Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2019-718-RC1, 2019

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modeling Trans-Pacific Transport and Stratospheric Intrusion of Tropospheric Ozone using Hemispheric CMAQ during April 2010: Part 2. Examination of Emission Impacts based on the Higher-order Decoupled Direct Method" *by* Syuichi Itahashi et al.

Anonymous Referee #1

Received and published: 1 October 2019

This paper presents high-order sensitivity analysis modeling of the impacts of emissions and stratosphere to troposphere transport on ozone. Modeling is done with a relatively recent hemispheric version of a widely-used photochemical model. While the methods are not fully novel, they pull together two relatively advanced techniques: HDDM and hemispheric modeling. Explanations are for the most part clear. I recommend publication after addressing the comments below.

Major comments: 1. It is difficult to reconcile the concentration and zero-out contribution estimates. In Figures 6 and 7 and Table 2, concentrations are much larger than the sums of the ZOCs. For example, over most mid- and high-latitude locations in the NH top of the free troposphere (Fig 6, top, c), ozone mixing ratios are over 75 ppb, but the zero-out contributions of East Asian emissions, USA emissions, and stratospheric ozone add to less than half that much. What's the source for the rest of the O3? Sure, there will be influences from other regions, but I would have expected these to be the largest contributions and for cross-sensitivity interactions to be small. More exploration and discussion are needed on this. 2. The results around the perimeter (i.e., tropics) in Figure 1 are peculiar, showing negative first-order sensitivity to VOC, positive second-order sensitivities, negative cross-sensitivity. All of these are opposite in sign to what the chemistry would typically suggest. Further investigation is needed to explore the role of boundary conditions or other factors in driving this, or if there is an error in the modeling.

Minor comments on text: p. 1, Line 32: The 250 to 50 hPa layer is actually in the stratosphere, so not "stratospheric intrusions" p. 4, line 4: typo p. 6, line 1: It is not clear to me how the sensitivity to stratospheric O3 is being calculated. p. 6, line 10: It should be noted that because the coarse grid resolution smears out NOx, you may be missing locations where O3 is actually VOC-limited, such as urban cores with intense NOx emissions at subgrid scales. p. 6, lines 21-22: It would be more appropriate to say: "... it can be concluded that ozone is more sensitive to NOx emissions than to biogenic VOCs emissions during April 2010." Also, at some point you should note that not all NOx is anthropogenic (e.g., lightning, soils). p. 10, line 2: Have you identified evidence of "active convection" in the meteorological model, or is this mere speculation? p. 12, line 29: It is difficult to follow where results are being presented on a MD8O3 or 24-hour average basis. Those sensitivities can be quite different.

Specific comments on figures: Fig 1: In the caption, clarify if these are for 8-hour maximum or daily-average results. Fig 9 and 10: Caption needs to say what the emissions

changes were.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-718, 2019.