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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The effects of intercontinental emission sources on European air pollution levels" *by* J. E. Jonson et al.

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We thank the reviewer for the effort to see into this multi-author paper. We apologise for oversights, partly due to the complex nature of the multi-model evaluation.

General comments

Lines 66-70: The structure in L66-70 seems not to match the actual content of the section (Section 5 became a very superficial discussion on model resolution, it seems that the authors forgot that they originally expected to suggest improvement in the experiment design in that section). It is unclear why comparison with measurements come back in Section 5, while it was introduced in Section 3. At the end of the introduction the reader is already sceptical to what extent the paper will address the problem at

hand.

We have changed this part of the paper, including a better motivation for Section 5 (in addition section 5 has also been improved). The motivation for section 5 is to sum up the results for the individual models and as such make the reader better prepared for the conclusions. These lines now reads: In this paper we first briefly discuss the model comparison to measurements in section 3. In section 4 we go on to describe the source receptor relationships for Europe, including a discussion on how and why the model results differ. Finally, in section 4 we sum up the results for the individual models. Based on model performance compared to measurements and where and when deviations in model results compared to the other models occur we try to indicate the origins of the differences in model behaviour. In the conclusions we then suggest some directions on how this information could be used to harmonize and improve future model calculations.

Specific comments

Line 23: define here the CTM acronym, which usually refers to chemistry transport models rather than chemical tracer models

Added CTM acronym

Line 25: TF-HTAP is organized under the EMEP programme of LRTAP

Added that HTAP2 reports to the EMEP stearing body

Line 50: add that the region targeted in that paper is Europe, but that (unlike in HTAP) the contribution is assessed by model tagging rather than sensitivity experiments.

We have added that tagging is used in his model.

Line 53-54: the sentence on additional papers is not relevant, suggest removing

We have removed the list and replaced in with a reference to the acp special issue.

Line 60-63 should be moved to the experiment description part (ex: L78)

This sentance is removed. This discussion is included elsewhere.

Line 63: rephrase “secondly we look at CO” to better introduce the actual chemical compound in opposition to the CO-like tracer.

We have changed to: Secondly we investigate CO as an interactive component of the atmosphere, participating in chemical reactions.

Line 87: For transparency and reproducibility concerns, but also with regards to the HTAP requirements, GFDM_AM3 should not be included if it is not part of the database.

We will now included GFDM_AM3 results in the database, note that these model results are present in a slightly different format. We have chosen to include these results as so few models have uploaded relevant results for this study.

Line 92: The Galmarini et al. article in the special issue is focused on the complementarity paper of global and regional models rather than model evaluation. In the version currently in discussion, only a Taylor diagram is given with models not labelled. Therefore it cannot be considered as a satisfactory reference regarding the capability of HTAP models in capturing surface ozone. Such an analysis should be included here if not covered elsewhere. The scatter plots in supplementary material is a good start, but further discussion is needed. The selection of Airbase sites is very questionable at this scale.

See also comments to reviewer 2 for this point. We have included ozone time series for several GAW sites in the supplementary material. Furthermore we have included a table with statistics similar to what is already included for CO. The mentioning of AirBASE as data source was wrong. The scatterplots in the supplementary material are based on European rural and remote sites from EMEP, as stored in the EBAS database.

Line 104: a reference is needed to conclude that GAW sites are affected by local

sources. Similarly, one could question why engaging in such analysis if global models are not capable to capture regional sources. If the author conclude that it is the case, it would be a major conclusion of the paper.

We agree with the reviewer that GAW sites are selected to represent regional background conditions. The argument was removed.

Line 116: what is the source of ozone profiles?

The original source for the ozone sondes is the "World Ozone and Ultraviolet Radiation Data Centre". This information is now included in the text. Data providers have been contacted and offered co-authorship.

Line 118: how “approximate” is the temporal matching between model and observations?

In HTAP2 model profiles are provided on an hourly basis. We have added "(to the nearest hour)" in the text.

Line 123: more quantitative results are needed to support the “tendency” for underestimation in tropospheric summertime ozone.

We have extended the section with the interpretation of the ozone profiles. This part now reads:

With a relatively inactive chemistry in the winter months the measured ozone profiles show little vertical variability, with ozone mixing ratios in the troposphere increasing gradually with height. Model calculated ozone profiles are close to the measurements. As the chemical activity increases in Spring and summer months the vertical variability increases, reflecting air masses of significantly different photochemical history at different levels. As was shown in Jonson (2010) the models are not capable of reproducing this vertical structure in ozone levels. Most of the models underestimate free tropospheric ozone in the summer months.

Line 154; why is GFDL_AM3 included in Figure 3 for CO_tracer but not in Figure 2?

Figure 2 requires data from the BASE, GlOALL and EURALL scenarios, whereas Figure 3 is based on the BASE and GLOALL scenarios only. For the GFDL_AM3 model we have no data for the CO tracer for the EURALL scenario. A comment has been added in the text.

Line 159: Why would EMEP have a too strong convection? If the comparison with measurement suggest that EMEP performs better than other models (L348), maybe the other models have a too weak convection ?

We have deleted the word too, subsequently changing the meaning so that we now say that the EMEP model has strong convention and not "too" strong convection.

Line 162: the larger vertical mixing seems to occur mainly in winter for EMEP, isn't that conflicting with the hypothesis about the role of convection? Maybe more discussion would be needed on the vertical diffusivity and resolution of the various models.

Running the global EMEP model with and without spinnup we see marked differences (in ozone) lasting all the way into Spring. We believe that this is caused by ozone lifted into the free troposphere increasing the free tropospheric reservoir of ozone in the following winter and spring. We believe this also the case for the CO tracer.

The text has been made more clear on this point: Differences in mixing ratios peak in the first part of the year when emissions are high and the exchange between the boundary layer and the free troposphere over Europe is weak. Differences in the free troposphere may reflect CO tracer advected from regions upwind with convective activity also in winter, or in the preceding autumn months increasing the free tropospheric reservoir in the following winter and spring.

Line 183: The difference between CO_tracer and CO seems larger for EMEP than for the other models. Would it also be the case in terms of relative increase, and if so, why?

The widening gap between EMEP end the IFS model is commented below in the same section. It is attributed to the possible lower OH levels in the EMEP model compared to the IFS model. (even though OH is not provided by the IFS model).

Line 193: the model differences for OH are very impressive (Fig 9 of the supplementary material). To the extent that one may wonder the relevance (and need) to produce a multi-model mean. Further discussion and external references are needed for that section. The sensitivity to upper boundary conditions, especially for EMEP that seems to behave differently.

A more detailed discussion on differences and of the effects of OH is now included in several places in the manuscript.

Line 213: a reference is needed to support the statement on the relative contribution of stratospheric/tropospheric ozone.

References to the HTAP 2010 report and Stevenson et al. 2006 now included.

Line 245: the discussion on aircraft emissions is interesting, but it seems that there are more important differences, such as the role of surface titration (why EMEP seems the less sensitive despite the higher resolution). Or the fact that the O3 response of Chaser is actually very close to that of CO_tracer (or is it a mistake in the Figure?)

Discussion on aircraft emissions strengthened following comments also from reviewer 2. Regarding the GFDL_AM3 (not CHASER), the figure is corrected. We believe that the reason why the EMEP model is less sensitive to titration must be sought in the chemistry schemes. This is now discussed later in the paper (see also comments from reviewer 2).

Line 256: the fact that CH4 is excluded from the experiments should appear before in the experiment description (Section 2).

The statement that CH4 is not included in the experiment is moved to section 2.

Line 259: The explanation of figure 5 needs to be re-worked in the text, in particular to explain that even if some sources are not isolated in some models, their contribution is still accounted for in the “remaining” fraction.

This section now reads: For each model the contribution from ROW (Rest Of the World) is calculated by subtracting the sum of the contributions from from available world regions from the BASE - GLOALL contribution. Thus the portion related to ROW includes a varying mixture of world region definitions depending on the model.

Line 266: more quantification is needed regarding the relative role of external/European sources. Figure 5 indicates that the external contribution seems indeed to exceed European contribution, but they are actually not that far. The percentage contribution (with error bar) should be given.

We have added an additional table listing the percentage contributions to annual ozone and summer ozone to Europe from Europe, North America and East Asia. In addition we also list the contributions to SOMO35 and POD forest calculated by the EMEP model. The numbers are a subset of those displayed in Figure 5.

Line 275: the comparison the HTAP1 is too weak. It is very frustrating not to better understand the added value of the new experiment and to what extent the earlier conclusions still hold. The benefit of having engaged in a complete new experiment should be better assessed. For instance by looking at a subset of models having participate to both and investigating clusters of regions for the reference/sensitivity simulations to conclude on the importance of (i) emission changes, (ii) region definition, (iii) participating models. Conclude on emission changes:

The paragraph has been rewritten and now reads: In comparison to HTAP1, HTAP2 regions are better defined. In addition emissions as well as models are up-to-date. To disentangle whether the changes from HTAP1 to HTAP2 are due to emissions, a changed model ensemble or changes in receptor regions is unfortunately not possible in a fully quantitative way. Source and receptor regions have been chosen in HTAP2

to cover the land-only politically connected regions accurately on a 0.1 degree grid. In HTAP1 the EUR region was a simple latitude - longitude box, also including parts of North Africa, the Middle East, Russia, Belarus, Ukraine and large sea areas, all of these identified as non European regions in HTAP2. In HTAP2 the European region is smaller, thus exporting larger fractions to nearby regions, but most major HTAP1 source regions are located within the smaller HTAP2 region, thus making this region more sensitive to titration effects. As a result the effects of emissions on ozone levels from the EUR region to itself is reduced.

The ensemble mean contribution to ozone levels from Europe to itself has decreased from 0.82 +- 0.29 ppb in HTAP1 to just 0.11 +- 0.32 ppb in HTAP2. Also - total and regional distribution of emissions for the base year changed from HTAP1 (2001) to HTAP2 (2010). Gaudelet al. (2018) have analysed the ozone trends between the years 2000 and 2014. Over Europe. They found a general ozone increase in the winter months (December, January, February) and a general decrease in the summer months (June, July, August). The emission trends in the HTAP1 world regions are given in Turnocket al. (2018) between 2001 (the base year for HTAP1) and 2010. The changes in measured ozone are consistent with the reductions in European (and North American) emissions of NOx (along with other ozone precursors) over the same period resulting in less titration and thus increased ozone levels in some areas mainly in the winter months, and simultaneously less net ozone production in summer. Likewise emissions in North America have decreased and may explain the 0.37 +- 0.10 to 0.22 +- 0.07 ppb decrease in the ensemble mean contributions from North America to European ozone levels. Over the same period emissions in other world regions as East Asia have increased. This increase may explain the 0.17 pm 0.05 to 0.22 +- 0.13 ppb ensemble mean increase from HTAP1 to HTAP2 in the East Asian contribution to European ozone levels. Contributions from South Asia are small in both HTAP1 and HTAP2 (0.07 versus 0.05).

Line 301: it is surprising to say that a comparison should not be made, when it is

made by the authors. There were no similar discussions on the realistic aspect of sensitivity experiments for non-CH4 species, so from an academic perspective regarding chemical sensitivity the comparison does hold.

Thanks. The point is that the effects of direct changes in concentrations is not the same thing as changing the emissions. This is now clarified and rewritten: However, comparing a 20% change in CH_4 concentrations, and the effects of the GLOALL emission scenario requires careful interpretation. Because of its relatively long lifetime of the order of 10 years in the atmosphere, a 20% change in concentration corresponds to an approximate 40 year long historic CH4 trend (Meinshausen 2011). The GLOALL scenario is not accounting for the full impact of a continued 20% reduction in emissions. With a continued emission reduction scenario, the overall ozone reductions would be larger, while the methane attributable fraction, relatively, would be smaller.

Line 330: the results related to ozone indicators are interesting and worth being highlighted in the abstract. It is frustrating that only one model can be used here, especially given the apparent different behaviour with regards to titration. More efforts should be given to investigate the HTAP database in order to include more models for a comparison of summertime mean of daily ozone maxima, or at least summertime mean ozone.

Unfortunately hourly data with attribution to source regions are only available for the EMEP and the CHASER model. We have thus instead included a table comparing annual average and summertime ozone based on the numbers in Figure 5. (see comments to line 266). SOMO35 and POD forest from the EMEP model are included in the same table. As SOMO35 and POD are mainly added up in the summer months the percentages are similar in the EMEP model and the table gives an indication on the differences between the models.

In addition to the table we have added some additional text: The regional contributions, expressed by these metrics, are also listed in table (new table). The figure and table

clearly shows that the choice of metric matters, in particular for the effects of European Emissions. POD forest is accumulated in the growing season in summer. A large portion of SOMO35 is also accumulated in the summer months. Table (new table) also lists the percentage contributions to summer ozone for all models. The similarities in the percentages for summer ozone and the ozone metrics in EMEP_rv48 is an indication that also for the other models these percentages are comparable.

Line 350: according to Section 3.2, EMEP is not the only model to display an overestimation of tropospheric ozone.

Yes, text is changed to: The model is one of the models with highest overestimation of ozone in the free troposphere in the winter and spring months.

Line 360: how can a lower diffusion can lead an underestimation of surface CO, the opposite would be expected.

Thanks - we agree. If the low RERER is caused by too much CO remaining in the PBL it should result in an overestimation of surface CO compared to measurements. We have deleted this statement.

Line 367: what is the rationale for a relaxation to zero in the GEOS-Chem adjoint?

The GEOS-Chem model has only ozone chemistry in the troposhere, and stratospheric levels should be disregarded.

Line 372: Section 5 is very descriptive and lacks a clear outcome

We have added more text, including text with additional motivation for this section: Here we try to point out if, and at what stage, the results from the individual models deviate from the other models. It should be stressed that such a deviation does not necessarily imply that the results from a particular model is wrong.

For the individual models we have tried to identify one ore more feature where the individual models differs from the other models.

Line 399: From the results provided in Section 5, it can not be concluded that the results are not sensitive to model resolution.

Yes, this is probably phrased too general - This part now reads: The model resolution differs between the individual models. Model results from the two CHASER models, differing in model resolution only, are qualitatively similar when compared to measured CO and O3} at background measurement sites and very similar in RERER for CO and ozone, suggesting that resolution differences at the scales investigated here, are not important to explain RERER differences between the global models. Still, it is difficult to conclude in general to what extent horizontal resolution affects the source receptor calculations at intercontinental scales.

Line 406: it is rather convection that is put forward rather than chemistry. The following sentence (L407) also goes in that direction. It is quite surprising to read that the conclusion and the content of the paper seem contradictory.

This part now reads: The joint and consistent analysis of a CO tracer, CO and O3 in this paper is a tool in understanding where and why (right or wrong) the models differ, however, it has a potential for wider use, enhancing our understanding of the result and also as a tool for model improvements, reducing the overall uncertainty in future model calculations. We believe that in order to close the gap in model results, and subsequently improving the reliability of the model output, possible future model inter-comparisons should be more process oriented (transport, depositions, chemistry etc). Our study shows that models differ already for CO and the inert CO tracer, where differences were established with 2 models, but that differences are amplified as more chemistry is added. Note that the CO RERER and O3 RERER values are not correlated taken the models as samples. The big additional spread in model results for ozone is clearly induced by differences in model chemistry and for instance treatment of titration in the winter boundary layer. However, differences in chemistry may well also be induced by differences in advection/convection as the level of exchange will inevitably affect the chemical regime in both the free trposphere and in the boundary

layer. We believe therefore that further process oriented evaluations (comparing advection/convection, chemistry, dry and wet deposition etc separately) should be made, making use of relevant meteorological and chemical measurements.

Line 419: from Figure 5, it seems that about half of ozone can be mitigated with European sources, isn't that "sizeable" already? More precise quantifications must be given in the conclusion and abstract with key figures and associated error bar across the multi-model ensemble.

We now include a comparison and discussion of RAIR in HTAP1 and HTAP2 in section 4.4. Furthermore RAIR is also included in the conclusions and the abstract.

The (almost) first parts of the conclusion section now reads: The model calculated relative contributions to surface ozone levels from non European sources is much larger in HTAP2 compared to HTAP1. Mainly because the contributions from Europe to it selves has decreased from 0.82 ppb to just 0.11 ppb. As a result RAIR has increased from 43 to 82\%. In parts differences could be explained by decreasing emissions in Europe and increased emissions in most other regions as East Asia from year 2001 to 2010. However, the results from the two HTAP phases can not easily be compared, ...

Table 1: what is given on the right part of the table? From the text, it appears to be correlation, but that should be stated clearly. Is CHASER_rel actually Chaser_t42 according to table 1 of the supp. Mat. A uniform model labelling would be appreciated.

We have added a line at the top of the table explaining what is calculated concentrations and what is correlations. Model labeling corrected.

Table 2: ibid about CHASER_rel. The labels of regions should be consistent with Fig 1.

Labels for regions now as in Figure 1.

Table 3: swap the first and second sentences. Use boldface rather than larger signs

for important deviations.

First and second sentence swapped. Large + and - replaced by bold face.

Figure 2. GFDM_AM3 missing for CO_Tracer

The calculation of RERER require BASE, GLOALL and EURALL. EURALL calculations are not available from GFDL_AM3.

Figure 3: panels g) and i) appear identical

g and i now corrected.

Figure 4 and 5: Truncation fixed

Figure 6: add that the results to 20% perturbation are plotted.

We have added that the the GLOALL scenario is calculated with with 20% reductions in anthropogenic emissions.

Supp. Table 1: Table 1: all models are in bold, not the first 7, since 7 models are displayed. The information about spin up should be given in the experiment description, not in Table 1. Footnote #2 is not references in the text.

We have removed bold face for the last model. The information about spinnup now in the experiment description. Footnote 2 deleted

Supp. figure 1: What happened in Heimaey in May in observations?

The peak is actually the IFS model. We have changed the colour scale for the models so that it is easier to tell models and measurements apart. In the IFS model it could be an artefact of the GFAS fire emissions (April/May 2010 is the time of EEyjafjallajökull eruption but it is not clear how this can effect CO).

Supp. figure 4: More details are needed in the legend about the source of data and the indicator displayed.

More information about data source added. Editing errors corrected.

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