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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Strong impacts on aerosol indirect effects from historical oxidant changes" *by* Inger Helene Hafsahl Karset et al.

Anonymous Referee #3

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This excellent atmospheric modeling paper presents the revival of an important, too neglected, topic: how gas-aerosol interactions influence aerosol climate impacts. The study quantifies the sensitivity of aerosol-cloud radiative forcing (ERFaci) or "aerosol indirect effect" (preindustrial to present day) to the use of preindustrial versus present-day oxidant fields/input data. The study finds a substantial sensitivity (20-30%), a smaller ERFaci estimate when the preindustrial oxidant fields are correctly applied. The upshot is that when using PI oxidant fields, clouds are brighter in the PI era. Many published and active global modeling studies of aerosol-cloud interactions continue to apply off-line unchanging oxidant fields in the different climatic states. Over a decade ago, a line of research first showed how the anthropogenic aerosol direct radiative forcing is sensitive to changing oxidants (e.g. Berglen et al., 2004; Bell et al., 2003;

Unger et al., 2006). It is timely and exciting to demonstrate quantitatively the sensitivity of the ERFaci to atmospheric photochemistry. The study does a fine job to identify the chemical mechanisms responsible for this sensitivity via a set of 8 perturbation runs altering the PI or PD state of a single or set of oxidants. NO3 radical changes drive most of the sensitivity. The paper is well written and structured. The figures are all necessary, clear and properly labeled. I highly recommend publication once the following issues are addressed.

1. The preindustrial oxidant fields are from Lamarque et al., 2010. Is it correct to assume that these oxidant fields were generated without on-line aerosol-cloud radiative interactions i.e. this preindustrial gas-phase chemistry does not "see" the brighter preindustrial clouds found in the present study with CAM5.3-Oslo? This question has broader implications. The changes in aerosol-cloud interactions and associated meteorology in PI versus PD state will have an influence on the resultant oxidant levels, not least through altering photolysis rates. How does the application of off-line oxidants here versus fully 2-way coupled on-line oxidants affect the main results?

2. It is not clear how long the simulations are run for in total? However, it is reported that the last 3 years of the run are used for the analyses. ERF allows all feedbacks between land-atmosphere and the land-atmosphere system to come into steady-state with the imposed radiative perturbation. Is the land-atmosphere system in steady-state after only 3 years of running the model? Many of the global chemistry-climate model frameworks seem to run for much longer (even with fixed SSTs and sea ice) to allow for the land-atmosphere system to come into steady-state i.e. more than 20 years.

3. Is it methodologically correct to 'nudge' a simulation and calculate ERF?

4. Is it possible to use the 3 model run years to generate a standard error estimate of uncertainty based on interannual internal variability – thus not providing naked numbers e.g. -1.32 W/m2 and -1.07 W/m2. The numbers may appear somewhat meaningless within the context of ERFaci without any uncertainty range information.

5. Does the preindustrial simulation include a preindustrial land cover map? A few recent studies show a substantial net decrease in BVOC emissions between preindustrial and present day due to the historical cropland expansion (e.g. Heald et al., 2016; Unger, 2013). Temperate zone forests and grasses have been replaced with crops and pasture that represents a loss of BVOCs from the Earth system. The PI-PD SOA and cloud changes are sensitive to the BVOC emission changes. How will the results be affected in the case of higher PI BVOC emissions? In turn, the higher PI BVOC emissions will influence oxidant levels (reducing them further?). It is unlikely that the higher PI BVOC emissions were included in the oxidant simulations in Lamarque et al., 2010.

6. Δclean from Ghan (2013) is introduced on Page 4. Readers from gas-phase chemistry community may appreciate a bit more explanation here (1-2 sentences) on the meaning of Δclean.

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