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

Anonymous Referee #1

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Reviewer's report on the manuscript by Beekmann and Vautard (2009) 'A modelling study of photochemical regimes over Europe: robustness and variability', ACPD-2008-0463

This manuscript reports a modelling study of O₃-NO_x-VOC sensitivity over Europe using a regional CTM, CHIMERE, at 0.5 deg x 0.5 deg resolution. The study looks at the geographical distribution of the different O₃-NO_x-VOC sensitivity regimes, with respect to various ozone objectives/targets, and examines relationship between the O₃-NO_x-VOC sensitivity and the various commonly used indicator species/ratios both in the context of Europe as a whole and for different regions. The authors also attempted to assess the changes in O₃-NO_x-VOC sensitivity due to the changes in emission during the recent past and the projected near future over Europe. While I think that the work

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is topical particularly for policy applications, I do hope that the authors will be able to respond to the comments that I have in the following.

General comments:

Model evaluation

1. Although the authors referred to several existing evaluations of model simulations using CHIMERE, it is not clear whether the evaluations were done for the same base simulation carried out for this study and whether the same model configurations were used in the existing model evaluations. It would be necessary to include some model evaluation results (e.g., a plot of modelled averaged O₃ daily max., Figure 1a, compared to the same from observations) or to provide specific reference for the model evaluation of the base simulation used for this study.
2. It would also be important to provide the readers with some sense as how the model is able to simulate ozone precursors and the 'indicator' species, if possible, when discussing the O₃-NO_x-VOC sensitivity based on model results.

Robustness of O₃-NO_x-VOC sensitivity regimes with regard to different ozone targets

1. While the authors showed general similarity in the regions of mostly NO_x- or VOC-sensitivity amongst different ozone targets, it can be seen that there is a distinction between the targets which reflect peak and/or more extreme ozone values (O₃ daily max., O_x daily max., AOT₆₀, and AOT₉₀) and those which reflect more of an average (O₃ daily mean, AOT₄₀, and SOMO₃₅).
2. Most of the uncertainty in this O₃-NO_x-VOC analysis is the transition region from VOC-sensitive area (mostly urban, source) to NO_x-sensitive area (more rural). The authors focused mostly on the area of mainly VOC- or mainly NO_x-sensitivity but did not address the variability in the transition area for different ozone targets.

3. It would be helpful for the discussion on robustness (and on variability also) if some suitable statistics were carried out (mean, median, standard deviation, probability dis-

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tribution, for example) for selected sub domains (e.g., the three regions that the authors have focused their discussions on).

Indicator species/ratios

1. The analysis concerning indicator species/ratios is done for 2001 only while the chemical regime analysis is done for the three year period. Is there a reason for this?
2. It would be good to do some scatter plots where one can get a sense of the spread/range rather than just single values for thresholds. How are those single value thresholds determined?
3. How much of the difference in the critical indicator values between this study and some other existing studies can be attributed to the difference in model resolution?
4. It may be better to move the discussion on the day-to-day variability in indicator species/ratios to 3.2 rather than at where it is in 3.3.

Variability (inter annual, seasonal, and day-to-day)

1. The inter-annual variability is examined based on the simulations of the three summer seasons. Is there a longer term analysis on ozone over Europe available (based on monitoring data), so that one can get a sense as whether this particular three-year period (2001 - 2003) is a fair representation of inter-annual variability?
2. Again, it would be helpful for the discussion if statistics is calculated for the three selected areas/sub-domains so that the comparison can be more qualitative/objective (see comment # 3 under 'robustness' above).

Decadal variability

1. When discussing the model-predicted O₃-NOx-VOC sensitivity over Europe for the projected future emission, it is important to state that the simulation does not take into account of future climate/meteorology, which may also have an impact on the regional distribution of O₃-NOx-VOC sensitivities.

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Impact of model uncertainty

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1. The authors looked at three areas of model uncertainty: anthropogenic VOC emission, chemical mechanism, and vertical model layers (i.e., the second lowest, 50 - 200 m, versus the lowest, 0 - 50 m). Do they represent the most important model uncertainty (concerning O₃-NO_x-VOC sensitivity)? Why are these three chosen to be the focus (the authors stated in the manuscript that these were 'well chosen sensitivity simulations' but said nothing to back this statement up)?
2. With regard to VOC emission, the sensitivity run involves increased anthropogenic VOC emissions across the board by 40%. Does this represent the typical uncertainty in emission inventory? Does the uncertainty mainly come from certain emission sectors or particular groups of VOC (which would not be across the board)?
3. On the sensitivity to chemical mechanism, the current study is limited to looking at the comparison between the more condensed MELCHIOR mechanism and the more extensive one which MELCHIOR is derived from. This may not be a fair representation of the variability in chemical mechanisms stemming from different VOC lumping methodologies.
4. Regarding the sensitivity to vertical mixing/transport, the model vertical resolution and diffusion scheme may be most important. Deeper model layer will effectively enhance mixing which may affect the O₃-NO_x-VOC sensitivity particularly closer to the sources. Simply looking at the second model layer (50 - 200 m) versus the first (0 - 50 m) does not reflect fully the impact of vertical mixing/transport.

Specific comments:

Page 1523, line 22: 'using twin simulation experiments' - explain or reference.

Page 1526, line 15: MM5 simulation is carried out by pieces of 5 days and 6 hours - how big is the domain for the MM5 runs? It seems a little too long (126 hr) for regional simulation (though it is constrained somewhat by data assimilation).

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Page 1527, second paragraph: The evaluation of Honoré et al. (2008) is for 2004 - 2006; van Loon et al. (2007) is for 2001. Is the 2003 summer simulation (Vautard et al., 2005) the same as the present base case (i.e., model version, configuration, input, boundary condition, etc.)? This is related to the general comment (#1 under 'model evaluation').

Page 1530, line 25 - 27: The sentence is unclear. Is it meant to explain why the degree of VOC sensitivity with respect to Ox max is less than that with respect to O₃ max? Ox is supposed to mask the effect of O₃ titration by NO (since Ox = O₃ + NO₂). While, over the O₃ titration region, a reduction in NO_x would lead to an increase in O₃ (less titration), which would in turn contribute to a positive D_{O₃}, the same can not be said for Ox (i.e., less titration -> more O₃ but less NO₂). One would expect D_{Ox} < D_{O₃} over the titration region, everything else being equal.

Page 1530, last two lines: It is stated that the chemical regime structure with respect to O₃ daily mean is similar that for O₃ daily max, but one can notice a definite difference in the orientation of the transition regions in the two cases. Here (and following on to the next page) the discussion on similarity or difference between different O₃ target is rather subjective. It would be good to have some more quantitative measure (as suggested in the general comments).

Page 1531, last paragraph: The discussion on AOT90 is somewhat tentative. It is based on very few events, and to say that it resembles the chemical regimes for all the rest of O₃ targets is very subjective.

Page 1533, line 3 - 4: What do you mean by 'the information on the chemical regime is lost faster than the regime itself'?

Page 1533, last paragraph (carrying over to page 1534): Should make explicit reference to Figure 3d. Also, is the 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 or other factors? The author argues that one of the important findings from the

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study is the ability to use these indicators to distinguish chemical regimes at regional scale. This is only useful if the threshold values determined by the modelling study are reliable for inferring the O₃-NO_x-VOC sensitivity in the real world.

Page 1534, line 12 - 13: The statement 'the restriction to analysis of a three-summer climatology appears to be justified for this work' is not supported by any observational evidence (see general comment, # 1 under 'Variability').

Page 1534, line 22 - 28: On explaining why more NO_x-sensitive over the Mediterranean region found in August, the authors pointed out two contributing factors: increased H₂O₂ and lower NO_x emission. The latter may be the main factor, but for both it would be good to include some bulk numbers to back up the statement.

Page 1535, line 25: The readers should be reminded, either here or in the figure caption, that this analysis is based on the 2001 summer season.

Page 1536, last paragraph: I find the discussion on whether high ozone days are associated with a particular O₃-NO_x-VOC sensitivity somewhat misleading and confusing. The practical purpose of carrying out this analysis is perhaps to be able to say whether a particular control (NO_x vs. VOC) will be more effective on the high ozone days. However, the O₃-NO_x-VOC sensitivity here is based on across-the-board reduction in NO_x and VOC which is not necessarily equivalent to local emission reductions. For NEG region to have more NO_x sensitivity on high ozone days may be more indicative (or as a result) of a particular synoptic pattern (transport from a particular source region) than anything else. The arguments towards the end of the paragraph (line 23 and onwards) on the compensating factors are confusing and not particularly meaningful.

Page 1537, line 7 - 8: This statement is not clear. Particularly, 'as a function of the remote or polluted character of a region' - what does this mean?

Page 1538, line 7 - 9: Needs a qualifier here that this sensitivity study only reflects the projected changes in emission under the current climate.

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Page 1538, line 26: Suggest removing 'well chosen' (see general comments above regarding impact of model uncertainty).

Page 1541, line 1 - 2: 'Chemical regimes appear also robust over various time scales: year to year, month to month, day to day' - this may be true for the regions mainly VOC- (e.g., NWEU) or mainly NOx-sensitive (e.g., MED) but less so for the transition regions (e.g., NEG). Particularly, as seen from Figure 6, the chemical regime (defined as D_O3 in 2.2) varies widely from day to day for NEG region.

Page 1541, 3rd paragraph: Need to state the limitations in the sensitivity tests regarding model uncertainty carried out in this study, with respect to emission, chemical mechanism, and vertical transport (see general comments).

Technical corrections (typo, figures):

Page 1524, line 6: Delete the first 'agglomerations'?

Page 1541, line 3: '2008' should be '2003'?

Figure 2j: the colour scale seems odd (different from the rest).

Figures in general: It is difficult to see the political boundaries on many of the map plots. It may be helpful to add the three regional boxes to all the chemical regime plots.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 1521, 2009.

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