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Impact of emissions and +2 °C climate change upon future ozone and nitrogen dioxide over Europe

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HIGHLIGHTS

- The impacts of a +2 °C climate change upon future O_3 and NO_2 were studied.
- Long (30-year) simulations were performed with a set of 4 chemical transport models.
- Unique approach: future time period centred on +2 °C warming, rather than fixed years.
- Reductions of anthropogenic emissions significantly improved future air quality.
- Isolated impacts of climate change upon O_3 were not statistically significant.

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ABSTRACT

The evolution of ozone and nitrogen dioxide over Europe between the present day and a future period with a +2 °C global warming relative to the pre-industrial climate was studied using four offline chemistry transport models, each driven by a different climate model. Given the recent outcome of the COP21 negotiations, understanding the implications of climate change around the +2 °C threshold has never been more pressing or relevant. One of the objectives of this study was to show how changes in anthropogenic emissions and +2 °C climate change are expected to affect future air quality, which may have important implications upon human health. It was found that a +2 °C climate change alone was responsible for a modest, and not statistically significant, increase in surface O_3 concentrations (of between −0.1–0.8 ppb in the summer averaged over the European domain) compared to the present climate. Two different emission scenarios were used for the future time period in order to provide an estimate of the extent of air pollution reductions that could occur if (a) all currently planned air quality legislation is implemented and (b) all maximum technologically feasible emission reductions are implemented. The results showed that summer O_3 could be reduced by between 4 and 5 ppb under a current legislation scenario, with at least 3 ppb of further reductions under the maximum mitigated scenario. Calculations of summer ozone enhancement were used as a metric to analyse the results after having removed background ozone level changes. In conclusion it was found that future air quality on a regional scale will depend upon the implementation of effective emission reduction policy; the positive effects of which should not be hindered by a +2 °C global warming.

1. Introduction

It is well-recognised that air pollutants, such as particulate matter (PM) and ozone (O_3), pose risks to human health when

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present in concentrations above specified target values (Guerreiro et al., 2014). The evolution of air pollutant concentrations over the next few decades will depend upon many factors, such as population and economic growth (Parrish et al., 2011); controls upon anthropogenic emissions (Butler et al., 2012); changes in biogenic emissions, precipitation and cloud cover; and the severity of climate change events, such as more frequent heat waves.

This paper focuses on the evolution of tropospheric O_3 under climate and emission changes, with PM discussed in a related paper (Lacressonnière et al., 2016b). O_3 is a secondary pollutant formed by the reactions of nitrogen oxides (NO_x) with carbon monoxide, methane (CH_4) and volatile organic compounds (VOCs) (Monks et al., 2009). The concentrations of O_3 in the planetary boundary layer are affected by meteorological factors such as precipitation, wind speed and direction, mixing height, relative humidity, and temperature. It is, therefore, reasonable to assume that climate change will affect the distribution and deposition of this air pollutant (Kinney, 2008; Jacob and Winner, 2009).

O_3 production in the atmosphere is correlated with temperature, so the increased temperatures associated with climate change would logically be expected to coincide with increased future O_3 concentrations (Jacob and Winner, 2009); however the relationship between O_3 and temperature is complex and depends upon many factors, hence the need to investigate the effects of climate change on O_3 using comprehensive models. The link between tropospheric O_3 and climate change in Europe has already been extensively investigated using regional chemical transport models (CTMs) (Andersson and Engardt, 2010; Langner et al., 2012; Colette et al., 2013; Lacressonnière et al., 2014). For example, Langner et al. (2012) compared the results of four offline CTMs and one online integrated chemistry-climate model (CCM) for simulations of the present climate (2000–2009) and a selected future period (2040–2049), using constant anthropogenic emissions for both time periods in order to isolate the impacts of climate change under an A1B climate scenario from the Special Report on Emissions Scenarios (SRES). They discussed how climate change impacts upon O_3 would counteract, at least in part, future reductions in anthropogenic emissions, particularly in the summer in southern Europe. Similar conclusions have been obtained for regional studies performed in other parts of the world, such as Penrod et al. (2014), who showed that although decreases in NO_x emissions in the future lead to reductions in summer O_3 over most of the US, increased future temperatures and associated increases in biogenic emissions can also lead to increases in wintertime O_3 of up to 5 ppb in the eastern US.

One of the key aims of the present study is to expand upon the research that has already been done with regards to the relative effects of climate change and emission reduction policies upon future air quality. But unlike previous work, this study, which was performed as part of the EU FP7 project IMPACT2C (http://impact2c.hzg.de/), is unique due to the fact that the future time period for each CTM is centred on the 30-year period when global average warming reaches +2 °C above pre-industrial times, rather than using the same fixed future time period for all models. This is an important distinction, as climate models currently exhibit a large variability when attempting to determine the time period when global temperatures will reach a certain level (Vautard et al., 2014). The +2 °C period is currently of particular relevance because countries agreed to stabilise global climate warming below this level in the December 2015 Paris agreement. The work presented in this paper also differs from previous studies because it uses long (30-year) simulations, which, particularly in the case of O_3, reduce the uncertainty of the results by accounting for long-term decadal variability (Lacressonnière et al., 2016a). This study is also unique in the fact that it uses four different CTMs, each driven by a different climate model. Although each model is designed to simulate a +2 °C world, the geographical distribution of temperature and other climate parameters may vary significantly between the four models. With 4 models, 30-year-long simulations and 4 scenarios, this paper presents robust results from 480 years of CTM modelling.

This paper follows on from a previous paper that assessed how climate model output modifies the results of an ensemble of four CTMs (Watson et al., 2015). Watson et al. (2015) showed that when the climate models were forced on the boundaries of the European domain by best available reanalysis data, all four of the CTMs used in this study were shown to overestimate observations of O_3 (annual mean biases ranged between 0.7 and 6.6 ppb) and three out of the four underestimated observed annual nitrogen dioxide (NO_2) (mean biases ranged between −3.1 and −5.2 ppb). In addition, Watson et al. (2015) showed that the use of climate models to force the GCMs, instead of using reanalysis data, introduced an additional bias to the O_3 and NO_2 results, but that this bias tended not to be significant in the majority of cases. Similar biases are therefore expected in the future results presented in this paper.

A description of the models used in this study, the emissions data and the types of simulations performed is provided in Section 2. The results for O_3 and NO_2 under a future +2 °C climate change scenario are discussed in Section 3. Section 4 explores the effects of climate change under future scenarios with fixed emissions, and Section 5 looks at the results of a highly mitigated emission scenario. Section 6 presents the O_3 summer enhancement for the climate change and emission scenarios discussed in this paper, and conclusions are then presented in Section 7. Results are presented throughout this paper for the summer (June, July and August) and winter (December, January and February) seasons. Annual results are provided in the supplemental materials to this article.

2. Methodology

2.1. Models

Here we use an ensemble approach with four CTMs: CHIMERE (IPSL), EMEP MSC-W (version rv4.4, hereafter 'EMEP', MET.NO), MATCH (SMHI), and MOCAGE (Météo-France). All four CTMs have already been extensively validated in previous studies (Schulz et al., 2013; Terrenoire et al., 2012; Solazzo et al., 2012a,b; Andersson and Engardt, 2010; Bousserez et al., 2007) and are described briefly in a previous paper (Watson et al., 2015). A summary of the CTMs used, their driving global climate models (GCMs) and their boundary conditions is shown in Table 1. Except in the case of MOCAGE, the driving GCM data for each CTM is dynamically downscaled with a regional climate model (RCM). MOCAGE does not require downscaled data, because it uses GCM output in a 50 km resolution across the globe to force a global simulation with a 2° × 2° horizontal resolution, which is then coupled with 2-way nesting to a regional domain with a 50 km resolution. Therefore, the chemical boundary conditions for MOCAGE also come from its global simulation, which vary from year to year. For the other three CTMs, the lateral boundary conditions are downscaled from LMDz-INCA with a resolution of 3.75° longitude by 1.9° latitude (Hauglustaine et al., 2004; Folberth et al., 2006; Szopa et al., 2013). The monthly mean climatologies from LMDz-INCA are averaged over 11 years, and the chemical boundary conditions include O_3, NO, NO_2, nitric acid, peroxyacetyl nitrate, hydrogen peroxide, CO, CH_4, formaldehyde, ethane, alkanes, ethene, propene and aromatics. The climatologies are centred on 2006 for the present and 2050 for the future, using the RCP 4.5 emissions and global climate scenario as described in Szopa et al. (2013). The global surface ozone change corresponds to a 6 ppb decrease in the future climatology compared to the present day. The tropospheric and surface O_3 changes simulated by LMDz-

Table 1
Description of the model chains used, from GCM simulations to regional CTMs.

Institute	Driving GCM	RCM used for downscaling	Lateral boundary conditions	CTM
CNRS-IPSL	IPSL-CM5A-MR	WRF	LMDz-INCA	CHIMERE
MET.NO	NorESM	WRF	LMDz-INCA	EMEP
SMHI	EC-EARTH	RCA4	LMDz-INCA	MATCH
Météo-France	ARPEGE-Climat	N/A	MOCAGE (global)	MOCAGE

N/A: Not applicable.

INCA over the 21st century are close to the multi-model ensemble mean in the ACCMIP intercomparison project (Young et al., 2013).

All CTMs simulate comprehensive atmospheric chemistry schemes and processes. The model results were provided for a European domain with a 50 km horizontal resolution in accordance with the EURO-CORDEX domain, but the vertical resolutions of the models vary between 8 and 47 layers. The lowest model layer ranges from 25 to 90 m. MOCAGE is the only model that explicitly includes a complete scheme to describe the physical and chemical processes in the lower stratosphere; the other three models rely on top boundary conditions coupled with transport processes to account for stratospheric influence. In order to reflect the uncertainty in our current understanding of atmospheric processes, each team was allowed to select their own input parameters (such as boundary conditions, vertical resolution, convection and advection schemes, biogenic emissions and deposition velocities). Further information about the models can be found in Watson et al. (2015). For post-processing, a masked domain was chosen to represent the European land mass, with oceans and the majority of north Africa excluded (Fig. 1).

2.2. Emissions and simulations

Representative Concentration Pathway (RCP) 4.5 was chosen for the climate model runs that drive the CTMs as it is a scenario in which greenhouse gas (GHG) emissions stabilize after mid-century with temperatures close to, but slightly above, +2 °C above pre-industrial times, which was decided to be the most appropriate RCP for the purposes of the IMPACT2C project. Although RCP 4.5 was used for the GHG emissions in the GCM runs, the emissions of anthropogenic air pollutants (ammonia, carbon monoxide, oxides of sulphur and nitrogen, PM and VOCs) used in the CTMs in this study come from the ECLIPSE project (http://eclipse.nilu.no/), not from RCP emissions. ECLIPSE is a EU FP7 collaborative project, in the frame of which the International Institute for Applied Systems Analysis (IIASA) has provided future emission data sets using the GAINS model (Amann et al., 2011). ECLIPSE v4a emissions were chosen for this study because data covering the EURO-CORDEX domain at a resolution of 0.5° × 0.5° were available for both 2005 and 2050 (corresponding approximately with the present-day and the period of +2 °C warming), with two different emission scenarios for the future (e.g. with and without stringent air quality legislation). The ECLIPSE emissions were developed using very different assumptions about air quality legislation and they are considered to be more appropriate for air quality modelling than the RCP emissions. The ECLIPSE emissions are based on an energy scenario close to RCP 6.0 and RCP 4.5, which both have very similar radiative forcing for the year 2050 and are therefore coherent with the GCM runs.

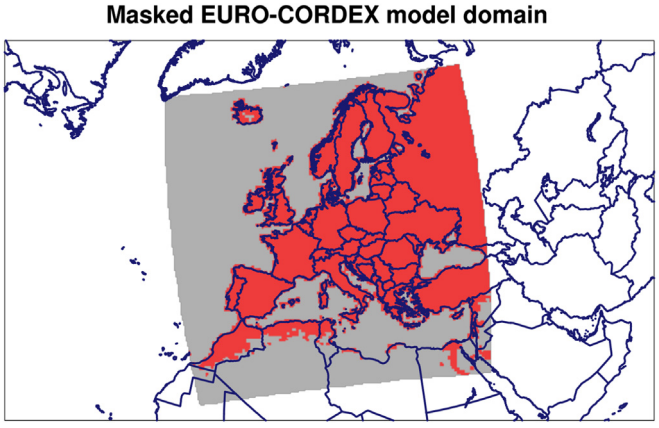

Fig. 1. The red area shows the part of the EURO-CORDEX domain that was used for calculations of surface NO_2 and O_3, with oceans and the majority of north Africa excluded (in grey). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Even though air pollutant emissions vary from year to year, fixed 2005 emissions were used for the present-day simulations and fixed 2050 emissions were used for the future simulations in order to remove inter-annual emission variations and simplify the interpretation of the results.

This paper will discuss the results of four different simulations, as presented in Table 2. The control simulation, HISTORICAL, uses current (2005) emissions for a 30-year simulation that is forced by GCM output for the control period (1971–2000), which already corresponds with approximately 0.5 °C of warming since pre-industrial times. The HISTORICAL simulation is used as a reference for the future scenarios and has been previously evaluated in Watson et al. (2015). The standard future scenario is called S1, which consists of a run forced by GCM output for the 30-year period when global average warming under the RCP 4.5 scenario reaches +2 °C with respect to the pre-industrial. 30-year running means were calculated for each GCM, and then the +2 °C period was defined as the first 30-year period for which the global 2-m temperature difference with respect to the 1971–2000 period was 1.54 °C (assuming that 0.46 °C is the 2-m temperature difference with respect to pre-industrial times, as found in observations) (Vautard et al., 2014). This +2 °C time period is different for each GCM (Vautard et al., 2014). For the IPSL GCM, the +2 °C time period is 2027–2056; while for EC-EARTH (SMHI), the time period is 2041–2070. NorESM (MET.NO) reaches the +2 °C threshold during the 2056–2085 time period, and ARPEGE (Météo-France) reaches this threshold during 2038–2067. All four models were also run for a common 30-year period centred on 2050, but those results are not shown in this paper. The fixed +2 °C runs show less uncertainty for temperature than the common period runs (Vautard et al., 2014). Analysis of the common period shows how air quality will change by 2050 as a result of climate change, taking into account the uncertainty in predicted warmings for that period. This is a valid question, but it is not the objective of IMPACT2C, and not of this paper either. Simulation S1 uses ECLIPSE v4a emissions for 2050 under the current legislation emission (CLE) scenario, which assumes full implementation of all currently planned emission reduction policy. The CLE emission scenario shows reductions in all anthropogenic air pollutants in comparison with 2005 emissions, aside from NH_3 (Fig. 2), whose emissions are linked to agricultural

Table 2
Description of the simulations performed.

Name	Regional climate	Climate of boundary conditions	Emissions
HISTORICAL	1971–2000	1971–2000	ECLIPSE v4a 2005
S1	+2 °C RCP4.5	+2 °C RCP4.5	ECLIPSE v4a 2050 CLE
S2	1971–2000	+2 °C RCP4.5 (1971–2000 for MOCAGE)	ECLIPSE v4a 2050 CLE
S3	+2 °C RCP4.5	+2 °C RCP4.5	ECLIPSE v4a 2050 MFR

activities and evaporation from sources such as animal manure (Simpson et al., 2014). Simulation S2 also uses ECLIPSE v4a CLE emissions for 2050, but with GCM and RCM forcing from the control period (1971–2000). The difference between S2 and S1 provides a way of studying the impacts of climate change in the absence of changes in emissions. Finally, in order to study the magnitude of future air quality improvements that may be obtained with ambitious anthropogenic emission reductions, simulation S3 uses ECLIPSE v4a 2050 emissions under a maximum feasible reduction (MFR) scenario, which assumes all technically feasible emission reductions will be implemented, regardless of cost. The MFR emission scenario foresees very large reductions in air pollutant emissions with respect to 2005 (Fig. 2).

ECLIPSE emissions for 2050 were used in the preparation of boundary conditions for scenarios S1, S2 and S3 for all models. For CHIMERE, EMEP and MATCH future climate conditions were also used to prepare the boundary conditions for these three simulations. But due to its different set-up, boundary conditions for MOCAGE in the S2 simulation came from a global model run using 2050 emissions with past (1971–2000) meteorological forcing (Table 2). Hence, the difference between S2 and S1 for MOCAGE represents the impacts of a global +2 °C warming above the pre-industrial climate in comparison to current climatic conditions. For the other three models, the difference between S2 and S1 represents the impacts of regional climate change in the EURO-CORDEX domain in isolation from the rest of the globe.

In terms of CH_4, MOCAGE used RCP 4.5 emissions fixed at the surface as zonal means across the globe and adjusted to the appropriate year. For the other three models, CH_4 concentrations at the lateral boundaries of the European domain came from the LMDz-INCA model. For EMEP, CH_4 within the modelling domain was set at observed 2005 levels for all simulations. MATCH and CHIMERE used LMDz-INCA CH_4 concentrations throughout their model domains, with future concentrations around 5% higher than the concentrations used for the present day. Under RCP 4.5, an increase in CH_4 concentrations of roughly 5% between 2005 and 2050 would translate into an ozone increase of approximately 0.25 ppb in Europe from 2005 to 2050, when using the formula from Wild et al. (2012). The individual contribution of CH_4 to O_3 in this study cannot be quantified without performing additional runs, but it would be reasonable to assume that future CH_4 in CHIMERE, MATCH and MOCAGE would serve to increase O_3 by a small amount. As noted in Young et al. (2013), future CH_4 has a relatively small impact on future O_3, apart from in the case of RCP 8.5 where CH_4 is projected to double by the year 2050.

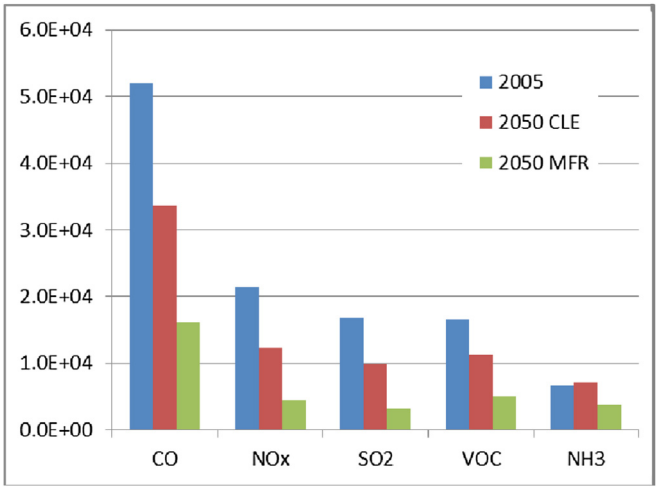

Fig. 2. ECLIPSE v4a emissions in Gg(species)/yr for the present-day (2005), future CLE (2050), and future MFR (2050) scenarios integrated over the whole EURO-CORDEX domain.

3. Future results (scenario S1)

Fig. 3 shows the surface NO_2 and O_3 concentrations for the future CLE scenario (S1), averaged over each model's +2 °C time period for the summer (June, July and August) and winter (December, January and February) seasons, respectively. The plots show that NO_2 concentrations are higher in the winter months than in the summer months, which is consistent with NO_2's observed seasonal variation. CHIMERE, EMEP, and MOCAGE have very similar average concentrations of NO_2 in the summer (between 0.7 and 0.8 ppb averaged over the masked European land domain), while the average concentration simulated by MATCH is much lower (0.4 ppb). In the winter, MOCAGE shows much higher concentrations of NO_2 (3.1 ppb on average) than the other three models (between 1.1 and 1.7 ppb on average). These results are consistent with what was seen in the HISTORICAL results in Watson et al. (2015), where MATCH experienced the lowest levels of NO_2 in the summer and MOCAGE experienced the highest levels of NO_2 in the winter. This inter-model variability is linked to numerous differences in the chemical and/or physical processes between the models. For example, MOCAGE has a lower planetary boundary layer height than the other three models, which may help to explain why its values of NO_2 are more concentrated at the surface in the winter. Due to the fact that in Watson et al. (2015) MOCAGE was shown to overestimate observations of winter NO_2 when using a GCM forced by the best available reanalysis data, whereas the other three models were shown to underestimate observations of winter NO_2, it would be logical to expect that the actual future winter concentrations of NO_2 would lie somewhere in between the concentrations simulated by MOCAGE and the concentrations simulated by the other three models. Similarly, due to the fact that all models had a negative bias for NO_2 in the summer, future concentrations on NO_2 in the summer would be expected to be slightly higher than the concentrations simulated here.

The summertime O_3 concentrations for CHIMERE and EMEP are very similar in distribution and magnitude (34 and 37 ppb averaged over the masked European land domain, respectively), whereas the average O_3 concentrations simulated by MATCH and MOCAGE are notably lower (28 and 29 ppb, respectively) (Fig. 3). This is because the O_3 concentrations simulated by MATCH and MOCAGE reach a peak in the springtime and then decrease over the summer months, whereas the concentrations of O_3 in CHIMERE and EMEP remain sustained throughout the summer season, consistent with the

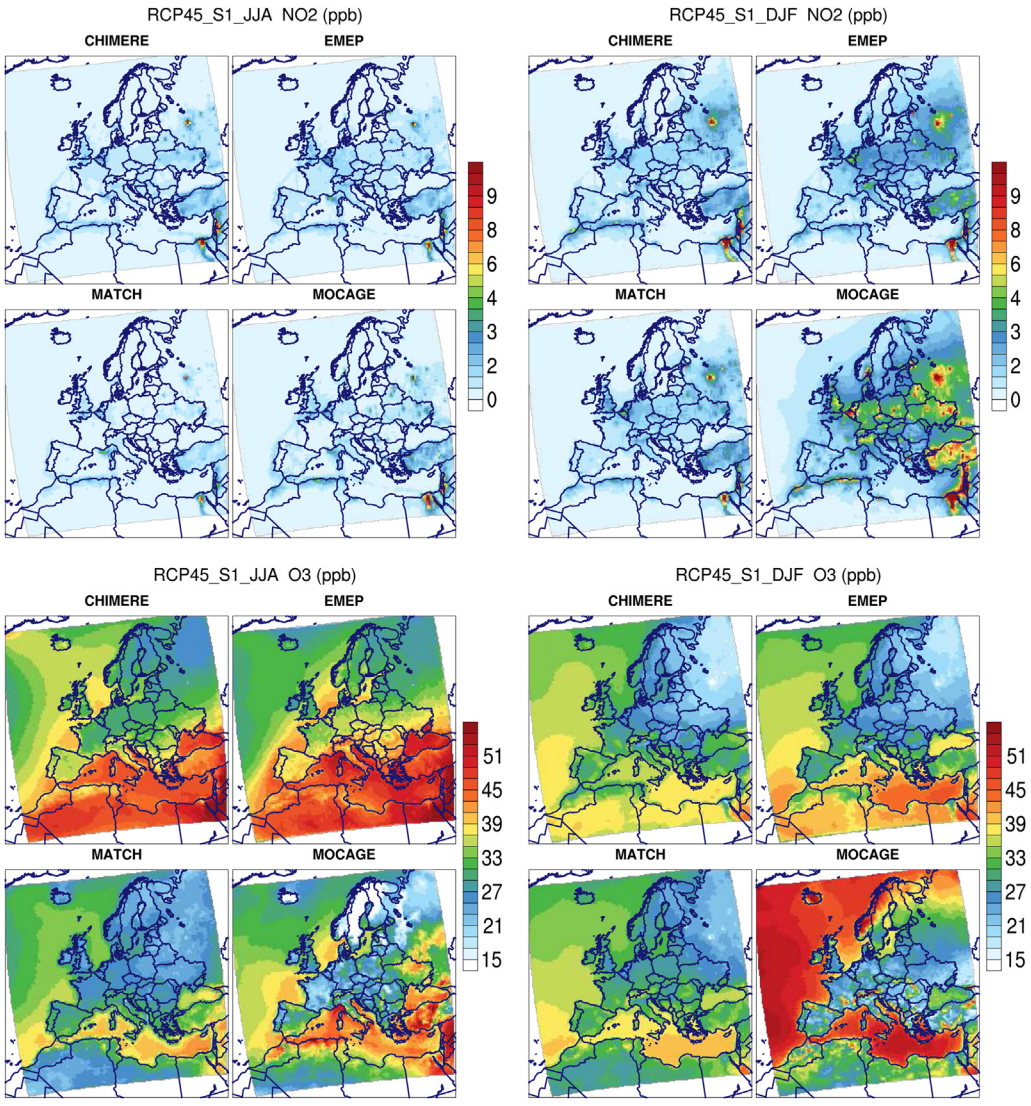

Fig. 3. Average summer (left) and winter (right) change in NO_2 (top) and O_3 (bottom) in ppb simulated by CHIMERE, EMEP, MATCH and MOCAGE for the RCP 4.5 + 2 °C scenario S1.

HISTORICAL results presented in Watson et al. (2015). In the winter, the concentrations of O_3 for CHIMERE, EMEP and MATCH are very similar (between 27 and 29 ppb averaged over the European land mass), whereas MOCAGE shows slightly higher values for O_3 (32 ppb), which are clearly linked to the fact that MOCAGE has a much higher concentration of O_3 over the ocean. Ordónez et al. (2010) postulated that MOCAGE's tendency to produce relatively high O_3 can be explained by the combined effects of lower dry deposition rates and higher chemical reactivity. Other processes in MOCAGE, such as its parameterisation of vertical diffusion, may also play a role (Ordónez et al., 2010). In addition, MOCAGE uses different lateral chemical boundary conditions than the other three models in this study. As mentioned previously, CHIMERE, EMEP, and MATCH all use output from the LMDz-INCA model as lateral chemical boundary conditions (Hauglustaine et al., 2004; Szopa et al., 2013), whereas MOCAGE uses chemical boundary conditions generated by a global version of MOCAGE with a 2° × 2° resolution. It was shown that, out of the four models, MOCAGE had the highest positive bias for O_3 in comparison to a simulation using a GCM forced by the best available reanalysis data, particularly in the winter (Watson et al., 2015), meaning that the future results presented here for MOCAGE are also expected to contain a positive bias.

3.1. Comparison of future results with the baseline case (S1-HISTORICAL)

The concentrations of NO_2 in the future CLE scenario are between 0.3 and 0.6 ppb (33–47%) lower than HISTORICAL in the summer months and between 1.0 and 3.0 ppb (46–51%) lower in the winter months (Table 3), with the largest reductions over the areas that were the most polluted in the baseline case, such as the UK, Benelux, Germany, Moscow, and the Po Valley (Fig. 4). These results represent the expected improvements to air quality that could be attained if all currently planned air pollutant emission legislation is implemented between now and 2050. The percentage reductions in NO_2 for the future S1 scenario in comparison with HISTORICAL are reasonably consistent between the models, particularly in winter when NO_2 is more clearly linked to emissions and less involved in photochemistry (Table 3). Despite the fact that all four models used the same ECLIPSE v4a CLE emissions, the absolute reductions in NO_2 seen in the future vary between the models to a certain extent. MATCH, which had the lowest concentrations of NO_2 out of the four models in the HISTORICAL

Table 3
Changes in average NO_2 (ppb) across the masked EURO-CORDEX land domain.

Model	S1-HISTORICAL		S1−S2		S3−S1	
	JJA	DJF	JJA	DJF	JJA	DJF
CHIMERE	−0.52 (−43%)	−1.19 (−51%)	0.01 (2%)	−0.08 (−7%)	−0.36 (−52%)	−0.69 (−60%)
EMEP	−0.38 (−33%)	−1.50 (−46%)	0.02 (3%)	−0.03 (−2%)	−0.37 (−48%)	−1.01 (−59%)
MATCH	−0.29 (−40%)	−1.03 (−48%)	0.00 (0%)	−0.04 (−4%)	−0.27 (−61%)	−0.72 (−64%)
MOCAGE	−0.63 (−47%)	−2.99 (−49%)	−0.01 (−1%)	0.14 (5%)	−0.50 (−70%)	−2.03 (−65%)

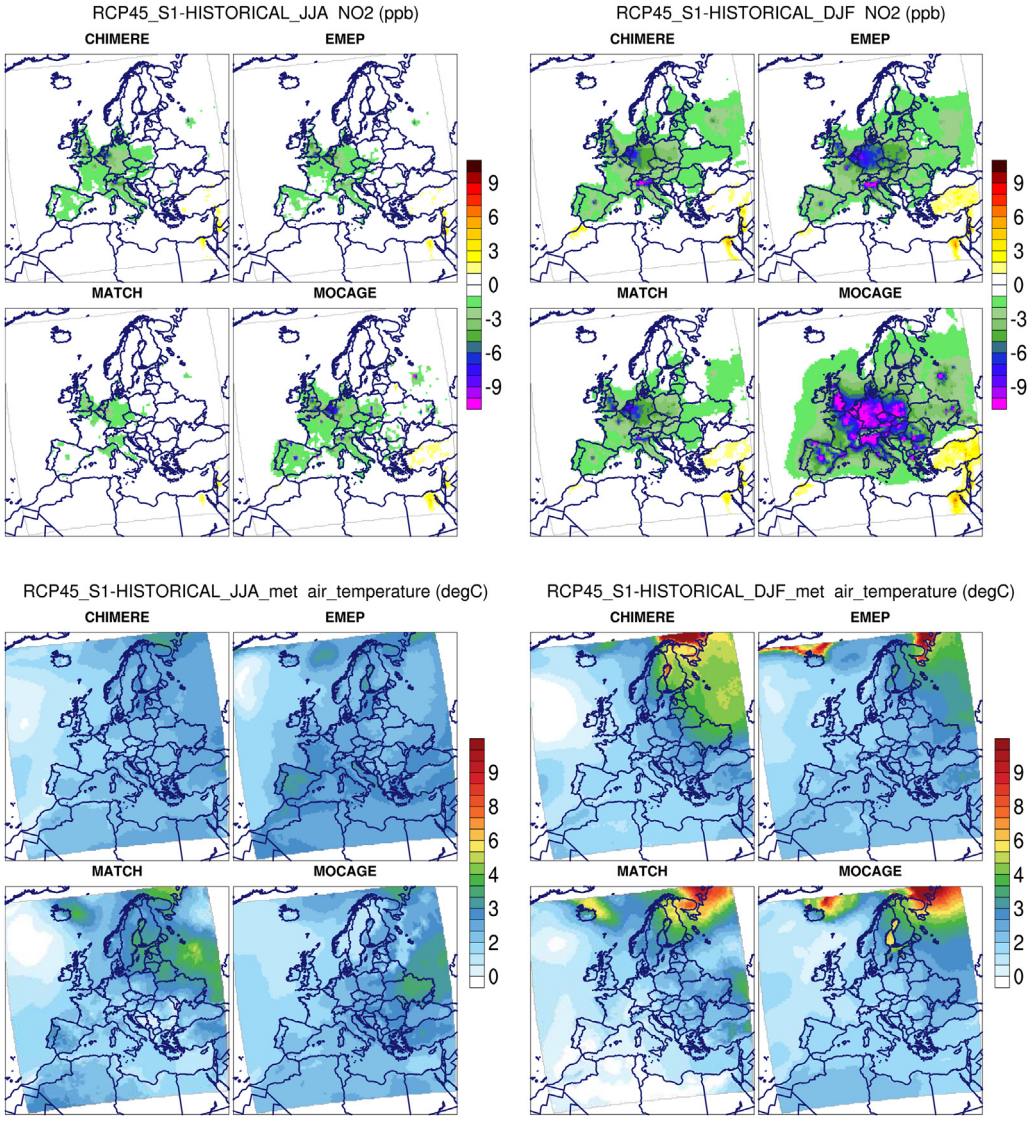

Fig. 4. Average summer (left) and winter (right) change in NO_2 (top) in ppb and temperature (bottom) in degrees C in CHIMERE, EMEP, MATCH and MOCAGE for the RCP 4.5 + 2 °C scenario S1 in comparison to HISTORICAL.

simulation (Watson et al., 2015), shows the smallest decrease in NO_2 concentrations in the future; whereas MOCAGE, which had higher concentrations of NO_2 than the other three models in the HISTORICAL simulation (Watson et al., 2015), shows the largest decreases in NO_2 in the future scenario (Table 3). As discussed in the previous related paper, physical differences between the models, such as differences in boundary layer height, may be responsible for the inter-model variation in NO_2 in the HISTORICAL scenario (Watson et al., 2015).

Fig. 4 also shows the average temperature increases for scenario S1 in comparison to the baseline simulation (HISTORICAL) for the four models, averaged over the thirty-year +2 °C time period for the summer (June through August) and the winter (December through February) seasons, respectively.

Although the global average temperature increase at the midpoint of each of the GCM simulations that drive S1 is +2 °C above pre-industrial times, geographical variations in temperature can been seen across the European domain, between the four models, and between the summer and winter seasons. The largest increases in temperature (that can get up to above 10° in certain

isolated areas) are seen in the northeastern corner of the domain during the winter season.

Although temperature is often correlated with O_3, future concentrations of O_3 in the summer months, as shown in Fig. 5, are actually around 4–5 ppb lower on average than the baseline case (see Table 4), as a result of the reduction in anthropogenic emissions in the CLE scenario (Fig. 2). This observation is consistent with previous studies, which have highlighted the importance of reducing future emissions of O_3 precursors in order to improve air quality (Colette et al., 2012). In wintertime, however, ozone is seen to increase over the majority of the European continent under the future scenario (Fig. 5). This is due to the fact that there is less sunlight and less photochemical production and destruction of ozone at this time of year, so reactions of NO_x with O_3 and OH become more important pathways for the destruction of O_3. As there is less NO_x under the CLE future scenario, O_3 levels are higher than in the baseline case due to a reduction in reactions of O_3 and OH with NO_x. The differences across the four models provide an idea of the uncertainty in the results. In both the winter and summer seasons, MOCAGE shows a greater variation in O_3 decreases and increases than the other three models. The results for CHIMERE, EMEP, and MATCH are remarkably similar over continental Europe, which may be due to the fact that these three models use the same lateral boundary conditions for chemical species. MOCAGE is the only model that uses its own lateral boundary conditions that vary from year to year. Fig. 7 shows an approximation of the difference in vertical ozone profiles between S1 and HISTORICAL for the MOCAGE boundary conditions averaged over 30 years alongside the LMDz-INCA boundary conditions and the ACCMIP boundary conditions (Young et al., 2013) for reference. These profiles were horizontally averaged along the borders of the regional domains. Although the difference in surface O_3 for MOCAGE's boundary conditions is often reasonably close to the changes in surface O_3 shown for LMDz-INCA, MOCAGE shows a particularly large difference in surface O_3 between S1 and HISTORICAL at the northern boundary of the regional domain in winter, which explains why there is a large difference in O_3 at the north of the domain in Fig. 5. Numerous studies have shown that chemical boundary conditions can have significant impacts upon O_3 results of regional model runs even far away from boundaries (Akritidis et al., 2013; Giordano et al., 2015; Schere et al., 2012). For example (Giordano et al., 2015), showed that that the negative bias for O_3 in winter that was present in the monitoring atmospheric composition and climate (MACC) re-analysis of the global IFS-MOZART model was mimicked by the regional models that used the MACC re-analysis as lateral boundary conditions. Despite the fact that the MOCAGE results stand out as being quite different from the other three models, a Student's t-test (following Chervin and Schenider (1976)) calculated over the model ensemble shows a 97% level of significance throughout Europe in the summer, meaning that there is a robust signal for the reduction in future summer O_3 across the four models in comparison to the inter-annual variability of the 4-model ensemble (Fig. 6). The increase in O_3 in the winter is also a robust result across northern Europe, but there are patches of southern and western Europe (Ireland, Spain, France, Greece, etc.) that do not meet the 97% level of significance (Fig. 6). Annual results can be seen in the supplementary materials.

4. Climate change effects (scenario S1–S2)

In previous studies, the isolated impact of climate change has often been calculated as the difference between two simulations with future and current climate conditions, with anthropogenic emissions fixed at current levels (Doherty et al., 2013; Fang et al., 2013; Colette et al., 2015). The approach for this paper differs from previous studies because emissions in the S1 and S2 scenarios are fixed at future 2050 levels instead of present-day levels. This provides a realistic assessment of future climate change under a future emission scenario, which has lower emissions than a present-day scenario. Although we are evaluating the projected impacts of a +2 °C world, the present-day climate already represents approximately 0.5 °C warming since pre-industrial times, so the difference between S1 and S2 is approximately 1.5 °C. Fig. 8 shows the annual profile of surface O_3 for scenarios S1 and S2, calculated from monthly averages over the masked model domain and then averaged over the 30 years of each simulation. These results show that future climate change, in isolation from changes in emissions, increases average summer O_3 by up to 0.83 ppb over the masked European land domain in the summer and up to 0.26 ppb in the winter in comparison with present-day climate (Table 4). However, in the case of MATCH, the future S1 scenario actually has

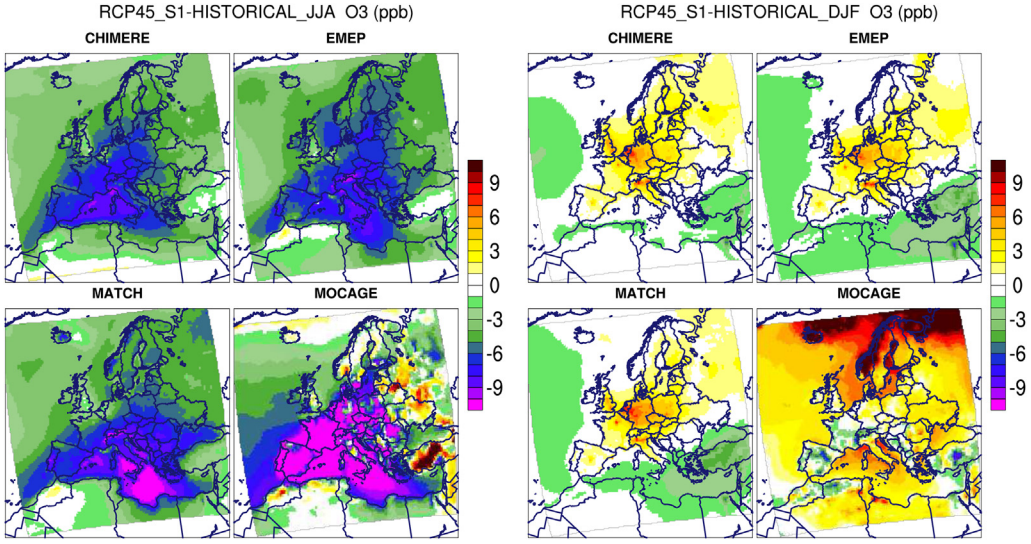

Fig. 5. Average summer (left) and winter (right) change in O_3 in ppb simulated by CHIMERE, EMEP, MATCH and MOCAGE for the RCP 4.5 + 2 °C scenario S1 in comparison to HISTORICAL.

Table 4
Changes in average O_3 (ppb) across the masked EURO-CORDEX land domain.

Model	S1-HISTORICAL		S1−S2		S3−S1	
	JJA	DJF	JJA	DJF	JJA	DJF
CHIMERE	−4.44 (−11%)	+1.12 (4%)	+0.36 (1%)	+0.25 (1%)	−2.61 (−8%)	+0.59 (2%)
EMEP	−4.68 (−11%)	+0.80 (3%)	+0.33 (1%)	−0.11 (0%)	−3.10 (−8%)	+0.53 (2%)
MATCH	−5.08 (−16%)	+0.71 (3%)	−0.11 (0%)	−0.21 (−1%)	−3.25 (−12%)	+0.26 (1%)
MOCAGE	−3.85 (−12%)	+3.60 (13%)	+0.83 (3%)	+0.26 (1%)	−9.27 (−32%)	−0.26 (−1%)

Values in parentheses are the percentage differences with respect to HISTORICAL for S1-HISTORICAL and with respect to S1 in the case of S1−S2 and S3−S1. JJA = June, July and August. DJF = December, January and February.

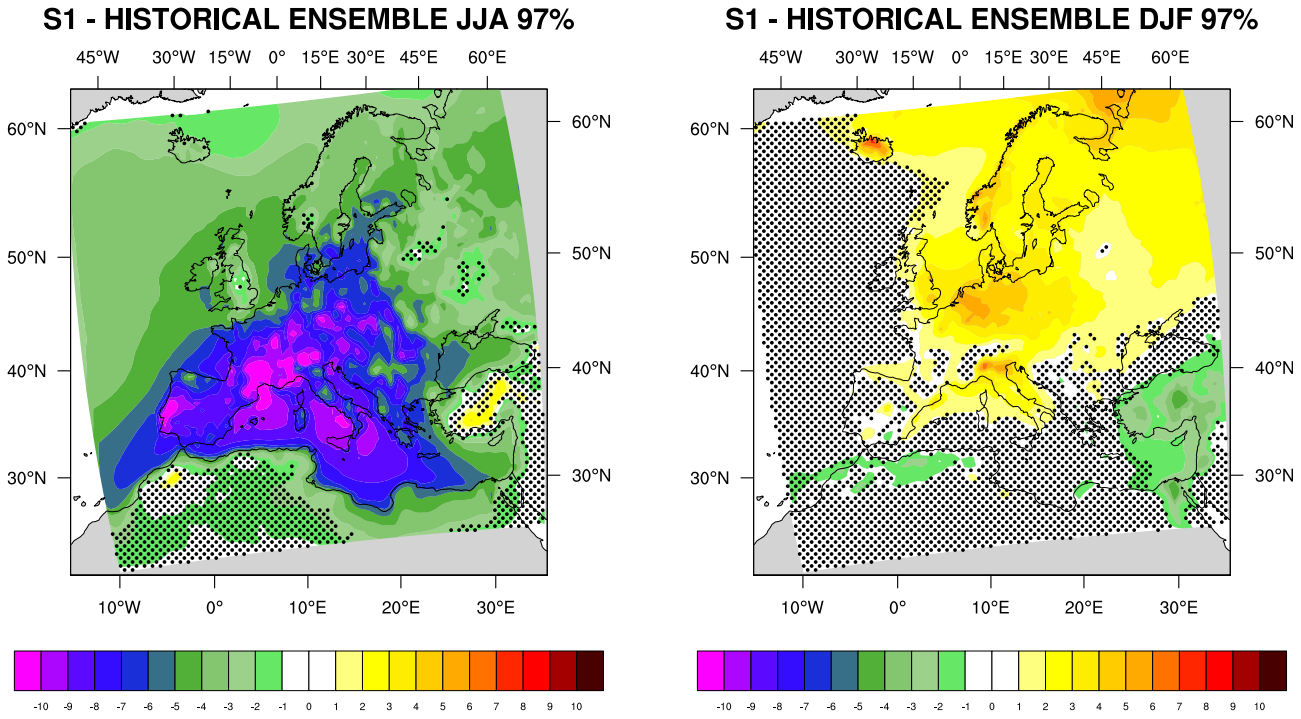

Fig. 6. Average summer (left) and winter (right) change in O_3 in ppb simulated by the 4-model ensemble for the RCP 4.5 + 2 °C scenario S1 in comparison to HISTORICAL. Black dots mark grid points that do not satisfy the 97% level of significance.

lower O_3 than S2, particularly in the spring and in October and November, due to changes in photochemical regimes (changes between NO_x-sensitive and VOC-sensitive conditions). In a NO_x-sensitive regime (with relatively low NO_x and high VOC), O_3 increases with increasing NO_x and changes little in response to increasing VOC. In a VOC-sensitive regime O_3 increases with increasing VOC. Changes between these two types of photochemical regimes can occur due to changes in the relative emissions of VOCs and NO_x and due to changes in the reactivity of the atmosphere (due to changes in season and incoming solar radiation, etc.) (Beekmann and Vautard, 2010). Although increased summertime temperatures are traditionally correlated with increased O_3, because the conditions that cause higher temperatures (such as low winds and high solar radiation) also cause O_3 levels to increase, the temperature changes in the future S1 scenario may not be that highly correlated with O_3 because they are caused by increased greenhouse gas (GHG) emissions, which do not have direct impacts upon O_3.

The geographical distribution of the difference in surface O_3 between simulation S1 (future emissions with future climate forcing) and simulation S2 (future emissions with present-day climate forcing) is shown in Fig. 9. In the summer, the climate impacts upon O_3 are not as strong in the north (Norway, Sweden, Finland, Ireland, and Iceland), where the differences between S1 and S2 can even be negative for certain models. In the case of MATCH, the minor increases in O_3 due to future climate change seen over France, Spain, and other areas of Europe, are offset by minor decreases in parts of northern Europe (such as Norway, Finland and Sweden), resulting in a net negative value for the masked domain as a whole (Table 4). The other three models demonstrate an overall increase in surface O_3 across the domain (Table 4). MOCAGE shows the highest increases for summer O_3, particularly in eastern Europe and Russia, which may be linked to the fact that MOCAGE used different chemical lateral boundary conditions than the other three models, representing the impacts of global climate change upon the regional domain, rather than the impacts of regional climate change in isolation from the rest of the globe. However, the average climate change effect upon summertime O_3 is still less than 1 ppb even for MOCAGE. The ensemble average, shown in Fig. 10, shows that the results averaged across the four models are not statistically significant. There is a large amount of variability between the results for the four models, meaning that robust conclusions cannot be drawn (annual results are provided in the supplemental materials). The differences between S1 and S2 also result in both positive and negative changes to precipitation (which affects wet deposition) and cloud cover (which affects

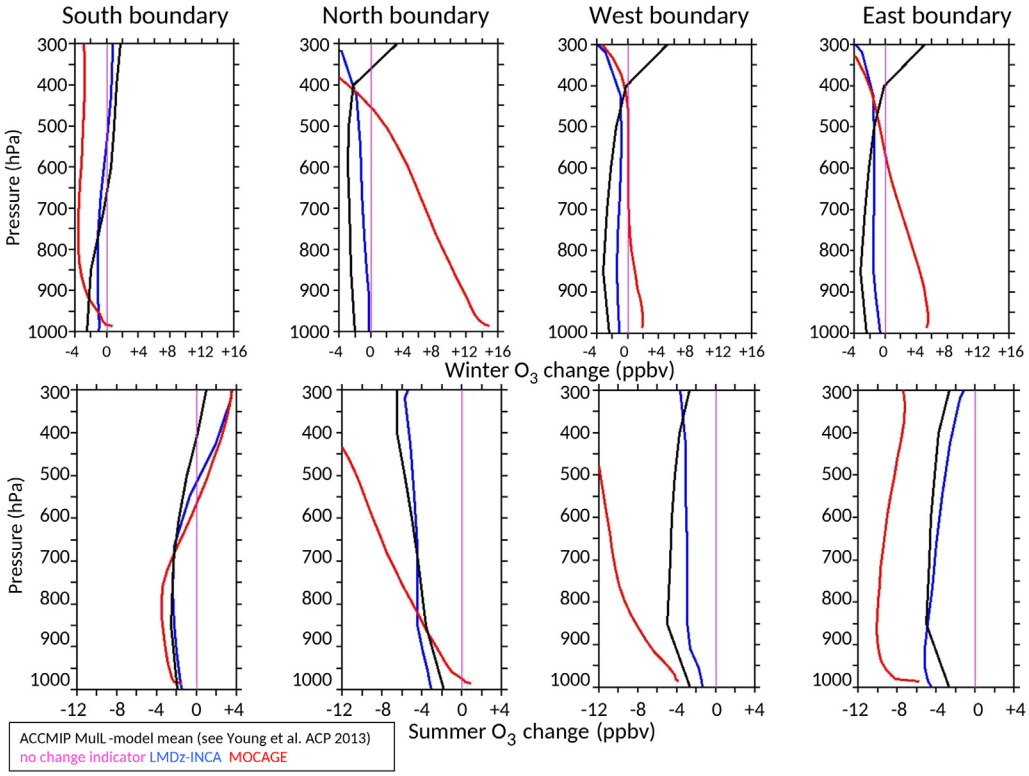

Fig. 7. Winter (top) and summer (bottom) ozone changes between 2050 and 2006 as simulated for RCP 4.5 by the global models: LMDz-INCA (in blue), MOCAGE (in red), and the ACCMIP multi-model mean considering linear changes between 2030 and 2000 (in black). Changes were averaged spatially over the four boundaries of the regional domains in order to provide an approximation of the boundary conditions used for the regional models. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

photo dissociation rates and heterogeneous chemistry), with large geographical, seasonal and inter-model variations (not shown). Changes in cloud cover and precipitation due to climate change would be expected to have impacts upon the concentration of O_3. However, as seen in Fig. 10, the change in O_3 between S1 and S2 is not statistically significant; therefore, the impacts of precipitation and cloud cover upon O_3 due to climate change also cannot be considered to be statistically significant. Although a +2 °C warming above pre-industrial times may increase future O_3 in the summer with respect to current (1971–2000) conditions for three out of four models, these climate impacts are smaller and less significant than impacts due to changes in anthropogenic emissions.

5. Mitigation scenario (scenario S3–S1)

The mitigated emission (MFR) scenario shows significant reductions in NO_x emissions in comparison to the CLE scenario throughout all of Europe, although certain pollution hotspots remain (in Benelux, for example) (a plot of NO_x emissions is provided in the supplemental materials). Due to reduced emissions of anthropogenic precursors such as NO_x, the MFR scenario presents an opportunity for significant reductions in future ozone in the summertime in comparison to the CLE scenario (Fig. 11). O_3 reductions are also generally seen in winter over the southwestern part of the domain under the MFR scenario (Fig. 11); however, the results for winter actually show an increase in O_3 in the northeast due to the fact that at this time of the year, when there is much less sunlight and less photolytic destruction of O_3, titration of O_3 with NO_x becomes a very important pathway for the removal of O_3 from the atmosphere. Therefore, the reduction in NO_x emissions in the mitigated scenario for the areas that do not have a large amount of sunlight leads to less removal of O_3 with NO_x, and therefore higher concentrations of O_3 during the winter season. However, this increase should not pose too much of a concern for human health, as wintertime O_3 is less of a health hazard than summertime O_3. Fig. 12 shows that, aside from regions in northern Scandinavia and Iceland, the reduction of O_3 during the summer months due to the MFR emission scenario has a 97% level of significance across the four models. During the winter, the increase in O_3 in the northeast is significant across the four models, but impacts on O_3 are not significant in large parts of central and northern Europe (Fig. 12). Overall, the reductions in O_3 (of between 3 and 9 ppb) seen during the summer under the MFR scenario are larger than the increases (up to 0.6 ppb) observed during the winter season (Table 4), meaning that the MFR scenario has an overall potential to significantly reduce tropospheric O_3 pollution (annual results are provided in the supplemental materials). These results do not take into account future reductions in the emissions of CH_4, which could serve to reduce tropospheric O_3 levels even further.

6. O_3 enhancement

In order to better assess changes in O_3 that are caused by climate change and different emission scenarios, the "summer O_3 enhancement" defined in Schnell et al. (2015) was considered. Firstly, it was assumed that the 30th percentile of all maximum daily 8-h averages of surface ozone (MDA8) in a year represents the background O_3 level. The background O_3 level represents the cleanest air possible over the summer season, which would not be expected to be influenced by recent, locally emitted or produced pollution (United Nations, 2010). Secondly, it was assumed that in Northern Hemisphere mid-latitudes, the MDA8 values during the

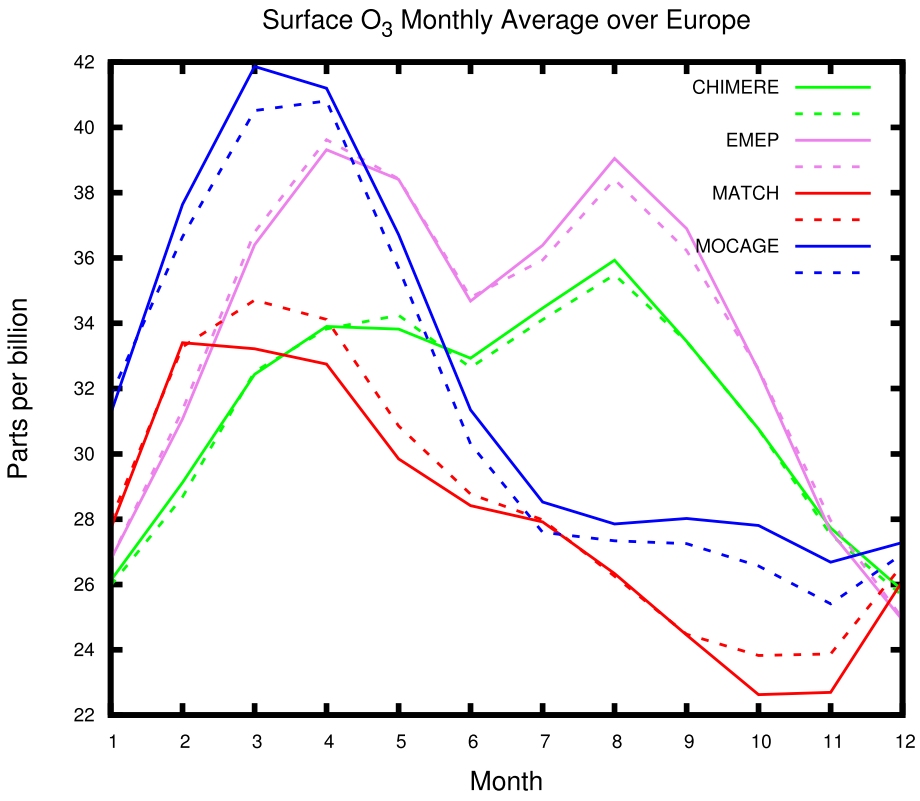

Fig. 8. Time series of monthly mean values of O_3 in ppb (averaged over the regional masked domain with sea surfaces excluded) simulated by CHIMERE (in green), EMEP (in purple), MATCH (in red), and MOCAGE (in blue). Solid lines for S1 and dashed lines for S2. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

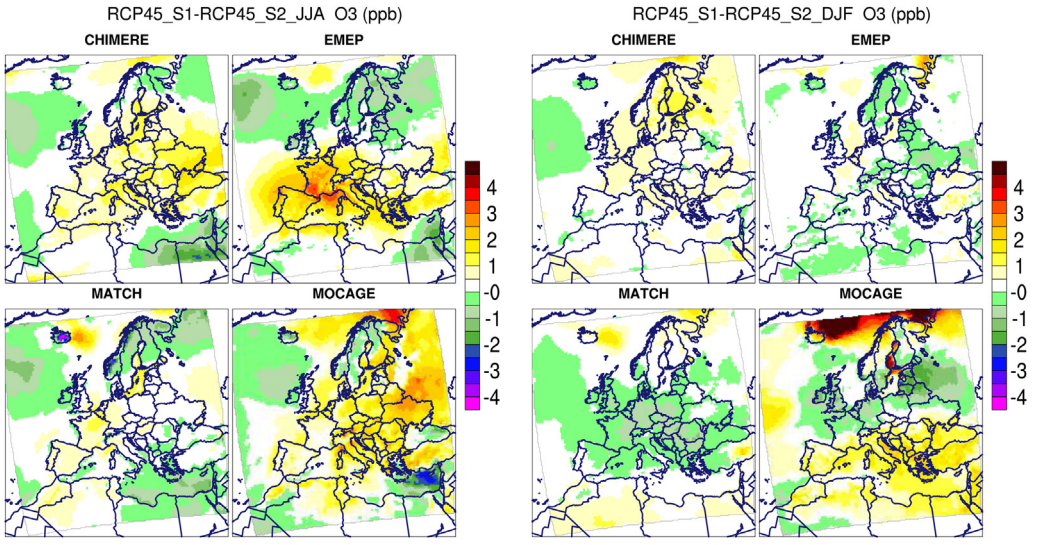

Fig. 9. Average summer (left) and winter (right) change in O_3 in ppb simulated by CHIMERE, EMEP, MATCH and MOCAGE for the RCP 4.5 + 2 °C scenario S1 in comparison to S2.

JJA season (92 days), averaged over multiple years, represent the highest values during a year. Their median is then represented by the 87th percentile of MDA8 values during a year, and the difference between the 87th and 30th percentiles is defined as the 'summer O_3 enhancement', i.e. the increase in O_3 during summer due to the combination of anthropogenic emissions and high solar radiation. Fig. 13 shows the changes in summer O_3 enhancement due to climate change and changes in emissions over Europe, as calculated by the four models for the different scenarios discussed in this paper. The spread between the models gives an indication of the uncertainty of these results.

In the case of S1-HISTORICAL, which is the future RCP4.5 CLE scenario minus the present day, the four models all show a substantial decrease in O_3 enhancement in the future over the majority of the domain (Fig. 13), due to the reduction in anthropogenic emissions under the CLE scenario compared to the present day. This

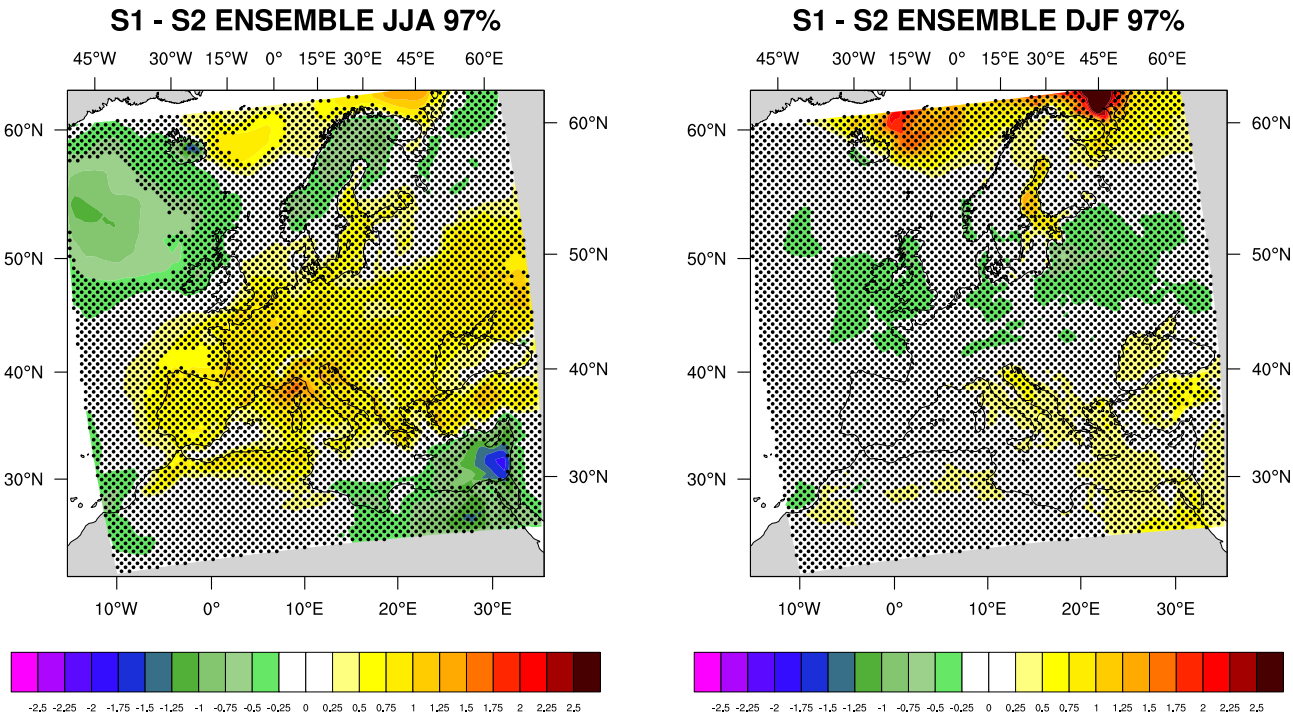

Fig. 10. Average summer (left) and winter (right) change in O_3 in ppb simulated by the 4-model ensemble for the RCP 4.5 + 2 °C scenario S1 in comparison to HISTORICAL. Black dots mark grid points that do not satisfy the 97% level of significance.

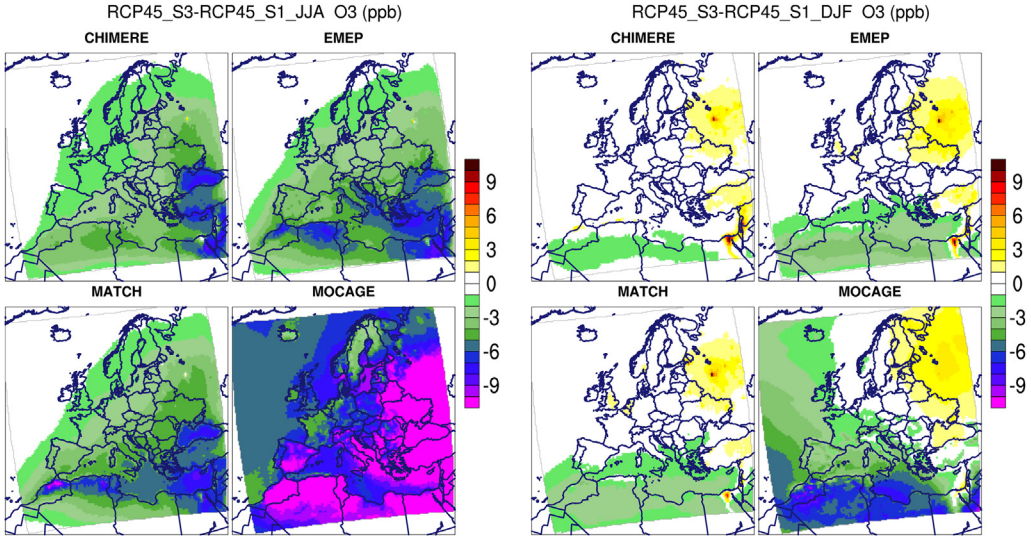

Fig. 11. Average summer (left) and winter (right) change in O_3 in ppb simulated by CHIMERE, EMEP, MATCH and MOCAGE for the RCP 4.5 + 2 °C MFR scenario S3 in comparison to CLE scenario S1.

result is consistent with what was seen in Section 3.1, except that now the background O_3 levels have effectively been removed from the analysis. The increase in O_3 enhancement simulated by MOCAGE in Iceland and over the Norwegian coastline is due to the unusually high levels of O_3 simulated over the ocean and at the northern boundary of the domain under the future scenario. It is interesting to note that MOCAGE now shows a reduction in O_3 enhancement throughout all of central and eastern Europe (apart from Turkey), whereas previously this model showed patches of O_3 increases in eastern Europe in the summer in the future scenario (refer back to Fig. 5). This is due to the fact that MOCAGE has a higher inter-annual variability than the other three models; therefore, the calculation of O_3 enhancement, which effectively removes the maximum and minimum MDA8 values from the analysis, has a significant impact upon MOCAGE's overall results. When averaged across the masked domain for each model, the O_3 enhancement is reduced by between 2.3 and 3.7 ppb for S1 in comparison to the present day.

The differences in O_3 enhancement for S1−S2, which represents the climate change effect under future conditions, are fairly close to zero over the majority of the domain for all four models (Fig. 13). When averaged over the masked model domain, the change in O_3

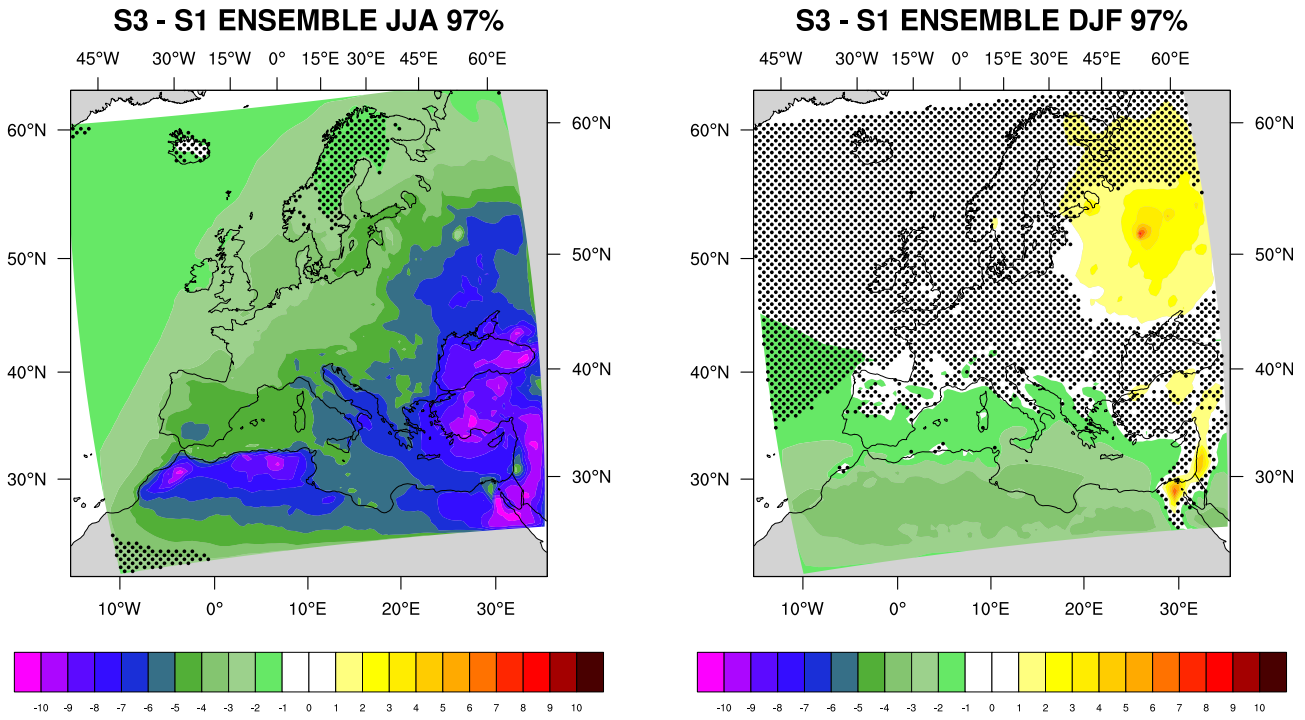

Fig. 12. Average summer (left) and winter (right) change in O_3 in ppb simulated by the 4-model ensemble for the mitigated emission scenario S3 in comparison to S1. Black dots mark grid points that do not satisfy the 97% level of significance.

enhancement does not exceed 0.5 ppb for any of the four models. The impacts of climate change upon O_3 should theoretically fall within the background levels of O_3, as climate change has a gradual and indirect impact upon O_3; therefore, it is logical that removal of background O_3 would lead to similar results for simulations S1 and S2, which both use the same anthropogenic emission data. But regardless of whether the O_3 differences occur in the background or within the 87th-30th percentile, the differences between S1 and S2 are small and statistically insignificant as discussed in Section 4.

Fig. 13 also shows the O_3 enhancement for the mitigation scenario S3 in comparison to the present day (HISTORICAL) and the future CLE scenario (S1). The decrease in O_3 enhancement caused by using the MFR mitigated emission scenario is captured by all four models to varying extents. When averaged across the masked domain for each model, the O_3 enhancement is reduced by between 0.5 and 2.1 ppb for S3 in comparison to S1, and by 4.1–5.8 ppb for S3 in comparison to the present day. These reductions in O_3 enhancement represent a significant improvement in air quality, which would correspond with important beneficial impacts upon human health.

Additional figures showing the difference between the 30th percentile (the background) have been included in the article's supplemental information in order to provide additional information about how background O_3 levels were affected in each scenario.

7. Discussion and conclusions

This study provides further evidence to support the finding that the future of European summertime air quality in the coming decades will primarily be controlled by reductions of anthropogenic emissions resulting from the implementation of national and European policy. The present study uses a novel approach by examining the 30-year period when global average warming reaches +2 °C above pre-industrial times, rather than using the same fixed future time period for all models. The use of 30-year simulations instead of 10-year simulations reduces the uncertainty of the results (Lacressonnière et al., 2016a) and the use of GCM simulations centred on a +2 °C time period shows less uncertainty for temperature than using fixed-year simulations (Vautard et al., 2014). This study uses a set of four CTMs, each driven by a different GCM, in order to provide an idea of the range of uncertainty in simulated gaseous air pollutants (O_3 and NO_2). Using robust results from 30-year simulations with the four models, it was shown that if all currently proposed legislation is properly implemented, significant reductions in summertime NO_2 (of 0.3–0.6 ppb) and surface O_3 (of 4–5 ppb) could be attained by the year 2050, when it is estimated that global temperatures will have reached +2 °C above pre-industrial times. This increase in temperature associated with global climate change was shown to induce small, and not statistically significant, changes in tropospheric O_3 (between −0.1–0.8 ppb in summer), which would not be expected to greatly hinder the beneficial effects of air quality legislation. This conclusion is in agreement with other studies, which have also concluded that the effects of climate change upon future O_3 are relatively small in comparison to changes in precursor emissions (Coleman et al., 2013; Hedegaard et al., 2013; Stevenson et al., 2013).

This study also showed that if all feasible mitigation measures could be implemented regardless of cost, as represented by the MFR scenario, a very large improvement in air quality, specifically a reduction in NO_2 of between 0.6 and 1.1 ppb and a reduction in tropospheric O_3 of between 7 and 13 ppb in the summer, could be experienced by the year 2050 in comparison with the present-day scenario. These results will be useful for future work regarding the impacts of future air quality upon health.

Although air quality studies of the Northern Hemisphere often focus on summertime O_3 due to the impacts of summer O_3 upon health, this paper also presents results for the winter season. Due to the low solar radiation in winter, combined with changes in the

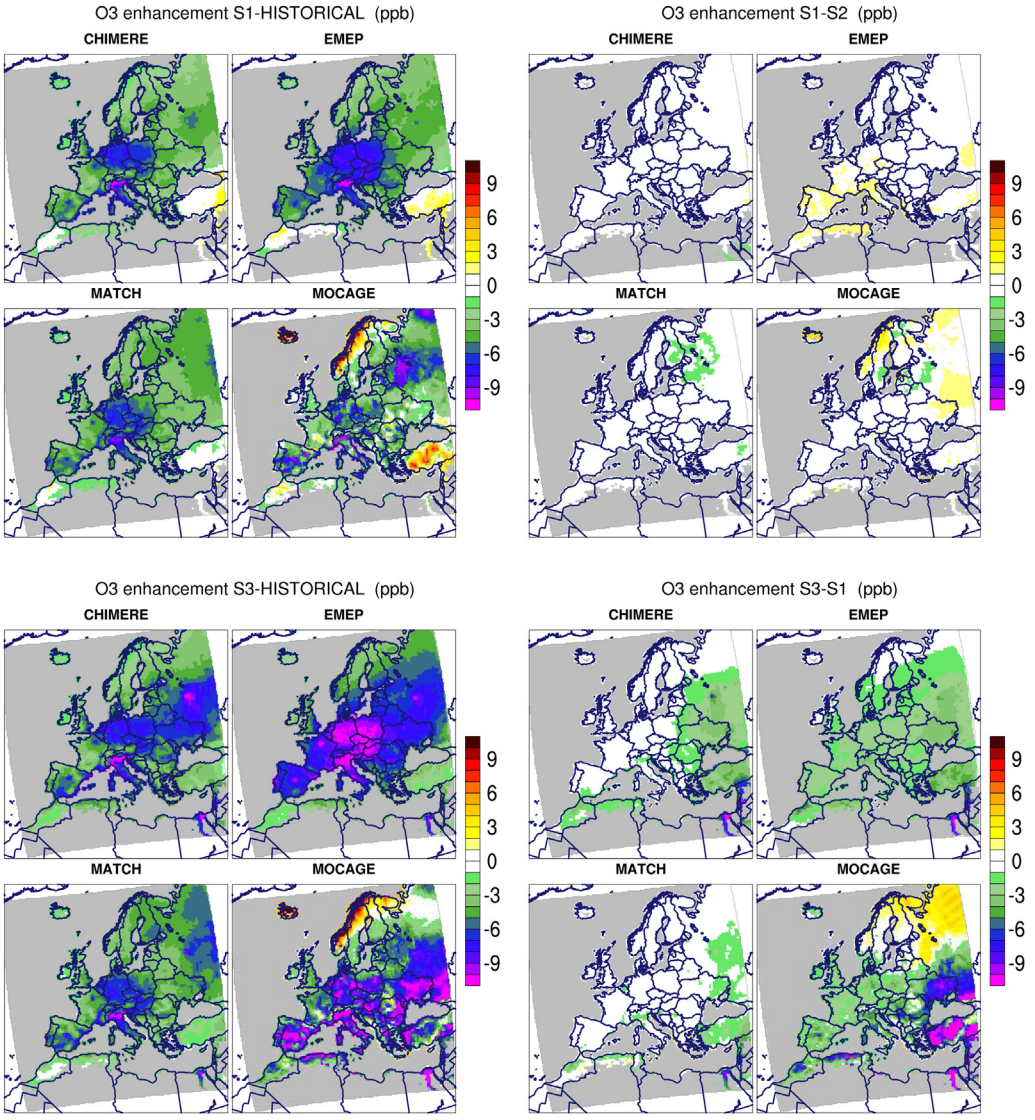

Fig. 13. Change in ozone enhancement (the 87th percentile minus the 30th percentile of the MDA8 value) in ppb for CHIMERE, EMEP, MATCH and MOCAGE for S1-HISTORICAL, S1−S2, S3-HISTORICAL and S3−S1. Grey areas have been masked out of the calculations.

VOC to NO_x ratio, the future CLE emission scenario S1 causes O_3 concentrations to increase in the winter in the northeastern part of Europe. These increases (of between 0.7 and 3.6) ppb are not as large as the decreases (of between 3.8 and 5.1 ppb) observed in the summer, therefore the overall annual impact of the future S1 scenario is still an improvement in O_3 air quality. However, it is interesting to note how the impacts upon air quality due to a reduced emission scenario have a large seasonal variation. There is also an increase in O_3 observed during the wintertime under the future mitigated emissions scenario S3, but this increase is more limited to a northeastern area of the domain and is greatly offset by the O_3 reductions during the rest of the year.

In order to better understand the impacts of climate change and emission changes upon O_3, the O_3 enhancement was calculated in order to remove background levels of O_3 from the analysis. Background levels of O_3 will change in the future due to large-scale changes in GHG emissions and climate, so it is interesting to see how the models simulate O_3 enhancement once background levels have been taken out. Consistent with the emission reductions seen under the CLE emission scenario, all four models showed reductions in O_3 enhancement in the future S1 scenario over regions that had significantly higher levels of O_3 precursor emissions in the present day (Germany, Po Valley and Benelux, for example). These improvements to air quality were more marked in the future mitigated emission scenario, S3. Inter-model differences can be attributed to differences in boundary conditions, differences in the reactivity of the models' chemical mechanisms, and physical differences in the models' set-ups. The O_3 enhancement for S1−S2, the climate change effect, was less than 0.5 ppb for all models when averaged across the domain, meaning that the effects of climate change upon O_3 are relatively small and/or lie within the background levels of O_3.

The fate of air quality in Europe is clearly highly dependent upon our implementation of emission reduction legislation, which will not be greatly obstructed by the effects of a +2 °C rise in global temperatures. This result is a good news if policies follow the December 2015 Paris agreement. However, if climate change causes global temperatures to increase beyond 2°, the impacts of climate change upon air quality may be more significant.

Acknowledgements

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2016.07.051.

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