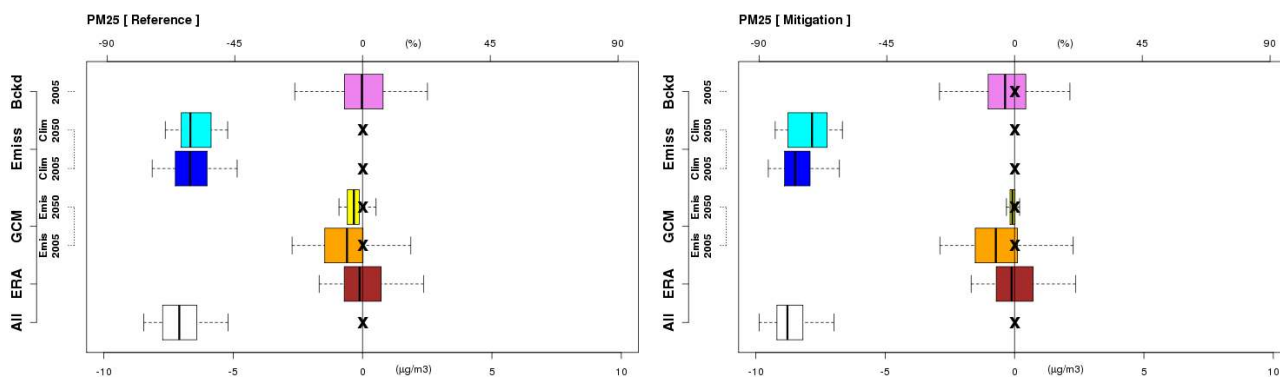
change (boxes in orange shadings). For SOMO35, the relative change attributed to climate ranges from 3 ( $\pm 8$  %) to 5 % ( $\pm 11$  %), while the response when changing emissions ranges from  $-24$  ( $\pm 10$  %) to  $-43$  % ( $\pm 7$  %). Recent studies on ozone projections relying on air pollutant emissions prescribed by the RCPs also reported that anthropogenic emission changes dominate over the effect of climate (Fiore et al., 2012; Hedegaard et al., 2013; Katragkou et al., 2011; Langner et al., 2012a, b; Lei et al., 2012; Manders et al., 2012). The fact that we use air pollutant emission projections based on explicit mitigation policies adds robustness to this finding.

Regional climate is found to constitute a significant penalty on ozone under present-day emissions (dark orange box) according to all metrics and scenarios. However, the response is not that large: below about  $1 \mu\text{g m}^{-3}$  for  $O_3^{\max}$ . This moderate impact is not surprising compared to the figures reported elsewhere where differences rarely exceed a few  $\mu\text{g m}^{-3}$  (Andersson and Engardt, 2010; Katragkou et al., 2011; Langner et al., 2012a, b), only Manders et al. (2012) found increases that could reach  $5$ – $10 \mu\text{g m}^{-3}$ . A more innovative finding lies in the assessment of the climate penalty under future AP emission (light orange box). We find that the penalty will decrease in magnitude and even become a net benefit for  $O_3^{\max}$  under the mitigation scenario. The fact that projected climate change can contribute to decrease ozone levels on average over Western Europe was never reported before and highlights the need to account for the AQ policies when addressing the climate penalty. Whereas there are examples in the literature of assessments including combined climate and emission changes, sensitivity attribution studies are systematically performed under present-day conditions. We show here that the future context must be accounted for, even in the sensitivity analysis.

Ozone projections over Western Europe are actually more sensitive to background concentrations changes than to the penalty/benefit brought about by regional climate change. The tropospheric background ozone change constitutes a penalty under the reference scenario and a benefit under the mitigation case. These opposite trends stem from the joint evolution of global emissions and global climate and were also reported in the global chemistry–climate projections for these scenarios (Szopa et al., 2012; Young et al., 2013). It is worth emphasising that in the global CTM, chemistry and climate are addressed jointly, it is therefore not possible to isolate to what extent these opposite trends are a result of AP emission changes in distant areas or a result of global climate on chemistry. In the mitigation scenario the decreasing background ozone burden contributes to increasing the benefit already obtained thanks to the reduction of AP emissions. But in the reference scenario, the compensation between a lower magnitude of AP emission changes and a penalty brought about by the increasing ozone background yields the penalty seen for SOMO35 in the net response.



**Fig. 6.** Contribution of the background air pollution (Bckd, violet), regional emissions (Emiss., blue shadings), and climate change (GCM, orange shadings) to the total projected changes (white) in  $O_3$  concentration as summer average of the daily max ( $O_3^{\max}$ ), and SOMO35 averaged over Western Europe in 2050 according to the reference (left) and mitigation (right) scenarios. In each case, we display net differences compared to a control selected to isolate one of the factors. The sensitivity to the meteorological driver (either GCM-historical climate or ERA-hindcast) is also given (ERA, brown). The sensitivity to emission and climate is investigated for both present and future conditions, hence the duplicate blue and orange boxes. A cross is marked when the distribution is significantly different from zero. The bottom axis provides the absolute difference and the relative difference with regards to the GCM-historical simulation is given as percentages on the top axis.



**Fig. 7.** Contribution of the background air pollution (Bckd, violet), regional emissions (Emiss., blue shadings), and climate change (GCM, orange shadings) to the total projected changes (white) in annual mean  $PM_{2.5}$  over Western Europe in 2050 according to the reference (left) and mitigation (right) scenarios. In each case, we display net differences compared to a control selected to isolate one of the factors. The sensitivity to the meteorological driver (either GCM-historical climate or ERA-hindcast) is also given (ERA, brown). The sensitivity to emission and climate is investigated for both present and future conditions, hence the duplicate blue and orange boxes. A cross is marked when the distribution is significantly different from zero. The bottom axis provides the absolute difference and the relative difference with regards to the GCM-historical simulation is given as percentages on the top axis.

**Table 3.** Synthesis of the model sensitivity experiments designed to isolate the role of the factors given on the first column. The simulations with the characteristic given in the middle columns are subtracted from those given in the right columns. The climate driver is the downscaled GCM unless otherwise stated as ERA (for downscaled ERA-interim). The set of unique simulations are bold. The results are provided in Figs. 6 and 7 as box plots.

	Emissions	Climate	Inter-continental transport	minus	Emissions	Climate	Inter-continental transport
Intercontinental transport	<b>2005</b>	<b>1995–2004</b>	<b>2045–2054</b>	–	2005	1995–2004	1996–2005
Emissions (2050 climate)	2050	2045–2054	1996–2005	–	2005	2045–2054	1996–2005
Emissions (2005 climate)	2050	1995–2004	1996–2005	–	2005	1995–2004	1996–2005
Climate (2050 emissions)	<b>2050</b>	<b>2045–2054</b>	<b>1996–2005</b>	–	<b>2050</b>	<b>1995–2004</b>	<b>1996–2005</b>
Climate (2005 emissions)	<b>2005</b>	<b>2045–2054</b>	<b>1996–2005</b>	–	2005	1995–2004	1996–2005
ERA-hindcast vs. GCM-historical	<b>2005</b>	<b>1998–2007 (ERA)</b>	<b>1996–2005</b>	–	2005	1995–2004	1996–2005
All	<b>2050</b>	<b>2045–2054</b>	<b>2045–2054</b>	–	<b>2005</b>	<b>1995–2004</b>	<b>1996–2005</b>

As mentioned in the introduction, regional climate change acts on ozone through several pathways: (1) it favours the emission of important precursors such as biogenic volatile organic compounds, (2) it has an impact on both chemical kinetics (through temperature and water vapour availability) as well as photochemical processes (through the incoming short wave radiation) and (3) it drives the frequency of weather patterns favourable for the build-up of pollutants and the turbulent mixing in the planetary boundary layer.

Katragkou et al. (2010) demonstrated, on the basis of sensitivity simulations, that biogenic emission, temperature and radiation have a comparable contribution to simulated changes. With regards to biogenic emissions, isoprene is a major factor (Meleux et al., 2007). Here we found that the isoprene emission increases by 2050 are of 15 and 34 % for the mitigation and reference scenario, respectively. These numbers are moderate compared to existing estimates of end-of-the-century changes reaching 100 % (Andersson and Engardt, 2010; Katragkou et al., 2011; Meleux et al., 2007) but they are in line with the 20 to 25 % increase in 2050 compared to 2000 for the RCP4.5 in Langner et al. (2012b). The sensitivity studies of Andersson and Engardt (2010) and Katragkou et al. (2011) report an increased production of O<sub>3</sub> of 1–2 µg m<sup>-3</sup> for a 30 % increase of biogenic emissions. While we cannot give comparable estimates without performing a dedicated sensitivity study, we can infer from existing studies that biogenic emissions constitute probably a significant fraction of the O<sub>3</sub><sup>max</sup> change attributed to climate change.

A last important feature in Fig. 6 regards the role of the meteorological driver. We pointed out in Sect. 4.3 that switching from a reanalysis to a climate simulation had a strong impact on modelled air quality. The brown boxes in Fig. 6 give a more quantitative view of this sensitivity where we find that SOMO35 is 28 % (±12) higher when using reanalyses compared to a GCM-historical regional climate model. Such behaviour was not unexpected: similar findings were mentioned in the few studies that proceeded to a such

a comparison (Katragkou et al., 2011; Manders et al., 2012). Nevertheless this bias raises serious concern on the uncertainty of such assessments.

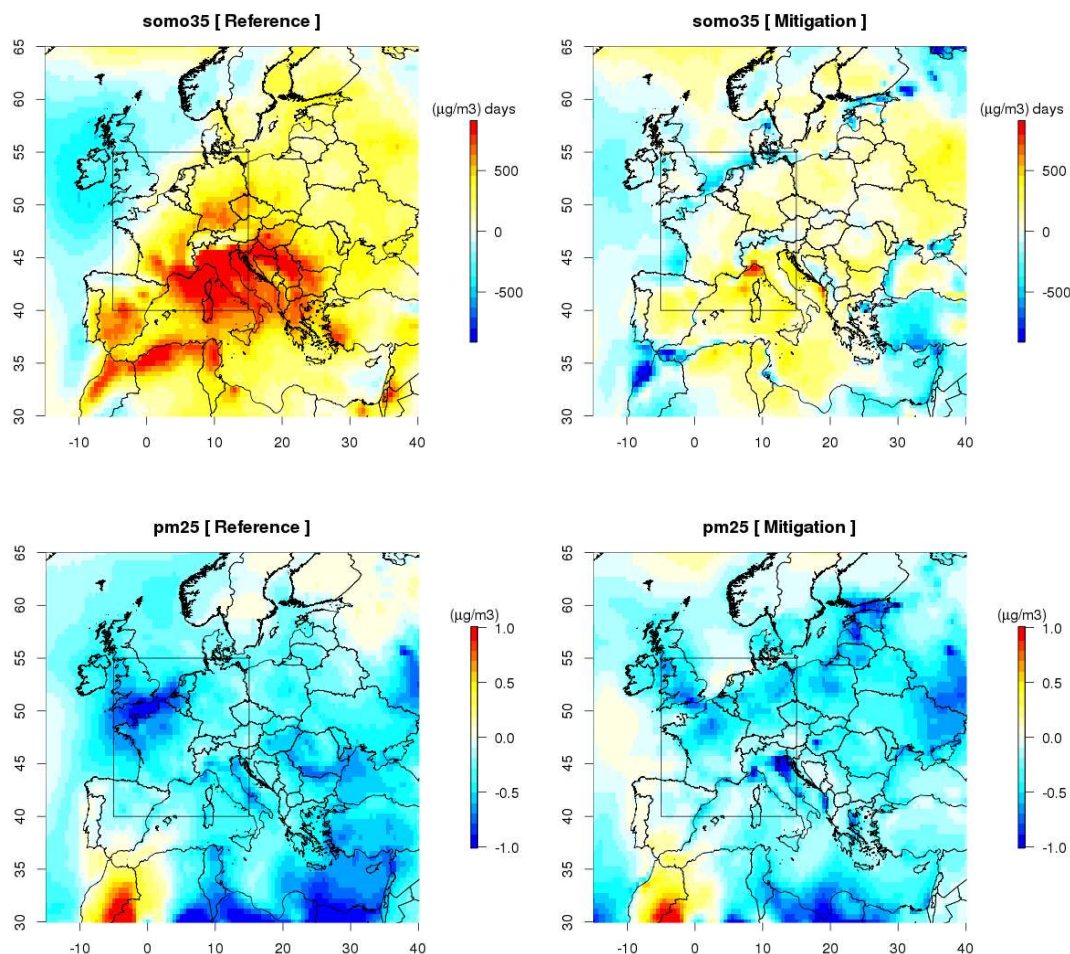
### Particulate matter

The same framework is applied to disentangle the different driving factors in the projections of particulate matter (Fig. 7).

Again, the contribution of AP emissions is found to largely dominate over the regional climate signal, in agreement with Hedegaard et al. (2013). This feature is even more pronounced than for ozone and the net decrease for PM<sub>2.5</sub> attributed to AP emission reaches –60 (±7 %) and –75 % (±7 %) for the reference and mitigation scenarios, respectively, out of a net change (all factors considered) of –65 (±8 %) and –79 % (±7 %). The contribution of intercontinental transport of pollution for PM<sub>2.5</sub> is small because of their shorter lifetime but not negligible for the mitigation scenarios.

With the present set of climate forcing, we find that regional climate change constitutes a slight benefit for PM<sub>2.5</sub> concentrations. The increase in precipitation in the future (Fig. 2) certainly contributes to this trend. But the magnitude of this benefit will decrease gradually in the future.

This climate benefit for PM<sub>2.5</sub> is contradictory compared to the penalty reported in previous studies (Hedegaard et al., 2013; Manders et al., 2012; Nyiri et al., 2010) The lack of robustness regarding the impact of climate on PM<sub>2.5</sub> was pointed out for the US by Tai et al. (2012) and Jacob and Winner (2009). The spread of precipitation projections in regional climate models (Christensen and Christensen, 2007) constitutes a major challenge in narrowing the uncertainty of the impact of climate on particulate matter.



**Fig. 8.** Map of the contribution of the regional climate to the projected change in air quality (from top to bottom: SOMO35, and  $PM_{2.5}$ ) for the reference (left) and mitigation (right) scenarios. A positive sign (red) indicates a climate penalty (increased air pollutant concentrations), whereas a negative sign (blue) shows that future climate tends to reduce detrimental air pollution levels.

The attribution analysis for individual particulate components (not shown) is in line with the main findings for  $PM_{2.5}$ : a domination of AP emissions, moderate contribution of boundary conditions and benefit of climate change. The only feature that is worth mentioning is a stronger impact of climate for nitrate and ammonia: from  $-5$  ( $\pm 11\%$ ) to  $-9$  ( $\pm 15\%$ ) depending on the scenarios, as compared to sulphates: from  $-2$  ( $\pm 8\%$ ) to  $-2$  ( $\pm 9\%$ ). This sensitivity highlights that beyond precipitation and the related washout (that do not discriminate particulate components), other climate factors (temperature, relative humidity) play a role in the formation of secondary aerosol species. If a significant sensitivity to climate was expected for sulphate (through water vapour changes and subsequent availability of the OH radical) (Hedegaard et al., 2008), our results show that ammonium nitrate is also affected by climate change (through the temperature dependence of its formation process) (Bessagnet et al., 2004).

#### 4.4.3 Mapping the indicator of the climate penalty/benefit

The isolated contribution of regional climate change to the net projected air pollution change is given for both scenarios as maps in Fig. 8. These maps are a composite of the differences obtained when comparing the decadal experiments under future and current climate with future and current emissions, i.e. an average of the orange boxes in Figs. 6 and 7. The indicator is coloured in red (blue) to indicate the penalty (benefit) brought about by climate change when it leads to increasing (decreasing) air pollutant concentrations.

In Fig. 8, we only discuss SOMO35 since its response is very similar to  $O_3^{\max}$ . A penalty dominates over continental Europe with the largest increases found over southern Europe, while more modest increases are found over northern Europe, even leading to decreases over the British Isles, corroborating the findings of Langner et al. (2012b) and Andersson and Engardt (2010). The largest changes are not

systematically located over areas exhibiting high ozone levels such as the Mediterranean, emphasising the role of biogenic precursors (Meleux et al., 2007).

For PM<sub>2.5</sub>, regional climate change constitutes mostly a benefit by decreasing the concentrations. Over Morocco a penalty is found, which can be related to the decrease of precipitation in this area (Fig. 2). The penalty over the North Atlantic under the mitigation scenario is likely attributed to sea salts since significant increases of surface wind speed are found in this area in winter and spring under the RCP2.6.

## 5 Conclusion

We presented an analysis of combined projections of air quality and climate impact at the regional scale over Europe under the latest CMIP5 climate scenarios produced for the Fifth Assessment Report of IPCC. The regional modelling system includes global and regional climate as well as global and regional chemistry. The global fields are those delivered in the context of well established international exercises (CMIP5 for the climate and ACCMIP for the chemistry). Emissions of trace species follow the recent representative concentration pathways for the global models while an update is used over Europe by using the scenarios developed for the Global Energy Assessment.

The use of recent emissions and consistent suite of models offers the opportunity to confront our findings with the literature and identify robust features in the overall projections of air quality and possible penalty and benefits brought about by climate change. The present set-up also allows performing sensitivity simulations in order to disentangle the respective contribution of climate, air quality mitigation and background changes.

An important approximation in the design of the modelling chain is that we used an offline regional air quality model, hence neglecting feedbacks of chemistry onto climate at the regional scale. The whole issue of short lived climate forcers is thus excluded from the present assessment (Löndahl et al., 2010; UNEP, 2011; Grell and Baklanov, 2011).

The first prerequisite when using an air quality model to investigate the impact of climate consists in switching the meteorological driver from reanalyses or forecast to a climate model. This step has significant consequences on the impact model. The climate fields that we used in the present study suffer from a cold and wet bias, as a result of a flaw in the North Atlantic oceanic circulation. When using climate fields instead of reanalyses, daily mean summertime ozone decreases from 90 to 84  $\mu\text{g m}^{-3}$ . Similar biases were reported before (Katragkou et al., 2011; Manders et al., 2012), but this feature is of course sensitive to the climate model selected and others had more satisfactory results (Hedegaard et al., 2008).

Using an ensemble of climate models (such as the forthcoming CORDEX ensemble of regional projections) would allow minimising the biases attributed to the climate model. Whereas it is common practice in climate impact studies, it raises a significant computing challenge for air quality projections that are often as demanding as the climate modelling itself. Alternatively, emerging initiatives proposing statistical adjustments of the climate model could be contemplated (Colette et al., 2012c).

Our air quality and climate projections indicate that exposure to air pollution will decrease substantively by 2050 according to the mitigation pathway (that aims at keeping global warming below 2 °C by the end of the century) where exposure weighted SOMO35 and PM<sub>2.5</sub> will be reduced by 80 and 78 %, respectively. For the reference scenario (ignoring any climate policy) the perspective is more balanced with a slight increase of SOMO35 (7 %) while PM<sub>2.5</sub> nevertheless decreases (by 62 %).

As far as the impact of climate alone on the net projected change is concerned, some of the features obtained with this new modelling suite appear robust when compared to the literature (Hedegaard et al., 2008, 2013; Katragkou et al., 2011; Langner et al., 2012a, b; Manders et al., 2012; Meleux et al., 2007; Szopa et al., 2006). The geographical patterns of projected impact of climate on ozone indicate an increase over southern continental Europe and a decrease over northern Europe and the British Isles. The decrease in the north western part of the domain is a very robust feature. As pointed out in Langner et al. (2012b), the increase over the southern part of the domain is more sensitive as it shifts from the continental surfaces (Hedegaard et al., 2013; Manders et al., 2012; Meleux et al., 2007) to a maximum over the Mediterranean (Andersson and Engardt, 2010). Our results are somewhat half-way between the two options.

The geographical patterns of the impact of climate on particulate matter appear much less robust, as emphasized by Tai et al. (2012). With the set of climate forcing used here, we obtain a benefit for PM<sub>2.5</sub> whereas penalties were reported by Hedegaard et al. (2013) and Manders et al. (2012). This lack of robustness may be related to the spread of precipitation projections that is very significant in regional climate models (Christensen and Christensen, 2007) but other climate features such as weather regimes can play a role.

A quantitative comparison of the driving factors has been conducted. The climate penalty is compensated by the projected changes in precursor emissions and to a lesser extent by intercontinental transport of pollution. Whereas the first studies on the sole impact of climate on ozone pointed toward a strong penalty brought about by climate change (Meleux et al., 2007), more recent assessments including air pollutant emission projections already emphasised the larger role of the latter (Hedegaard et al., 2013; Langner et al., 2012a). As far as intercontinental transport of pollution is concerned, a significant contribution was already envisaged by Langner et al. (2012a) and Szopa et al. (2006).



We conclude that the overall climate penalty bearing upon ozone is confirmed, and its geographical patterns present some degree of robustness. At the same time, its importance should not be overstated. On a quantitative basis, we find that the air quality legislation being envisaged today should be able to counterbalance the climate penalty. On the contrary, the sensitivity to background changes (resulting from both intercontinental transport of pollution and the impact of global climate change on the ozone burden) was overlooked in the literature, whereas its impact competes even more than the climate penalty with the beneficial air quality legislation.

For particulate matter, the small benefit brought about by climate change is largely dominated by the response attributed to changes in air pollutant emissions, while the contribution of boundary conditions is moderate. We note however that there is no consensus whether climate change constitutes a penalty or a benefit for particulate matter (Jacob and Winner, 2009). At the same time a considerable attention is devoted to the investigation of direct and indirect impact of aerosols on climate. Increasing the robustness of the anticipated impact of climate change on particulate matter should become a key research priority in the coming years.

At this stage, our evaluation of uncertainty remains limited since the results are confined to a comprehensive and up-to-date but *individual* suite of models and scenarios. Our study calls for a more coordinated approach using an ensemble of models for both climate and air quality. While such a coordinated exercise has been conducted before at global scale (e.g. ACCMIP), there is no equivalent at the regional scale.

Would such an experiment be implemented in the future, there are good chances that the modelling community could provide more quantitative inputs in terms of climate penalty/benefit for ozone over Europe. It remains difficult to give an estimate of the level of understanding of the contribution of distant sources because of the lack of significant coverage in the literature devoted to regional air quality projections. However, this contribution is expected to be significant for ozone, perhaps even more so than the impact of regional climate change. The issue of particulate matter is much less mature, if the contribution of intercontinental transport of pollution is probably limited, the impact of climate is uncertain.

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## References

- Amann, M. and Lutz, M.: The revision of the air quality legislation in the European Union related to ground-level ozone, *J. Hazard. Mater.*, 78, 41–62, 2000.
- Amann, M., Bertok, I., Borcken-Kleefeld, J., Cofala, J., Heyes, C., Höglund-Isaksson, L., Klimont, Z., Nguyen, B., Posch, M., Rafaj, P., Sandler, R., Schöpp, W., Wagner, F., and Winiwarter, W.: Cost-effective control of air quality and greenhouse gases in Europe: Modeling and policy applications, *Environ. Modell. Softw.*, 26, 1489–1501, 2011.
- Andersson, C. and Engardt, M.: European ozone in a future climate: Importance of changes in dry deposition and isoprene emissions, *J. Geophys. Res.*, 115, D02303, doi:10.1029/2008JD011690, 2010.
- Bessagnet, B., Hodzic, A., Vautard, R., Beekmann, M., Cheinet, S., Honore, C., Liousse, C., and Rouil, L.: Aerosol modeling with CHIMERE – preliminary evaluation at the continental scale, *Atmos. Environ.*, 38, 2803–2817, doi:10.1016/j.atmosenv.2004.02.034, 2004.
- Bessagnet, B., Menut, L., Curci, G., Hodzic, A., Guillaume, B., Liousse, C., Moukhtar, S., Pun, B., Seigneur, C., and Schulz, M.: Regional modeling of carbonaceous aerosols over Europe – focus on secondary organic aerosols, *J. Atmos. Chem.*, 61, 175–202, 2008a.
- Bessagnet, B., Menut, L., Curci, G., Hodzic, A., Guillaume, B., Liousse, C., Moukhtar, S., Pun, B., Seigneur, C., and Schulz, M.: Regional modeling of carbonaceous aerosols over Europe – focus on secondary organic aerosols, *J. Atmos. Chem.*, 61, 175–202, doi:10.1007/s10874-009-9129-2, 2008b.
- Butler, T. M., Stock, Z. S., Russo, M. R., Denier van der Gon, H. A. C., and Lawrence, M. G.: Megacity ozone air quality under four alternative future scenarios, *Atmos. Chem. Phys.*, 12, 4413–4428, doi:10.5194/acp-12-4413-2012, 2012.
- Christensen, J. and Christensen, O.: A summary of the PRUDENCE model projections of changes in European climate by the end of this century, *Climatic Change*, 81, 7–30, 2007.
- Cohan, D. S. and Napelenok, S. L.: Air Quality Response Modeling for Decision Support, *Atmosphere*, 2, 407–425, 2011.
- Colette, A., Granier, C., Hodnebrog, Ø., Jakobs, H., Maurizi, A., Nyiri, A., Bessagnet, B., D'Angiola, A., D'Isidoro, M., Gauss, M., Meleux, F., Memmesheimer, M., Mieville, A., Rouil, L., Russo, F., Solberg, S., Stordal, F., and Tampieri, F.: Air quality trends in Europe over the past decade: a first multi-model assessment, *Atmos. Chem. Phys.*, 11, 11657–11678, doi:10.5194/acp-11-11657-2011, 2011.
- Colette, A., Granier, C., Hodnebrog, Ø., Jakobs, H., Maurizi, A., Nyiri, A., Rao, S., Amann, M., Bessagnet, B., D'Angiola, A., Gauss, M., Heyes, C., Klimont, Z., Meleux, F., Memmesheimer, M., Mieville, A., Rouil, L., Russo, F., Schucht, S., Simpson,

- D., Stordal, F., Tampieri, F., and Vrac, M.: Future air quality in Europe: a multi-model assessment of projected exposure to ozone, *Atmos. Chem. Phys.*, 12, 10613–10630, doi:10.5194/acp-12-10613-2012, 2012a.
- Colette, A., Koelmeijer, R., Mellios, G., Schucht, S., Péré, J.-C., Kouridis, C., Bessagnet, B., Eerens, H., Van Velze, K., and Rouil, L.: Cobenefits of climate and air pollution regulations, The context of the European Commission Roadmap for moving to a low carbon economy in 2050, ETC/ACM – EEA, Copenhagen, 78, 2012b.
- Colette, A., Vautard, R., and Vrac, M.: Regional climate downscaling with prior statistical correction of the global climate forcing, *Geophys. Res. Lett.*, 39, L13707, doi:10.1029/2012GL052258, 2012c.
- Curci, G.: European Biogenic Isoprene Emissions Constrained by Satellite Observations of Formaldehyde, PhD, Univ. of L'Aquila, 2006.
- Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M. A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N., Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Hólm, E. V., Isaksen, I., Kållberg, P., Köhler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M., Morcrette, J. J., Park, B. K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J. N., and Vitart, F.: The ERA-Interim reanalysis: configuration and performance of the data assimilation system, *Q. J. Roy. Meteor. Soc.*, 137, 553–597, 2011.
- Dufresne, J.-L., Foujols, M.-A., Denvil, S., Caubel, A., Marti, O., Aumont, O., Balkanski, Y., Bekki, S., Bellenger, H., Benschila, R., Bony, S., Bopp, L., Braconnot, P., Brockmann, P., Cadule, P., Cheruy, F., Codron, F., Cozic, A., Cugnet, D., de Noblet, N., Duvel, J.-P., Ethé, C., Fairhead, L., Fichefet, T., Flavoni, S., Friedlingstein, P., Grandpeix, J.-Y., Guez, L., Guilyardi, E., Hauglustaine, D., Hourdin, F., Idelkadi, A., Ghattas, J., Jous-saume, S., Kageyama, M., Krinner, G., Labetoulle, S., Lahellec, A., Lefebvre, M.-P., Lefevre, F., Levy, C., Li, Z. X., Lloyd, J., Lott, F., Madec, G., Mancip, M., Marchand, M., Masson, S., Meurdesoif, Y., Mignot, J., Musat, I., Parouty, S., Polcher, J., Rio, C., Schulz, M., Swingedouw, D., Szopa, S., Talandier, C., Terray, P., and Viovy, N.: Climate change projections using the IPSL-CM5 Earth System Model: from CMIP3 to CMIP5 *Clim. Dynam.*, 40, 2123–2165, 2013.
- EEA: Air pollution and climate change policies in Europe: exploring linkages and the added value of an integrated approach, European Environment Agency, Copenhagen, 2004.
- EEA: Assessment of ground-level ozone in EEA member countries, with a focus on long-term trends, European Environment Agency, Copenhagen, 56, 2009.
- Fichefet, T., and Morales-Maqueda, A.: Modelling the influence of snow accumulation and snow-ice formation on the seasonal cycle of the Antarctic sea-ice cover, *Clim. Dynam.*, 15, 251–268, 1999.
- Fiore, A. M., Naik, V., Spracklen, D. V., Steiner, A., Unger, N., Prather, M., Bergmann, D., Cameron-Smith, P. J., Cionni, I., Collins, W. J., Dalsoren, S., Eyring, V., Folberth, G. A., Ginoux, P., Horowitz, L. W., Josse, B., Lamarque, J.-F., MacKenzie, I. A., Nagashima, T., O'Connor, F. M., Righi, M., Rumbold, S. T., Shindell, D. T., Skeie, R. B., Sudo, K., Szopa, S., Takemura, T., and Zeng, G.: Global air quality and climate, *Chem. Soc. Rev.*, 41, 6663–6683, 2012.
- Folberth, G. A., Hauglustaine, D. A., Lathière, J., and Brocheton, F.: Interactive chemistry in the Laboratoire de Météorologie Dynamique general circulation model: model description and impact analysis of biogenic hydrocarbons on tropospheric chemistry, *Atmos. Chem. Phys.*, 6, 2273–2319, doi:10.5194/acp-6-2273-2006, 2006.
- Forster, P., Ramaswamy, V., Artaxo, P., Berntsen, T., Betts, R., Fahey, D., Haywood, J., Lean, J., Lowe, D., Myhre, G., Nganga, J., Prinn, R., Raga, G., Schultz, M., and Van Dorland, R.: Changes in atmospheric constituents and in radiative forcing, in: *Climate Change 2007: The physical science basis: Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change* edited by: Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K., Tignor, M., and Miller, H., Cambridge University Press, Cambridge, UK, 2007.
- Giorgi, F., Jones, C., and Asrar, G. R.: Addressing climate information needs at the regional level: the CORDEX framework, *WMO Bulletin*, 58, 175–183, 2009.
- Grell, G. and Baklanov, A.: Integrated modeling for forecasting weather and air quality: A call for fully coupled approaches, *Atmos. Environ.*, 45, 6845–6851, 2011.
- Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), *Atmos. Chem. Phys.*, 6, 3181–3210, doi:10.5194/acp-6-3181-2006, 2006.
- Hamaoui-Laguel, L., Meleux, F., Beekmann, M., Bessagnet, B., Génernont, S., Cellier, P., and Léinois, L.: Improving ammonia emissions in air quality modelling for France, *Atmos. Environ.*, 2012.
- Hauglustaine, D. A., Hourdin, F., Jourdain, L., Filiberti, M. A., Walters, S., Lamarque, J. F., and Holland, E. A.: Interactive chemistry in the Laboratoire de Météorologie Dynamique general circulation model: Description and background tropospheric chemistry evaluation, *J. Geophys. Res.*, 109, D04314, doi:10.1029/2003JD003957, 2004.
- Hedegaard, G. B., Brandt, J., Christensen, J. H., Frohn, L. M., Geels, C., Hansen, K. M., and Stendel, M.: Impacts of climate change on air pollution levels in the Northern Hemisphere with special focus on Europe and the Arctic, *Atmos. Chem. Phys.*, 8, 3337–3367, doi:10.5194/acp-8-3337-2008, 2008.
- Hedegaard, G. B., Christensen, J. H., and Brandt, J.: The relative importance of impacts from climate change vs. emissions change on air pollution levels in the 21st century, *Atmos. Chem. Phys.*, 13, 3569–3585, doi:10.5194/acp-13-3569-2013, 2013.
- Honoré, C., Rouil, L., Vautard, R., Beekmann, M., Bessagnet, B., Dufour, A., Elichegaray, C., Flaud, J. M., Malherbe, L., Meleux, F., Menut, L., Martin, D., Peuch, A., Peuch, V. H., and Poisson, N.: Predictability of European air quality: Assessment of 3 years of operational forecasts and analyses by the PREV'AIR system, *J. Geophys. Res.-Atmos.*, 113, D04301, doi:10.1029/2007jd008761, 2008.
- Hourdin, F., Musat, I., Bony, S., Braconnot, P., Codron, F., Dufresne, J.-L., Fairhead, L., Filiberti, M.-A., Friedlingstein, P., Grandpeix, J.-Y., Krinner, G., LeVan, P., Li, Z.-X., and Lott, F.: The LMDZ4 general circulation model: climate performance and sensitivity to parametrized physics with emphasis on tropical convection, *Clim. Dynam.*, 27, 787–813, 2006.

- Hourdin, F., Foujols, M.-A., Codron, F., Guemas, V., Dufresne, J.-L., Bony, S., Denvil, S., Guez, L., Lott, F., Ghattas, J., Braconnot, P., Marti, O., Meurdesoif, Y., and Bopp, L.: Impact of the LMDZ atmospheric grid configuration on the climate and sensitivity of the IPSL-CM5A coupled model, *Clim. Dynam.*, 40, 2167–2192, doi:10.1007/s00382-012-1411-3, 2012.
- IPCC: Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge, UK and New York, NY, USA, 2007.
- Jacob, D. J. and Winner, D. A.: Effect of climate change on air quality, *Atmos. Environ.*, 43, 51–63, 2009.
- Katragkou, E., Zanis, P., Tegoulas, I., Melas, D., Kioutsioukis, I., Krüger, B. C., Huszar, P., Halenka, T., and Rauscher, S.: Decadal regional air quality simulations over Europe in present climate: near surface ozone sensitivity to external meteorological forcing, *Atmos. Chem. Phys.*, 10, 11805–11821, doi:10.5194/acp-10-11805-2010, 2010.
- Katragkou, E., Zanis, P., Kioutsioukis, I., Tegoulas, I., Melas, D., Krüger, B. C., and Coppola, E.: Future climate change impacts on summer surface ozone from regional climate-air quality simulations over Europe, *J. Geophys. Res.*, 116, D22307, doi:10.1029/2011JD015899, 2011.
- Krinner, G., Viovy, N., de Noblet-Ducoudré, N., Ogée, J., Polcher, J., Friedlingstein, P., Ciais, P., Sitch, S., and Prentice, I. C.: A dynamic global vegetation model for studies of the coupled atmosphere-biosphere system, *Global Biogeochem. Cy.*, 19, GB1015, doi:10.1029/2003GB002199, 2005.
- Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi, K., and van Vuuren, D. P.: Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, *Atmos. Chem. Phys.*, 10, 7017–7039, doi:10.5194/acp-10-7017-2010, 2010.
- Lamarque, J.-F., Shindell, D. T., Josse, B., Young, P. J., Cionni, I., Eyring, V., Bergmann, D., Cameron-Smith, P., Collins, W. J., Doherty, R., Dalsoren, S., Faluvegi, G., Folberth, G., Ghan, S. J., Horowitz, L. W., Lee, Y. H., MacKenzie, I. A., Nagashima, T., Naik, V., Plummer, D., Righi, M., Rumbold, S. T., Schulz, M., Skeie, R. B., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., Voulgarakis, A., and Zeng, G.: The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP): overview and description of models, simulations and climate diagnostics, *Geosci. Model Dev.*, 6, 179–206, doi:10.5194/gmd-6-179-2013, 2013.
- Langner, J., Engardt, M., and Andersson, C.: European summer surface ozone 1990–2100, *Atmos. Chem. Phys.*, 12, 10097–10105, doi:10.5194/acp-12-10097-2012, 2012a.
- Langner, J., Engardt, M., Baklanov, A., Christensen, J. H., Gauss, M., Geels, C., Hedegaard, G. B., Nuterman, R., Simpson, D., Soares, J., Sofiev, M., Wind, P., and Zakey, A.: A multi-model study of impacts of climate change on surface ozone in Europe, *Atmos. Chem. Phys.*, 12, 10423–10440, doi:10.5194/acp-12-10423-2012, 2012b.
- Lathière, J., Hewitt, C. N., and Beerling, D. J.: Sensitivity of isoprene emissions from the terrestrial biosphere to 20th century changes in atmospheric CO<sub>2</sub> concentration, climate, and land use, *Global Biogeochem. Cy.*, 24, GB1004, doi:10.1029/2009GB003548, 2010.
- Lei, H., Wuebbles, D. J., and Liang, X.-Z.: Projected risk of high ozone episodes in 2050, *Atmos. Environ.*, 59, 567–577, 2012.
- Löndahl, J., Swietlicki, E., Lindgren, E., and Loft, S.: Aerosol exposure versus aerosol cooling of climate: what is the optimal emission reduction strategy for human health?, *Atmos. Chem. Phys.*, 10, 9441–9449, doi:10.5194/acp-10-9441-2010, 2010.
- Madec, G., Delecluse, P., Imbard, M., and Levy, C.: Ocean general circulation model reference manual 3., LODYC, Paris, 91, 1997.
- Manders, A. M. M., van Meijgaard, E., Mues, A. C., Kranenburg, R., van Ulft, L. H., and Schaap, M.: The impact of differences in large-scale circulation output from climate models on the regional modeling of ozone and PM, *Atmos. Chem. Phys.*, 12, 9441–9458, doi:10.5194/acp-12-9441-2012, 2012.
- Marti, O., Braconnot, P., Dufresne, J. L., Bellier, J., Benshila, R., Bony, S., Brockmann, P., Cadule, P., Caubel, A., Codron, F., de Noblet, N., Denvil, S., Fairhead, L., Fichefet, T., Foujols, M. A., Friedlingstein, P., Goosse, H., Grandpeix, J. Y., Guilyardi, E., Hourdin, F., Idelkadi, A., Kageyama, M., Krinner, G., Lévy, C., Madec, G., Mignot, J., Musat, I., Swingedouw, D., and Talandier, C.: Key features of the IPSL ocean atmosphere model and its sensitivity to atmospheric resolution, *Clim. Dynam.*, 34, 1–26, 2010.
- Meleux, F., Solmon, F., and Giorgi, F.: Increase in summer European ozone amounts due to climate change, *Atmos. Environ.*, 41, 7577–7587, 2007.
- Menut, L.: Adjoint modeling for atmospheric pollution process sensitivity at regional scale, *J. Geophys. Res.*, 108, 8562, doi:10.1029/2002JD002549, 2003.
- Menut, L., Tripathi, O. P., Colette, A., Vautard, R., Flaounas, R., and Bessagnet, B.: Evaluation of regional climate model forcing with an air quality perspective, *Clim. Dynam.*, 40, 2515–2533, doi:10.1007/s00382-012-1345-9, 2012.
- Menut, L., Bessagnet, B., Khvorostyanov, D., Beekmann, M., Blond, N., Colette, A., Coll, I., Curci, G., Foret, G., Hodzic, A., Mailler, S., Meleux, F., Monge, J.-L., Pison, I., Siour, G., Turquety, S., Valari, M., Vautard, R., and Vivanco, M. G.: CHIMERE 2013: a model for regional atmospheric composition modelling, *Geosci. Model Dev.*, 6, 981–1028, doi:10.5194/gmd-6-981-2013, 2013.
- Messner, S., and Strubegger, M.: User's guide for MESSAGE III, IIASA, Laxenburg, 1995.
- Nenes, A., Pandis, S., and Pilinis, C.: ISORROPIA: A New Thermodynamic Equilibrium Model for Multiphase Multicomponent Inorganic Aerosols, *Aquat. Geochem.*, 4, 123–152, 1998.
- Nyiri, A., Gauss, M., Tsyro, S., Wind, P., and Haugen, J. E.: Future Air Quality, including Climate Change in Transboundary Acidification, Eutrophication and Ground Level Ozone in Europe in 2008, EMEP, Oslo, 2010.
- Pankow, J. F.: An absorption model of the gas/aerosol partitioning involved in the formation of secondary organic aerosol, *Atmos. Environ.*, 28, 189–193, 1994.
- Rao, S., Chirkov, V., Dentener, F., Dingenen, R., Pachauri, S., Purohit, P., Amann, M., Heyes, C., Kinney, P., Kolp, P., Klimont, Z., Riahi, K., and Schoepp, W.: Environmental Modeling and Methods for Estimation of the Global Health Impacts of Air Pollution, *Environ. Model. Assess.*, 17, 613–622, doi:10.1007/s10666-012-

- 9317-3, 2012.
- Riahi, K., Gruebler, A., and Nakicenovic, N.: Scenarios of long-term socio-economic and environmental development under climate stabilisation, *Technol. Forecast. Soc.*, 74, 887–935, doi:10.1016/j.techfore.2006.05.026, 2007.
- Riahi, K., Dentener, F., Gielen, D., Grubler, A., Jewell, J., Klimont, Z., Krey, V., McCollum, D., Pachauri, S., Rao, S., van Ruijven, B., van Vuuren, D. P., and Wilson, C.: Energy Pathways for Sustainable Development, in: *Global Energy Assessment: Toward a Sustainable Future*, edited by: Nakicenovic, N., IIASA, Laxenburg, Austria and Cambridge University Press, Cambridge, UK and New York, NY, 2012.
- Rouil, L., Honore, C., Vautard, R., Beekmann, M., Bessagnet, B., Malherbe, L., Meleux, F., Dufour, A., Elichegaray, C., Flaud, J. M., Menut, L., Martin, D., Peuch, A., Peuch, V. H., and Poisson, N.: PREV'AIR An Operational Forecasting and Mapping System for Air Quality in Europe, *B. Am. Meteorol. Soc.*, 90, 73–83, doi:10.1175/2008bams2390.1, 2009.
- Shindell, D. T., Lamarque, J.-F., Schulz, M., Flanner, M., Jiao, C., Chin, M., Young, P. J., Lee, Y. H., Rotstajn, L., Mahowald, N., Milly, G., Faluvegi, G., Balkanski, Y., Collins, W. J., Conley, A. J., Dalsoren, S., Easter, R., Ghan, S., Horowitz, L., Liu, X., Myhre, G., Nagashima, T., Naik, V., Rumbold, S. T., Skeie, R., Sudo, K., Szopa, S., Takemura, T., Voulgarakis, A., Yoon, J.-H., and Lo, F.: Radiative forcing in the ACCMIP historical and future climate simulations, *Atmos. Chem. Phys.*, 13, 2939–2974, doi:10.5194/acp-13-2939-2013, 2013.
- Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L. D., Fagerli, H., Flechard, C. R., Hayman, G. D., Gauss, M., Jonson, J. E., Jenkin, M. E., Nyíri, A., Richter, C., Semeena, V. S., Tsyro, S., Tuovinen, J.-P., Valdebenito, Á., and Wind, P.: The EMEP MSC-W chemical transport model – technical description, *Atmos. Chem. Phys.*, 12, 7825–7865, doi:10.5194/acp-12-7825-2012, 2012.
- Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Duda, M. G., Huang, X. Y., Wang, W., and Powers, J. G.: A Description of the Advanced Research WRF Version 3, NCAR, 2008.
- Solazzo, E., Bianconi, R., Pirovano, G., Matthias, V., Vautard, R., Moran, M. D., Wyat Appel, K., Bessagnet, B., Brandt, J., Christensen, J. H., Chemel, C., Coll, I., Ferreira, J., Forkel, R., Francis, X. V., Grell, G., Grossi, P., Hansen, A. B., Miranda, A. I., Nopmongcol, U., Prank, M., Sartelet, K. N., Schaap, M., Silver, J. D., Sokhi, R. S., Vira, J., Werhahn, J., Wolke, R., Yarwood, G., Zhang, J., Rao, S. T., and Galmarini, S.: Operational model evaluation for particulate matter in Europe and North America in the context of AQMEII, *Atmos. Environ.*, 53, 75–92, 2012a.
- Solazzo, E., Bianconi, R., Vautard, R., Appel, K. W., Moran, M. D., Hogrefe, C., Bessagnet, B., Brandt, J. r., Christensen, J. H., Chemel, C., Coll, I., Denier van der Gon, H., Ferreira, J., Forkel, R., Francis, X. V., Grell, G., Grossi, P., Hansen, A. B., Jercevic, A., Kraljevic, L., Miranda, A. I., Nopmongcol, U., Pirovano, G., Prank, M., Riccio, A., Sartelet, K. N., Schaap, M., Silver, J. D., Sokhi, R. S., Vira, J., Werhahn, J., Wolke, R., Yarwood, G., Zhang, J., Rao, S. T., and Galmarini, S.: Model evaluation and ensemble modelling of surface-level ozone in Europe and North America in the context of AQMEII, *Atmos. Environ.*, 53, 60–74, 2012b.
- Stevenson, D. S., Dentener, F. J., Schultz, M. G., Ellingsen, K., van Noije, T. P. C., Wild, O., Zeng, G., Amann, M., Aher-ton, C. S., Bell, N., Bergmann, D. J., Bey, I., Butler, T., Co-fala, J., Collins, W. J., Derwent, R. G., Doherty, R. M., Drevet, J., Eskes, H. J., Fiore, A. M., Gauss, M., Hauglustaine, D. A., Horowitz, L. W., Isaksen, I. S. A., Krol, M. C., Lamarque, J. F., Lawrence, M. G., Montanaro, V., Müller, J. F., Pitari, G., Prather, M. J., Pyle, J. A., Rast, S., Rodriguez, J. M., Sanderson, M. G., Savage, N. H., Shindell, D. T., Strahan, S. E., Sudo, K., and Szopa, S.: Multimodel ensemble simulations of present-day and near-future tropospheric ozone, *J. Geophys. Res.*, 111, D08301, doi:10.1029/2005JD006338, 2006.
- Szopa, S., Hauglustaine, D. A., Vautard, R., and Menut, L.: Future global tropospheric ozone changes and impact on European air quality, *Geophys. Res. Lett.*, 33, L14805, doi:10.1029/2006GL025860, 2006.
- Szopa, S., Balkanski, Y., Schulz, M., Bekki, S., Cugnet, D., Fortems-Cheiney, A., Turquety, S., Cozic, A., Déandreis, C., Hauglustaine, D., Idelkadi, A., Lathière, J., Lefevre, F., Marchand, M., Vuolo, R., Yan, N., and Dufresne, J. L.: Aerosol and ozone changes as forcing for climate evolution between 1850 and 2100, *Clim. Dynam.*, 40, 2223–2250, doi:10.1007/s00382-012-1408-y, 2012.
- Tai, A. P. K., Mickleby, L. J., and Jacob, D. J.: Impact of 2000–2050 climate change on fine particulate matter (PM<sub>2.5</sub>) air quality inferred from a multi-model analysis of meteorological modes, *Atmos. Chem. Phys.*, 12, 11329–11337, doi:10.5194/acp-12-11329-2012, 2012.
- Taylor, K. E., Stouffer, R. J., and Meehl, G. A.: An Overview of CMIP5 and the Experiment Design, *B. Am. Meteorol. Soc.*, 93, 485–498, 2012.
- UNEP: Integrated Assessment of Black Carbon and Tropospheric Ozone: Summary for Policy Makers, UNEP, 2011.
- United Nations, D. o. E. a. S. A., Population Division: World Population Prospects: The 2008 Revision, Highlights, UN, New York, 2009.
- van Loon, M., Vautard, R., Schaap, M., Bergström, R., Bessagnet, B., Brandt, J., Builtjes, P. J. H., Christensen, J. H., Cuvelier, C., Graff, A., Jonson, J. E., Krol, M., Langner, J., Roberts, P., Rouil, L., Stern, R., Tarrasón, L., Thunis, P., Vignati, E., White, L., and Wind, P.: Evaluation of long-term ozone simulations from seven regional air quality models and their ensemble, *Atmos. Environ.*, 41, 2083–2097, 2007.
- van Vuuren, D., Edmonds, J., Kainuma, M., Riahi, K., Thomson, A., Hibbard, K., Hurtt, G., Kram, T., Krey, V., Lamarque, J.-F., Masui, T., Meinshausen, M., Nakicenovic, N., Smith, S., and Rose, S.: The representative concentration pathways: an overview, *Climatic Change*, 109, 5–31, 2011.
- Vautard, R., Honoré, C., Beekmann, M., and Rouil, L.: Simulation of ozone during the August 2003 heat wave and emission control scenarios, *Atmos. Environ.*, 39, 2957–2967, 2005.
- Vautard, R., Builtjes, P. H. J., Thunis, P., Cuvelier, C., Bedogni, M., Bessagnet, B., Honore, C., Moussiopoulos, N., Pirovano, G., Schaap, M., Stern, R., Tarrason, L., and Wind, P.: Evaluation and intercomparison of Ozone and PM10 simulations by several chemistry transport models over four European cities within the CityDelta project, *Atmos. Environ.*, 41, 173–188, doi:10.1016/j.atmosenv.2006.07.039, 2007.

- Vautard, R., Brankovic, C., Colette, A., Deque, M., Fernandez, J., Gobiet, A., Goergen, K., Nikulin, G., Guettler, I., Keuler, K., Warrach-Sagi, K., Teichmann, C., and Halenka, T.: The simulation of European heat waves from an ensemble of regional climate models within the EURO-CORDEX project, *Clim. Dynam.*, doi:10.1007/s00382-013-1714-z, 2012.
- Vial, J., Dufresne, J.-L., and Bony, S.: On the interpretation of inter-model spread in CMIP5 climate sensitivity estimates, *Clim. Dynam.*, 1–24, doi:10.1007/s00382-013-1725-9, 2013.
- Wilkinson, M. J., Monson, R. K., Trahan, N., Lee, S., Brown, E., Jackson, R. B., Polley, H. W., Fay, P. A., and Fall, R. A. Y.: Leaf isoprene emission rate as a function of atmospheric CO<sub>2</sub> concentration, *Glob. Change Biol.*, 15, 1189–1200, 2009.
- Wu, S., Mickley, L. J., Leibensperger, E. M., Jacob, D. J., Rind, D., and Streets, D. G.: Effects of 2000–2050 global change on ozone air quality in the United States, *J. Geophys. Res.-Atmos.*, 113, D06302, doi:10.1029/2007JD008917, 2008.
- Young, P. J., Archibald, A. T., Bowman, K. W., Lamarque, J.-F., Naik, V., Stevenson, D. S., Tilmes, S., Voulgarakis, A., Wild, O., Bergmann, D., Cameron-Smith, P., Cionni, I., Collins, W. J., Dalsøren, S. B., Doherty, R. M., Eyring, V., Faluvegi, G., Horowitz, L. W., Josse, B., Lee, Y. H., MacKenzie, I. A., Nagashima, T., Plummer, D. A., Righi, M., Rumbold, S. T., Skeie, R. B., Shindell, D. T., Strode, S. A., Sudo, K., Szopa, S., and Zeng, G.: Pre-industrial to end 21st century projections of tropospheric ozone from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), *Atmos. Chem. Phys.*, 13, 2063–2090, doi:10.5194/acp-13-2063-2013, 2013.
- Zanis, P., Katragkou, E., Tegoulas, I., Poupkou, A., Melas, D., Huszar, P., and Giorgi, F.: Evaluation of near surface ozone in air quality simulations forced by a regional climate model over Europe for the period 1991–2000, *Atmos. Environ.*, 45, 6489–6500, 2011.
- Zyryanov, D., Foret, G., Eremenko, M., Beekmann, M., Cammas, J.-P., D’Isidoro, M., Elbern, H., Flemming, J., Friese, E., Kioutsioutkis, I., Maurizi, A., Melas, D., Meleux, F., Menut, L., Moinat, P., Peuch, V.-H., Poupkou, A., Razinger, M., Schultz, M., Stein, O., Suttie, A. M., Valdebenito, A., Zerefos, C., Du-four, G., Bergametti, G., and Flaud, J.-M.: 3-D evaluation of tropospheric ozone simulations by an ensemble of regional Chemistry Transport Model, *Atmos. Chem. Phys.*, 12, 3219–3240, doi:10.5194/acp-12-3219-2012, 2012.