

Interactive comment on “Sensitivity of tropospheric loads and lifetimes of short lived pollutants to fire emissions” by N. Daskalakis et al.

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We thank the reviewer for his careful reading of the manuscript and the constructive comments. We have addressed all of them as below explained.

The paper reads smoothly and the results seem logical and valid. However, both the validation and the discussion of the results fall a bit short. After reading the paper, it remains unclear what we have learned from the paper. I think the authors should try to improve both aspects of the paper along the lines discussed below.

The validation part has been extended to include satellite (TES) observations and ozonsondes (new section 4.2). Clarifications on surface observational stations have been provided in our reply to reviewer 1. The discussion of the results has been also

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extended by comparing with earlier relevant studies and by developing the section on the feedback mechanism. Finally, the conclusion has been rephrased to highlight the key finding that concern the sensitivity of tropospheric loads and lifetimes of pollutants to the different biomass burning emissions, the isoprene-biomass burning feedback and the recommendations for locations for new measurements that could better constrain biomass burning emission inventories.

Some validation with satellite data (MOPITT, IASI, OMI) would certainly differentiate the different model runs and could hint towards quality differences: which emission inventory performs best, and which emission height distribution leads to the best comparison?

Unfortunately from our comparisons none of the studied biomass burning emission inventories stands out to perform the best. This is now discussed in the new section 4.2.

One aspect that is particularly interesting in the paper, is the indirect effect of the biomass burning emissions. For instance, the lifetime and burden of isoprene is impacted, because biomass burning seems to enhance the oxidizing capacity of the atmosphere. However, once the results become a bit more complicated, the discussion in the paper tends to stop. For instance, figure 6e and 6f show what happens with OH and isoprene when biomass burning emissions are omitted from the model. Over biomass burning areas this leads to reductions in OH and increases in isoprene. However, outside the biomass burning regions, OH increases, e.g. at high northern latitudes. It might be, that these differences (in

The discussion relevant to isoprene and biomass burning emissions interactions has been further elaborate as explained in our reply to reviewer 1. With regard to the high percentage OH changes computed in the high northern latitudes, the last sentence at the end of section 4.3.1 (new number) now reads: ‘As a consequence of the NO_x and O₃ reductions when fire emissions are omitted, the computed hydroxyl radical (OH)

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load (Fig. 6e) is significantly reduced (5-10

In general, the authors should comment on what we have learned, and also quantify better how additional measurement strategies should look like.

The conclusion of the manuscript has been rephrased to highlight the key findings of the study and recommendations on additional measurement strategies have been also made as explained in our earlier replies.

So, also sonde, and satellite observations should be involved in the validation and discussion. A new section 4.2 is dedicated to present and discuss these comparisons..

Comments on textual issues and suggestions can be found in the annotated manuscript. All comments have been taken into account in the revised manuscript.

P22642, Line 12. Good also to mention the FRP approaches. P22642, Line 15. Much more about this< e.g. Rio Freitas work

These lines now read: 'Several biomass burning emission inventories have been constructed based on burned area, active fire detections, and plant productivity from satellite observations (van der Werf et al., 2010) or on assimilated Fire Radiative Power derived from satellite observations (Kaiser et al., 2012) and experimentally determined pollutant emission factors (Andreae and Merlet, 2001) and assumptions on the state of burning of the biomass (smoldering or flaming, van der Werf et al. (2006)).'

P22649, Line 10. In general: How and why are some stations included? For O3, it would be very interesting to include also vertical information, e.g. of sondes.....

See comments on reviewer1

P22651, Line 21. But outside the region OH goes up!

It is now discussed (section 4.3.3) "In the presence of fires, for the same isoprene emissions from vegetation (Fig. 7e) more nitrogen oxides (NOx) (Fig. 7c) are emitted leading to higher OH radicals in the extended biomass burning region (up to 20

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P22653, Line 2. Well, I think also the OH + O3, etc should play a role....how much?

It is well known that NOx-driven chemistry is the major contributor to O3 losses under high NOx conditions as here studied. We did not perform a reaction-by-reaction budget analysis with our model. Thus we did not quantify the contribution of the HOx and O3 reaction cycle for O3 depletion.

P22653, Line 5. I note that the lifetimes of OC and BC are affected in a different way than CO, isoprene, O3. Why is that? Not discussed at all....!

Aerosols species, like OC and BC, have significant primary emissions from biomass burning and are removed from the atmosphere by dry and wet deposition, while carbon monoxide, isoprene and O3 loads and lifetimes are driven by strong chemical production and loss terms. Thus aerosol species behave differently than these short lived chemically reactive gases. This is now added in section 4.4 (new section number).

P22654, Line 9. how does this depend on resolution?

As above mentioned, we have performed a set of low resolution simulations to investigate the dependence of this increase in the apparent SOA yield from isoprene. While the computed loads of isoprene₅O and isoprene itself are affected by the model resolution, the percent change in the ap

P22654, Line 11. Is this the reason that the OC lifetime responds different compared to the isoprene etc.??? (i.e. burden increases more than the loss, since species are produced above?

See discussion above

P22667, Unclear what the unit Tg stands for (N, S, C, CO, SO2, NO2, ??)

Now it is stated in the caption of the figure.

P 22673, caption: Based on total column?

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This is tropospheric column, now stated in the caption.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 22639, 2014.

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