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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.

Reply to general comments:

Most of the general comments are addressed in the detailed comments below, eg error in figure 3, fig 3i, AM reference etc.

At the time of the submission we believed that another paper would address the validation of ozone. As it turned out this was not the case, and as a result the ozone validation has been extended in this paper. We have reorganised the paper. The content of in particular section 5 has been expanded, and a motivation for this section is

included in the introduction.

The issue of ozone titration is discussed in more detail. The reviewer is right that as in particular European NOx emissions have decreased from 2001 to 2010, and as a result European controls have been offset by by removal of local suppression. These considerations are now discussed in section 4.4.

Detailed comments: ————————

Line 16: capitalization error.

Comma replaced by .

Line 25: Missing parenthesis

Added right)

Line 29: Verb agreement. Replaced is with are.

Line 46-54: The list of published papers should be used to provide context. Here it is simply a list.

We have deleted this list and replaced it with this: A large number of papers from HTAP2 have been published in the ACP (Atmospheric Physics and Chemistry) special issue: "Global and regional assessment of intercontinental transport of air pollution: results from HTAP, AQMEII and MICS"

Line 60: Details like model count would be better in the methods.

Model count deleted here.

Line 68-70: Differ should be differences?

Not applicable. The description of the sections later in the paper is changed.

Line 72-74: Poorly written.

Now corrected to: The HTAP2 model experiment was set up by the Task Force on

Hemispheric Transport of Air Pollution (TF∼HTAP). A project work plan, a description of the model experiments etc can be found on the TF∼HTAP web page (\url{ http://www.htap.org/}).

Line 82: "etc" seems particularly poor when later you will refer to advection schemes as a causal difference.

Advection added to list, and a reference to the supplementary material. These models have different resolutions, advection schemes, chemical mechanisms etc (see supplementary material and references therein).

Line 87: Space added

Line 92: Space added

Line 95: Replaces in by is

Line 95: How does evaluation of upwind sources affect conclusions about transport to Europe?

We have not included an evaluation of upwind sources here. Several other HTAP2 papers are addressing this.

Line 99: GAW (Global Atmospheric Watch) spelled out.

Line 101: How "high" correlations are expected given the resolutions of the models?

We have included some more text and references here: Correlations shown here are in the same range as correlations with MOPITT satellite measurements as reported by Naik et al.2013. However, as shown in Table 3, all models except IFS_v2 underestimate annual CO levels by 13% or more. Similar underestimations was also shown Strode et al. 2015.

Line 101-102: resolutions of all the models should be provided in the methods rather than the comparison to measurements.

The information about the resolution of the CHASER models is provided here as explanatory information for how the versions of the CHASER model differ. Information on model resolution for all models is given in a table in the supplementary, referenced in section 2.

Line 98-109 How is it that CO deserves a site-by-site comparison and ozone?

Unfortunately it was communicated to us until a few days before the manuscript had to be submitted that this would be included in the Galmarino et al. paper. Therefore is was not included in this ACPD submission. We are now including a site-by-site surface ozone evaluation based on GAW data as already included for CO.

Line 112. The authors should mention that they do have some surface evaluation in this paper. Currently, Table 3 in this manuscript is not referenced until Section 5.

We now say that there is additional surface evaluation in the Galmarini paper, and we refer to Table 3 also in this section and too all supplementary material on ozone evaluation.

Line 114: There is currently no discussion of ozone results except to say they exist somewhere in the supplement. Why is this sufficient?

We have added a more complete paragraph on the ozone comparisons made in chapter 3.

Lines 122 - 123: There must be more discussion of the basic results that will clearly affect transport.

This section has been extended and now reads: The profile comparison allows to identify differences between the models in vertical mixing of ozone useful for further interpretation in inter-hemispheric transport efficiency. Note that the GEOS-Chem model only simulates ozone in the troposphere and its ozone levels above 300 hPa should be disregarded. 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 in general 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 \cite{Jonson2010} 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 138: Here and elsewhere the definition of regions is incorrect. Here you have NW, SW, SE, GR+TU. In the Figure, you have NW SW, E, GR+TU. Other places you have NW, SW, E, SE. Choose one, and be consistent.

The region notations are now consistent throughout the paper.

Lines 139 - 140: Is this source apportionment the same as contribution in sections 4.4 and 4.5?

Yes. We have now included references to the subsections.

Line 142: rate of decay is later explained, but here seems completely arbitrary.

We disagree. We think that the rate of decay is useful information/reminder here.

Line 182: Numbering of Figure 3 and 4 corrected.

Lines 185 - 189: The reasonableness of this should be discussed.

Differences between the individual models are very similar for CO and the CO tracer. Differences in the CO tracer can only be caused by advection as there is no chemistry for this species. The similarity between CO and the CO tracer for two models leads us to believe that the causes for the differences are the same.

This argument is included in the text.

Line 205: This gets discussed in several places and is really part of the methods.

OK, shortened here, but this information is also repeated here as part of the interpre-

tation of the results.

Lines 217 - 2019: Web citation is inappropriate. Further, the lifetime of ozone is expected to vary with respect to season and altitude (Wang et al. 1998; Brasseur, Orlando, and Tyndall 1999). Estimates of lifetime at 500hPa range from 15-160d and from 40-300d at 10km. Your upper bound of 18days is misleading. Table 1.1 of the HTAP 2010 report cites weeks to months in the free troposphere. The IPCC range of values do not acknowledge the complexity of ozone transport.

In acknowledgement of the complexity of ozone chemistry and transport, we now refer to the HTAP 2010 report for the lifetime of ozone. In addition we have replaced the web citation with a reference to the IPCC report.

Line 242: AM3?

The GFDL_AM3 model is added to the list of models not perturbing aircraft emissions.

Lines 246 - 247: Provide some reference or evidence.

We are now referring to a paper by Cameron et al. (2016} for the effects of aircraft emissions on surface ozone calculated by several global models. See updated discussion in the manuscript for details.

Line 247: here = PBL?

This part has been rewritten.

Lines 254 - 284: Is this contribution from a simple mean within seasons? What months were included in each season? Are the numbers in the text ensemble means? What about ensemble mean RBU? MDE? EU? 290-291: Did they "too" calculate smaller "than in this study" or did they "too" calculate "smaller as in this study"?

This section has been rewritten. See also comments from reviewer 1. In Figure caption 5 we now specify which months are included in WI, SP, SU and AU. 0.37 NA to EU and 0.17 EA to EU are from Table 4.2 in the HTAP1 report. The numbers are ensemble

means. This is now noted in the text. We have chosen not to compare the numbers for EU as the definition of the European domain is so different. We now also list list the numbers for the remaining regions. They are also shown in Figure 5.

Line 269: MDE appears to always be small.

We now say that contributions from the Middle East and North Africa are small.

Lines 290 - 291: Did they "too" calculate smaller "than in this study" or did they "too" calculate "smaller as in this study"?

The text is changed to make this clearer: They calculate a much smaller contribution from non European sources than in this study, similar to the contributions calculated in HTAP1.

Lines 272 - 273: Did these other studies use the same model?

We have added that in Jonson et al. (2015) the EMEP model was used. Brandt et al. (2013) used a different model.

Line 277: Right parenthesis added.

Lines 274 - 280: Methods?

This part is now rewritten.

Line 305: HTAP1?

We have added that the Fiore et al. paper was based on the HTAP1 model experiment.

Lines 306 - 335: There needs to be a clearer connection to the previous section. In fact, you could just add two bars to Figure 5a. That would help to connect the of POD and SOMO35 to the seasonality of titration.

We have added more material to this section following the recommendations also from reviewer 1

Line 361 - 371: Terse and uninformative

This section is rewritten bringing more information.

Line 390 - 392: See previous comments about ozone lifetime.

We now refer to discussions on lifetimes in previous section.

Line 400: Probably deleted. Improved text.

Line 405: added for ozone.

Comments regarding Table 1: If mountain sites are used at readers peril, consider making room for ozone evaluation by moving them from the first data result.

We have included a similar table as Table 1 with ozone. We have not included mountain sites in the ozone table as the "peril" is much larger for ozone as the dry deposition is faster and lifetime shorter.

Comments Table 2: Update region definitions to be consistent with figures and text.

Regional definitions updated.

Figure 1: update region names to be consistent. Also, too many extra colors so it is hard to tell what is included. Is the Baltic Sea part of Eastern Europe? Black Sea? Caspian Sea? Mediterranean?

Not changed. Difference in colour is visible both on the screen and on printout. The European seas are part of the OCN region.

Figure 2: Necessary?

We would definitely like to keep the figure. We think it illustrates very well the evolution in RERER going from a simple CO tracer to CO and finally ozone with a multi model ensemble.

Figure 3: lettering needs to be updated in the figure and in the text. What was the com-

mon grid and how was it treated when a grid cell at 1000hPa was below the surface?

Lettering is updated and is now the same as in the text. All model data has been interpolated to a common vertical grid. For gridcells below 1000hPa values at the lowest model level was used.

Figure 3: 3i is AM3 CO not ozone. Column 3: consider a scale that does not saturate in so much of the figure.

Panel 3i corrected.

Figure 4: North and south boundaries are unnecessarily different from figure 3. Further, this highlights that no meaningful discussion of the boundaries was made. In fact, 50E includes a lot of Russia and a lot of ocean. Column 3: consider a scale that does not saturate in so much of the figure.

North and south boundaries changed corresponding to Figure 3. This resulted virtually no visual changes in the figures. We have added a discussion on the boundaries: This area roughly corresponds to the European regions as shown in Figure 1, but also some additional land and sea areas. The main focus of the figure is in the free troposphere where horizontal gradients in concentrations are small. Liu et al. 2009} calculated the correlations between nearby pairs of sonde stations. They found low correlations near the surface indicating that local and regional effects are important here. From the surface correlations rose sharply to a local maximum in the lower troposphere. We therefore conclude that the selected area is a good representation of the atmosphere above Europe.

Figure 5: There is no discussion about the CHASER model being the only one without apparent titration, and this should be discussed somewhere. Region definitions should be consistent with the text or the text should be consistent with the figure. The units are cutoff on the first row.

We have commented the low level of ozone titration for the CHASER model in section

4.3: For all models, except the CHASER_re1 model, ozone titration dominates the overall European contributions when summed up over the three winter months. However, for all the models, including also the CHASER_re1 model, the net European contributions includes regions of net ozone production and net ozone destruction in winter.

Regional definitions now consistent with text.

Figure 6: Region definition nomenclature. I recommend showing as 3 stacked-bars (or adding to Figure 5). If I am interpreting this right, the RAIR is 84% compared to 43% from HTAP1. I suspect that all models provided annual and I think reporting RAIR would be useful (maybe in Figure 2).

Region definition nomenclature fixed.

Figure 6 is complemented by a table with results for summer ozone from the models following the recommendations from reviewer 1. For ozone this table lists the annual (and summer) percentage contributions to Europe from several regions, including Europe to it selves. We have also calculated average RAIR for the models in Figure 5. The HTAP2 RAIR of 82% compared to 43% in HTAP1 is discussed in section 4.4, and these numbers are also repeated in the conclusions. RAIR for the individual models proved difficult with European contributions to it selves was close to zero and even negative for some models.

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