**Atmospheric
Chemistry
and Physics
Discussions**

# *Interactive comment on* "Interannual variability of tropospheric composition: the influence of changes in emissions, meteorology and clouds" *by* A. Voulgarakis et al.

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We wish to thank Referee 1 for the thorough and very useful review. The suggestions have stimulated us to investigate several issues further and to make detailed improvements to our manuscript. We believe that the general comments made by the reviewer at the beginning of their response are answered step-by-step in our replies to their specific comments, as described below:

SPECIFIC COMMENTS:

*Abstract*:

S1) We have added "1996-2000" in 2 places in the abstract in order to highlight that the period of study is limited to this 5-year period. In the second sentence of the abstract, we now highlight the 1997–98 El-Niño and the intense biomass burning events during this period.

*1. Introduction:*

S2) As noted above, we now refer specifically to the "1996-2000" period. We agree that it would clearly have been valuable to extend the runs for a longer period, but this was not possible for logistical and computational reasons. We thus try to exploit the 5-year data that we obtained from the experiments to the largest extent possible. In the conclusions, we have added two sentences which describe this limitation of the study, related to the choice of period for the experiments (we believe that discussing this in the conclusions is more useful for the reader than adding this to the introduction).

S3) Sentence "Apart from the strongly anomalous conditions…" which did not make sense, has been replaced as suggested by the reviewer. In the 1st paragraph, we add a sentence clarifying the difference between interannual variability and trends. We added a paragraph and now we outline in a clearer way how each meteorological parameter can affect composition. Also, we removed "significant/significantly" from where they could have been misleading.

S4) We rephrased (stating that the issue is still under exploration) and added references of the studies, which have looked at the effect of meteorology on tropospheric composition interannual variability.

S5) We added the word "exceptionally" to point out that we are referring to the very strong El Niño events.

*2. Model set-up*

S6) Having interannually varying biogenic emissions in the model is one of our main future goals. In this case, we note that, especially over Indonesia, the inclusion of higher

isoprene emissions during 1997 would most likely have led to ozone increases, which would not change the sign of the ozone change in 1997 over Indonesia in the model (something which could have been a problem if isoprene emissions were expected to decrease in 1997 in this geographical area). We have added a caveat that biogenic emissions in these model runs do not vary from year to year. In Section 3.2, when discussing Indonesia, we now mention that ozone could have been higher had we included varying isoprene emissions in our simulations. And then we briefly mention that the results of a recent study suggest that even if we had taken interannual variability of isoprene emissions into account, the effects could have been small.

S7) A table including surface emissions for all species, for 4 geographical areas (global, northern extratropics, tropics, southern extratropics) and for all different types of emissions (anthropogenic, biomass burning, biogenic) would provide too much extraneous detail and would be very large. Since anthropogenic emissions in the RETRO dataset did not change greatly in the 5–year period that we examine, we expect that the main contribution to emission changes would be from biomass burning. Taking into account that biogenic emissions do not vary from year to year in our simulations, we do not think that splitting emissions by source type in this table would provide much interpretive information to the paper. However, we agree that showing values for different geographical zones would add value to this table and help the reader understand the contribution of each region to the global emissions better. We have thus changed this table to include emissions from the Northern Extratropics, the Tropics and the Southern Extratropics.

*3.1 Tropospheric NO$_2$*

S8) We have now added a paragraph which briefly gives examples of regions/years where NO$_2$ columns are lower than average (paragraph starting "There is a variety of continental. . ."), and states what we attribute the low columns to.

S9) We have added some more material to the paper, and since the reviewer suggests

that these figures (previous Figures 3 and 4) do not add much value to the paper we have removed them and made the related discussion shorter.

S10) We have rewritten the paragraph in order to make its content clearer.

S11) We have now included a new figure (Figure 3) which shows how the model captures various variables which are immediately related to El-Niño meteorology: a) Middle tropospheric radon concentration anomalies over Indonesia, as an indicator for convection; b) Total lightning NOx emission anomalies, also an indicator for convection and a useful figure for discussion on the emission changes over the region; c) Water vapor anomalies to demonstrate that the model captures dryness over the Western Pacific and high water vapor concentrations over the Central and Eastern Pacific.

S12) This part of the discussion has been removed as we think that it does not add much to the discussion and makes the section too diffuse (the overall picture of what drives changes over the oceans – meteorology - is mentioned in the previous paragraph). But we note that both higher humidity and HNO$_3$ deposition are indeed found in the model results.

S13) Figure 4 has now been removed, as suggested.

*3.2 Tropospheric ozone*

S14) We have added two short sentences to the end of this paragraph which make the connection with (and the transition from) the previous section better.

S15) We removed "related to El-Nino" as we agree on the comment of the reviewer. Also, we have added a sentence to the end of this paragraph to clarify the effect outside the tropics in 1997.

*4. Quantitative analysis*

S16) We have now added some sentences in the Introduction (in the part discussing the impact of various meteorological parameters on chemistry) which outline how

clouds affect chemistry via photolysis.

*4.1 Analysis of global ozone IAV drivers*

S17) We have made this clearer in the caption of the figure.

*4.2.1 Tropospheric ozone IAV*

S18) The figure has been moved to Section 2, as suggested.

*4.2.2 Tropospheric CO and OH IAV*

S19) On further consideration of the issue, we suspect that the origin of these differences may lie in the use of different metrics - we are comparing variability in regional burdens while Szopa et al. (2007) were looking at variability at individual stations. The variability at a station could have a much higher component from meteorological sources than a regional mean would. For this reason, we think that the models may not give greatly different results if we use the same metrics.

*Conclusions*

S20) We agree that such a statement would add value to the paper and we add a sentence to the conclusions which points to the relevant results from the analysis.

TECHNICAL CORRECTIONS

*1. Introduction:*

T1) Done.

T2) Changed the first sentence to help avoid confusion.

T3) Done.

T4) Done.

T5) Deleted.

*2. Model set-up*

T6) Done.

T7) Done.

T8) Rephrased.

T9) Of course there are a lot of factors that can be driving the discrepancies. However, here we underline the obvious ones – the factors that *we know* do not vary interannually in the model.

*3.1 Tropospheric $NO_2$*

T10) Removed.

T11) Corrected.

T12) This part of the discussion has already been removed/replaced.

T13) The shipping emissions are part of the aggregated RETRO anthropogenic emissions that we have used, so we do not have access to them separately. The most that we can say is that the trend in the shipping emissions is expected to be non-negligible in 1996–2000 (as also briefly presented by the RETRO emissions report) but that there is no spatial pattern detectable in our results, due to the existence of other more important factors. This is mentioned in the discussion in our paper.

*3.2 Tropospheric ozone*

T14) Done.

T15) In the case of Indonesia ozone loss is 19% higher in 1997 (El-Niño) than in other years, although humidity is lower. This is mainly driven by the higher ozone concentrations (and to a smaller extent by higher radiation and thus faster photolysis), due mainly to higher precursor emissions. Therefore the destruction term does not adequately reveal the underlying negative effect of reduced humidity. In addition, the reader may be

confused by this additional detail in Table 4, and we think that the point is sufficiently demonstrated using the net chemistry term.

T16) Sentence removed.

T17) Paragraph was re-written.

T18) Rephrased.

T19) This paragraph discussed extratropical features, not tropical. We have added a few words to clarify this.

*4.1 Analysis of global ozone IAV drivers*

T20) Done.

*4.2 Regional scale analysis*

T21) Done.

*4.2.1 Tropospheric ozone IAV*

T22) We appreciate the potential confusion here, and have amended the beginning of the second sentence.

T22) Deleted contents of parenthesis.

*4.2.2 Tropospheric CO and OH IAV*

T24) Added.

T25) We appreciate the suggestion to center the maps at 180 degrees, but believe that this would marginalize and split the European region, a major focus region for this study, and therefore prefer to retain a Greenwich-centred view.

T26) Done.

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