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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

Anonymous Referee #2

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The paper deals with longâĂŘterm changes in summer time ozone in the planetary boundary layer in North America (USA) and Western Europe (1990âĂŘ2005), the main aim is to describe and understand the effects of anthropogenic ozone precursor changes (strong decreases in Europe, decreases in North America and strong increases in Asia). The GEOSâĂŘCHEM model is used for numerical simulations. The most important part of the study is the innovative and inâĂŘdepth analysis and comparisons of the numerical results with many monitoring measurements of rural and remote sites (using maximum 8 hour average concentrations as metrics also considering (changes) in their cumulative probability distributions). The model is capable to describe in a semi quantitative way the decrease in highest ozone values whereas the

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model is not able to describe the changes at the low percentile range in a way consistent with the measurements. The study also clearly shows that the observed ozone increase at low concentrations in Europe ("background ozone") is not attributable to Asian emission increase. The reasons discussed to be potentially responsible for the discrepancies with the measurements at low concentrations include (i) problems of the model to describe long range (intercontinental) transport (plumes) in an adequate way; (ii) significant deficiencies of description of anthropogenic ozone precursor longâĂŘterm changes; (iii) changes in flux of ozone from the stratosphere to the troposphere (not well captured by the model); (iv) changes in key meteorological variables. These processes need further study. One might ask whether the use of global numerical simulations (with a horizontal grid of 2x2.5o) is a promising approach to describe PBL ozone changes. Nevertheless, I got the impression that state of the art regional models (see item 1) have also difficulties to describe measured O3 changes in an appropriate way. I have the following comments and suggestions: 1. I suggest to include in the discussion (possibly in the conclusions) also regional modeling results (e.g. (i) Vautard R., Szopa S., Beekmann M., Menut L., Hauglustaine D. A., Rouil L., Roemer M., 2006. Are decadal anthropogenic emission reductions in Europe consistent with surface ozone observations, Geophysical Research Letters 33, L13810, doi:10.1029/2006GL026080. (ii) EEA, 2009, Assessment of groundâĂŘlevel ozone in EEA member countries, with a focus on longâĂŘterm trend technical report series: ISSN 1725âĂŘ2237 (Technical report No 7/2009); (iii) Wilson R.C. , Fleming Z.L. , Monks, P.S. ,Clain G., Henne, S., Konovalov, I.B., Szopa S., and Menut L., 2012. Have primary emission reduction measures reduced ozone across Europe? An analysis of European rural background ozone trends 1996–2005, Atmos. Chem. Phys., 12, 437–454; 2. p. 2027, line 5: I suggest to try to find more recent references for ozone trends at urban sites and at sites downwind of urban centers than those referenced by Vingarzan, 2004; 3. Fig. 3 and 4: I suggest to try to improve: It is difficult to read the very small symbols for the not significant trends and the color code is not suitable to discern trend magnitude; 4. Fig. 5 and 6: Did you check the "robustness" of the fre-

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quency distribution concerning individual sites ? Or: would the frequency distributions look much different when few sites would be excluded ? Or in other words: are the frequency distributions of the individual sites much different from the average ? 5. p. 2033. Last paragr., ff: I find it very useful to derive VOC sensitivity from the numerical simulation, but I don't believe that VOC sensitivity is the same as titration (fast reaction between NO and O3); 6. p. 2035, line 22: " . . .. Indicating performance issues when O3 levels are strongly influenced by background concentrations": In your discussion of the significant and sometimes large discrepancies between numerical simulations and measurements at low concentrations you stress the role of "background ozone" changes. I agree with this argument at high mountain sites but I am less convinced that this this is the only important reason for deviations at typical PBL sites: Here I think that titration could be important as well: At a "background" site NO emissions e.g. from a close road might occasionally influence ozone distributions: Even if the average is possibly well representative of the location the series might be affected by close vehicle emission; 7. p. 2036, last paragr.: You might consider to site and shortly discuss the paper of P.T. Martien, and R.A. Harley, adjoint sensitivity analysis for a three dimensional photochemical model: Application to Southern California, Env. Sci. Technol., 40, 4200âĂŘ4210 (2006); 8. p. 2040, line 4: The model overestimates the decrease in high concentrations of O3 and you list as possible reason a too strong decrease in O3 precursor emissions: Do you have independent information confirming an overestimate of the decrease in O3 precursors in the used emission inventory ? Are available primary pollutant monitoring data confirming the emission inventory changes ?

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