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Robert Vautard, Sophie Laval-Szopa, Matthias Beekmann, Laurent Menut, Didier Hauglustaine, Laurence Rouil, Michiel Roemer

Are decadal anthropogenic emission reductions in Europe consistent with surface ozone observations?

Robert Vautard,[1,2] Sophie Szopa,[3] Matthias Beekmann,[4] Laurent Menut,[1]
Didier A. Hauglustaine,[3] Laurence Rouil,[5] and Michiel Roemer[6]

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[1] The consistency between pollutant emission reductions in Europe during the 1990–2002 period and ozone observations is quantitatively verified by 13-year long simulations over the whole period using the regional chemistry-transport model and the EMEP emission inventory. A statistically significant decadal tendency of 0.65 ppb/year is found in the difference between simulated and observed summer 90th percentiles of ozone daily maxima when model emissions are kept constant from year to year. By contrast the use of yearly dependent emissions does not yield a statistically significant percentile difference tendency. The regional structure of the 90th percentile differences shows that emissions may have decreased with a higher rate than assumed in the U.K. and at a lower rate in central Europe. The observed 10th percentiles are also compatible with the assumed emission reductions in Europe during 1990–2002, but are of lesser agreement with simulations using a uniform trend in the baseline ozone.
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1. Introduction

[2] Understanding the history of tropospheric ozone (O_3) during the last decades is quite a challenge. The global increase of O_3 precursor emissions has progressively raised the mean global O_3 level [*Akimoto*, 2003]. However, the increasing number of ground-based, airborne or radiosounding observations shows a far more heterogeneous picture, even in the sign, of O_3 level trends [*Oltmans et al.*, 1998, *Vingarzan*, 2004, *Saraf and Beig*, 2004, *Zbinden et al.*, 2006]. In Europe, even though high surface O_3 events are decreasing, the O_3 baseline appears to have increased by up to few ppb per year at several surface sites [*Naja et al.*, 2003, *Monks et al.*, 2003; *Simmonds et al.*, 2005; *Carlsaw*, 2005], with uncertainty on rates.

[3] Significant trends in European surface O_3 concentrations are difficult to assess due to several antagonist processes, such as stratosphere-troposphere exchanges, stratospheric O_3 depletion, boreal biomass burning. The drastic ozone precursor emission reduction in Europe, of 25–30% [*Vestreng et al.*, 2004], tends to decrease O_3 maxima and increase urban minima because of reduced O_3 titration [*Lindskog et al.*, 2003; *Jonson et al.*, 2005; *Monks et al.*, 2003].

[4] Due to these multiple phenomena, the effective gain of the regulatory European effort to air quality improvement is hard to assess from the use of observations alone, and models are required. The first attempt to compare observed trends and simulated impacts of the 90's emission reductions in Europe was made in the EUROTRAC/TOR project where 6 regional air quality models gave the response to the emission changes for a given meteorological year, and found it consistent with the ozone statistics differences between Summer 2000 and 1990 [*Roemer et al.*, 2003]. Other studies, based on one or two meteorological years, found agreement between measured and simulated trends [*Monks et al.*, 2003; *Solberg et al.*, 2005; *Derwent et al.*, 2003]. Recently *Jonson et al.* [2005] simulated ozone in 1990 and in the 1995–2002 years and compared trends in observed and simulated ozone. Although a fair agreement was found between observed and simulated ozone trends, lack of time continuity in the simulation made it difficult to quantatively assess the statistical significance of the differences found in model and observed trends.

[5] The aim of the present paper is not to explain observed trends in ozone, but to quantitatively verify the consistency between assumed anthropogenic emissions and ozone behaviour during the last decade or so. We use a regional chemistry transport (CHIMERE) to simulate the entire 1990–2002 period and compare with surface O_3 observations. In order to enhance the effect of emissions relative to other trends, we use high percentiles of ozone daily maxima.

[6] Simulations and observations are briefly described in Section 2. In Section 3 the tendency in model biases are interpreted. Section 4 contains a conclusion and a short discussion.

2. Observations and Simulations

[7] Ozone observations, from the EMEP network (http://www.emep.int), at sites representative of the continental background surface atmosphere, are used throughout this work. Only 37 surface sites (see site locations in Figure 3)

[1]Laboratoire de Météorologie Dynamique (LMD)/IPSL Ecole Polytechnique, Palaiseau, France.
[2]Now at Laboratory of Sciences of the Climate and Environment (LSCE)/IPSL, CEA/CNRS/UVSQ, Gif sur Yvette, France.
[3]Laboratory of Sciences of the Climate and Environment (LSCE)/IPSL, CEA/CNRS/UVSQ, Gif sur Yvette, France.
[4]Laboratory Inter-Academic of the Atmospheric Systems (LISA), Faculté des Sciences et Technologie, Créteil, France.
[5]Institut National de l'Environnement Industriel et des Risques (INERIS), Verneuil-en-Halatte, France.
[6]Netherlands Organisation for Applied Scientific Research (TNO), Apeldoorn, Netherlands.

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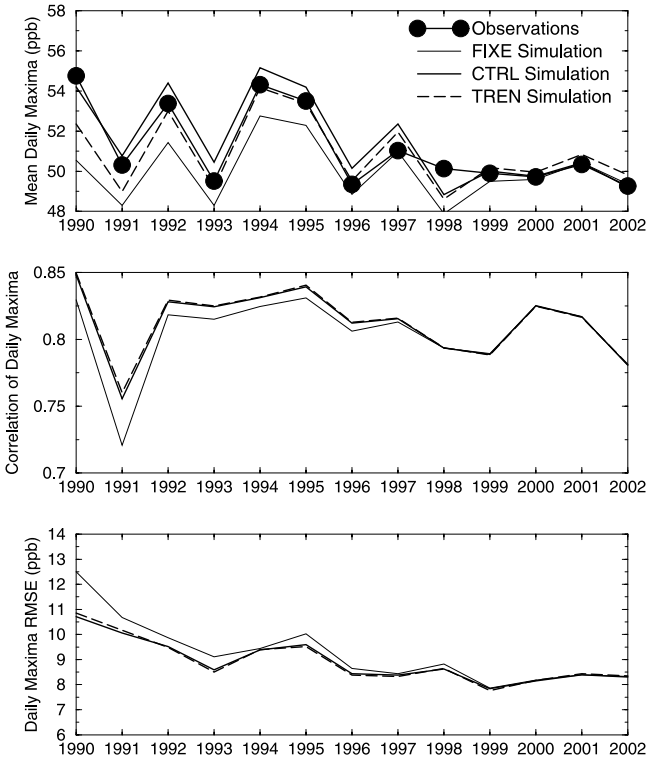

Figure 1. Skill measures of the simulations of daily O_3 maxima as a function of year, for the 3 simulations, CTRL, FIXE and TREN. (top) Evolution of the summertime daily maxima means (in ppb) for simulations and observations. (middle) Evolution of the correlation averaged over summer and all stations. (bottom) Evolution of the root mean square error (in ppb) averaged over summer and all stations.

are selected in such a way that data are continuous and homogeneous from 1990 to 2002. Since we are looking for responses to regional emission reductions, only summertime (April to September) O_3 daily maxima are considered.

[8] Regional simulations are carried out using the gas-phase version of the regional CHIMERE CTM described by *Schmidt et al.* [2001], with updates described by *Vautard et al.* [2005]. This model gives an accurate simulation of O_3 daily maxima in summer [e.g., *Vautard et al.*, 2005]. It is driven by hourly meteorological fields issued from the MM5 meso-scale model [*Dudhia*, 1993], which simulates meteorological variables on a grid with an approximate resolution of 40 km, and which is nudged to (and forced at the boundaries by) the 6-hourly ERA40 European Centre for Medium Range Weather Forecast reanalyses.

[9] The chemistry-transport model is forced at the boundaries by a climatology of O_3 and precursors issued from the global-scale LMDz-INCA model [*Hauglustaine et al.*, 2004, *Folberth et al.*, 2005], using monthly averages of a 5 year simulation with varying meteorology and biomass burning emissions from 1997 to 2001. CHIMERE uses hourly primary emissions derived from the EMEP inventory [*Vestreng*, 2003], which are available for the period under study on a yearly basis, except during the 1991–1994 period where a linear interpolation between 1990 and 1995 emissions is performed. As EMEP emission annual totals and their variation are given in 10 anthropogenic activity sectors, decadal reactivity changes in emissions are taken into account in a rough manner.

[10] A five day spin-up is considered before the first analysis day (1 April) for each of the 13 summertime periods. First, a reference (**CTRL**) simulation is performed using fixed (with year) boundary conditions and year to year variations in emissions according to the EMEP inventory. Second, in order to study the effect of assumed emission changes during the considered period, a simulation (**FIXE**) uses a fixed-year emission set, all other parameters being equal. The year 2001 is arbitrarily selected for reference. Finally, in order to evaluate the influence of possible trends in boundary conditions, another simulation (**TREN**) is carried out assuming a positive 0.4 ppb.year^{-1} O_3 trend added to the LMDz-INCA climatology, no additional ozone in 2001 being arbitrarily assumed. This value is an upper limit of summer trends in baseline O_3 deduced from observations at Mace Head by *Simmonds et al.* [2005] and *Carslaw et al.* [2005] who found respectively $+0.39 \pm 0.25$ and $+0.25 \pm 0.06$ ppb.year^{-1}. The 0.4 ppb.year^{-1} trend is arbitrarily applied at all model boundaries (side and top), and regional emissions inside the domain vary with year as in the CTRL experiment.

[11] Figure 1 shows the skill of the model in simulating the daily and interannual variability of summertime daily O_3 maxima. The simulated summertime ozone averages faithfully follow the observed ones. Mean daily maxima correlations lie around 0.8 and their root mean square (RMS) errors range from 8 to 12 ppb. During the 13-year period there is a general decrease of the RMS, probably due to emission decrease, as photochemistry is more sensitive to meteorological variability, and thus to meteorological errors as precursor emissions are higher. The CTRL and TREN simulations, both based on year-varying emissions, have a comparable best skill. The FIXE simulation has a clear negative bias in the early nineties, resulting in a larger RMS error, a first sign of a real impact of emission reductions on O_3 daily maxima.

3. Time Evolution of the 10th and 90th Percentiles

[12] As noticed by *Roemer et al.* [2003], the observed downward trend of O_3 is better captured in high percentiles of the distribution (Figure 2a). During the study period, the 90th percentile decreases by about 10 ppb. A trend of similar amplitude is obtained in the simulations with yearly changing emissions (CTRL), and no trend is found in the FIXE simulation. CHIMERE systematically underestimates the 90th percentile by at least 5 ppb. This discrepancy is not surprising as high percentiles are generally reached in reality in concentrated structures having sizes smaller than the model grid size (50km), like city or power plant plumes.

[13] The simulated 90th percentile difference evolutions (Figure 2b) display a clear, statistically significant positive tendency of 0.65 ppb/year ($p < 0.01$) for the FIXE simulation, while the slight negative tendencies (respectively -0.18 ppb/year and -0.07 ppb/year) for the CTRL and TREN simulations are not statistically significant ($p > 0.1$).

[14] The evolution of observed lower percentiles (bottom curves of Figure 2) does not exhibit any trend, and is not

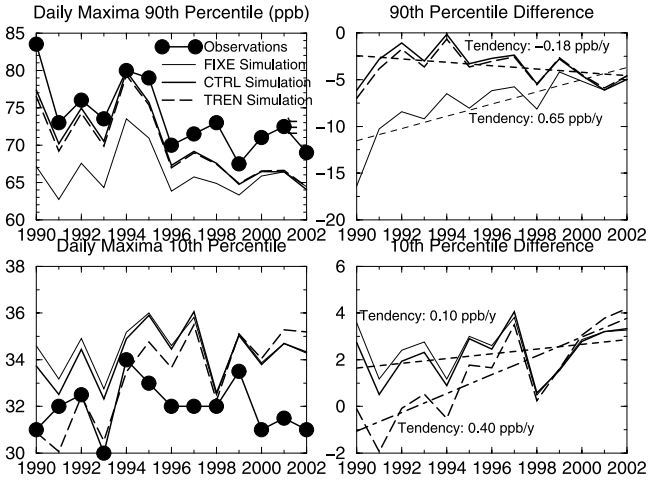

Figure 2. (left) Time evolutions of the simulated and observed O_3 daily maxima 90th and 10th percentiles (considering all stations), in ppb. (right) Differences (simulation−observations) of the percentiles, with regression lines.

sensitive to emission changes. The 10% percentile differences do not show either a significant tendency (respectively 0.03 ppb/year and 0.10 ppb/year). However the trend imposed in O_3 boundary conditions leads to an equivalent tendency in differences of the 10% percentile (0.40 ppb/year, $p < 0.001$). The model overestimates the O_3 daily maxima 10% percentile by about 3 ppb, which could be due to model deficiencies on physical processes.

[15] From Figure 2 we conclude that the highest daytime O_3 values are sensitive to emissions and insensitive to increasing ozone at boundary conditions. Most likely, high O_3 concentrations are obtained in episodic stagnant weather conditions where transport time is larger than deposition time. During these episodes, the O_3 formation results from local or regional photo-chemical production. In cloudy and windy conditions the situation is reversed: O_3 molecules largely come from outside Europe, which explains why the 10th percentile sensitivity to boundary conditions rather than to regional emissions. However we cannot reject the possibility that model deficiencies make its low percentiles too sensitive to boundary conditions at inland stations, explaining the discrepancies for the trended simulation.

[16] Of the three simulations CTRL is the one that best fits the observed 10th and 90th percentiles. Assuming no major model errors, one concludes that, (i) a uniform baseline O_3 increase of 0.4 ppb.year^{-1} at the whole domain boundaries is not consistent with observations, and (ii) the EMEP inventory emission changes, during the 13 year period, are consistent with the O_3 observations over North-Western Europe, on average. Further results shown in this paper only use the CTRL simulation.

[17] So far we have considered trend statistics for all stations taken together. The tendencies of the biases in 90th percentiles, calculated for each station, exhibit an interesting regional pattern (Figure 3a). Over the UK a marked positive tendency of about 1 ppb/year is found in the simulation-minus-observation difference. Figures 3b–3c show that, over the UK stations, the model strongly underestimates the percentile in the earlier part of the period (by about 5–15 ppb) while reasonable agreement is found in summer 2000 to 2002. Over Germany and central Europe, the agreement is rather found at the beginning of the period and a stronger underestimation is obtained at the end.

[18] Figures 3a–3c show that there are areas where the O_3 daily maxima behaviour is not consistent with the evolution of the EMEP emissions: the emission reduction would be larger than in the reported inventory for the UK and smaller over Germany and central Europe. Furthermore, the early nineties UK emissions would be underestimated in the inventory whereas central European emissions would be rather underestimated in late years.

[19] A similar analysis is carried out with nitrogen dioxide observations using daily averages instead of daily maxima (not shown) as given by *Jonson et al.* [2005]. For most of the NO_2 sites (located in Central Europe) a negative tendency is found in the model-minus-observation difference, indicating once more that the downward trend in reported EMEP emissions in central/northern Europe may be too large. Such regional differences are also consistent

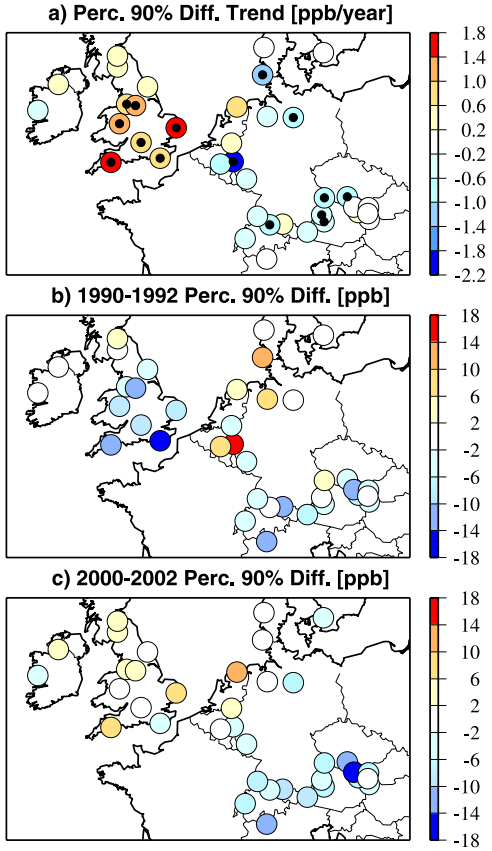

Figure 3. (a) 1990–2002 O_3 daily maxima 90th percentile bias (simulation minus observation) tendency at each station used in ug/m3/y. Stations where tendencies are significant at the $p <= 0.1$ level are marked with a solid circle inside. (b) 90th percentile of O_3 daily maxima bias (simulation minus observation) averaged over the summers 1990 to1992 in ppb; (c) As Figure 3b but for an average over the summers 2000 to 2002. For this period many more EMEP sites have O_3 observations than for the early nineties.

with the results of *Jonson et al.* [2005] and the inverse modelling of emission study of *Konovalov et al.* [2005].

4. Discussion and Conclusions

[20] Using a tendency analysis of the simulated-minus-observed differences in daily O_3 maxima 90th percentiles we found that the decadal evolution of the emissions of the EMEP inventory is quantitatively consistent with the observations when all available European observations are taken together. This can be considered as a verification of the ensemble of reported national emission estimates made in the framework of the international Convention on Long-Range Transboundary Air Pollution (CLRTAP). However when considering individual stations we found that observed 90th percentiles have a larger decrease rate than simulated over the UK, while the reverse occurs over Germany and central Europe. This behaviour probably results from an underestimation of precursor emissions in the early nineties in the UK. By contrast over Germany and central Europe the downward emission trend is overestimated in our simulations, suggesting too low inventory emissions in the latest years.

[21] This regional contrast gives one confidence that there is no systematic deficiency of the model regarding the sensitivity to emissions. However the specific climate of the UK and its upwind geographical location call for further discussion of our results. This maritime climate makes the model there less sensitive to errors in surface physics than over continental Europe. Oversensitivity to emissions may thus be due to a lack of dispersion over land, such as too thin daytime boundary layers over land or too weak winds. Although we cannot reject this possibility, it is to be noticed that MM5 winds are nudged to the global ERA40 analyses, themselves strongly constrained by radiosoundings and surface observations. Percentile difference tendencies over Western maritime Europe could not either be biased by a possible upward trend in Atlantic lower-atmosphere O_3 concentrations as it would oppose the general O_3 decrease.

[22] On average, the emission reductions during the 1990–2002 time period led to an O_3 maxima decrease that are consistent with the observations made at several sites over Northwestern Europe, which is the main result of this article. The model underestimates the impact of emission reductions over the UK and overestimates it over Germany and Central Europe, a hint that our above main result may not be valid on a country basis.

[23] Finally a side result of our study is that applying an artificial strong trend (0.4 ppb/year) all along European boundaries has no impact on strong episodic O_3 maxima, but leads to simulated low percentiles that are not consistent with observed ones.

[24] **Acknowledgments.** We are particularly thankful to Valérie Thouret for interesting discussions. The study was supported by the French national research program "Programme National de Chimie Atmosphérique" of the National Centre from Scientific Research.

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M. Beekmann, Laboratory Inter-Academic of the Atmospheric Systems (LISA), Faculté des Sciences et Technologie, 61 avenue du Général de Gaulle, F-94010 Créteil Cedex, France.

D. A. Hauglustaine, S. Szopa, and R. Vautard, Laboratory of Sciences of the Climate and Environment (LSCE)/IPSL, CEA/CNRS/UVSQ, F-91191 Gif sur Yvette Cedex, France. (robert.vautard@cea.fr)

L. Menut, Laboratoire de Météorologie Dynamique (LMD)/IPSL Ecole Polytechnique, Route départementale 36, F-91128 Palaiseau Cedex, France.

M. Roemer, Netherlands Organisation for Applied Scientific Research (TNO), Laan van Westenenk 501, NL-7334 DT Apeldoorn, Netherlands.

L. Rouil, Institut National de l'Environnement Industriel et des Risques (INERIS), Parc Technologique ALATA, BP 2, F-60550 Verneuil-en-Halatte, France.