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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Factors controlling variability in the oxidative capacity of the troposphere since the Last Glacial Maximum" *by* Murray et al.

Anonymous Referee #1

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This article presents an offline coupled GCM-Vegetation-Chemistry model study of the atmospheric oxidation capacity in the present-day, pre-industrial and Last Glacial Maximum atmospheres. It specifically investigates the roles of lightning and fire emissions as well as stratospheric chemistry. The focus on the effect of NO_x is especially valuable in my view as it has not attracted a lot of attention in previous studies. Presenting a complex multi-model setup and several sensitivity tests on 3 different time slices is challenging. I think the manuscript could be improved in order to allow a non specialized audience (e.g. from the paleo-climate community) to better grasp the overall main controlling factors and uncertainties, and major points of agreement versus differences

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with previous studies. The reliability of the main conclusions is currently difficult to assess although this could be improved. Suggestions are provided below.

General comments:

p24521 l6-9, p24543 l6-8 and p24556 l11: at least Crutzen and Brül, 1993, Karol et al., 1995 and Martinerie et al., 1995 have considered changes in the stratospheric ozone burden.

p24521 l28 - p24522 l3, p24551 l4 - p24552 l14 and p24557 l9-10: the discussions of $\Delta^{17}O$ to be published in Soften et al. in prep should be suppressed as no result is shown. Moreover the reasons why this study might lead to a different conclusion than Levine et al., JGR, 2011 regarding its utility as a proxy for oxidant variability are not explained.

p24524 l21 - p24525 l4, p24534 l22-28, p24535 l5-12, p24538 l3-21, and Figure 6: a detailed description of sulfur chemistry and aerosols treatment is provided, however their impact is only briefly and partially analysed (Section 6). Aerosol related perspectives could be provided in Section 8 (e.g. coupled climate-chemistry simulations).

p24527 l20 - p24528 l8; p24530 l5 - p24531 l2; p24541 l10 - p24542 l13; p24544 l24 - p24545 l2: The potential impact of the large O_3 biases for present-day atmosphere on the reliability of LGM results (especially ozone photolysis rates) should be commented. For example, would the conclusion on the strong effect of J_{O_3} be the same if ozone fields from Rind et al., 2009 were used?

Section 2.5 and Supplement: the modelled NO_x levels in present-day atmosphere are not evaluated although NO_x are a focus of this study. For example satellite data could be used as in e.g. Schindell et al., ACP, 2013 (www.atmos-chem-phys.net/13/2653/2013/) or van Noije, ACP, 2006 (www.atmos-chem-phys.net/6/2943/2006/).

p24536 l14 - p24537 l6 and Section 6: I was surprised to see no mention of methane

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isotopes ice core records in the discussions of fire emissions and comparison with ice core-record (see e.g. Sapart et al., Nature, 2012, doi:10.1038/nature11461; Fischer et al., Nature, 2008, Levine et al., JGR, 2011 and references therein).

Specific comments:

p24518 l26 and Section 5.4: I guess that NO_3 is used here as a night time proxy for NO_x ($NO+NO_2+NO_3$), this should be clarified as the term NO_x is used when speaking of emissions.

p24520 l11-12, l18-19, l27-29 and Table 1: the modelling studies agreement on ozone changes does not seem much better to me than for OH results in Table 1. Please rephrase. p24520 l29 - 24521 l5: The discussion of previous results on LGM OH should also mention the study of Levine et al., GRL, 2011 which concludes that methane concentration changes are essentially source driven as in this study.

p24528 l10 - 24529 l4: Different ice sheet topography and SST from the reference datasets in PMIP3-CMIP5 are used at LGM. The major differences between these boundary conditions should be better commented (especially for ice sheet topography), and the impact of these differences on the climate results should be commented.

p24534 l5-8 and p24535 l14 - p24536 l12: the study of Steinkamp and Lawrence, ACP, 2011 (www.atmos-chem-phys.net/11/6063/2011/) should be mentioned in the discussion of soil NO_x emissions, as well as the ACCMIP inter-comparison for present day and pre-industrial soil and lightning NO_x emissions (Stevenson et al., ACP, 2013, www.atmos-chem-phys.net/13/3063/2013/).

p24534 l26-28: DMS results should be compared with Castebrunet et al., GRL, 2006 (doi:10.1029/2006GL027681).

p24537 l8-10 and p24542 l14-18: The uncertainties on fire emission types and emission factors in the context of changing climate (e.g. humidity) and their possible impact on the reliability of the conclusion that OH is insensitive to variations in fire emissions

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should be introduced. The uncertainty analysis in van der Werf et al., 2010 could be used.

p24553 l9-11: the reliability of the LGM methane maximum shift should be commented. For example, is it consistent with bottom-up studies such as Weber et al., 2010?

Table 1: ΔOH results from Bock et al., Earth Planet. Sci. Lett., 2012 (http://dx.doi.org/10.1016/j.epsl.2012.06.052) using a similar design as Martinerie et al., 1995 but updated chemical reaction rates could be mentioned together with Martinerie et al., 1995.

Section 2.4 of the Supplement could be suppressed as it mostly repeats the article main text, or changed into a "Carbon monoxide" Section commenting better Figure 8.

Technical corrections:

p24519 l14: what is meant by "a new model framework"?

p24519 l23: "most oxidants are highly volatile" - unclear. Do you mean very short lived?

p24554 l16: "a Bolivian ice core" - please provide the reference.

The title of the Supplement is slightly different from the article title.

p7 of Supplement: "Liu et al." - incomplete reference

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 24517, 2013.