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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.

Anonymous Referee #1

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General comments

This paper presents model sensitivity simulations in order to determine the role of meteorology vs. emissions in explaining interannual variability (IAV) in tropospheric composition from 1996-2000. The paper predominately focusses on years 1997 and 1998 and the El Niño event that occurred during that period. Sensitivity runs are performed with either meteorological input data for temperature, winds, humidity and cloud water content fixed at 1996 values or with surface emissions fixed at 1996 values. These are compared to a "base" run that uses annually varying meteorology and emissions for 1996-2000. In the first part of the paper (5.5 pages) the interannual variability of tro-

pospheric column NO2 and ozone is described in terms of differences in spatial maps between the "base" and the "fixed" meteorology and emissions runs. The authors generally find that IAV in meteorology is more important than IAV in emissions for explaining the year-to year differences in column NO2 and ozone as compared to the 5-year 1996-2000 average. Biomass burning events in Siberia/Canada and Africa/Amazonia are also mentioned briefly. There is $\sim$1 page of description of high column NO2 over the UK in 1996 and over West Central Africa in 1999; these are related to meteorological conditions. For column ozone, the influence of high STE in 1998 is highlighted. In the second part of the paper (5 pages) the amount of variability explained by varying emissions vs. varying meteorology vs. varying clouds (subset of varying meteorology) are quantified for a) the global ozone burden and b) regional ozone and CO burdens and OH boundary layer (BL) concentrations for Europe and Indonesia. This analysis shows that the influence of varying meteorology is the dominant factor responsible for IAV in global and regional ozone burdens and for OH BL, but that varying emissions are also important for Indonesia. For BL OH IAV, the role of clouds is expounded on. This is perhaps the most interesting new result of this paper. In contrast for regional CO IAV, the influence of varying emissions is greater than that of varying meteorology. Global and regional ozone budget terms, in particular elevated STE in 1997-99, and changes in net transport and chemistry are described. All but the first 2-3 lines of the abstract and conclusions describe the results from the second part of the paper (minus the ozone budget results).

My recommendation is that the paper is accepted after revisions that account for the following major points and the specific comments given below:

a) The paper is lengthy given this abstract and conclusions set. The first and second parts of the paper are not well connected. A clearer write-up of the important results and figures would greatly improve the readability of this paper. b) Interannual variability is determined from just 5 years of model runs. Moreover, the time period was 1996-2000 and most of the text in this paper relates to the 1997/8 El Niño. The title or

abstract should reflect this. Furthermore, in this short time period how can the authors separate isolated individual events (e.g forest fires in Siberia in 1998) from periodic events that are important for year-to year variability (see specific comment S2)? c) The results comparing individual years to the 5-year average from the "emfix" and "metfix" runs may not be connected, therefore caution is needed in interpretation of these results (see specific comment S15). d) There are several places in the text that discuss meteorological conditions that appear speculative rather than confirmed from the analysis of model data. See specific comments S11), S12).

Specific comments

Abstract: S1) The abstract ends with the comment that the approach followed in this paper can explain observed tropospheric variability such as increases in ozone concentrations over Europe in 1998. However, at the end of the paper I'm not clear as to why tropospheric ozone increased in 1998. Increased STE is the extra-tropics is discussed in sections 3.2 and 4.1 but not connected with the conclusion of decreased net export in section 4.2.2.

1. Introduction: S2) A major issue is the model runtime and period. Interannual variability is determined from just 5 years of model runs. Moreover, the time period was 1996-2000 and most of the text in this paper relates to the 1997/8 El Niño. The authors must highlight in the abstract that their study focusses on the interannual variability due to the intensely strong 1997/8 El Niño, or else amend their paper title to include the time period of IAV or mention the 1997/8 El Niño. The authors can then justify their choice of only 5-years of model runs. Justification of a 5-year period w.r.t Richter et al. 2005 (page 4 line 10) is not valid since the latter is a satellite study and further years of data are simply not available. The model runs in this paper could be extended for a longer period as they do not rely on satellite measurements. This would allow more robust conclusions. For example in section 3.1 the authors state the emissions from fires in certain regions drive IAV of NO2 – however the fires are only associated with one particular year e.g Siberian fires in 1998 or the Iberian Peninsula in 2000, so how can

we be sure that these are not isolated individual events rather than periodic events that are important for year-to year variability on longer timescales? This limitation should at least be noted.

S3) The introduction should be written more clearly e.g. the sentence starting on page 3 line 25 makes no sense. The distinction between trends and IAV should be made and the connection between meteorological variables and how they affect transport vs. chemistry outlined. The word "significantly" should be used with caution.

S4) Page 3, line 18. "is debated" – elobarate on why the influence of meteorology on tropospheric composition is debated. The text below this doesn't suggest any debate.

S5) Page 3 line 25 and Page 5 line10. El Niño in itself is not extraordinary but a regular 2-7 year event. Also note that after April 1998 SSTs changed towards La Niña –like conditions so the "annual average" for 1998 represents the end of an El Niño event and the start of a La Niña event.

2. Model set-up

S6) Page 5, line 3. Biogenic isoprene emissions do not vary from year to year in the model, so the effect of IAV in temperatures on IAV in isoprene emissions and hence on tropospheric composition is not included. For example, higher temperatures in the maritime continent during the 1997/8 El Niño event may have enhanced isoprene emissions. This important caveat should be noted.

S7) Page 5, line 8. Table 1 is potentially very important for this paper as it tells us about IAV in surface emissions. Much of the discussion in the text discusses IAV due to emissions from biomass burning in certain regions. However, this table needs surface emissions to be divided into anthropogenic, biomass burning and natural emissions in order to be most useful. What would be even more useful is a breakdown by region – even as broad as tropics, extra tropics, mid-latitudes. To be comparable with Figures 2) and 5) the 5-yr mean and percentage change for each year should be tabulated.

3.1 Tropospheric NO2

S8) The authors only discuss the regions in Figure 2 where simulated NO2 column concentrations are larger in individual years as compared to the 5-year mean. Can they also comment on regions and years that exhibit large column NO2 decreases as compared to the 5-year mean?

S9) Figures 3 and 4 do not seem to add extra value to the paper. The NOx lifetime in the upper troposphere can be of the order of a week. Transport is already discerned from the "Emfix" plots in Figure 2.

S10) The paragraph starting on Page 7 line 26 is most confusing, especially the sentence that starts "When these emission are not taken into account. . ..". Re-write for clarity and refer to the appropriate figure panel/s.

S11) Page 8, line 14, Page 10, line 13. There is substantial description of the meteorological conditions associate with El Niño such as strong level divergence and suppressed convection around Indonesia, but there are no figures to verify that these conditions are represented in the ECMWF winds and other variables used to drive the model simulations. Figures that confirm the meteorological conditions described should be included.

S12) Page 8, line 27. The authors should confirm whether they find higher humidity and HNO3 deposition in their model results?

S13) Page 9, line 12. Why does Figure 4 and the associated text on page 9 not include Indonesia and tracer transport during the El Niño event?

3.2: Tropospheric ozone

S14) Page 10, line 10. At the end of this first paragraph, a few lines relating similarities and differences in column NO2 and ozone spatial patterns in Figures 2 and 5 would be helpful; STE could be mentioned in this context.

S15) Page 10 line 26. "Thus while IAV in emissions alone would cause ozone increases in tropical areas, it is the meteorology related to El Niño that makes this more than a regional feature. Much care is needed with such interpretation. The differences in ozone in 1997 compared to 5-year mean in the varying meteorology runs are not likely to be solely due to El Niño. Furthermore the ozone increases in 1997 compared to the 5-year mean in the varying emissions simulation may not be at all connected to the ozone increases in 1997 in the varying meteorology run. Could the ozone increases due to increased biomass burning over Indonesia be transported to the extra-tropical and mid-latitude regions in the same year?

4. Quantitative analysis

S16) Page 12, line 15. Could the authors describe the effects of clouds on tropospheric composition and relate that to Page 15, line 28 where they state that the impacts of clouds is expected to be greatest in the BL.

4.1 Analysis of global ozone IAV drivers

S17) Page 12, line 21. Figure 6 is interesting but needs careful explanation in light of the proceeding text and figures. For example in Figure 5 the results of the "Emfix" runs are displayed and the text discusses that "Emfix" runs represent the effects of varying meteorology. In Figure 6, it is the effect of changing emissions that is calculated from the "Emfix" runs. It is important to clarify that in Figure 6 the % variability explained by varying emissions is calculated from the difference in the "Emfix" run compared to the "Base" run. Stating this another way, the year to year variability in the "Emfix" fun is due to meteorology, but the difference between the "Base" run and the "Emfix" run is due to emissions.

4.2.1 Tropospheric ozone IAV

S18) Page 14, line 26. Figure 8 seems out of place here, it fits in better with section 2.

4.2.2 Tropospheric CO and OH IAV

S19) Page 15, line 19. The authors state that their results completely contradict those of Szopa et al. (2007). This is an important issue. Can the authors rule out that the meteorology or emissions were substantially different between the two studies?

Conclusions

S20) Page 17, line 4. "around 80% of the ozone variability can be explained by changes in meteorological conditions." As discussed in section 3.2, changes in meteorological conditions can influence ozone through changes in transport or changes in ozone chemistry. It would add value to the paper to understand whether changes in transport or chemistry were more important. Although further models runs would be required to quantify these effects, some qualitative statements can be made based on the ozone budget results in Tables 3 and 4.

Technical corrections

1. Introduction:

T1) Page 3: line 2. Delete "to" - "reaching to" a variety

T2) Page 3, line 8. "One factor . . ." in this paragraph be clear to not to confuse changes in emissions from year to year influence tropospheric composition from year to year with trends in emissions affect trends in composition.

T3) Page 3, line 25. Re-phrase this sentence, e.g. "long range transport events initiated by lifting of surface pollution by weather systems or deep convection".

T4) Page 3, line 28. "high amplitude changes"- re-word to increases?

T5) Page 4, line 12. "factors affecting IAV by . . .method based on Sauvage et al. (2008)- be more explicit or delete.

2. Model set-up

T6) Page 5, line 10. "intense wildfire events –where? Refer specifically to the tropics

and the 1997/8 El Niño event.

T7) Page 5, line 14. State that for the "Emfix: run lightning NOx emissions still vary.

T8) Page 6, line 13. Re-phrase "reported by" (van Nioje)

T9) Page 6, Line 26. Discrepancies in emissions or model processes may also be important.

3.1 Tropospheric NO2

T10) Page 7: line 10. "Recall . . ." this sentence is unnecessary.

T11) Page 7, line 18. Figure 2 suggests that only in 1998 were there important biomass burning events in Siberia/Canada. In 1997 there seems to be reduced emissions activity in this region compared to the 5-year average.

T12) Page 9, line 19 are a) and b) really distinct? I suggest re-phrasing this sentence. The wording "strongly" negative NAO phase is likely to be more appropriate since the NAO was also in its negative phase in 2000, but no increases in NO2 columns due to meteorology are simulated.

T13) Page 9, line 28. Is there any interannual variability in the shipping emissions data between 1996-2000 to be detectable? If there is and this paragraph is important text, then it should be included as an additional column in Table 1.

3.2 Tropospheric ozone

T14) Page 10, line 11. It would be helpful to split this sentence up to explain the different effects of meteorological processes i.e. transport vs humidity on column ozone.

T15) Page 10, line 15. Include ozone loss in Table 4 so that the amount or % can be referred to in the text here.

T16) Page, 10, line 16. "The effects of NOx . . .also". This sentence is misleading as changes in biomass burning emissions (Co and NOx) are already mentioned in the

previous sentence.

T17) Page 11, line 14. This whole paragraph (in particular the first two and last sentences) should be re-written more carefully and shortened for clarity.

T18) Page 12, line 1. Re-word as this is not strictly true. Humidity effects on ozone destruction have already been discussed in page 10.

T19) Page 12, line 5. Changes in humidity over the tropical Pacific and Indonesia should be described rather than hemispheric scale changes.

4.1 Analysis of global ozone IAV drivers

T20) Page 13: line 9. Specify that +7% is for the base run.

4.2 Regional scale analysis

T21) Page 14, line 2. “Europe as an example of the northern extra-tropics” – re-word.

4.2.1 Tropospheric ozone IAV

T22) Page 14, line 16. “Net chemistry was lower ..so the ozone peak is attributed to decreased export. Decreased export may be a result of decreased net chemistry”. These two sentences are circular, re-phrase.

T23) Page 14: line 21. “La Niña since” –This La Niña event actually began in 1998- re-word or delete text in ()

4.2.2. Tropospheric CO and OH IAV

T24) Page 15: Line 23. Differences in transport schemes should be included here also unless you can rule that out.

T25) Figures 2 and 5. The text on these figures focuses on the El Niño event. It would be clearer to the reader to centre these plots at 180 degrees in order to see the dipole in ozone between the East/Central and West Pacific/Indonesia in 1997.

T26) In Tables 3 and 4, provide the 5-yr mean value also so that the reader can easily assess if the value for a given year was higher or lower than the 5-yr average.

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