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Using statistical models to explore ensemble uncertainty in climate impact studies: the example of air pollution in Europe

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Abstract. Because of its sensitivity to unfavorable weather patterns, air pollution is sensitive to climate change so that, in the future, a climate penalty could jeopardize the expected efficiency of air pollution mitigation measures. A common method to assess the impact of climate on air quality consists in implementing chemistry-transport models forced by climate projections. However, the computing cost of such methods requires optimizing ensemble exploration techniques.

By using a training data set from a deterministic projection of climate and air quality over Europe, we identified the main meteorological drivers of air quality for eight regions in Europe and developed statistical models that could be used to predict air pollutant concentrations. The evolution of the key climate variables driving either particulate or gaseous pollution allows selecting the members of the EuroCordex ensemble of regional climate projections that should be used in priority for future air quality projections (CanESM2/RCA4; CNRM-CM5-LR/RCA4 and CSIRO-Mk3-6-0/RCA4 and MPI-ESM-LR/CCLM following the EuroCordex terminology).

After having tested the validity of the statistical model in predictive mode, we can provide ranges of uncertainty attributed to the spread of the regional climate projection ensemble by the end of the century (2071–2100) for the RCP8.5.

In the three regions where the statistical model of the impact of climate change on $PM_{2.5}$ offers satisfactory performances, we find a climate benefit (a decrease of $PM_{2.5}$ concentrations under future climate) of -1.08 (± 0.21), -1.03 (± 0.32), -0.83 (± 0.14) µg m^{-3}, for respectively Eastern Europe, Mid-Europe and Northern Italy. In the British-Irish Isles, Scandinavia, France, the Iberian Peninsula and the Mediterranean, the statistical model is not considered skillful enough to draw any conclusion for $PM_{2.5}$.

In Eastern Europe, France, the Iberian Peninsula, Mid-Europe and Northern Italy, the statistical model of the impact of climate change on ozone was considered satisfactory and it confirms the climate penalty bearing upon ozone of 10.51 (± 3.06), 11.70 (± 3.63), 11.53 (± 1.55), 9.86 (± 4.41), 4.82 (± 1.79) µg m^{-3}, respectively. In the British-Irish Isles, Scandinavia and the Mediterranean, the skill of the statistical model was not considered robust enough to draw any conclusion for ozone pollution.

1 Introduction

The main drivers of air pollution are (i) emission of primary pollutants and precursors of secondary pollutants, (ii) long-range transport, (iii) atmospheric chemistry and (iv) meteorology (Jacob and Winner, 2009). We can thus anticipate that air quality is sensitive to climate change taking as example the link between heat waves and large-scale ozone episodes (Vautard et al., 2005). But in addition to the direct impact of climate change on air pollution through the change in frequency and severity of synoptic conditions conducive to the accumulation of air pollutants we must also note that climate can have an impact on anthropogenic and biogenic emission of pollutants and precursors (Langner et al., 2012b) as well as on changes in the global background of pollution, and therefore long-range transport (Young et al., 2013). There is therefore a concern that in the future, climate change could jeopar-

dize the expected efficiency of pollution mitigation measures, even if the available studies indicate that if projected emission reductions are achieved they should exceed the magnitude of the climate penalty (Colette et al., 2013; Hedegaard et al., 2013).

The most widespread technique used to assess the impact of climate change on air quality consists in implementing regional climate projections in Chemistry Transport Models (CTM) (Jacob and Winner, 2009). The computational cost of such technique is substantial given that it involves multi-annual global climate simulations, dynamical downscaling through regional climate simulations and ultimately CTM simulations. Besides the computational cost, it also raises technical difficulties in collecting, transferring, and managing large amounts of model data. Unlike many climate impact studies, CTM projections require Regional Climate Model fields in three dimensions and at high temporal frequency, whereas many regional climate modelling groups only store a few vertical levels in compliance with the CORDEX data archiving protocols. Altogether, these difficulties led to the use of a single source of climate projections in the majority of future air quality projections (Meleux et al., 2007; Katragkou et al., 2011; Jiménez-Guerrero et al., 2012; Langner et al., 2012b; Colette et al., 2013, 2015; Hedegaard et al., 2013; Varotsos et al., 2013) or two at most in published studies (Huszar et al., 2011; Juda-Rezler et al., 2012; Langner et al., 2012a; Manders et al., 2012). There are examples where more than two climate forcings are used, but then they are implemented with different CTMs, so that the uncertainties in the spread of RCM and CTMs is aggregated, thereby offering a poor understanding of the climate uncertainty. In addition, it should be noted that the choice of the climate driver is generally a matter of opportunity rather than an informed choice. These studies capture trends and variability but their coverage of uncertainty is not satisfactory in the climate change context. This unsatisfactory handling of uncertainties is well illustrated by the divergence in the very sign of the impact of climate change on particulate matter (e.g. Lecœur et al., 2014, find a climate benefit for $PM_{2.5}$ in Europe while Manders et al., 2012, suggest the opposite). Thus the lack of multi-model approach in air quality projections is a serious caveat that needs to be tackled in order to comply with best practices in the field of climate impact research, where ensemble approaches is state of the art.

Hence, in order to assess the climate uncertainties on surface ozone and particulate matter over Europe in a changing climate, we developed an alternative method which does not require forcing a CTM with an ensemble of climate models. It consists in developing a statistical model fitted to a deterministic CTM simulation forced by a single RCM that can be subsequently applied to a larger ensemble of regional climate projections. This method allows selecting the members of the RCM ensemble that offer the widest range in terms of air quality response, somehow the "air quality sensitivity to climate change projections". These selected members should

be used in priority in future air quality projections. A byproduct of our statistical air quality projections is that we explore an unprecedented range of climate uncertainty compared to the published literature that relies, at best, on two distinct climate forcings. The confidence we can have in these statistical projections is of course limited by the skill of the statistical model. Our approach of using a simplified air quality impact model but with a larger range of climate forcing can therefore be considered complementary with the more complex CTMs used with a limited number of climate forcings. The use of such a methodology is inspired from earlier work in the field of hydrology, where Vano and Lettenmaier (2014) estimated future stream-flow by using a sensitivity-based approach which could be applied to generate ensemble simulations. Such a hybrid statistical and deterministic approach has also been used in the past in the field of air quality, but mostly for near-term and local forecasting, relying on statistical models of various complexity (i.e. Land Use Regression, Neural Network, Nonlinear regression, Generalized Additive Models etc.) (Prybutok et al., 2000; Schlink et al., 2006; Slini et al., 2006). The most relevant example in the context of future air quality projection is that of Lecœur et al. (2014), who used the technique of wind regime analogues, although they did not apply their approach to an ensemble of climate projections.

This paper deals with all the steps needed to build the proxy of ensemble. First (Sect. 2) we present the methods and input data: the design of the statistical model of the air quality response to meteorological drivers is presented as well as the deterministic modelling framework used to create our training data set. Section 3 focuses on results. The deterministic air quality projections are presented for ozone peaks and $PM_{2.5}$ in Sect. 3.1. The selected statistical models for each region are evaluated in Sect. 3.2 for ozone, $PM_{2.5}$ and each sub-constituent of the particulate matter mix. The relevance of the statistical method to evaluate climate uncertainties and optimize the exploration of the ensemble of climate projections is discussed in Sect. 4.

2 Methodology

2.1 Design

We consider ozone and $PM_{2.5}$ as the main pollutants of interest for both purposes: public health (Dockery and Pope, 1994; Jerrett et al., 2009) and climate interactions (IPCC, 2013). For both of them, we investigated the best correlation that can be found for various European subregions using the following meteorological variables as predictants: near-surface temperature (T_{2m}), daily precipitation, incoming short-wave radiation, planetary boundary layer (PBL) depth, surface wind (U_{10m}) and specific humidity.

The choice of these meteorological variables is based on an analysis of the literature on the chemical and physical processes linking air pollution and meteorology. For $PM_{2.5}$, turbulent mixing, often related to the depth of the planetary boundary layer, dominates (McGrath-Spangler et al., 2015). A decrease of the PBL depth lead to either (i) an increase of the concentration of pollutants because of the lower mixing volume (Jiménez-Guerrero et al., 2012) or (ii) a decrease of their concentrations because of their faster dry deposition to surface receptors (Bessagnet et al., 2010). The wind plays also multiple roles for $PM_{2.5}$. High wind speed favours the dilution of particulate matter (Jacob and Winner, 2009) but enhances sea-salt and dust mobilization (Lecœur and Seigneur, 2013). Precipitation is often reported as a major sink of $PM_{2.5}$ through wet scavenging (Jacob and Winner, 2009). Water vapour participates in aerosol formation during nucleation processes. Moreover, it can have an impact on the rates of certain chemical reactions, similarly to temperature. The overall impact of temperature on $PM_{2.5}$ is difficult to isolate because of the mix of components contributing to $PM_{2.5}$ (organic, inorganic, dust, sea-salt...) and possible compensating effects. For instance, according to Jacob and Winner (2009), a temperature rise has opposite effects for sulphate and nitrate (respectively an increase and a decrease of concentrations). But for the overall $PM_{2.5}$ mass, an increase in temperature will decrease the concentration as a result of higher volatility and subsequent higher aerosol to gas phase conversion (Megaritis et al., 2014).

As far as ozone is concerned, temperature is expected to play a major role as it catalyses atmospheric chemistry (Doherty et al., 2013). Moreover increasing temperature and solar radiation enhance isoprene emission which is a biogenic precursor of ozone (Langner et al., 2012b; Colette et al., 2013). Finally changing the amount of incoming short-wave radiation will play a role on ozone photochemistry, either by enhancing its photolysis by the hydroxyl radical in the presence of water vapour and short-wave radiation or by enhancing its production in the presence of photolysed nitrogen dioxide (Doherty et al., 2013). The impact of the PBL depth on ozone varies with the meteorological conditions. Increasing the depth of the PBL dilutes ozone concentrations, but it may also favour the dilution of nitrogen oxides close to the sources, therefore leading to an increase in ozone concentrations in NO_x saturated areas (Jacob and Winner, 2009). The amount of water vapour in the atmosphere mostly drives the abundance of the hydroxyl radical (OH). OH is involved in ozone destruction through several processes (i.e. photolysis, HNO_3 production) (Varotsos et al., 2013). It is also involved in ozone production via the formation of NO_2 and radicals (Seinfeld and Pandis, 2008).

Starting from the above list of meteorological predictants, we aim to develop a statistical model of ozone and particulate matter for each of the eight European climatic regions defined in the PRUDENCE project (Christensen and Christensen, 2007). These regions are: British-Irish Isles (BI), Iberian Peninsula (IP), France (FR), Mid-Europe (ME), Scandinavia (SC), Northern Italy (NI – referred to as the Alps in Climate studies but chiefly influenced by the polluted Po-Valley in the air quality context), Mediterranean (MD) and Eastern Europe (EA). For each of these regions, a spatial average of predictants (meteorological variables) and pollutant concentrations values is taken. The statistical model is based on daily averages for all meteorological and air pollutant concentrations except ozone for which the daily maximum of 8 h running means is used. The seasonality is removed by subtracting the average seasonal cycle over the historical period. It should be noted that focusing on aggregated quantities greatly improves the skill of the statistical model that would struggle in capture higher temporal frequency and spatial resolution. An analogy is presented in Thunis et al. (2015) who demonstrated that annual mean ozone and particulate matter responses to incremental emission changes were much more linear than previously thought.

For each region and each pollutant, we first select the two most discriminating predictants by testing all the possible couple of meteorological variable and selecting those that reach the highest correlation. In a second stage we design the actual statistical model that consists of a Generalized Additive Model based on the two most discriminating perdictants (Wood, 2006).

It is to facilitate the geophysical interpretation that we use two meteorological variables instead of a linear combination of multiple variables (i.e. prior principal component analysis axes). Limiting their number to two also allows remaining in a 2-D physical parameter space that supports the discussion as will be illustrated below.

2.2 Training and validation data sets

The data sets used to fit and test the statistical models are produced by the regional climate and air quality modelling framework presented in Colette et al. (2013). By using deterministic climate and chemistry models from the global to the regional scale, they could produce long-term air quality projections over Europe. The Earth System Model (ESM) which drives these simulations is the IPSL-CM5A-MR (Dufresne et al., 2013). The global data used in this study were produced for the Coupled Model Intercomparison Project Phase 5 initiative (CMIP5) (Taylor et al., 2012; Young et al., 2013). Then the climate data obtained by the ESM are dynamically downscaled with the regional climate model WRF (Skamarock et al., 2008). The spatial resolution is 0.44° over Europe (Colette et al., 2013). These simulations were part of the low-resolution simulations performed within the framework of the European-Coordinated Regional Climate Downscaling Experiment program (EURO-CORDEX) (Jacob et al., 2014). Whereas higher spatial resolution simulations are available in the EuroCordex ensemble, the 0.44 resolution was considered appropriate for air quality projections in agreement with other publications (Meleux et al.,

2007; Langner et al., 2012a, b; Manders et al., 2012; Colette et al., 2013; Hedegaard et al., 2013; Watson et al., 2015), and also because higher RCM resolution are not specifically performed to improve the climate features that are most sensitive for air quality purposes (temperature, solar radiation, stagnation events, triggering of low-intensity precipitation events etc.). Finally the regional climate fields are used to drive the CTM CHIMERE (Menut et al., 2013), for the projection of air quality under changing climate. Since we are only interested in the effect of climate change, pollutant emissions remain constant at their level of 2010, as prescribed in the ECLIPSE-V4a data set (Klimont et al., 2013). Similarly, chemical boundary conditions prescribed with the INCA model (Hauglustaine et al., 2014) as well as the land-use are also kept constant.

The Chemistry and Transport Model CHIMERE has been used in numerous studies: daily operational forecast (Rouïl et al., 2009), emission scenario evaluation (Cuvelier et al., 2007), evaluation in extreme events (Vautard et al., 2007), long-term studies (Colette et al., 2011, 2013; Wilson et al., 2012) and inter-comparisons models and ensembles (Solazzo et al., 2012a, b).

The model performances depend on the setup but general features include a good representation of ozone daily maxima and an overestimation of night-time concentrations, leading to a small positive bias in average ozone (van Loon et al., 2007). Concerning particulate matter, similarly to most state-of-the-art CTMs, the CHIMERE model presents a systematic negative bias (Bessagnet et al., 2014). Regarding more specifically its implementation in the context of a future climate, evaluations of the CHIMERE model are presented in Colette et al. (2013, 2015) and also Watson et al. (2015) and Lacressonniere et al. (2016).

The training data set used to build the statistical models consists of the historical air quality simulations (1976 to 2005), while projections of air quality under a future climate (RCP8.5 2071–2100) will be used for testing purposes.

In order to evaluate the uncertainty related to climate change, the statistical models should be skillful for both pollutant concentrations over the historical period (training period) and in predictive mode. Alternative RCM forcing of the CHIMERE CTM could be used to test the approach. Unfortunately, such alternatives are not available at this stage. The statistical ensemble exploration technique presented here will ultimately allow selecting the RCM that should be used in priority to cover the range of uncertainties in air quality and climate projections. When such simulations become available, we will be able to further test the skill of the statistical model. However, so far, the only validation that could be included here was to rely on a future time period as a validation data set. The underlying hypothesis is that the historical range of meteorological parameters used to train the model will be exceeded in the future, therefore offering an appropriate testing data set. The results of this validation are presented in Sect. 3.2.

2.3   Projection data set

To evaluate the uncertainty related to the climate forcing, and identify the RCM that should be used in priority for future air quality projections, the statistical model of air quality is used in predictive mode using the regional climate projections performed in the framework of the EURO-CORDEX experiment (Jacob et al., 2014). The combinations of global and regional climate models used here are the following: CanESM2/RCA4; CSIRO-Mk3-6-0/RCA4; CNRM-CM5-LR/RCA4; EC-EARTH/RACMO2; EC-EARTH/RC4; GFDL-ESM2M/RCA4; IPSL-CM5A-MR/RCA4; IPSL-CM5A-MR/WRF; MIROC5/RCA4; MPI-ESM-LR/RCA4; MPI-ESM-LR/CCLM; NorESM1-M/RCA4 (see Jacob et al., 2014, for details on the model nomenclature).

The performances of the global models used to drive the regional projections have been evaluated in Jury (2012) and Cattiaux et al. (2013). In the general EuroCordex evaluation, Kotlarski et al. (2014) finds a good reproduction of the spatial temperature variability even if the models exhibit an underestimation of temperature during the winter in the north Eastern Europe. In addition to this general feature, the specificity of the WRF-IPSL-INERIS member is an overestimation of winter temperatures in the southeast. In terms of precipitations, most of the models exhibit a pronounced wet bias over most subdomains.

When focusing on WRF members of the EuroCordex ensemble, Katragkou et al. (2015) points out that the IPSL-INERIS member offers one of the best balance between precipitation and temperature skills. Both studies are limited to the evaluation of RCM used with perfect boundary conditions (ERA-Interim forcing) and no published study has yet evaluated the various global and/or regional combinations. It should also be noted that the ensemble is poorly balanced in terms of GCM/RCM combinations (see the larger weight of the RCA regional model which raise important question regarding the representativeness of the ensemble).

3   Development and validation of the statistical model

In this part we studied the end (2071–2100) of the century, for one scenario (RCP8.5) which is an energy-intensive scenario (van Vuuren et al., 2011). This 30-year period is chosen to be representative regardless of the inter-annual variability (Langner et al., 2012a). We focus on the RCP8.5 and the end of the century on purpose to reach a strong climate signal.

3.1   Air quality projections

3.1.1   Fine particulate matter

Figure 1a shows the 30 years average $PM_{2.5}$ concentrations over the historical period (1976 to 2005). Higher concentrations are modelled over European pollution hotspots: the Benelux, the Po Valley, Eastern Europe and large cities. A
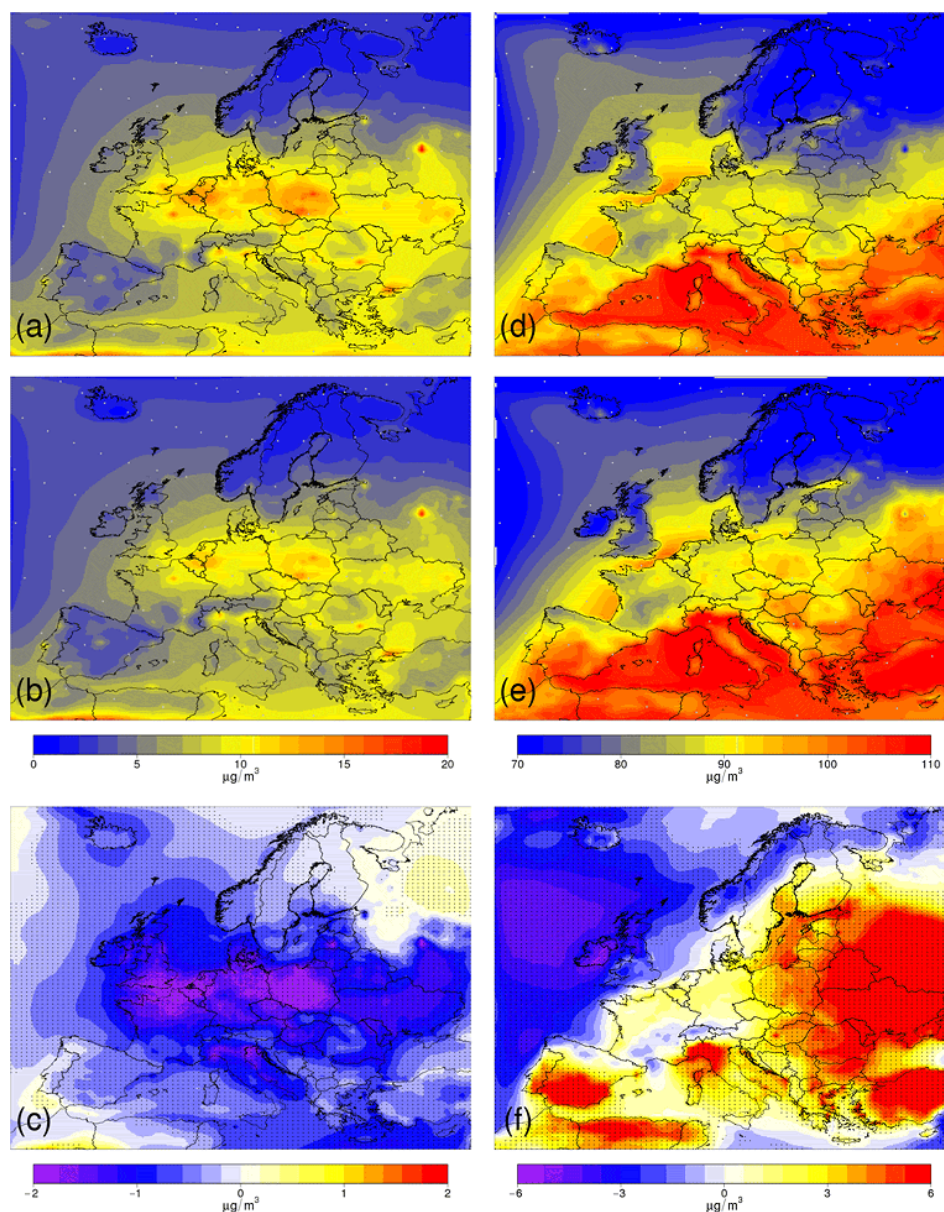
Figure 1. The left column represents daily average $PM_{2.5}$ concentrations for the historical (1976–2005) **(a)**, the end of the century (RCP8.5 – 2071–2100) **(b)** and the difference between the future and the historical **(c)**. The statistical significance of this difference is evaluated by a t test and represented by a black point. The right column presents the same figure for daily maximum ozone projections. For both pollutants, the CTM CHIMERE has been used to predict the concentration (Sect. 2.2).

similar pattern is found in the future (RCP8.5 – average over the period 2071–2100) albeit with lower concentrations (Fig. 1b). The difference (future minus historical) is given in Fig. 1c where the statistical significance of the changes was represented by black points at each grid points and evaluated by a Student t-test with Welch variant at the 95 % confidence level based on annual mean. The decrease is statistically significant over most of the domain.

Overall, we identify a climate benefit on particulate matter pollution similarly to Colette et al. (2013) and Lecœur et al. (2014) but in opposition to Manders et al. (2012). Hedegaard et al. (2013) find a decrease in high latitude and an increase in low latitude. The role of future precipitation projections and more efficient wet scavenging has often been pointed out to explain such a future evolution of particulate matter (Jacob and Winner, 2009). However, the lack of robustness in precipitation evolution over major European particulate pollution hotspots in regional climate models (Jacob et al., 2014) challenges the confidence we can have in single

model air quality and climate projection, supporting again the need for ensemble approaches.

3.1.2   Ozone peaks

Figure 1d represents the summer (JJA) average ozone daily maximum concentrations over the historical period (1976 to 2005). A north–south gradient appears with lower concentration in the north and higher concentration fields over the Mediterranean Sea. Figure 1e corresponds to the summer average ozone projection of the RCP8.5 at the end of the century (2071–2100) predicted by the model suite presented in Sect. 2.2. A similar pattern is found, with higher concentrations in the southern part of the domain (Fig. 1e). The map of the difference (RCP8.5 – actual), Fig. 1f, indicates an increase of ozone concentrations over Eastern Europe, Mediterranean land surfaces, and North Africa and a decrease over British-Irish Isles and Scandinavia. Most of the changes are statistically significant except over Western Europe. This concentration rise is frequently associated to an increase of temperature in the literature (Meleux et al., 2007; Katragkou et al., 2011), see Sect. 2.1 above for a review of physical and chemical processes underlying this association.

Following Langner et al. (2012b), Manders et al. (2012), Colette et al. (2013, 2015) these results confirm the fact that climate change constitutes a penalty for surface ozone in Europe.

3.2   Statistical models

Here we introduce the statistical models trained over the historical period and their evaluation over the future testing period. First we discuss the impact of key meteorological processes on pollutants concentration on the basis of the model correlation and put our results in perspective with the key driving factors reported in the literature. Then we evaluate the performance of statistical models over the future period in order to discard regions and pollutants where the skill of the statistical model is too small to draw robust conclusions on the uncertainties of projections.

3.2.1   Fine particulate matter

The skill and predictors for generalized additive models fitted for each region are given in Table 1. The depth of the planetary boundary layer is identified as the major meteorological driver for $PM_{2.5}$ which is a different finding compared to Megaritis et al. (2014) who reported a smaller impact for the PBL depth. Near-surface temperature is often selected as second predictor. The wind is pointed out as a relevant predictor twice but only for coastal regions (respectively BI and MD) where sea-salt is important. Last, precipitation is selected only once and as 2nd variable for the Iberian Peninsula (IP). It could be partly due to our choice of statistical model whereas a logical regression would have been more efficient given that PM correlations are sensitive

to the presence and/or absence of precipitation rather than their intensity. It is difficult to assess objectively whether the larger role of temperature than precipitation in our findings is an artifact related to the design of the statistical model. The importance of precipitation in the impact of climate change on particulate pollution is often speculated in the literature, with little quantitative evidence. The statistical model used here offers an objective quantification of that role. It should be added that the importance of temperature is well supported by the volatilization process for Secondary Inorganic Aerosol and Secondary Organic Aerosol. Moreover in the CTM CHIMERE, the volatile species in the gas and aerosol phases are assumed to be in chemical equilibrium. This thermodynamic equilibrium, computed by ISORROPIA (Fountoukis and Nenes, 2007), is driven by temperature and humidity and conditions the concentration of several aerosol species (ammonium, sodium, sulphate, nitrate and so on). This feature could explain the major role of temperature. It is also supported by the pattern of projected $PM_{2.5}$ change, which is spatially correlated with present-day $PM_{2.5}$ concentration. This spatial correlation suggests an impact of a uniform driver which points towards temperature rather than precipitation change that exhibits a strong north-south gradient in Europe.

Then the predictive skill of these models is tested over the period 2071–2100 by computing the Normalized Root Mean Squared Error (NRMSE) between the statistically predicted $PM_{2.5}$ (concentrations estimated with the statistical models), and the results of the deterministic regional air quality and climate modelling suite presented in Sect. 2.2 for 2071–2100. The NRMSE is defined as the root mean square error between statistically predicted and deterministically modelled concentrations changes aggregated by region and at daily temporal frequency, normalized by the standard deviation of the deterministic model. It allows describing the predictive power of a model, if the NRMSE is lower or equal to 1 then the model is a better predictor of the data than the data mean (Thunis et al., 2012).

Figure 2 shows, for each region, the scatter between R^2 over the historical period and the NRMSE in predictive mode for the RCP8.5 at the end of the century. We expect regions where the correlation over the historical period is low to be poorly captured by the statistical model in the future. The fact that the good correlation for EA and ME are associated with an NRMSE around 0.6 in the future indicates either that the main meteorological drivers in the future will remain within their range of validity or that extrapolation is a viable approximation. This feature gives confidence in using statistical models for these regions in predictive mode. For the NI region, the NRMSE is acceptable (below 0.8) even if the R^2 is low.

Considering that the model skill was satisfactory for the EA, ME and NI regions, we decided to focus on these regions for the uncertainty assessment in the remainder of this paper. The fine particulate matter concentrations have been poorly

Table 1. Statistical models per region that explain the average $PM_{2.5}$ concentrations during 1976–2005.

PM$_{2.5}$			
Regions	R^2	Meteorological variable 1	Meteorological variable 2
BI	0.327	PBL-height	Surface wind
IP	0.228	PBL-height	Specific humidity
FR	0.343	PBL-height	Near-surface temperature
ME	0.613	PBL-height	Near-surface temperature
SC	0.206	Specific humidity	Incoming short-wave radiation
NI	0.403	PBL-height	Near-surface temperature
MD	0.194	PBL-height	Surface wind
EA	0.595	PBL-height	Near-surface temperature
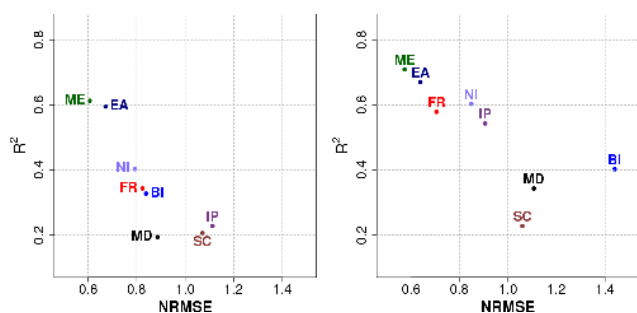
Figure 2. Statistical model evaluation for $PM_{2.5}$ (left) and ozone (right). The x axis represents the Normalized Mean Square Error applied to the delta (future minus historical) of the generalized additive model and CHIMERE. The y axis represents the R^2 of the statistical model (training period).

captured for the region BI, SC, FR, IP and MD. The associated bad NRMSE are explained by the poor performances of model over the historical. They are thus excluded from the uncertainty assessment.

3.2.2 Particulate matter composition

Because total $PM_{2.5}$ is constituted by a mix of various aerosol species, there is a risk of compensation of opposite factors in the statistical model. In order to assess that risk, we developed such models for each individual PM constituent in the chemistry-transport model. The performances of these statistical models in terms of correlation for the historical (training) period or in predictive mode for the future period (testing) are presented in Fig. 3.

For all regions, the statistical models are not able to capture the variability of mineral dust. This is because the design of the statistical model is exclusively local (i.e. average concentrations over a given region are related to average meteorological variables over the same region), whereas most of the mineral dust over any European region is advected from the boundaries of the domain, in North Africa. It should be noted however, that except for the regions IP and MD, the dust represents only a small fraction of the PM concentra-

tions (Fig. 4). That could explain why the statistical model for $PM_{2.5}$ performs poorly over IP and MD, but it will not undermine the confidence we can have in concluding about the robustness of the $PM_{2.5}$ model for the region selected above: ME, EA and NI.

All over Europe, primary particulate matter (PPM) is one of the smallest particulate matter fractions. Their variability is well captured by the statistical model for all regions except SC. But because of their small abundance in that region, they should have a limited impact on the $PM_{2.5}$ model performance.

The sea salts are well reproduced by the statistical model for all regions except NI and EA. These two regions have no maritime area, therefore sea-salt concentrations are lower and exclusively due to advection which, as a non-local factor, is not well captured by the statistical model.

Ammonium (NH_4^+) aerosols are satisfactorily captured by the statistical models for five regions out of eight including those selected for the overall $PM_{2.5}$ model (ME, EA, and NI).

The organic aerosol fraction (ORG) is well reproduced over the historical period and the predictive skill is satisfactory (NRMSE around 0.7) for ME, EA, and NI.

The statistical models are efficient to reproduce the nitrate (NO_3^-) concentrations over the historical period for ME, EA, AL, MD, FR, and BI regions but the predictive skills are only considered satisfactory for ME, EA, FR and NI, where nitrate constitutes a large fraction of $PM_{2.5}$.

Sulphate aerosols (SO_4^{2-}) are well represented by the statistical models for BI, EA, and ME. The performances are low in the NI region, but sulphates constitute one of the smallest particulate matter fractions for that region.

This analysis of the skill of statistical models for each compound of the particulate matter mix confirms that there is no compensation of opposite factors in the selection of skillful models for total $PM_{2.5}$ proposed in Sect. 3.2.1. The only cases were one of the particulate matter compound was not well captured by a statistical model, could be attributed to a low, and often non-local contribution of the relevant particulate matter constituent for the considered regions. We conclude that the selection of ME, EA, and NI as regions where
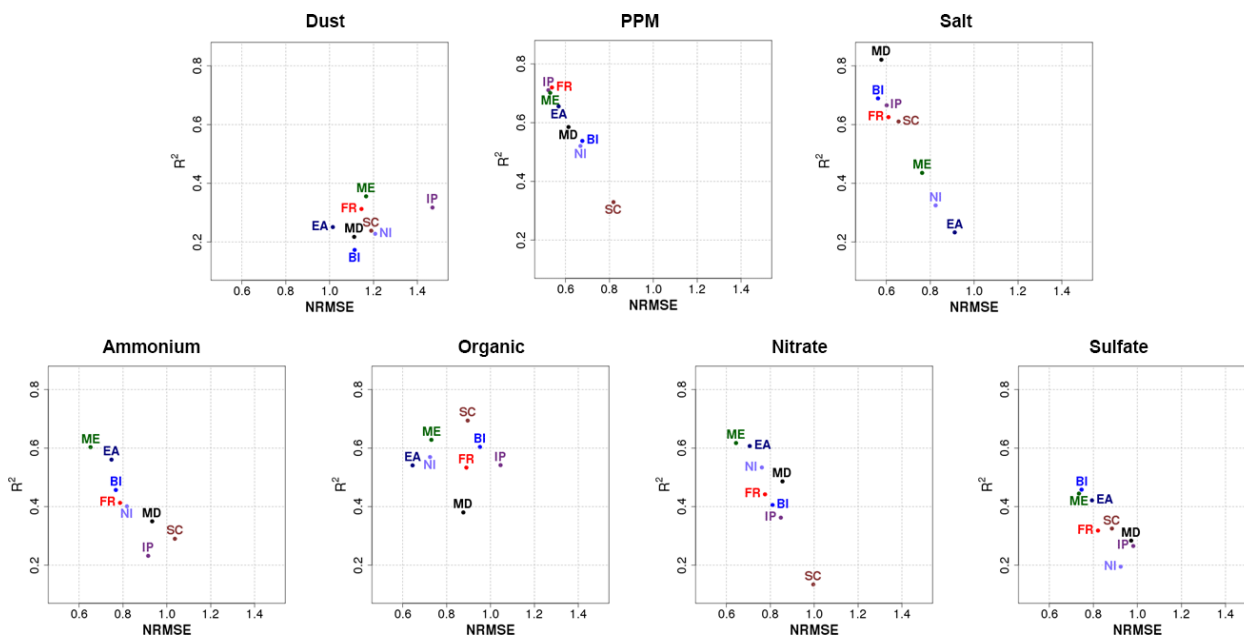
Figure 3. Statistical model evaluation for each particulate matter constituent (from left to right: Dust, Primary Particulate Matter, Sea-salt, Ammonium, Organic fraction, Nitrate, Sulphate). The *x* axis represents the Normalized Mean Square Error applied to the delta (future minus historical) of either the generalized additive model or CHIMERE. The *y* axis represents the R^2 of the statistical model (training period).
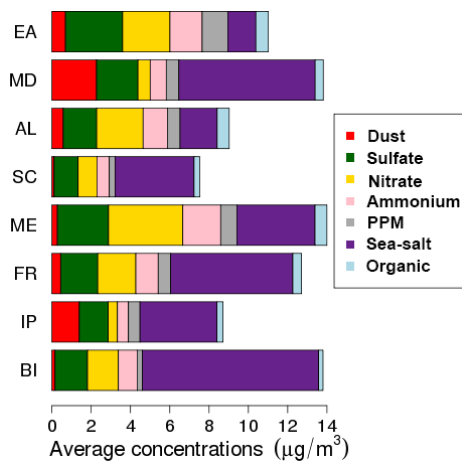
Figure 4. Average particulate matter composition for the historical period per region.

it is possible to build a statistical model of $PM_{2.5}$ variability using Generalized Additive Models based on meteorological predictants would hold if the model had been built for each constituent of the particulate matter mix.

3.2.3 Ozone peaks

For summertime ozone peaks, as expected, near-surface temperature and incoming short-wave radiation are identified as the two main meteorological drivers for most regions (Table 2). Concerning the region EA, the drivers which give

the best results are near-surface temperature and specific humidity. Nevertheless, when using specific humidity as second predictor, the statistical model is overfitted and has a low predictive skill (NRMSE = 0.9). Thus the use of short-wave radiation as second predictor appears much more robust (NRMSE = 0.6) even if the R^2 is lower. The skill of the statistical model is very low over the British-Irish Isles and Scandinavia. This is because ozone pollution in these regions is largely influenced by non-local contributions (long-range transport of air pollution). The poor performances of the statistical model over the Mediterranean region are more surprising. The lower variability of temperature and incoming short-wave radiation in this region compared to other parts of Europe (standard deviation of 12.5 °C and 150 W m^{-2} for MD; from 15 to 20 °C and from 220 to 300 W m^{-2} for the other regions) makes them less relevant as statistical predictants of ozone concentrations.

We conclude that the generalized additive models that can be considered efficient enough in terms of correlation to capture the ozone variability over the historical period are those of the following regions: EA, FR, IP, ME and NI.

This selection is further supported by investigating the predictive skill of the models assessed by computing their NRMSE against deterministic CTM simulations available for a future period. The regions mentioned above where the correlation of the statistical model is low (BI, SC and MD) also exhibit a large NRMSE (Fig. 2). So that, only the regions EA, FR, IP, ME and NI are selected for the remainder of this paper.

Table 2. Statistical models per region that explain the daily maximum summer ozone levels during 1976–2005.

Ozone max			
Regions	R^2	Meteorological variable 1	Meteorological variable 2
BI	0.402	Incoming short-wave radiation	Specific humidity
IP	0.543	Near-surface temperature	Incoming short-wave radiation
FR	0.579	Near-surface temperature	Incoming short-wave radiation
ME	0.709	Near-surface temperature	Incoming short-wave radiation
SC	0.228	Near-surface temperature	PBL-height
NI	0.603	Incoming short-wave radiation	Near-surface temperature
MD	0.343	Near-surface temperature	Surface wind
EA	0.671	Near-surface temperature	Incoming short-wave radiation

4 Exploring the ensemble of climate projections with the statistical model

The statistical models introduced in Sect. 2, developed in Sect. 3 and tested in Sect. 3.2 are applied here to the ensemble of regional climate projections presented in Sect. 2.3 to develop a proxy of ensemble of air quality and climate projections for each selected region. This proxy of ensemble will be used to identify the subset of regional climate projections that should be used in priority in the deterministic modelling suite, but it can also give an indication on the robustness of the climate impact on air quality where the skill of the statistical model is considered satisfactory.

4.1 Fine particulate matter

In order to assess qualitatively the robustness of the evolution of regional climate variables having an impact on air quality, we first design a 2-D parameter space where the isopleths of statistically predicted pollutant concentrations are displayed (background of Fig. 5). Then the distributions of historical and future meteorological variables as extracted from the regional climate projections are added to this parameter space. For each Regional Climate Projection, we show the average of the two driving meteorological variables as well as the 70th percentile of their 2-D-density plot, i.e. the truncation at the 70th quantile of their bi-histogram which means that 70 % of the simulated days lies within the contour. Both historical and future climate projections (here for the RCP8.5 scenario and the 2071–2100 period) are displayed on the parameter space. The climate projections are all centred on the IPSL-CM5A-MR/WRF member so that only the distribution of the latter is shown for the historical period.

As pointed out in Table 1, the main meteorological drivers are the depth of the PBL and near-surface temperature for the example of $PM_{2.5}$ over Eastern Europe region displayed in Fig. 5. The statistically modelled isopleths in the background of the figure show that $PM_{2.5}$ concentration decrease when the depth of the PBL increases (x axis), or when temperatures increase (y axis). The interactions captured by the GAM exhibit the strong influence of high vertical stability events

(with low surface temperature and PBL depth) in increasing $PM_{2.5}$ concentrations. On the contrary, for high temperature ranges, the depth of the PBL becomes a less discriminating factor. The comparison of historical and future distributions shows that both meteorological drivers evolve significantly in statistical terms (Student t test with Welch variant at the 95 % confidence level based on annual mean). However, even though the PBL depth constitutes the most important meteorological driver for $PM_{2.5}$, it does not evolve notably compared to the surface temperature in the future (Fig. 5). Thus the largest increase of the secondary driver (surface temperature) leads to a decrease of $PM_{2.5}$ concentrations. The largest and the smallest $PM_{2.5}$ concentrations decrease are found for CSIRO-Mk3-6-0/RCA4 and MPI-ESM-LR/CCLM, respectively. But the overall spread of RCMs in terms of both the evolution of PBL depth and temperature is limited, suggesting that this climate benefit on particulate pollution is a robust feature. Those isopleths present the same characteristics for ME and NI regions (Figs. S1, S4 in the Supplement). The qualitative evolution represented in Fig. 5 is further quantified by applying the GAM to the future meteorological variables in the regional climate projections. These results are represented by the probability density functions of the predicted concentrations of each GCM/RCM couple minus the estimated values for the historical simulation (e.g. 2071–2100 vs. 1976–2005, Fig. 6). For EA and ME, the longer tail of the probability density function of MPI-ESM-LR/CCLM compared to the average of the models reflects that stronger pollution episodes will occur in the future even if the mean of the concentrations is lower than the average of the ensemble (Fig. 6 for EA and Fig. S2 for ME).

Besides the distribution, the ensemble mean and standard deviation of the estimated projected change in $PM_{2.5}$ concentrations has been quantified (Table 3). All the selected regions depict a significant decrease of the $PM_{2.5}$ concentrations across the multi-model proxy ensemble indicating that according to the GAM model, the climate benefit on particulate matter is a robust feature in these regions. The magnitude of the decrease depends on the region, its ensemble mean (± standard deviation) is −1.08 (±0.21), −1.03

Table 3. Predicted concentrations evolution of summertime ozone and PM$_{2.5}$ (expressed in µg m^{-3}) per selected regions and per model. The ensemble mean and standard deviation are also calculated.

RCP8.5 2071–2100	Delta (future – historical)							
	Ozone max					PM$_{2.5}$		
GCM/RCM\Regions	EA	FR	IP	ME	NI	EA	ME	NI
CNRM-CM5-LR/RCA4	8.00	6.96	9.75	4.82	3.69	−0.77	−0.82	−0.71
CSIRO-Mk3-6−0/RCA4	11.26	16.03	13.30	14.15	5.81	−1.39	−1.72	−1.06
CanESM2/RCA4	17.97	19.03	15.07	21.20	7.46	−1.29	−1.56	−1.03
EC-EARTH/RACMO2	7.77	11.37	10.79	8.55	6.77	−1.16	−0.98	−0.77
EC-EARTH/RCA4	10.88	14.43	11.45	12.11	5.15	−0.92	−0.92	−0.75
GFDL-ESM2M/RCA4	7.26	7.79	10.28	5.85	4.54	−1.04	−0.90	−0.70
IPSL-CM5A-MR/RCA4	13.76	13.46	12.88	11.02	4.43	−1.28	−1.12	−1.04
IPSL-CM5A-MR/WRF	10.11	6.05	9.08	5.19	0.01	−1.32	−1.30	−0.86
MIROC5/RCA4	12.30	11.29	11.61	9.62	3.85	−1.16	−0.86	−0.85
MPI-ESM-LR/CCLM	6.40	9.63	11.03	6.01	5.58	−0.81	−0.58	−0.62
MPI-ESM-LR/RCA4	9.56	11.75	11.51	9.64	5.54	−1.02	−0.79	−0.83
NorESM1-M/RCA4	10.88	12.60	11.58	10.12	5.02	−0.79	−0.88	−0.76
Ensemble Mean	10.51	11.70	11.53	9.86	4.82	−1.08	−1.03	−0.83
Ensemble Standard Deviation	3.06	3.63	1.55	4.41	1.79	0.21	0.32	0.14
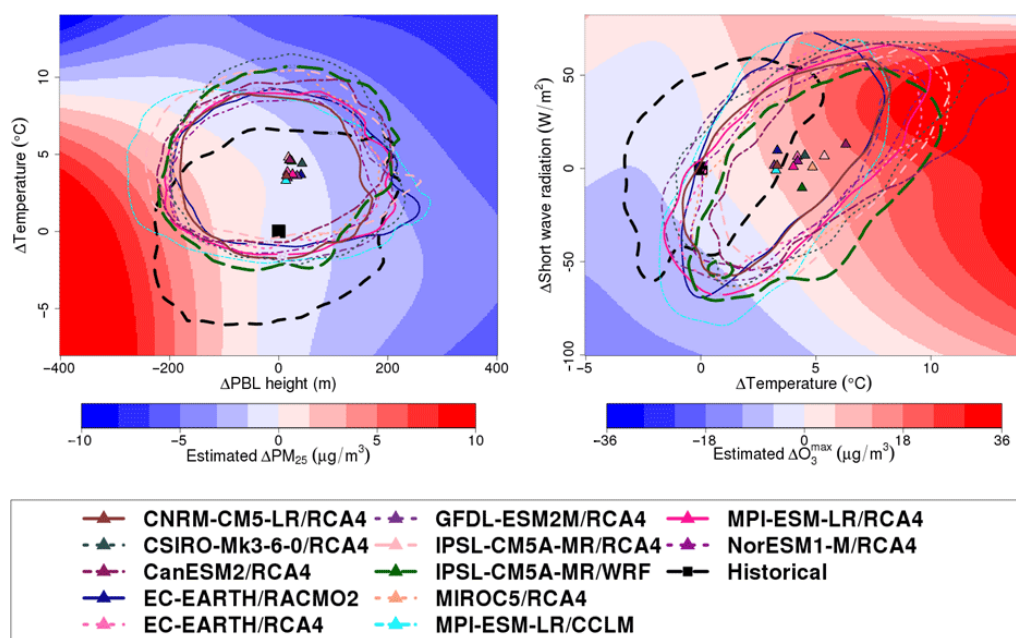
Figure 5. The left figure presents the proxy of ensemble projections for daily average de-seasonalized PM$_{2.5}$ concentrations in Eastern Europe. The right figure represents the proxy for daily maximum de-seasonalized summer ozone for Eastern Europe. For both figures, the shaded background represents the evolution of pollutants estimated by the statistical models. The contours are representing the regional climate projections and the triangles their mean. The black dashed contour represents the historical – IPSL-CM5A-MR/WRF – and the square its mean.

(± 0.32), $-0.83 \pm (0.14)\,\mu g\,m^{-3}$, for respectively EA, ME and NI (Table 3).

In order to explain the differences in the response of individual RCM in the ensemble, we need to explore the historical meteorological variables probability density functions (PDF, Fig. 7) and to compare them with the evolution of

IPSL-CM5A-MR/WRF (Fig. 7). The comparison of the historical distribution for the temperature reflects the stronger extremes of IPSL-CM5A-MR/WRF (e.g. colder than the others when it is cold). It is only for the NI region that IPSL-CM5A-MR/WRF lies in the mean of the ensemble. Concerning the PBL depth, the values are similar to the aver-
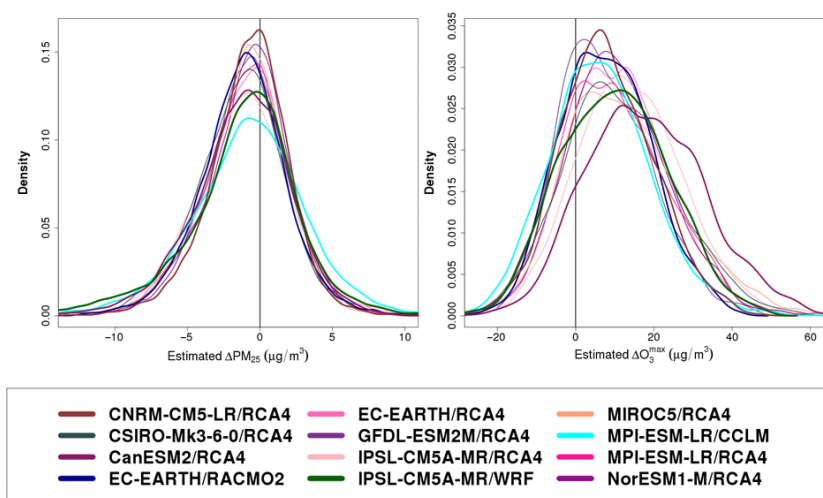
Figure 6. The left figure represents, for each regional climate model the probability density function (PDF) of the concentrations estimated with the generalized additive model at the end of the century minus the estimated concentrations of the historical period for daily average de-seasonalized PM$_{2.5}$ concentrations in Eastern Europe. The right figure presents the results for daily maximum de-seasonalized summer ozone for Eastern Europe.

age of the ensemble for ME even if MPI-ESM-LR/RCA4 and EC-EARTH/RACMO2 present the largest values. IPSL-CM5A-MR/WRF has a thinner boundary layer for NI and a deeper than the average for EA but the differences are limited Fig. 7).

It is for CSIRO-Mk3-6-0/RCA4 that we find the most important decrease of PM$_{2.5}$ for the selected regions (Table 3). This is related to a larger temperature rise compared to the other models and a larger boundary layer height increase compared to the other member of the ensemble for these regions Fig. 5). CanESM2/RCA4 and CSIRO-Mk3-6-0/RCA4 exhibit the same features for the ME region.

MPI-ESM-LR/CCLM presents the smallest decrease of PM$_{2.5}$ for each of the selected regions (e.g. over ME is almost 3 times smaller than the largest decrease) except EA where CNRM-CM5-LR/RCA4 presents a smaller decrease ($-0.77\,\mu g\,m^{-3}$ vs. $-0.81\,\mu g\,m^{-3}$). As already mentioned above, the particular tails of the statistically modelled PM$_{2.5}$ distributions for EA and ME indicate a larger contribution of large pollution episodes in the future for that RCM. But the historical distributions exhibit a larger boundary layer than the average models of the ensemble and a similar temperature. Thus, the low PM$_{2.5}$ concentration decrease is explained by the limited average evolution of the meteorological drivers as shown in Fig. 5.

Overall we conclude that a climate benefit is identified for the PM$_{2.5}$ for each of the selected regions. To the extent that the statistical model is skillful, as demonstrated in Sect. 3.2.1, this result is robust across the range of available climate forcings since the whole ensemble of regional climate projection present consistent features. The regional climate models that exhibit the largest and smallest responses are CanESM2/RCA4; CSIRO-Mk3-6-0/RCA4 and

MPI-ESM-LR/CCLM, which should therefore be considered a priority for further evaluation using explicit deterministic projections involving full-frame regional climate and chemistry models.

4.2 Ozone peaks

For most of the selected regions (FR, IP, ME and NI), the main drivers are the same (i.e. near-surface temperature and short-wave radiation). The isopleth in the background of Fig. 5 show that temperature and short-wave radiation have a similar impact on ozone peaks, except in the larger range of short-wave radiation anomalies, where temperature becomes less discriminating. All the isopleths (Fig. 5 for EA and Figs. S1, S4 and S7 for ME, NI, FR and IP) exhibit an increase in the distribution of temperatures because the projected future is warmer than the historical period. According to the ozone peak concentrations predicted by the GAM (displayed in the background of Fig. 5) these increases will lead to more ozone episodes. This trend appears for the entire models ensemble so that we can conclude with confidence that the climate penalty bearing upon ozone is a robust feature even if the specific distribution of some of the models stand out (CanESM2/RCA4; CNRM-CM5-LR/RCA4; CSIRO-Mk3-6-0/RCA4; IPSL-CM5A-MR/WRF).

The ozone increase of the ensemble reaches $+10.51$ (± 3.06), $+11.70$ (± 3.63), $+11.53$ (± 1.55), $+9.86$ (± 4.41), $+4.82$ (± 1.79) $\mu g\,m^{-3}$ for EA, FR, IP, ME and NI (Table 3). These values confirm the statistically significant climate penalty (the mean is at least two times larger than the standard deviation). However, as already mentioned for Fig. 5, we find minor differences among the models. The meteorological distributions are marginally different between
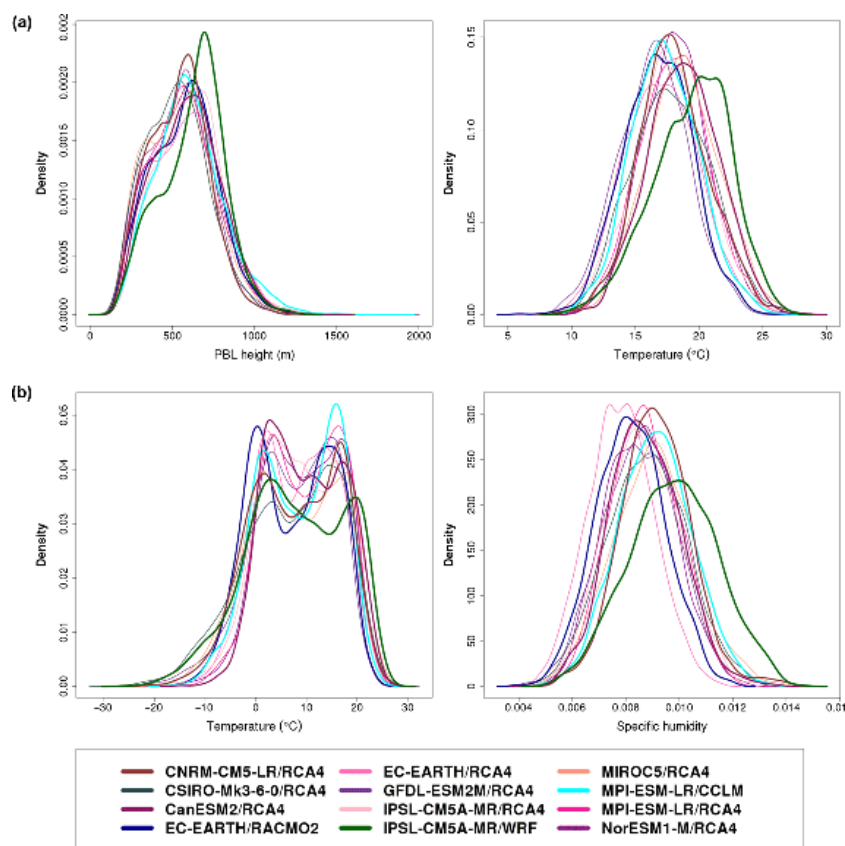
Figure 7. The first column of the panel represents the historical distribution of the meteorological variables identified by our statistical models as the two major drivers (**a** PBL Height; **b** near-surface temperature) for $PM_{2.5}$ in Eastern Europe. The second column represents the historical JJA distribution of the two main drivers for summer ozone (**a** near-surface temperature; **b** incoming short-wave radiation).

the models of the ensemble: the summertime temperature predicted by IPSL-CM5A-MR/WRF has stronger extremes than the other models. Moreover, it is warmer than the ensemble in EA. Concerning incoming short-wave radiation, IPSL-CM5A-MR/WRF lies in the average (Figs. S3, S6, S9) except for the region EA where the amount of incoming radiation is the highest among the ensemble (Fig. 7). Note that, only EC-EARTH/RACMO2 and MPI-ESM-LR/RCA4 exhibits lower values (around half of the average for MPI-ESM-LR/CCLM). The lower amount of summertime incoming short-wave radiation for the couple MPI-ESM-LR/CCLM is relevant for all the selected regions.

The magnitude of the ozone rise differs between the models and the regions. Note that CanESM2/RCA4 exhibits the largest difference (i.e. around 1.5 times the ensemble mean) followed by CSIRO-Mk3-6-0/RCA4 for each selected regions. This is explained by the larger temperature increase during summertime which is the major driver, as identified by the statistical models, of ozone concentration. Note that the value is skyrocketing for the region ME, 5 times the value of IPSL-CM5A-MR/WRF which shows one of the lowest increases. CNRM-CM5-LR/RCA4 presents the lowest increase.

On the contrary, the lower increase of the summer temperature and sometimes a decrease of the incoming short-wave radiation amount (e.g. IPSL-CM5A-MR/WRF in NI) lead to lower ozone concentration changes for IPSL-CM5A-MR/WRF and CNRM-CM5-LR/RCA4 for FR, IP, ME and NI (Table 3). Note the specific evolution for the region NI, where the IPSL-CM5A-MR/WRF model yields almost no increase of the ozone concentration compared to the other models while on the map of the differences in the deterministic model (Fig. 1f), the evolution was statistically significant. This absence of evolution reflects the limitation of the statistical models.

In Fig. S5, we can point out an outstanding pattern of the MPI-ESM-LR/CCLM distribution for the NI region with particularly large tails. The ozone rise would be more pronounced for the upper quantile which depicts more extreme ozone pollution episodes (note that this was also the case for that model in terms of $PM_{2.5}$ pollution).

Overall the climate penalty is confirmed even if some regional climate models stand out of the distribution, such as CanESM2/RCA4; CNRM-CM5-LR/RCA4 and CSIRO-Mk3-6-0/RCA4 which should therefore be considered for further deterministic projections.

5 Conclusions

An alternative technique to assess the robustness of projections of the impact of climate change on air quality has been introduced. Using a training data set consisting of long-term deterministic regional climate and air quality projections, we could build statistical models of the response of ozone and particulate pollution to the main meteorological drivers for several regions of Europe. Applying such statistical models to an ensemble of regional climate projections leads to the development of an ensemble of proxy projections of air quality under various future climate forcings. The assessment of the spread of the ensemble of proxy projections allows inferring the robustness of the impact of climate change, as well as selecting a subset of climate models to be used in priority for future explicit air quality projections, therefore proposing an optimized exploration of the ensemble.

The main meteorological drivers that were identified are (i) for $PM_{2.5}$: the boundary layer depth and the near-surface temperature and (ii) for ozone: the near-surface temperature and the incoming short-wave radiation. The skill of the statistical models depends on the regions of Europe and the pollutant.

For $PM_{2.5}$ and the regions Eastern Europe (EA) and Mid-Europe (ME), a generalized additive model captures about 60 % of the variance and 40 % for Northern Italy. But for the British-Irish Isles (BI) and Scandinavia (SC), where air pollution is largely driven by long-range transport, such a local approach is not able to reproduce the variability of pollutant concentrations.

The ozone concentrations are well reproduced by the statistical model for the following regions: Eastern Europe (EA), France (FR), Iberian Peninsula (IP), Mid-Europe (ME) and Northern Italy (NI). The meteorological variables are not discriminating enough for the Mediterranean region. For the regions where the performances of the statistical model were considered satisfactory, a proxy of the future pollutant concentrations could be estimated (i.e. **(i)**. EA, ME, and NI; **(ii)**. EA, FR, IP, ME and NI).

An overall climate benefit for $PM_{2.5}$ was found in the proxy ensemble of climate and air quality projections. The ensemble mean change is -1.08 (± 0.21), -1.03 (± 0.32), $-0.83 \pm (0.14)\,\mu g\,m^{-3}$, for EA, ME and NI, respectively. This beneficial impact of climate change for particulate matter pollution is in agreement with the deterministic projections of Huszar et al. (2011), Juda-Rezler et al. (2012) and Colette et al. (2013) but in opposition to Manders et al. (2012). These differences could be partly explained by the different time windows (i.e. 2060–2041 vs. 2100–2071), climate scenario (i.e. A1B which is similar to RCP6.0 vs. RCP8.5), and pollutant (i.e. PM_{10} vs. $PM_{2.5}$). This impact of climate change on particulate pollution should be put in perspective with the magnitude of the change that is expected from the current air quality legislation. Such a comparison was performed by Colette et al. (2013) who found (on av-erage over Europe) a climate benefit by the middle of the century of the order of 0–1 $\mu g\,m^{-3}$, therefore in line with our estimate but also much lower than the expected reduction of 7–8 $\mu g\,m^{-3}$ that they attributed to air quality policies.

For all the selected regions a robust climate penalty on ozone was identified: $+10.51$ (± 3.06), $+11.70$ (± 3.63), $+11.53$ (± 1.55), $+9.86$ (± 4.41), $+4.82$ (± 1.79) $\mu g\,m^{-3}$ for EA, FR, IP, ME, and NI, respectively. This finding is in line with previous studies (Meleux et al., 2007; Huszar et al., 2011; Katragkou et al., 2011; Jiménez-Guerrero et al., 2012; Juda-Rezler et al., 2012; Langner et al., 2012a, b; Colette et al., 2013, 2015; Hedegaard et al., 2013; Varotsos et al., 2013). It should be noted that when comparing the impact of climate change and emission reduction strategies, Colette et al. (2013) found a climate penalty of the order of 2–3 $\mu g\,m^{-3}$ (which is broadly consistent with our results given that they focused on the middle of the century) that could be compensated with the expected magnitude of the reduction of 5–10 $\mu g\,m^{-3}$ brought about by air quality policies.

The major strength of our approach is to account for the climate uncertainty in the recent EuroCordex ensemble of regional climate projections, whereas all the published literature relied on a very limited subset of RCM forcing (at best two for a given chemistry-transport modelling study). We therefore propose an unprecedented view in the robustness of the impact of climate change on air quality across an ensemble of climate forcing. However, this achievement is limited by the quality of the underlying statistical model that does not capture all the variance of the air quality response to climate change. These results should thus be ultimately compared with further deterministic projections using a range of climate forcings. Then, our approach can yield precious information in pointing out which regional climate models should be investigated in priority, therefore proposing a smart exploration of the ensemble of projections. The following models: CanESM2/RCA4; CNRM-CM5-LR/RCA4 and CSIRO-Mk3-6-0/RCA4 and MPI-ESM-LR/CCLM, have been identified as the climate models that should be used in priority for future air quality.

Finally, we should add that the method applied here for air quality projection also opens the way for other climate impact studies, where quantifying uncertainties using low computational demand is desirable.

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