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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Can a global model reproduce observed trends in summertime surface ozone levels?" *by* S. Koumoutsaris and I. Bey

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1. Highly variable trends in observations from site to site (for Europe) combined with relatively low model resolution (2x2.5)

We think that a global chemistry-transport model better suits the objectives of this study, notably to quantify the potential role of long-range (inter-continental) transport of pollutants. For our study, the global model (which includes a chemical mechanism as detailed as possible given current computing capabilities and resource) was run at a resolution of 2x2.5 over a rather a long period (15 years) and several sensitivity simulations were performed, which is also key if one wants to distinguish between different

processes. Therefore we believe that we used a tool that is often used to address such scientific questions and needs to be evaluated. Notice that the high variability in O3 trends over Europe is seen in the observations is sometimes related with error measurements. For example, we mention in page 2032 that the two nearby stations of Langerbrugge (10.75E,52.8N) and Waldhof (10.77E,52.8E) in Germany show very different trends in the 5th percentile. In fact, Dr. Solberg has contacted us indicating that Langerbrugge (from EMEP database) and Waldhof (from WDCGG database) are in fact the same station and therefore should get the same trend. We examined this discrepancy and we found that the difference results because the WDCGG ("Waldhof") data for this station has been recently revised and several O3 values in the beginning of the record (summer 1991) have been dismissed because they were too low and inconsistent with the rest of the dataset. However, these low values in the beginning of our study period are present in the EMEP dataset for "Langerbrugge" dataset and are responsible for the much larger trend that we found. As also discussed in Logan et al. (2012), biases as small as few ppb for a few years can lead to very different trends for relative short periods of 10-15 years. Notice that we have removed "Langerbrugge" station from our results as suggested by Dr. Solberg.

2. Ignoring changes in the spatial distribution of emissions (very important for the US);

We have revised the text to clarify that changes in the spatial distribution of emissions are not ignored as erroneously stated by the reviewer. Spatial changes in US emissions are indeed applied in our model for the period from 1990 to 1998. Unfortunately, emission data were not available after 1999 for our model, when we were planning our model simulations. Therefore, for the period from 1999 to 2005, we have chosen to scale the US emission inventory using national emission trends from EPA. We therefore applied a uniform decreasing trend in our emissions which qualitatively agrees with the decreasing trends found in both western and eastern US during the same period (Kim et al., 2009). Note that this procedure is not unusual in global & regional chemistry modeling since emission inventories are rarely available for multiple years (e.g.

Lin et al. 2012). The effects of using for short time period spatially uniform trends is discussed in our results (see section 4.2, page 2036).

3. Ignoring changes in biomass burning emissions.

Indeed, we have examined the effects of year-to-year biomass burning emissions. We performed a sensitivity simulation with climatological biomass burning emissions. The differences between show only minor, insignificant at the 0.05 level, effects on O3 trends in both Europe and the U.S. and this is why we have not included them in the present document. However, we agree that this should be mentioned and we have added an extended paragraph in the revised document. For more details on the effects of biomass burning trends please also read our reply for comment 6 below.

4. Analysis only goes thru 2005. Five additional years would add a lot of insight. While I recognize that it would be a major undertaking to implement these changes, this study would be improved substantially by addressing the points above.

Unfortunately this was not possible at the time the study was done (for example emission inventories were not available) and is now beyond the scope of the study.

5. In summary, the model appears to capture O3 trends in regions where there are robust, spatially broad trends from numerous sites. This mainly applies to the Eastern US, where emissions are known to be decreasing. In Europe, it seems the trends are more variable. As such, we would not expect a global model to be able to reproduce these well at all.

Indeed, the resolution of our model is relatively large to capture the highly variable ozone trends as we also already mention in the paper. However, the model does reproduce qualitatively the overall ozone changes. In addition, the focus of this study also lies on the changes in the background ozone levels, which is associated with changes in long-range transport of pollution and which we think that a global chemistry transport model can more realistically resolve.

6. In the western US, anthropogenic emissions are probably not decreasing (in contrast to your presumed distribution) and biomass burning emissions may be increasing. There is at least one paper that claims a summer increase in O3 due to increasing biomass emissions in the western US.

Please also refer to our reply in comment 2 above. In the western US, anthropogenic emissions were slightly increasing during the period from 1990 to 1999 according to EPA and this is taken into account in our model: until the year 1998 US emission trends vary interannually and spatially based on EPA statistics. However, anthropogenic emissions have decreased in most of the western US cities the following years (Kim et al 2009) which at least qualitatively aggress with the scaling in the emissions that we have applied.

Fires in the Northern Hemisphere are mostly of natural origin (over two thirds are ignited naturally by lightning) and as a result show large interannual variability and their trends are difficult to assess (Giglio et al. 2010). O3 changes in the western U.S. have been suggested to be influenced by local fire emissions (Jaffe et al 2008) and from long-range transport of pollution from Siberian/Canadian fires (Jaffe et al 2004). Our model shows only minor influence (not significant at the 0.05 level) on summer O3 trends over the western US over our study period. The influence is somewhat more important in the low percentiles of the ozone distribution but still not significant during our study period. This is probably related to long-range transport of forest fire emissions that affect the background O3 levels but have less influence on pollution episodes, which occur under stagnant meteorological conditions (high percentiles in the distribution).

7. So while there are some useful results here, the analysis is not nearly as useful as it could be. My detailed comments below are meant to be helpful should the authors decide to submit a revised manuscript. Pg. 2026, line 15: The authors imply that long-range transport is thought to be an important source of O3. There is ample evidence of this for spring, but most studies suggest the impact is minimal in summer (see for

example US NAS 2009).

Indeed long-range transport in the NH is maximum in spring and minimal in summer. Nevertheless, long range transport of pollution can affect ozone background and as such influence O3 exceedances during the summer months. This is a reason why we focus our study in summer, like other studies have also done so in the past (see for example Fiore et al. 2002, Wang et al. 2009, Hogrefe et al. 2011).

8. 2027, line 7: There are much more recent references for surface O3 trends. While recent, the Cooper (2010) paper focuses on free trop O3, not surface. Jaffe (2008) claims that increasing O3 in the western US trends in summer is linked to increasing biomass burning.

Please refer to our reply to our comment above on biomass burning. We are also going to include more references for surface O3 trends in the revised document as suggested by the Reviewer.

9. Line 12:There are also suggestions that CH4 may be important in the changing background O3.

The effect of global rise in methane levels is mentioned in line 14. We specifically discuss the influence of methane in section 5.2.

10. Line 21: There have been other changes since 2005. Why do you constrain yourself to this period. The data are readily available?

Please refer to our reply to the comment 4 above.

11. 2028, line 22: “Interannual varying:::” In the discussion below, you mostly focus on trends in emissions.

We do not understand this comment. We describe in this section the emission inventories and their interannual variation as they are implemented and used in this study (see also Figure 1). Since, we focus mostly on O3 trends in this paper, we find it logical

to discuss in detail the actual trends in the emissions during our study period.

12. 2029: line 6: Uniform scaling across the US??? This is an important mistake for the US.

As stated previously, we did not use a uniform scaling throughout the US and this is now clarified.

13. Line 8: Unclear if Mexico and Canada are still only 10%. US NOx emissions coming down, Mexican emissions going up.

Mexico and Canada emissions account for approximately 10% according to data of 2001 (BRAVO and CAC respectively) (see Wang et al 2009). Mexican emission have been decreasing during the following years according to EPA (http://www.epa.gov/ttn/chief/net/mexico/1999data/mexico_summary_32_states.xls).

14. 2030, line 1: I am unclear how biomass burning emissions through 2005 are included. Text says interannual variations, but cites papers from 1999 and 2003. This is important for summer O3.

Interannual variation of biomass burning emissions are included for all the simulation years (1991 to 2005) using AATSR DATA (following the method by Generoso et al (2003) with few modifications) as described in details in Koumoutsaris et al. (2008). This has been now clarified in the paper.

15. 2031, line 2: While a seasonal average of June, July and August is commonly used, it should be noted that June MDA8s are typically much higher than August.

This is not a problem since we are also looking at the distribution of ozone not only the JJA means.

16. 2032, line 1-10: To what extent is the model-observation correlation driven by seasonal cycle?

There is no seasonal cycle. We use JJA averages.

17. 2033, line 25: “As these data are not available:::” Really?

NOy (total reactive nitrogen) is defined as the sum of nitrogen oxides NO, NO2, NO3, HONO2, and HNO3. As far as we could find, hourly observations of NOy (concomitant to that of ozone) are only available at 3 stations and none in the northwestern part of Europe (all in the Swiss and Austrian Alps) for both EMEP and WDCGG datasets.

18. 2034, lines 1-8: This discussion is confusing. How can the sites be rural, yet have high NOx/VOC. Seems contradictory.

As we mention in section 4.1.1, it has been demonstrated by several studies that even though stations may be designated as rural, they can be affected by anthropogenic local pollution (see Vingarazan et al 2004). It has been shown for example that upward mixing may occur to a greater extent than expected in high elevated locations (which area usually assumed as representative of background ozone conditions), because of convective or orographic lifting, which delivers boundary layer air to such locations (Reid, 2007, Cui et al. 2011). In addition, stations which were classified as rural some years back may have changed in characters as a results of urban expansion and densification.

19. 2037: “5.1 Long-range transport from Asia” Unclear why the authors focused on this. Past work has shown very little impact on surface O3 in summer, with some impact on free trop. Much greater impacts demonstrated in spring. Using more or less the same type of model, why do you expect a different result?

It is true that long-range transport in the NH is minimal in summer. Nevertheless, its influence in exceeding O3 limits can be very important in the summer months and this is the reason why we focus our study in summer, like other studies have also done so in the past (see for example Fiore et al. 2002, Wang et al. 2009, Hogrefe et al. 2011). In addition, the influence of long-range transport of pollution on the trends can be very important on the low percentiles of O3 distribution as discussed in the text.

20. Line 15-16: GEOS-Chem has done well on long-range transport in the past. Many examples of this (eg Jaegle et al 2003; Zhang et al 2008).

That is true that there are many examples and papers showing that GEOS-Chem can represent some features of the long-range transport. However the opposite is also true, and there are some papers that show that the model (like any global model) may have difficulties in representing such processes. At the end of the section 5.1, we discuss some studies that discuss the reasons of the difficulties in the model to represent the long-range transport of pollution. We have extented this discussion in the revised document.

21. 2040, lines 25-27: Not clear what this is referring to. The free tropospheric increase documented by Cooper 2010 is only for spring. The sentence seems to mix modeled and observed results so it is not clear what the sentence is saying.

We have corrected this sentence to make it clearer.

22. Figure 2: Too many lines.

We have reduced the number of lines.

23. Figure 5: I find this figure confusing. There is an inconsistency between number of lines and caption.

We have corrected this figure to make it clearer.

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