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Future global tropospheric ozone changes and impact on European air quality

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[1] A global chemistry climate model is used in conjunction with a regional chemistry-transport model dedicated to air quality studies to investigate the impact of anthropogenic emission changes, under several scenarios, on western European summertime surface O₃ levels in 2030. The implementation of presently decided emission control legislation in the individual countries worldwide leads to a geographically heterogeneous impact on summertime surface O₃ levels over Europe. A decrease of the averaged O₃ mixing ratio reaching −3 ppbv is predicted in southern areas whereas an increase reaching up 4 ppbv is calculated in northwestern Europe. The benefit of European emission control measures is found to be significantly counterbalanced by increasing global O₃ levels and subsequent long range transport since both are of the same magnitude (up to 4 ppbv) but opposite in sign. However, the net effect of both global and European emission changes is a significant decrease of O₃ extreme episodes during summertime. **Citation:** Szopa, S., D. A. Hauglustaine, R. Vautard, and L. Menut (2006), Future global tropospheric ozone changes and impact on European air quality, *Geophys. Res. Lett.*, 33, L14805, doi:10.1029/2006GL025860.

1. Introduction

[2] Ozone (O₃) is a trace gas species resulting, in the troposphere, from the oxidation of carbonaceous species (CO, hydrocarbons) in the presence of NO_x both emitted by anthropogenic and natural processes. Attention is paid to this molecule for its greenhouse effect as well as for its impact on air quality close to inhabited areas during smog events, causing both human health problems and vegetation damages. Consequently, as far as CH₄ and O₃ are concerned, policy initiative should consider both global warming and sustainable air quality to define a strategy of emission control legislation [Fiore *et al.*, 2002].

[3] The emission of O₃ precursors is expected to decline in the European Union (EU-25) until 2020 even under the assumption of accelerated economic growth [Amann *et al.*, 2005; Dentener *et al.*, 2005]. However, air quality depends on both regional production and global background levels. For O₃, this latter component is controlled by emissions in remote countries and long-range transport of O₃ and its precursors [Parrish *et al.*, 1993; Li *et al.*, 2002; Auvray and

Bey, 2005]. Mainly due to economic growth in emerging countries, the distribution and intensity of worldwide emissions will be strongly modified during the next decades [Cofala *et al.*, 2004]. Consequently, the assessment of the future European air quality requires the consideration of the global scale chemistry.

[4] From a global perspective, the impact of worldwide emission changes on tropospheric O₃ and the associated radiative forcing was investigated by 25 state-of-the-art global chemistry-transport models in the framework of the PHOTOCOMP experiment [Stevenson *et al.*, 2006; Dentener *et al.*, 2005]. Three emission scenarios were tested, representing respectively the implementation of current air quality legislation in each individual country around the world (CLE scenario), the maximum reduction of emissions currently technically feasible (MFR scenario) and the scenario SRES-A2 developed by the Intergovernmental Panel on Climate Change. These scenarios can be considered respectively as ‘likely perspective’, ‘most optimistic’, and ‘most pessimistic’ [Dentener *et al.*, 2005]. Stevenson *et al.* [2006] indicate that the 25 model ensemble mean tropospheric O₃ burden changes between 2000 and 2030, are 6%, −5% and 15% for the CLE, MFR and SRES-A2 scenarios, respectively. These changes are accompanied by a decrease of global surface O₃ of 1.8 ± 0.5 ppbv in the most optimistic scenario (MFR) and an increase both in the CLE and SRES-A2 scenarios of respectively 1.7 ± 0.3 ppbv and 4.4 ± 1.1 ppbv [Dentener *et al.*, 2006].

[5] The aim of this study is to examine the impact of these future scenarios on western European surface O₃, using a down-scaling approach from a global into a regional chemistry-transport model. In addition, the relative contribution of long-range transport of O₃ and its precursors with respect to the European emission control strategy is investigated.

2. Modeling Set-Up

[6] The global simulations are performed using the LMDz-INCA chemistry-climate model. The LMDz atmospheric General Circulation Model is run with a horizontal resolution of 3.75° in longitude and 2.5° in latitude and uses 19 vertical σ -p levels extending from the surface to 3 hPa. A detailed description and evaluation of LMDz-INCA is given by Hauglustaine *et al.* [2004] and Folberth *et al.* [2006]. The monthly global emission data sets generated by IASA [Cofala *et al.*, 2004] for CO, NO_x and CH₄, at 1° × 1° resolution, are used in this study. They were extended to non methane hydrocarbons (NMHC) emissions by Dentener *et al.* [2006]. For each scenario, the simulations were spun up for 3 months and performed over one year. The meteorological fields are relaxed toward the 2001 ECMWF ERA40 reanalysis. The global LMDz-INCA results have already been evaluated by Dentener *et al.*

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[2006] and *Stevenson et al.* [2006]. The response in term of O₃ changes lies in the middle of the range of the models.

[7] The daily averaged LMDz-INCA concentrations are taken as boundary conditions for the CHIMERE regional model [*Schmidt et al.*, 2001; *Bessagnet et al.*, 2004], which has been used in many air quality studies [e.g., *Vautard et al.*, 2001]. For this study, CHIMERE calculates gaseous chemical concentrations over Europe within a horizontal domain ranging from -10.5°W to 22.5°E and from 35°N to 57.5°N . The horizontal resolution ($0.5^{\circ} \times 0.5^{\circ}$) allows to roughly capture the local O₃ maxima in large city plumes and downwind. Eight vertical hybrid σ -p levels represent the atmospheric column from the surface to 500 hPa. The dynamical fields are computed using the MM5 model (PSU/NCAR mesoscale model) driven by the ECMWF ERA40 reanalysis (see auxiliary material¹).

[8] The regional simulation for present-day uses the anthropogenic EMEP 2002 emissions [*Vestreng*, 2003]. For each of the three 2030 scenarios (CLE, MFR and SRES-A2), these emissions were rescaled, for CO, NMHC and NO_x, using, at each grid point, the ratios between the 2030 and the present day emissions from the IIASA global inventories (see auxiliary material). Using the LMDz-INCA boundary conditions, CHIMERE was applied to investigate the response of European summer pollution to (1) both global and European changes in anthropogenic O₃ precursor emissions (runs EW1, EW2, EW3); (2) changes only in global anthropogenic emissions (present day emissions for Europe) (runs W1, W2, W3); (3) changes only in western European emissions (present day emissions for global chemistry in LMDz-INCA) (runs E1, E2, E3). The performed simulations and the corresponding configurations for the anthropogenic emissions are listed in Table 1.

3. Present-Day Simulation

[9] In order to check the relevance of the present-day regional simulation performed with the CHIMERE model (Ref), the simulated daily O₃ maxima are compared with observations over 244 stations spread all over Europe and located in rural, suburban, as well as urban areas. At these stations, a root mean square error (rms) of 9.14 ppbv and a correlation (r) of 0.826 between observed and simulated daily maximum O₃ are obtained over the “summer” period (1st of May to 31st of August). Table 1 shows a standard deviation ratio ($\sigma_{mod_Ref}/\sigma_{obs}$) of 0.88, illustrating the ability of CHIMERE to simulate the O₃ variability over Europe with confidence. The averaged present-day O₃ maximum and standard deviation compare well with ground based stations. Nevertheless, we note that the number of days with O₃ exceeding the 90 ppbv standard value (threshold from which governments are required by European Union directive to inform the public) is underestimated by 50% in the model due to the lack of resolution which does not allow to adequately represent city and industrial plume cores.

4. Impact of 2030 Worldwide Emissions

[10] Figure 1a depicts the surface O₃ change calculated by the CHIMERE model in July 2030 (run EW1), by

Table 1. Daily O₃ Maximum (ppbv) During “Summer” at 244 European Ground Stations for the 10 CHIMERE Runs: Mean (m), Standard Deviation (σ) and the Mean (Over the 244 Stations) Number of Days With Daily Maximum Exceeding 90 ppbv (n) With the Number of Concerned Location (l)^a

	m	σ	n (l)
Measurements	51.9	16.0	2.69 (147)
<i>Reference Simulation = Global[Present] + Regional[Present]</i>			
Ref Present-day	51.2	14.1	1.26 (138)
<i>2030 Anthropogenic Emission Changes = Global[2030] + Regional[2030]</i>			
EW1 2030 CLE	51.4	11.7	0.53 (68)
EW2 2030 MFR	45.1	10.0	0.04 (8)
EW3 2030 SRES-A2	61.4	18.2	9.52 (232)
<i>2030 Anthropogenic Emission Changes Worldwide Except Europe = Global[2030] + Regional[Present]</i>			
W1 World 2030 CLE	53.4	14.0	1.62 (149)
W2 World 2030 MFR	49.3	14.2	1.08 (130)
W3 World 2030 SRES-A2	55.8	13.9	2.04
<i>2030 Anthropogenic Emission Changes Over Europe Only = Global[Present] + Regional[2030]</i>			
E1 Eur 2030 CLE	49.4	11.8	0.39 (53)
E2 Eur 2030 MFR	46.9	9.9	0.05 (9)
E3 Eur 2030 SRES-A2	57.1	18.6	7.48 (230)

^aConsidering the photochemical activity in Europe, summer is assumed to be from the 1st of May until the 31st of August.

comparison with July 2001 (run Ref), for the CLE scenario. Whereas the future emissions lead to an increase of worldwide averaged surface O₃ [*Denstener et al.*, 2006], the CHIMERE runs indicate that these changes are more contrasted over Europe. In the EW1 run, an increase of more than 4 ppbv is calculated over highly populated areas (e.g., in the vicinity of Paris, London, Manchester, Düsseldorf, Bruxelles, Milan, Cracovie). A general increase of 0.1 to 4 ppbv is predicted over northern Europe whereas a decrease of 0.1 to 3 ppbv is calculated over most of southern Europe. For this CLE scenario, the mean O₃ daily maximum at European stations is almost unchanged whereas the number of days reaching 90 ppbv is decreased by a factor of 2.4 (Table 1). Figure 2 shows the percentiles of daily O₃ maximum for the different scenarios simulated with CHIMERE and supports this decrease in the occurrence of extreme values (minima as well as maxima) for the CLE scenario. Hence, the interval between the 10th and 90th percentiles is decreased by 6.3 ppbv from the Ref run to the EW1 run (with differences of respectively 2.7 and 3.6 ppbv for the 10th and 90th percentiles) indicating, as far as O₃ levels are concerned, a general improvement of the air quality.

[11] The MFR and SRES-A2 scenarios provide far more homogeneous but extreme responses (Figure 1b and 1c). The MFR scenario (EW2) induces a reduction of surface O₃ in 2030 larger than 2 ppbv over most of Europe. Nevertheless, the O₃ levels are, as in the CLE scenario (EW1), increased by a few ppbv over northern high emission spots. The general decrease is also accompanied by a 6.1 ppbv reduction of the averaged O₃ daily maximum at European monitoring stations (Table 1). In this “optimistic case” (EW2), the improvement of air quality is characterized by a weaker dispersion of O₃ values due to an important

¹Auxiliary material data sets are available at <ftp://ftp.agu.org/append/g1/2006gl025860>. Other auxiliary material files are in the HTML.

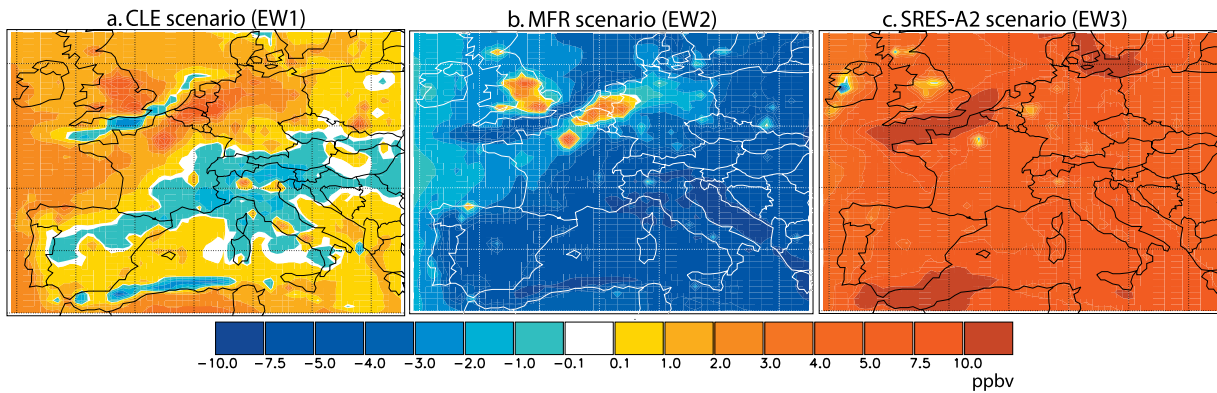


Figure 1. Monthly mean surface O_3 changes in July 2030 (compared with July 2001) simulated over Europe, with CHIMERE for (a) the “current legislation” scenario; (b) the “maximum feasible reductions” scenario; (c) the SRES-A2 scenario.

decrease of the number of high O_3 mixing ratio events. Comparing the EW2 and Ref simulations in Table 1 shows that both the number of exceedance per station and the number of concerned stations decrease, meaning that the spatial extent of higher values as well as their occurrence are reduced in this MFR case. Also supporting this air quality improvement, the 90th percentile reaches 57.9 ppbv against 70.4 ppbv in the present-day experiment (Ref). By contrast, the SRES-A2 (EW3) scenario leads to an O_3 increase, exceeding 5 ppbv in July over Europe. The averaged O_3 daily maximum over European stations grows by 10.2 ppbv. The number of stations concerned by the exceedance increases from the Ref to the EW3 runs but to a lesser extent than the number of occurrences of these exceedances per station. The dispersion of the results is confirmed by the increase of the 90th percentile reaching 86.2 ppbv.

5. Relative Impacts of European Emissions and Long-Range Transport

[12] In the CLE scenario, the response of mean surface O_3 in July to (1) change in global emissions (with present-day European emissions) (W1) and (2) changes in European emissions only (E1) are displayed in Figure 3. The future global emission changes (increase of CH_4 and NO_x and decrease of CO and NMHC) induce a slight increase of continental surface O_3 generalized over Europe (typically 1 to 3 ppbv) through long-range transport and background chemistry (W1). In contrast, the European legislation adopted to improve air quality leads to a reduction of O_3 by 0.1 to 4 ppbv, relative to the 2001 level, over most parts of continental Europe, except in the North (E1). The northern highly inhabited areas (in England, Belgium, the Netherlands and France) is characterized by a rise of mean O_3 levels from typically 0.1 to 5 ppbv. These areas (unlike the rest of Europe) were shown to be VOC limited due to both high emissions and low VOC/ NO_x ratio due to low biogenic VOC emissions [Tarasson *et al.*, 2003]. Consequently, as this VOC/ NO_x ratio increases in all Europe with the 2030 CLE emissions, the O_3 production is enhanced. For the most upwind areas (Portugal, Ireland, England and western France), the global emission changes and subsequent inter-continental transport are responsible for an O_3 increase of 5 to 7.5% in July 2030 (W1) compared to the level obtained

considering European emission changes only (E1). Therefore, the emission reduction efforts achieved by the European Union appear significantly competed by the global emission changes and long-range transport of O_3 and its precursors. These efforts are even entirely counterbalanced over a significant part of western Europe (Portugal, parts of Spain and France, southern Germany, Poland, Denmark, Czech republic, western Hungary) leading to an O_3 increase despite the downward trend of European emissions.

[13] In the MFR scenario (runs W2 and E2), both European and global emission changes lead to a general decrease of O_3 . Hence, the efforts done at European levels are reinforced by the downward trend of background O_3 . In the SRES-A2 scenario (runs W3 and E3), convergent effects of local and global emissions changes are also simulated, both leading to a degradation of air quality in Europe.

6. Conclusion

[14] In this study, we used a global and a sub-continental chemistry-transport models to investigate the relative

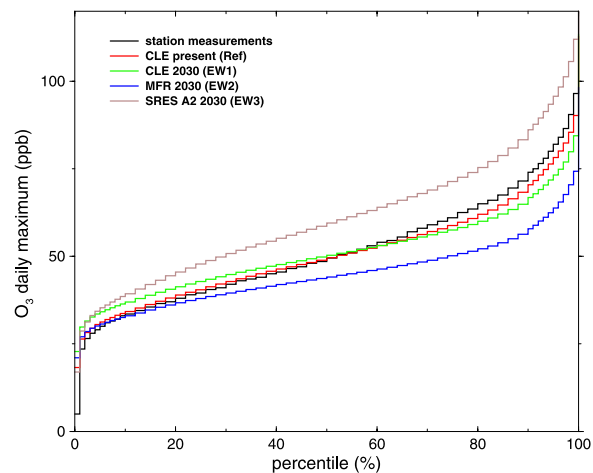


Figure 2. Distribution of daily O_3 maximum percentiles computed over 244 European ground based stations using 2001 measurements (black) and for CHIMERE results for present-day run (red); 2030 CLE run (green); 2030 MFR run (blue); 2030 SRES-A2 run (brown).

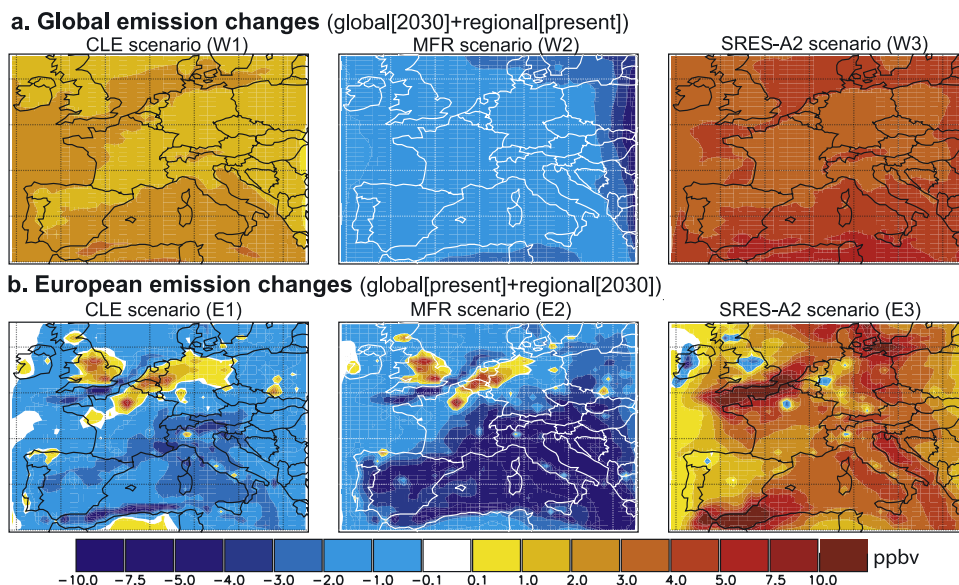


Figure 3. Monthly mean surface O_3 changes in July 2030 (compared with July 2001) for the CLE, MFR, SRES-A2 scenarios simulated with CHIMERE (a) considering global emission changes and current emissions in Europe; (b) considering European emission changes only.

impacts of European emission changes and long-range transport on the future (2030) evolution of summertime O_3 over western Europe. The emission changes increase globally averaged surface O_3 for the most plausible scenario based on the application of current legislation. Over Europe, the surface O_3 response results from the competing effects of long-range transport of pollutants and reduced regional photochemical production. Over northern Europe highly populated areas, where the O_3 production is VOC limited, surface O_3 increases in July by more than 4 ppbv. On the contrary, the O_3 levels in Switzerland, Austria, Slovenia, Croatia and parts of Spain, France and Italy benefit from the reduction policy of European emissions in spite of the global emission changes which tend to increase background ozone. Over these areas, the summertime surface O_3 is reduced by up to 2 ppbv. Over the rest of Europe, the intercontinental transport of pollution offsets the decrease of O_3 levels achieved with European emission changes solely. A resulting increase (up to 2 ppbv) of summertime O_3 is obtained when both effects are considered. However, in all cases, O_3 extrema, often encountered in the vicinity of high emission areas where city pollution plumes develop, decrease significantly as they result mostly from regional emissions. The prediction of future air quality remains an open topic since the anthropogenic emission scenarios are quite uncertain. Furthermore, climate itself is supposed to change in the future, inducing also changes in biogenic emissions [Hauglustaine et al., 2005], factors which have not been accounted for in the present study.

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