

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

Anonymous Referee #1

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This paper evaluates the impact of changing concentrations of oxidant between the preindustrial to present day on aerosol-cloud interactions. The change in oxidant, changes the oxidation pathways of emitted gas phase compounds (SO₂, DMS, Isoprene, Monoterpenes) leading to change is the concentration of low/semi volatile compounds and so onto aerosol and clouds. The paper uses an offline calculation of chemistry where oxidants are specified from another model simulation. A highly simplified chemical scheme is then used to evaluate the influence of these changes on a range of aerosol and cloud properties and concludes that there is a relatively large change in aerosol indirect effects due to the change in oxidants. The paper attributes much of this change to the large increase in NO₃ concentrations between the pre-industrial and the present day.

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The paper is in general well written and I think the paper is potentially a useful contribution to the field. However, I have some significant issues with some of the methods used and hence the conclusions. Until these issues are resolved I feel the publication is unwarranted.

Major issues.

Diurnal Cycle of NO₃. The paper identifies the increase in NO₃ between the pre-industrial and the present day as the major change in oxidants. This is most likely true. However, their model treats NO₃ in a relatively unsophisticated manner. The text explicitly says that OH and HO₂ concentrations have a diurnal cycle imposed on them whereas NO₃ is not mentioned suggesting that it does not. In reality NO₃ does have a significant diurnal cycle. Its rapid photolysis leads to low concentrations during the day and high during the night.

Thus, in the real atmosphere NO₃ anti-correlates with isoprene and monoterpenes (this depends to an extent upon the monoterpene speciation) thus NO₃ likely only plays a minor role in the oxidation of these species. By not having a diurnal cycle in the NO₃ concentration this anti-correlation is lost in the model which then likely favours the NO₃ oxidation route over the other oxidations routes.

Given the central importance of NO₃ to the primary conclusion of the paper this lack of anti-correlation provides significant problem that needs to be resolved before the paper can go forwards to publication.

Attribution of impacts The chemistry scheme in the model is fundamental to the magnitude of the impacts simulated. There is little explanation for the choice of yield from the VOCs or DMS. A section explaining these choices and previous work to justify these choices would be useful.

There are couple of ways that the change in oxidants could impact the oxidation of the precursor compounds, and so the production of low volatile products. They could

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change the net yield of compound (SOA.SV vs SOA.LV), or they could change the location of the oxidation. It would be useful to know which process is occurring here. Given that the yield of SOA.SV is the same for all isoprene oxidation routes, any change in the cloud-aerosol feedback due to isoprene can only be occurring because of the change in the location of oxidation - the globally total production of SOA.SV from isoprene is the same in all simulations ($0.15 \times$ total isoprene emission). Monoterpenes behave differently. Oxidation by O₃ leads to the production of SOA.LV whereas oxidation by other methods leads to SOA.SV. Here a change in the oxidant may lead to significant changes in the production of the different SOA times. A similar change might occur with DMS oxidation. The authors attribute all of the changes they see to changes in the location of the oxidation, but they don't really give much evidence to support this. Figure 9 shows the change in the fractional oxidation as a function of height, but this doesn't show in absolute terms how much of the precursor is oxidized in different places. This is a little misleading - very little of the monoterpene / isoprene is oxidized in the upper troposphere compared to the lower troposphere. Perhaps showing the absolute change in oxidation would help with this? This would probably need to be on a log scale.

It would be useful to have a table or figure which shows how much H₂SO₄, SOA.LV, and SOA.SV is produced globally by each route for each simulation.

Given the length of the description of the impact on the aerosol and clouds there should be an increase in the description of how the chemistry is making this impact. This explanation of why the emissions are changing the SOA.LV, SOA.SV and H₂SO₄ tracers is somewhat weak.

Minor comments.

Can the total mass of isoprene and monoterpene be included into a table somewhere? The MEGAN scheme can lead to significantly varying emissions depending upon the implementation into the transport model. It would be useful to know these values. It

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would also be useful to state that the assumption all of the mono-terpene emission is considered to be alpha-pinene which seems to be the implication of the rate constants chosen.

The chemistry scheme also doesn't seem to include aqueous $\text{SO}_2 + \text{H}_2\text{O}_2$. Although this is occurring within the cloud phase it is still a chemical reaction and for completeness I think it should be included.

Figure 9: Figure Caption. The language in the figure caption should be re-examined as it doesn't make sense

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