

Interactive comment on “Modeling the impact of solar “brightening” on summer surface ozone over Europe between 1990 and 2010” by Emmanouil Oikonomakis et al.

Anonymous Referee #2

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This study reports on impacts of solar brightening on summer-time ozone levels across Europe through analysis of model simulations examining the impact of changes in radiation on photolysis rates and biogenic emissions. Several studies have previously examined the impact of aerosol induced radiation perturbations on photolysis and subsequent atmospheric chemistry (Dickerson et al., 1997; Benas et al., 2007; Bian et al., 2007; Anger et al., 2016; Wang et al., 2016; Xing et al., 2017) and similar to the current study suggest that the aerosol induced reduction in solar irradiance leads to lower photolysis rates and less O₃ production. Such interactions and feedbacks are a potentially important consideration for design of multi-pollutant control strategies seeking to simultaneously reduce O₃ and particulate matter pollution. Thus studies that help quantify

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the magnitude of these impacts relative to actual changes in composition of the atmosphere are of interest. Though the results reported are along expected lines, I think the manuscript needs to be strengthened to provide the context in which the results should be interpreted. In my assessment the current manuscript will benefit from some additional work in: (i) a clearer description of the design and methodology employed in the sensitivity experiments; (ii) clearer articulation of the assumptions and limitations of these experiments; and (iii) acknowledging that the study does not comprehensively examine the process changes induced by solar brightening between 1990 and 2010, but rather presents model sensitivity analyses that approximate the impact of aerosol burden changes on photolysis rates and biogenic emissions. The following comments and suggestions are offered:

1) The suggestion that the study examines changes in ozone between 1990 and 2010 due to solar brightening is misleading. Multiple atmospheric processes can be impacted by the direct radiative effects associated with brightening in addition to changes in photolysis rates (e.g., thermal reactions, atmospheric ventilation, changes in dry deposition). No specific simulations were conducted to fully represent conditions in 1990. Instead, AOD and surface shortwave radiation conditions “representative” of 1990’s, approximated from changes in measured surface PM at a few sites, were used to perturb one component of the aerosol-radiation system, i.e., photolysis rates. Further, since no comparison of SSR changes over the time period are presented, it is difficult to ascertain whether the induced changes are actually representative of the brightening observed during this period. Thus, I would be careful in characterizing these results as trends or changes over the two decades. The analysis is essentially a sensitivity study and should be portrayed that way, so that the results can be conveyed and interpreted in an accurate manner. In addition to changes in the text, the authors could also consider an alternate, more representative title for the manuscript.

2) The model set-up and sensitivity simulation specifics could benefit from additional clarification. From what I understand, simulations with the CAMx model driven by mete-

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orological fields from WRF and emissions representative of 2010 were first conducted (BASE run). Then a series of photolysis sensitivity simulations were conducted in which AOD used in the TUV photolysis code was somehow perturbed – this description currently is confusing and contradictory across the text.

a) Line 20-24 on page 5 first suggests that the TUV is used “externally” to estimate clear sky photolysis which are then adjusted in the model for clouds. It is also suggested that dry extinction efficiencies and SSA at 350nm are provided to the model. How is the AOD calculation then used to modify the already estimated clear sky photolysis rates? Line 26 on page 7 then suggests that the study used an in-line version of TUV? Which one is it? It seems that an in-line version of the TUV code in CAMx would be needed to conduct the PHOT sensitivities described in Table 1, but from the current description it is not clear. Since much of the analysis focuses on these sensitivities, it is important that the model setup and the experimental design be clearly described.

b) Equation 2 shows how the AOD is estimated and modified. Some parts of the text suggest that sensitivities are approximating the impact of changes in aerosol burden on radiation and photolysis, which may lead readers to assume that the aerosol concentrations in the equation are being modified. However, I believe in the PHOT experiments the AOD is solely perturbed by the adjustment factor (pf). Please clarify.

c) If the perturbation is only induced through the adjustment factor, then the photolysis changes are only estimating the impacts on a chemical regime representative of 2010. I would imagine if (higher) emissions representative of the 1990s were used then the estimated changes in ozone due to the corresponding changes in photolysis would have been even larger.

d) Were photolysis rates through the model column perturbed by the same amount? Were the perturbations at the surface (or within the boundary layer) different from those in the free troposphere?

3) The authors should better explain the criteria for the choice of the observation loca-

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tions used in estimating the trends in PM (section 3). Why were sites only in Switzerland and Netherlands (3 each) used and how they can be considered to be representative of regional PM trends across Europe?

4) Lines, 15-20 on Page 10 discuss the estimated trends in PM at the observation sites and quantify the changes to be 41-44%. How are these changes then used to estimate the 50% and 65% perturbations to the AOD for the sensitivity tests described on page 8?

5) Evaluation statistics for the BASE calculation are provided in Table 4, without much information on the measurements themselves -location, time, etc. Without such information it is difficult to gauge what these statistics represent. I believe correlation coefficient shown here is representative of the spatial variability captured by the model and not the “temporal evolution” as suggested on Page 19, line 30.

6) Section 4.3.2 and Figure 5: Please provide more details on how the SSR values are estimated. Are they from the WRF simulation or TUV? How different are the SSR from the two? Please emphasize and clarify the assumption that the changes in radiation only impact the photolysis rates and no other aspect of the modeled chemistry and transport.

7) Page 14, lines 1-2: I think the authors should caveat the conclusion that feedback chains associated with secondary aerosol formation and subsequent aerosol burden have negligible impact on photolysis rates. Direct radiative effects on temperature and boundary layer ventilation are also important effects that can modulate secondary organic aerosol production – since these effects are not accounted for in this study, I would caution against a broad conclusion.

8) The impacts of photolysis changes on seasonal average ozone mixing ratios are estimated to be rather modest (a few percent). I would imagine the impacts on daily maximum ozone values will be larger and would be of greater interest. It appears the authors have analyzed those impacts also, but have not presented them here. I think

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many readers would be interested in impacts on daily maximum ozone.

9) The discussion in section 4.6 involving conversion of the change in ozone from the sensitivity runs to a trend over two decades and comparison with other reported trends is not convincing, especially given the range in the trends (0.06-0.16 ppb/yr). Given that the current study only examines the change induced by a single DRE process (i.e., photolysis) on a chemical state representative of 2010, I do not see how it can be converted and compared to a trend inferred from observations that have been influenced by many more chemical and physical processes that are not even approximated in this analysis.

10) A recent study by Xing et al. (2017) analyzes the impacts of aerosol direct effects on tropospheric ozone through changes in atmospheric dynamics and photolysis rates. For summertime conditions in China they report comparatively larger impacts on ambient ozone induced by DRE impacts on atmospheric dynamics (through stabilizing of the atmosphere and modulation of dry deposition) than photolysis. Their results suggest that reducing the aerosol DRE (as would happen in a brightening scenario) will benefit the reduction of maximum O₃ in summer driven both by changes in photolysis and to a larger extent atmospheric dynamics. Could similar impacts of DRE changes also have occurred over Europe during the 1990-2010 brightening period?

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