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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A modelling study of photochemical regimes over Europe: robustness and variability" *by* M. Beekmann and R. Vautard

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Reply to the first referee We thank the referee for his constructive remarks which led, we hope, to a significant improvement of the paper. Among the many suggestions made by the referee, the idea to introduce more objective metrics to better characterise the chemical regime in a given region was very helpful in making the paper more quantitative.

General comments A) Model evaluation : 1. Question of specific model evaluation The model version used for this study is the version V200501. It is the same version than the one used in the studies of Vautard et al. 2006 and 2005 for summers 2001/2002 and August 2003, and in Van Loon et al., for summer 2001. Also input data used in our and the Vautard et al. studies are equal: for meteorology MM5 (Dudhia, 1993, version 2.3) simulations forced by ECMWF analysis, EMEP emissions for years 2001

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and 2002 (Vestreng et al., 2005), climatological chemical boundary conditions from the LMDZ/INCA model (Hauglustaine et al., 2004). Changes in later versions of the CHIMERE model concerned in particular the aerosol module (but which was not activated for this study), parallelisation, and the output format (Netcdf). In Vautard et al., 2006, daily ozone maxima in summers 2001 and 2002 from CHIMERE simulations have been compared to observations rural EMEP sites: bias with surface ozone is below 2 ppb, correlation is 0.82 and 0.78 respectively, RMSE is 8.5 ppb for both years. These results are consistent with the Honoré et al. evaluation study for 2006, but with NCEP / MM5 meteorology. In this latter study, bias for daily ozone maxima with rural AIRBASE sites was on the average below + 1 ppb, correlation 0.83 and RMSE 9 ppb. These model evaluations are judged as very satisfying. As suggested by the referee, in order to make sure, that referenced former comparison exercises are relevant for the current study, model version and input data used, and period treated, will be specifically mentioned in the revised version of the text. We also put more emphasis on evaluations for summers 2001 to 2003 corresponding to the years studied here.

2. Question about evaluation of “indicator” species evaluation We agree with the reviewer that a model evaluation with precursor and indicator species is desirable. However, observational data sets with precursor or indicator species, that would be representative on a regional/continental scale, are scarce over Europe. The most useful comparison with precursor species for the continental scale simulations is with satellite data, which show good spatial coverage of the continental scale model domain. An important comparison has been made between satellite and model derived tropospheric NO2 columns (Konovalov et al., 2005). The bias between simulated and observed tropospheric NO2 columns, averaged over Western Europe and summer 2001, was about -10%. For individual grid cells, normalised root mean square error for summer 2001 averages is about 30%. Indeed, the tropospheric NO2 column variability is strongly forced by that of boundary layer NO2 concentrations. For VOC species, satellite derived formaldehyde columns have been used for comparison with CHIMERE simulations (Dufour et al., 2009). Formaldehyde is an oxidation product of many VOC species,

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but its summer column variability over Europe is mostly constrained by that of biogenic VOC emissions. The normalised root mean square for differences between observed and simulated columns (summer 2003 averages) is about 25%. These comparisons with precursor species show are satisfying, but can not guarantee that the models O3-NOx-VOC sensitivity is correct. In particular, the uncertainty in anthropogenic VOC concentrations and emissions can not be assessed from these comparisons. To test the impact of uncertainty in anthropogenic VOC emissions, a dedicated sensitivity experiment was already included in the initial version of the paper. For indicator species, no direct comparison has been performed at continental scale, due to the absence of suitable observations. During the ESQUIF experiment in the Paris region in summer 1999, the O3/NOz and O3/NOy slopes simulated with a high resolution version of CHIMERE were low compared to the observed ones (Sillman et al., 2003). Based on this finding, the authors suggested a possible bias of the simulated chemical regime towards VOC sensitivity. Other comparisons with indicator species (for other models than CHIMERE) have been performed in particular in the frame of the LOOP campaign over the Po valley in summer 1997 (e.g. Martilli et al., 2001). However, these results for the Paris and Milano regions are relative to local conditions in a particular region and can not be extrapolated to a continental scale. As a conclusion, a comparison of simulated and observed indicator species representative for a continental scale is currently not possible for Europe. The continental scale comparisons with satellite data and the local scale ones during the ESQUIF experiment are briefly summarised in the revised text.

B) Robustness of chemical regimes 1. Differences between different ozone targets 2. Detailed analysis of transition areas 3. Use of objective statistics The reviewers remarks have been taken fully into account and are found to be very helpful. Differences in the chemical regimes for different targets are now analysed in a more quantitative way. This allows both to confirm the similarities between chemical regimes for different targets, but also to better identify differences. Following the referees suggestion, this analysis is carried out for the three regions already used in the initial version of the pa-

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per: North-Western Europe with an always VOC sensitive regime, the Mediterranean region with a NOx sensitive regime for most of the grid cells, and North-Eastern Germany, which has been chosen a s a transition region. For each of these regions, the following metrics characterising the chemical regime were calculated: its average along with the standard deviation, its minimum and maximum, and the fraction of grid cells with a VOC sensitive chemical regime. These data are presented in a new table 1. The following major results can be drawn from this analysis: • three target species show a more NOx sensitive regime than the daily maximum ozone (O3max) target: the daily maximum OX (O3 + NO2) OXmax, AOT 60 and AOT90; • three others show a more VOC sensitive regime : daily average ozone (O3moy), AOT40, SOMO35 (excess of daily maximum 8 h mean above 35 ppb); • for the North-Western Europe region, all targets except AOT90 show a VOC sensitive regime for all grid cells, for the Mediterranean region all targets except AOT90 show a NOx sensitive regime. This confirms and underpins the qualitative statement made in the paper about the robustness of chemical regimes with respect to the target; • differences are most important for the transition region in North Eastern Germany and are most manifest in the metrics “fraction of grid cells with a VOC sensitive regime”. This fraction is 0.36 for O3max, but zero or near zero for OXmax, AOT 60 and AOT90. It is respectively 0.97, 0,59 and 0.48 for O3moy, AOT40, SOMO35. Thus, the groupings of targets already noticed for the other region are confirmed in this third region. The text in section 3.1 (from Page 1530, line 21 “Next, we investigate how the chemical regime depends on the considered target”) is reorganised in the revised version in order to emphasize groupings and differences between different targets, along the lines given above. It is concluded, that for all targets, except partly AOT90, the chemical regimes are robust with some differences, except for the transition region.

C) Indicator species/ratios 1. Time period for indicator species analysis. The analysis was indicator species and ratios was undertaken for simplicity for a subset of data only (summer 2001), but results are very similar for the whole three year period. Another reason was to restrict the number of points (corresponding to days) in Figure 7 to about

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120, which still assures a good readability of figures.

2. Chemical regime vs. indicator scatter plots; how are indicator thresholds determined? Scatter plots for the three indicators NOy, H2O2/NOz and O3/NOz are added as suggested by the referee (new figure 3), in order to show the spread between individual points (representing grid cells). This backs up the old Table 1, where general statistical properties for the relation between chemical regimes and indicators are given (correlation, thresholds). In the revised version, a sentence was added to better explain, how the threshold values are determined : “A threshold value of 7.6 ppb can be determined which optimally discriminates about 83% of the chemical regimes (i.e. 83% of the grid cells assigned to a particular regime as a function of the indicator really indicate the correct chemical regime, while 17% indicate the opposite one).”

3. Impact of model resolution on differences in the critical indicator values between this study and some other existing studies? We think that differences in the horizontal model resolution are not a major reason for differences in the indicator thresholds. While the limited model resolution in our work does not allow properly resolving chemical regimes for isolated urban areas, it is appropriate for regions with a suite of large and neighbouring agglomerations like the one of roughly 500 x 300 km comprised by South-Eastern England, Benelux and the Rhine Ruhr area. The threshold values of the indicator species derived in this study are determined by those values occurring at the edge of this large region. The studies of Sillman (1997) and Martilli et al. (2001) are relative to a smaller scale, typically of several tenths to 100 km around an agglomeration. Thus, differences in the thresholds are expected to be due to the different scale of the problem, rather than due to model resolution. A larger scale possibly decreases the NOy threshold and increases the O3/NOz threshold through rapid HNO3 dry deposition. This has been pointed out already in the initial version of the paper and is emphasized in the revised version.

4. Suggestion to move the discussion on the day-to-day variability in indicator

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species/ratios to 3.2 rather than at where it is in 3.3. The authors wish to keep this plan as it is. The rationale of this plan is to first present summer average properties of chemical regimes in section 3.3 and then time variability in 3.3, starting from discussion of year to year differences in the chemical regimes and going down to day to day differences.

C) Variability (inter annual, seasonal, and day-to-day) 1. Is particular three-year period (2001 - 2003) is a fair representation of inter-annual variability? 2. Discussion for the three selected areas/sub-domains should be added. The referee questions, if summers 2001 to 2003 chosen for this analysis are representative for a longer time scale. The general answer, based on surface ozone observations, is that summers 2001 and 2002 were typical, but summer 2003 was exceptional: • Konovalov et al., 2008 calculated summer average 90% percentiles of daily ozone maxima for 36 EMEP sites representative for European rural conditions for summers 1996 to 2004. Summer 2003 ranked in the first position (146 μg/m3), summers 2001 and 2002 ranked in the 4. (132 μg/m3) and 5. position (130 μg/m3) out of 9 summers. • An EEA report (Air pollution in Europe 1990–2004, European Environment Agency Report No 2/2007) analyses European surface ozone observations again for years 1996 to 2004; SOMO35 values (see explanation above) are very similar for all years (about 6000 μg/m3 day), except for 2003 (about 8000 μg/m3 day). Over both periods 1990 to 2004 and 1996 to 2004, trends were insignificant. Given these differences between the extreme summer 2003, and the typical summers 2001 and 2002, it is interesting to analyse if significant differences in chemical regimes can be observed. In section 3.3, page, line 1534, line 9, we stated : “Differences between average results for summers 2001, 2002 and 2003 are rather weak, below 1 ppb in absolute terms and below 20% in relative terms for O3max. …… Thus the restriction to analysis of a three-summer climatology appears to be justified for this work.” This statement can be better quantified by stratifying the chemical regime statistics using the metrics defined above for different years. For all three analysed regions, characteristics for chemical regimes only span a small range of values for different years

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(May – August 2001, 2002, 2003) for O3max : • over North-western Europe, 97 – 98 % of the grid cells show a VOC sensitive regime with an average between +3.65 – +4.06 ppb, • over the Mediterranean Sea, 97 – 99% of the grid cells show a NOx sensitive regime with an average of -1.92 – -2.40 ppb, • over North-Eastern Germany, the range is somewhat larger, 57 – 73% of grid cells show a NOx sensitive regime with an average of -0.09 – -0.42 ppb; for this region, the hot year 2003 turns out to be slightly more NOx sensitive than years 2001 and 2002. Thus, the initial statement keeps valid in light of the new analysis. In the revised version, we refine the initial discussion : • by stating that summers 2001 and 2002 were typical, but 2003 extreme, making reference to Konovalov et al., 2008, and EEA, 2007. • by making the initial statement about summer to summer differences in the chemical regime more quantitative by citing the above given numbers. Also differences in the chemical regimes for different months are again analysed in a more quantitative way, using the same regions and metrics as for the discussion of differences with respect target species. A new table 3 is added. Results about the robustness of the chemical regimes with respect to months over North-West Europe and the Mediterranean region are confirmed. A less pronounced VOC sensitive regime over North-West Europe in May and a more pronounced NOx sensitive chemical regime for the Mediterranean region in August (both already mentioned in the discussion paper) are also confirmed. A new feature coming out from this analysis is the transition from a VOC sensitive regime over North-Eastern Germany during May to July over most of the grid cells to a NOx sensitive regime in August. This has been added to the revised text.

D) Decadal variability: Simulation does not take into account future climate/meteorology. The following sentence has been added at the end of section 3.4. “It has to be kept in mind that changing meteorological conditions also could influence ozone concentrations and the chemical regime. Taking these complicated effects into account is beyond the scope of this paper.”

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E) Impact of model uncertainty 1. Choice of choosing pertinent sensitivity tests 2. Is VOC emission uncertainty of 40% across the board? 3. On the sensitivity study between extended and condensed MELCHIOR mechanism may not be a fair representation of variability in chemical mechanisms ? 4. Sensitivity to vertical mixing/transport

The authors admit, that performing a completely systematic uncertainty analysis of chemical regimes was beyond the scope of the paper. Instead, we choose several sensitivity scenarios that we thought to be representative to get a general impression of model uncertainty with respect to chemical regimes. The three particular experiments were chosen, because: • changes in VOC emissions directly act on the VOC/NOx ratio which is one of the well recognised driving forces for chemical regimes in polluted areas (e.g. Sillman, 1999, and references therein). Also, it has been pointed out before that the evaluation of a continental scale model with respect to VOC emissions and concentrations is difficult, leaving us with the necessity to perform sensitivity tests with different emissions. • The 40% uncertainty in VOC emissions is “across the board”. It represents a typical uncertainty in emission inventories as explained in Beekmann and Derognat, 2003 and in Deguillaume et al., 2007. The estimation comes from expert hearings (Hanna el al., 2001), from comparison of different independent emission inventories, from specific uncertainty analysis and from specific emission inversion exercises over Augsburg (Mannschreck et al., 2001) and over Paris (Vautard et al., 2003). As this estimate is in itself uncertain, and as estimates for specific sources would be even more uncertain, it was felt that a simple sensitivity experiment with an “across the board” perturbation makes most sense in this work. • Testing differences in behaviour of chemical mechanisms would have been interesting, but was beyond the scope of the paper. Here, we simply intended to test, if the reduction hypotheses for the condensed with respect to the extended MELCHIOR code, in particular the use of the operator concept (Carter et al., 1990) impacted results of the study (which was not the case). However, we have tested the impact of using different chemical mechanisms in an earlier work (Beekmann, unpublished results): in EKMA plots with three chemical mechanisms (MELCHIOR-

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extended, EMEP, RACM), differences in the chemical regimes were significant under urban polluted conditions. The interesting observation was that these differences corresponded to a shift in of the magnitude of VOC emissions of about 30%. Thus it seems that under some conditions, a “chemical mechanism” experiment can be replaced by an “emission experiment”. • As stated in the text, comparing results for the first (0-50 m height) and second model layer (50 – 200 m height) intended to identify regions where uncertainty in vertical mixing is important. When differences between the two layers are negligible, vertical mixing is strong (case of the continental well mixed boundary layer), and uncertainty to the formulation in vertical mixing is expected to be weak (i.e. also for somewhat weaker and stronger mixing, the concentration profile would be the same). However, for the case of strong differences between the two layers (observed especially over large water surfaces, even during afternoon), the impact of uncertainty in vertical mixing is expected to be important. Thus the differences between the two layers are used as a kind of qualifier for regions where important uncertainties in vertical mixing are expected, but without pretending to quantify them. These regions turn out to be rather limited.

Corresponding to the reviewers request, the initial statement of well chosen sensitivity experiments was put to a lower level in the revised manuscript (end of 1. paragraph of section four, bottom of page 1538): “In the present work, as a first approach the evaluation of model robustness is simply made by performing a certain number of specific sensitivity simulations”. Justification for sensitivity experiments, following the above discussion, has been extended in the revised text.

Specific comments: Page 1523, line 22: “using twin simulation experiments” - explain or reference. The term “twin experiments” has been better explained in the text :

Page 1526, line 15: MM5 simulation time too long ? The domain for MM5 is about 3000 km large both in West-East and South-North direction. In order to avoid divergence during the 126 hours of simulation, MM5 is constrained by large scale fields (winds,

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temperature, humidity), through grid nudging with a three to six hour time step.

Page 1527, second paragraph: model evaluation As suggested by the referee, in order to make sure, that referenced former comparison exercises are relevant for the current study, model version and input data used, and period treated, will be specifically mentioned in the revised version of the text (see our response to general remarks).

Page 1530, line 25 - 27: The sentence is unclear. The sentence is made clearer by replacing with the following (corresponding also to the referees argument): “This (i.e. D_Ox $<$ D_O3 over the titration region) results from the fact that ozone titration with fresh NO emissions adds to a VOC sensitive regime for O3 (through O3 loss with NO), but is neutral for Ox (following the Ox definition as O3 + NO2). The effect is most pronounced in regions with large NO emissions as in North-Western Europe.”

Page 1530, last two lines. As suggested by the reviewer in the general comments, the comparison between the chemical regimes for different types is put on a more quantitative basis.

Page 1531, last paragraph, discussion on AOT90. As explained above, the presentation of results for AOT90 has been put on a more objective basis using statistics for three selected zones. It confirms the basic result of a VOC sensitive regime for North-Western Europe. For the Mediterranean region, it confirms the NOx sensitive regime at least in the Genoa Golf, but for the rest of the region AOT90 is zero, so the chemical regime is undefined for the AOT90 target. For the transition region over North-Eastern Germany, AOT90 is also close to zero for most of the grid points. On the basis of this, the text is rewritten putting more emphasis on the differences between AOT90 and other target species.

Page 1533, line 3 - 4: What do you mean by “the information on the chemical regime is lost faster than the regime itself”? It means that, because NOy dry deposition is faster than ozone dry deposition, the chemical regime tracer (NOy) is less well conserved in time than the chemical regime itself. As the sentence in page 1533 may be misleading,

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it was replaced by the following text simply stating that the NOy indicator value tends to get lower with time for a given chemical regime due to rapid HNO3 deposition : “a larger domain considered in our study making loss of the NOy tracer through rapid HNO3 dry deposition more critical (i.e. deposition processes tend to lower the NOy indicator value for a given chemical regime, because ozone deposition, the major PBL ozone loss, is slower than NOy deposition)”

Page 1533, last paragraph (carrying over to page 1534): reference to Figure 3d; difference between the threshold value found here and that of Martilli et al. (for Po Valley region) mainly due to the difference in model resolution? Reference to Figure 3d is added. As we have argued above, we think that a difference in the horizontal model resolution is probably not a major reason for differences in the indicator thresholds. We further argued, that the indicator thresholds determined in our study are mainly forced by the transition of a VOC sensitive chemical regime over North-Western Europe to a NOx sensitive regime in the surroundings, and that this transition is well taken into account in our model with a typical resolution for continental scale models. However, we have to agree then with the reviewer that our derived indicator thresholds do not have predictive character for chemical regimes over individual agglomerations like the Milano area, for which our model resolution is too coarse. To make this clear, we added a cautionous note in the conclusion paragraph of section 3.2 : “However, it has to be kept in mind that thresholds derived in our work are relative to larger scale features, in particular to differences in chemical regimes in North-Western Europe and the regions around, and that these thresholds can not be directly applied to urban plumes.”

Page 1534, line 12 - 13: Justification for restriction to analysis of a three-summer climatology. This point has been discussed already above, supporting the restriction to at three summer analysis.

Page 1534, line 22 - 28: Reasons for more NOx-sensitive over the Mediterranean region found in August. Bulk numbers for average H2O2 and NOy concentrations during August, as compared to the rest of the summer are given in the revised text, and

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support both larger H2O2 and lower NOy in August over the Mediterranean region.

Page 1535, line 25: The readers should be reminded, that this analysis is for summer 2001. This is done; either in the text and in the figure 6 caption.

Page 1536, last paragraph: discussion on whether high ozone days are associated with a particular O3-NOx-VOC sensitivity. We kept in the text the finding of different more NOx sensitive chemical regime for high ozone days as this is important for air quality issues, as agreed also by the referee. To take into account its remark on “across-the-board” reductions in emissions, we put a reminder in the text about this fact. Clearly, it is understood and pointed out several times in this paper that flat continental emission reductions are tested and not local ones. However, also the impact of continental emission reductions is an air quality issue ! We added the following sentence: “This is an important issue for air quality management, although it has to be pointed out, that emission reductions applied in this study are flat over the whole European domain.”

Following the referees recommendation, we removed much of the explanatory part concerning the chemical regimes / ozone dependence, and introduced the potential importance of transport patterns in the revised text: “Giving an exact explanation for these features is difficult. Clearly, enhanced ozone values trigger enhanced radical production by ozone photolysis, and may shift the chemical regime to more NOx sensitive. However, days with large ozone values may also be indicative for particular transport patterns and source regions, triggering particular chemical regimes.”

Page 1537, line 7 - 8: statement is not clear. The statement has been replaced by the more specific statement: “Finally, the chemical regime can also depend on ozone itself. Especially for North-Western Germany and to less extent for the Mediterranean region, the regime is more NOx sensitive in presence of large ozone levels.”

Page 1538, line 7 - 9: Needs a qualifier here that this sensitivity study only reflects the

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projected changes in emission under the current climate. The required qualifier has been added.

Page 1538, line 26: Suggest removing “well chosen” (see general comments above regarding impact of model uncertainty). OK, this is done.

Page 1541, line 1 - 2: “Chemical regimes appear also robust over various time scales” statements needs to be refined for transition regions.. Yes, we agree that the statement made was too strong. We modified it as follows: “Chemical regimes appear also robust over various time scales (year to year, month to month, day to day) for regions with pronounced VOC sensitive (North-West Europe) or NOx sensitive (Mediterranean region) chemical regimes. However, for transition regions as North-Eastern Germany, both month-to-month and day-to-day differences are strong.”

Page 1541, 3rd paragraph: Need to state the limitations in the sensitivity tests. We added a note on the limitation of sensitivity analysis : “The robustness of chemical regimes with respect to model uncertainty (in emissions, chemistry, vertical transport) has also been assessed, without claiming to address overall model uncertainty of the global model or of specific modules.”

Technical corrections (typo, figures): Items have been corrected as suggested by the referee.

Indeed, colour scale in Figure 2j is slightly different than for other figures. This was done on purpose, to have white colour for a chemical regime close to zero. For this case, AOT90 is in general zero itself and the chemical regime undefined.

Three regional boxes are added to all chemical regime plots, as asked be the referee. Political boundaries are not strongly emphasized in the figures, because they are not of major interest for the paper. Depending on the colours used, political boundaries are indeed sometimes difficult to discern, but can clearly seen on other figures. Since they are identical for all figures and since they are not the major focus of the paper, this

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should be all right.

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