

Interactive comment on “Reanalysis of tropospheric sulphate aerosol and ozone for the period 1980–2005 using the aerosol-chemistry-climate model ECHAM5-HAMMOZ” by L. Pozzoli et al.

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We thank Referee #1 for his/her insightful and constructive remarks, which improved our manuscript. Below we try to answer the various remarks.

Anonymous Referee #1 Review of “Reanalysis of tropospheric sulphate aerosol and ozone for the period 1980- 2005 using the aerosol-chemistry-climate model ECHAM5-HAMMOZ” by Pozzoli et al. In this paper the authors presented results from two hind-cast simulations: one with varying anthropogenic emissions and one with constant

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emissions. They compared simulated results to measurements, in particular surface measurements over North America and Europe. I found this to be a very nice paper. Multiyear analysis of chemical hindcast simulations is a difficult undertaking. It involves sifting through an enormous amount of data, synthesizing the data, and presenting the results in a clear and cohesive manner. I think the authors succeeded extremely well in this. I found the analysis and presentation of these simulations to be clear and insightful. The figures and tables included in the paper were well chosen. They are easy to read, and yet present a great deal of useful information. The text was well written. I have a few comments and suggestions below, but would recommend this paper for publication in ACP with only minor changes.

1. I think the paper could have tied its results in better with some of the ozone trend work already published. While the authors addressed simulated and measured ozone trends globally, I feel they have not been very diligent in referencing some of the previous work on these trends. Some of the papers that come to mind is the work of Dan Jaffe and Dave Parrish in the Western U.S.; Jonson and others in Europe; Lamarque et al. in their recent ACC-mip simulations.

As suggested by the reviewer we compared our estimated trends over Europe and North America with recent studies. Despite the difficulty in comparing trends calculated with different methods and for different periods of time, our observed trends are qualitatively in good agreement with previous studies. In Section 4.2.1, where we described our findings on regional O₃ changes in Europe, we now refer to the studies of Lamarque (2010) and Cui (2011). In these studies were reported observed O₃ trends of 0.32–0.40 ppbv/year for stations in central Europe, which are comparable with the observed trends calculated in our study (see Figure A2 in the APPENDIX). In Mace Head Lamarque (2010) reported an increasing trend of 0.18 ppbv/year, comparable to our study (see Figure A2 in the APPENDIX). Nevertheless the annual trends from model simulations in our study are not statistically significant for almost the entire Europe. Statistically significant trends are found only for seasonal means (Figure 6

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and Table A1). In section 4.2.2, where we discussed O₃ changes in North America, we now compare the observed O₃ trends calculated in our study with previous findings. Over Western US the observed O₃ trends found in our study are in a qualitatively good agreement (see Figure 6, Table A1, and Figure A2) with the following studies: Oltmans (2008) observed positive trends at some sites, but no significant changes at others; Jaffe (2007) estimated positive O₃ trends of 0.21–0.62 ppbv yr⁻¹ in winter and 0.43–0.50 ppbv yr⁻¹ in summer; Parrish (2009) found 0.43±0.17 ppbv yr⁻¹ in winter and 0.24±0.16 ppbv yr⁻¹ in summer O₃ trends; Lamarque (2010) found annual increasing O₃ trend of 0.33 ppbv yr⁻¹; Chan (2010) found larger positive daytime O₃ summer and winter trends, close to 1 ppbv yr⁻¹, for the period 1997–2006. Over Eastern US our study qualitatively agrees with Chan (2010) who found decreasing significant trends over all Eastern US in summer, and mainly no significant trends in winter.

2. The meteorology was apparently changed from reanalysis to operational analysis in 2000. It would be surprising indeed if this has no impact on the meteorological variables. While I understand the authors had little choice, I think there is some cause for concern here. While the authors comment on it briefly later in the paper, I think the possible ramifications should be clearly addressed near the beginning of 10196 where the authors first discuss the model simulations.

We agree with Referee #1, and despite we have no evidence of discontinuities in our analysis, we have explicitly highlighted this point to warn the readers in Section 2 when describing the model simulations.

3. The simulated emissions were apparently kept constant after 2000, or were based on trends. In some locales there is now enough information to assess how good these projections are based on more recent inventories. I think this would be valuable. To what extent are the simulated emissions after 2000 consistent with more recent measurements? I don't think an extensive evaluation is needed here, but it would be helpful in interpreting the results if the authors could consider their emissions in the context of more recent results.

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We added a paragraph on the comparison between the anthropogenic emissions used in our study and the more recent inventory from Lamarque et al. (2010). We also included in Figure 2 the regional emission changes between 1980 and 2005 for the Lamarque et al. (2010) emission inventory which includes regional emission trends in Europe, North America and Asia derived from recent studies (e.g. Richter et al., 2005; Zhang et al., 2009; and Klimont et al., 2009). For the period 1980-2000, our global amounts of emission from anthropogenic sources, which are based on RETRO, are lower by more than 10% for CO and NO_x, and they are higher by more than 40% for VOCs, when compared to the new emission inventory prepared by Lamarque et al. (2010) in support of the Intergovernmental Panel on Climate Change (IPCC) Fifth Assessment Report (AR5). For the period 2001-2005 several studies have recently shown significant changes in regional emissions, especially in Asia (e.g. Richter et al., 2005; Zhang et al., 2009, and Klimont et al., 2009). In our study, compared to the projection for year 2005 of Lamarque et al. (2010), which includes the references cited above, we applied: significantly lower emission reductions between 1980 and 2005 for CO and SO₂ in EU; larger for BC and OC in both EU and NA; in EA the emissions changes are generally lower in our simulation, except for CO; in SA we applied similar emission changes except for NO_x and SO₂ (see also new version of Fig. 2).

4. Forcing a general circulation model (CAM) with interannual SST variations (10210:Line 20) as in Hess and Mahowald should still incorporate significant meteorological variability. Thus, I think it is rather speculative to assume that the rather low variability found in the Hess and Mahowald runs is due to the SST forcing. I think the conclusions (page 10220) raising questions about the applicability of this technique to future climate experiments are too strong. It will be interesting to see if this is a general result when more models use this type of simulation for chemical analysis.

We highlighted the differences in inter-annual variability by using different nudging methods and re-analysis datasets. In particular we noticed how our analysis, which use ECMWF ERA40 re-analysis data, is closer to the experiment of Hess and Ma-

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howald, which use NCEP re-analysis data. While prescribing only SSTs Hess and Mahowald observed generally lower inter-annual variability. By using a full nudging methodology the modelled dynamics in a GCM should be closer to the observed meteorology during the simulated period and therefore we believe it is also more consistent when we want to compare simulated chemical composition with observations. However, since we have not performed the SST simulations ourselves, we will reformulate the sentence under consideration, referring to the opportunities that the forthcoming ACC-Hindcast and ACC-MIP are offering to shed light on this matter.

5. I would have liked to see a little more discussion in the conclusions as to the general importance of meteorological variability on chemical constituents. The conclusions mention global meteorological variability, but not the importance of the meteorological variability on the chemical record. I think this was an important and interesting component of your study.

The role of meteorological variability on the chemical composition of the atmosphere is emphasized in different points of the conclusions: impact of meteorology on natural emissions; 75% of O₃ variability is explained directly or indirectly by processes related to meteorology; we found for OH a linear trend well correlated with lightning emissions. We will try to further improve the manuscript by more systematically identify when talking about meteorological variability and chemistry.

6. I think it would be helpful if you could summarize some of the aspects of interannual variability the model really does not capture well. While I think you addressed this somewhat, it would be helpful to have a better understanding of the unsolved problems that remain: what aspects of the observations do we really not understand?

We answered this question by extending the discussion in the beginning of Section 8 'Outlook'. We can state that generally models -even if they were perfect- underestimate variability, in particular if we compare to well resolved surface measurements. The reason is that the model averages spatially and temporally. Our re-analysis study

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showed that several of the overall processes determining the variability and trends of O₃ and aerosols are qualitatively understood- but also that many of the details are not well included. For example the variability due to stratosphere/troposphere O₃ fluxes is not included in our study, and the description of lightning and biogenic emissions are characterized by large uncertainties. We have seen that aerosol variability, and in particular sulphate, is mainly driven by emissions. Improving the description of anthropogenic and natural emission inventories and the description of secondary formation of aerosols, such as for secondary organic aerosols (SOA) not included in this study, may improve our understanding of aerosol variability.

Minor Comments:

-An additional very careful reading of the paper would be helpful. There are a number of places where words were missing or there is a grammatical mistake. Some of these are listed below along with other minor comments.

-Page 10193 Abstract, line 15: I found this sentence confusing. Are the numbers in parenthesis standard deviations? At this point it is not clear what you mean by natural and total variability. A little bit of explanation here would be helpful.

Sentence changed into: " Our model analysis indicates an increase of 1 ppbv (corresponding to 0.04 ppbv yr⁻¹) in global average surface O₃ concentrations due to anthropogenic emissions, but this trend is largely masked by the larger O₃ anomalies due to the variability of meteorology and natural emissions. The changes in meteorology and natural emissions account for the 75% of the total variability of global average surface O₃ concentrations."

-Abstract, line 20: "Ozone increases" should probably read "Simulated ozone increases"

Corrected.

-Page 10194 Line 23-24: "To our knowledge . . ." I would leave this sentence out or

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make it clearer. What do you mean by consistent?

The sentence was removed.

-Page 10195 Line 7: I think “antagonistic” is the wrong word here.

“Antagonistic” changed with “concurrent”

-Page 10196 Line 11: “Luca Pozzoli 27 March 2011 10:39 a.m” Line 26-27: “The relaxation technique . . . to retain . . . cloud and aerosol “. I’m not sure what you mean here.

Sentence changed into “The relaxation technique forces the large scale dynamic state of the atmosphere as close as possible to the re-analysis data, thus the model is in each time in a consistent physical state but it calculates its own physics.”

-10198: Line 15: “derived emission trends”. Please give where these emission trends are derived from. Also, I assume from figure 2, that the derived emission trends change on an annual basis. Please clarify in the text.

This question is already partially answered in lines 16-19. The sentence is changed into: “The emission ratios between years 2001-2005 and year 2000 from the USEPA (<http://www.epa.gov/ttn/chieftrends/>), EMEP (<http://www.emep.int/>), and REAS (<http://www.jamstec.go.jp/frcg/research/p3/emission.htm>) emission inventories were applied to year 2000 emissions used for this study over the US, Europe, and Asia.”

-10201 Line 6. Please change “from” to “are from”

Corrected.

-10203 Line 25. Discussion about the influence of model bias on model variability and trends: did I miss this discussion or was it omitted from the paper?

We removed the sentence as we left only few general considerations in Section 8,

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Outlook. (Page 10223, lines 16-23). It was indeed omitted from the paper for length reasons.

-10204 Line 3. It is not clear exactly what the EU region is. I believe it may be the same as the EU HTAP region, but this should be clarified. Line 9. “Augments” by 9 ppbv. I think you mean increases by (augments by is not very good English).

The EU region corresponds to the EU HTAP region. One sentence was added in the introduction to clarify that the regions were defined as in the HTAP study. (Page 10196, line 5). Augments changed to increases.

-10205 Line 5. 20% reduction. I presume you mean 20% reduction over Europe of all HTAP emissions. Line 6. “were resulting” Line 22. Chage “Despite” to “Despite the fact”

Sentence changed to: “They found that the 20% reduction over EU of all HTAP emissions determined an increase of O₃ by 0.2 ppbv in winter, and an O₃ decrease by -1.7 ppbv in summer (average of 21 models)”

-10207 Line 6: “125%”. This estimate seems small compared with satellite measurements of increases in NO₂ and estimates from other emission inventories (e.g., Klimont et al., 2009).

A comparison with more recent trends is added in the section describing anthropogenic emissions. See also answer to point 3 above.

-10208: Line 16: “We further remark...”. This comment is rather out of place here. Instead it should be given up above where describe the model evaluation.

Sentence moved to Section 2 ‘Model description’. Page 10196, Line 15.

-10209: Line 2: Is stratospheric influx determined as a residual?

Definition added to page 10208. $S_{inf}=L+D-P$. It is indeed an inferred residual.

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-10211 OH variability. Please briefly specify the model processes here that contribute to the OH variability. In particular I think it should be mentioned that interannual variability in the ozone column is included. Are the effects of Pinatubo included?

The main processes that contribute to the OH variability are both meteorological and chemical. Dentener et al. (2003) found that OH variability for the period 1979-1993 was mainly driven by meteorological processes, i.e. humidity/temperature and wet removal/precipitations. They found only a small total contribution from the changes in chemical species like CH₄, O₃, and emissions of NO_x, VOCs, and CO. Differently from Dentener et al. (2003), in our study the effect of meteorological processes (SFIX) includes also changes in emissions of CO, NO_x, and VOCs from biomass burning and lightning emissions, and it results in a negative trend of OH tropospheric concentrations. We found a good correlation between the OH decreasing trend and the decreasing trend in our NO_x emissions from lightning (R=0.78), while lower correlations are found with water vapor (R=-0.48) and photolysis rates at surface (e.g. JNO₂, R=-0.65; JO_{1D}, R=-0.56). In our study the variability from stratospheric O₃ is not included as well as the effect of Pinatubo eruption.

-10214 Line 11: "interannual variability" – do you mean trends? Line 16: ".05-.08" I believe this should be -.05 to -.08.

We refer here to inter-annual seasonal mean anomalies. I added reference to APPENDIX B. On line 16, the trends are negative, -.05 to -.08.

-10218: Line 1: "... correlation ..." It is not clear from the sentence what you are correlating with what. Line 9: the global number given in Table 5 is -1.32, much smaller than the range -26 to -48 given here. Is the global number correct?

We improved the paragraph. The correlation is calculated between the anthropogenic aerosol radiative perturbation (RP) and the changes in AOD due to changing anthropogenic emissions. The RP is calculated as the difference between instantaneous radiative forcings of the 2 simulations SREF and SFIX. The definition is given in the

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paper just before Section 6.1 (Page 10217, Line 9). A high correlation coefficient is found globally between RP and the change of AOD at the top of the atmosphere. RP is changing by -17 W/m² for an AOD increase of 1 unit. This high correlation coefficient is not found globally for RP at surface and in the atmosphere. RP is changing by -1.32 W/m² and -4.62 W/m² for an AOD increase of 1 unit. These linear relationships, with very high correlation coefficients, can be used to have a quick estimate of aerosol radiative perturbations (RP) corresponding to AOD changes from anthropogenic emissions.

-Tables and Figures

-Figure 2. What is graphed here are changes in emissions, not the emission trends. Thus the title of the figures is somewhat misleading.

We corrected the titles of Figure 2, including also a comparison to recent estimates of emission changes between 1980 and 2005 from Lamarque, 2010 and others.

-Table 5. It is not clear which correlations are significant and which are not.

High correlations will be highlighted in bold in Table 5.

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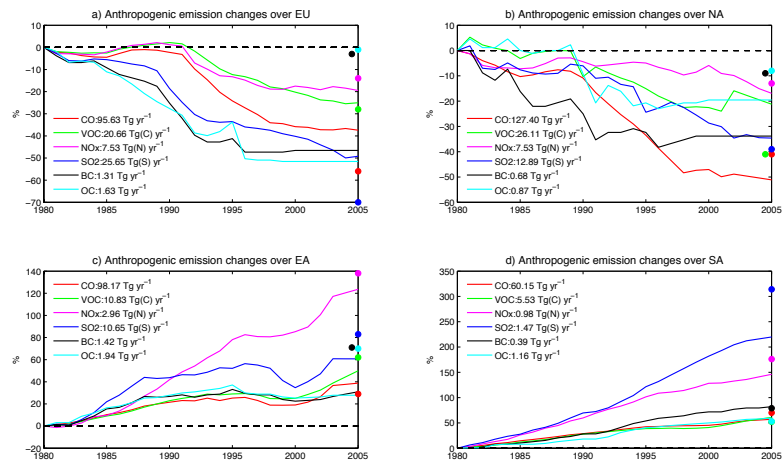


Fig. 1.