Answers to reviewers on "Sensitivity of tropospheric loads and lifetimes of short lived pollutants to fire emissions" by N. Daskalakis et al.

We would like to thank the three reviewers of the manuscript for their careful reading and their comments that helped improving significantly this study. All comments have been taken into account in the revised version of the manuscript, inconsistencies have been removed and discussion has been rephrased for clarity.

In particular, the FINN aerosol emissions have been recalculated based on additional instructions provided by the owners of the database. Thus, all related simulations have been performed again and Tables and Figures have been appropriately corrected. Figure and Table captions have been further detailed where necessary for clarity. The discussion has been modified for clarity, where needed, and that on the feedback mechanism between isoprene and biomass burning emissions has been extended as requested.

We further provide a point-by-point reply to the reviewers' comments.

Reviewer #1

General comments

The authors perform sensitivity studies using 3 biomass burning emission inventories, and a study on the height distribution of these emissions. The results are reasonably well presented and discussed. However, I am missing an in-depth discussion on the model dependency of the results (what did other studies find?), and how that together with the uncertainty in inventories would translate in overall uncertainties. The authors should think about their scoping: what they want to evaluate and why? Biomass burning versus fossil fuel? Human controlled versus wildfires? What would the consequence of this work for more impact related work; e.g. climate modeling as was performed in ACCMIP. What are the lesson to be learned?

While surface measurements are explored, I wonder why no similar attempt has been made to compare to satellite observations (CO, aerosol, perhaps O3), which could at least give information on spatial extents of biomass burning plumes. Finally, I thought the isoprene-biomass burning relation is interesting and could be explored a bit deeper.

I give some suggestion in the detailed comments. Despite my criticism, I think this work deserves to be published in ACP as a welcome addition to the literature.

We thank the reviewer for his/her pertinent comments that helped improving the content and the presentation of our results by further clarifying the description of the simulations and the key results and correcting inconsistencies in the manuscript.

In this respect, although the scope of the paper is not to investigate the inter-model dependence of the uncertainties in computing the impact of biomass burning on atmospheric chemistry; we have now further developed the discussion on earlier published studies (in the introduction) that investigated the impact of biomass burning emissions on atmospheric composition and compared our findings with earlier studies (in section 4).

In the revised version of the manuscript, in the last paragraph of the introduction it is now clarified that the aim of the study is to evaluate uncertainties in model estimates of biomass burning impacts on atmospheric composition that are associated with the use of different emission inventories in the same model. The study also aims to identify locations where additional observations can provide constrains for biomass burning emission estimates.

With regard to the comparisons between model results and observations at surface stations, while all available data have been used for these comparisons as mentioned in the manuscript (first paragraph of section 4.1), '*only comparisons at stations that have been selected to make evident differences between the simulations using different biomass burning emission inventories are shown for OC, CO and O$_3$)'*.

Furthermore, detailed comparison of our base case model results with satellite observations is part of a paper in preparation by Myriokefalitakis et al. that is focusing on European pollutant budget analysis. However, to satisfy the reviewer as well as reviewer's #2 general comments, we have performed comparisons of model results with O$_3$ and CO mid-tropospheric columns as observed by TES. None of the performed simulations stands out as more performant when model results are compared to observations. This is now discussed in the revised manuscript in the new sub-section 4.2 and documented by scatter plots of comparison between model results and observations provided in the supplement (Figure S7) of the revised version.

Finally, we further analyze the isoprene-biomass burning relationship seen in our model results in section 4.3.3 as explained in detail in our reply to specific comment by the reviewer.

Specific comments

22640 l 10 to be able to introduce=>to lead to

Modified

22640 l 12 lifetimes, I think one could also express this a load- or is there a specific reason why in l. 10 loads and l. 12 lifetimes are discussed?

We agree that lifetime and load are linked but we want to introduce the lifetime as a measure of pollutant persistence in the environment. This is now mentioned in the beginning of the new section 4.4 (old 4.3) in the revised manuscript. In addition, the explanation of how lifetimes are calculated has been moved from the beginning of the second paragraph of this section to the beginning of the section. "*The lifetimes of pollutants provide a measure of pollutant persistence in the atmosphere. They are here computed as the ratio of the tropospheric load to the loss rate (sum of chemical loss and deposition fluxes) for each model column (first 22 vertical layers of the model). Global mean tropospheric lifetimes are derived from the computed global burdens and losses.*"

22640 l 13 it would be interesting to evaluate and discuss which component are specifically responsible for 'transferring' the changes in oxidant concentrations from biomass burning regions to the much larger regions that have isoprene emissions.

The link between isoprene chemistry and biomass burning emissions is done by NOx-driven oxidant and isoprene chemistry and by the presence of primary biomass burning aerosols that provide surface for partitioning of semi-volatile organics.

The abstract has been slightly modified to described the above analysis: " Computed changes in lifetimes point to a strong chemical feedback mechanism between emissions from biomass burning and isoprene emissions from vegetation that are linked via NOx-driven oxidant chemistry, NOx-dependent changes in isoprene oxidation products, aerosol emissions and atmospheric transport." And in the last sentence, the 'apparent' aerosol yield is defined: "This feedback is shown to be able to increase the apparent secondary aerosol yield from isoprene, defined as the ratio of tropospheric loads of secondary aerosol from isoprene oxidation to that of isoprene, by up to 40%."

22640 l 19 this is an interesting finding, which was probably present in all models, but not as such analysed. What would be the enhanced factor of biomass burning aerosol emissions, but inducing larger isoprene-aerosol yields? Could you define a feedback factor (see below).

We have already calculated and provided an upper limit for this feedback factor of 40% that is the increase in the apparent SOA yield from isoprene computed in S0 (with biomass burning) simulation compared to S4 (no biomass burning).

The feedback impacts on the effective (for clarity this has been changed to 'apparent') secondary organic aerosol yield from isoprene that is defined as the ratio of the tropospheric load of secondary organic aerosol from isoprene oxidation to the tropospheric load of isoprene itself (section 4.3.3 of ACPD version page 22651, lines 15-18). This sentence has been rephrased for clarity and the apparent yield is now also defined in the abstract (last sentence).

The feedback is linking isoprene destruction and aerosol formation via the oxidants (hydroxyl-OH- and nitrate radicals and ozone) that consume isoprene and produce semi-volatile organics but also via primary biomass burning aerosols that provide surface for organics to condense on. In the presence of fires, for the same isoprene emissions from vegetation (Fig. 7e) more nitrogen oxides (NO$_x$) (Fig. 7c) are emitted leading to higher OH radicals in the extended biomass burning region (up to 20% regionally) and slightly lower over northern hemisphere regions with intensive anthropogenic NOx emissions and their outflow. Thus isoprene ambient levels are reduced with the highest reduction over and downwind tropical forested areas. Isoprene global tropospheric column is calculated to be lower by 15% in S0 than in S4 (fig. 7f). However, due to the NOx dependence of the semi-volatile organic compounds formation from isoprene oxidation the total (gas and particulate) isoprene_SOA concentrations change little (1%). This implies an overall 14% reduction in semi-volatile organic compounds formation yield from isoprene oxidation that comes to compensate for the increased isoprene oxidation. In addition, the primary organic aerosols (POA) emitted by biomass burning provide surface for partitioning of semi-volatile compounds, thus significantly increasing the partitioning of organic vapors to the aerosol phase that in turn also stimulate further partitioning to the aerosol phase. Thus the isoprene-SOA partitioning to the aerosol phase increases by 19% in depletion of the gas phase isoprene-SOA precursors. This enhancement is consistent with, although much lower than derived from results by Kanakidou et al. (2000) on the enhancement of SOA formation from biogenic VOC due to partitioning on POA from pollution sources. That earlier study was using higher aerosol yields from BVOC than here and did not account for the later studied NOx-dependence of these yields; it also presented changes due to both combustion and fossil fuel POA. It has also shown that the use of different parameters in the two product yield representation of SOA formation from BVOC can lead to up to 70% of differences in the computed SOA tropospheric burden depending on atmospheric conditions. Tsigaridis et al. (2006) have evaluated the importance of the consideration of the NOx-dependent SOA formation by calculating changes in the SOA burden and characteristics and found that in the current troposphere about 72% of the total SOA mass is formed under NOx-driven chemistry

while in the past this fraction was lower (48%). Note however that large uncertainties and gaps in knowledge exist in the kinetics of isoprene-aerosol formation. Rollins et al. (2009) studying the NO_3 radical-driven chemistry of isoprene-SOA formation, have demonstrated the complexity of isoprene chemistry with respect to SOA formation with a drastic increase in aerosol yield when both double bounds of isoprene are oxidized, thus documenting the aerosol yield dependence on the level of oxidation of the precursors. Ervens et al. (2008) investigations have shown that isoprene aqueous phase chemistry is more efficient (about 40% aerosol yield) than gas phase chemistry (about 3% of aerosol yield) in forming SOA and depends on the water content in the atmosphere and the pH. Carlton et al. (2009) review of laboratory measurements, field experiments and modeling studies concerning SOA formation from isoprene, documented differences in SOA yield parameterizations that most rely on a single set of chamber experiments, while aerosol yields are known to depend on various factors including the relative importance of NOx versus peroxide chemistry, temperature (that affects aerosol components volatility based on their enthalpy of vaporization) and pre-existing aerosol loading. They have calculated differences in SOA load induced by the NOx dependence parameterizations that are up to 30% of the total simulated OA over Eastern USA.

This discussion has been added in section 4.3.3, as suggested by all reviewers.

In addition comments have been added on the impact of vegetation and biomass burning emissions co-location that is linked to the model grid size since co-location area increases with lowering the horizontal resolution of the model. To further investigate the impact of the feedback strength to the model resolution, a lower resolution set of simulations has been also performed. These low resolution simulations give results similar to the higher resolution with regard to the feedback strength (relative changes between S0.0 and S4.0) while the computed tropospheric loads of isoprene and secondary organic aerosol differ between the high and low resolution simulations with low resolution simulation computing about 10% lower SOA and 4% lower isoprene loads.

22640 l 4-l 6 the sentence on function of biomass burning is overcomplete when referring to atmospheric chemistry, and not very comprehensive when discussing overall issue.

Our study is, indeed, atmospheric chemistry oriented as also reflected in the title of the paper.

22641 l 24 biomass burning 'emissions'?

Corrected

22642 l 29. Probably refer to some newer references, as source of both anthropogenic and biomass burning emissions have been changing a lot in the last 25 years, and the views have been changing from CH4 only chemistry to more comprehensive VOCs.

We have included references in the introduction to complement these old but pioneering studies with more recent works by (Freitas et al. (2007); Jaffe and Wigder (2012); Kaiser et al. (2012); Keywood et al. (2013))

22643 l 9-13 check grammar.

Corrected

22643 l 14 Pacic Northwest USA?

USA has been added

22643 l. 22 compared to a standard inventory?

Compared to the simulation using GFEDv3 biomass burning emission inventory.

22643 l. 27 can help in reducing uncertainties?

Changed as suggested.

22644 meteo data: any particular year or years was considered?

For all simulations we use the 2008 data (either meteorological or emissions). This is now added at the end of the 1st paragraph of section 2.

22644 Describe the vertical resolution of the model in the boundary layer, as important for the experiments.

The model's first 4 vertical layers are between the surface and 900hPa. This is now included at the end of the introduction of section 2.

22645 l. 11 this sentence reminds that it is not entirely clear what is actually evaluated, and why? If the purpose is to evaluate only naturally occurring fires, the authors may run in problems, because there is a human influence in many types of fires. The double counting issue is tricky- as there are many small scale waste burning activities that may not be picked up by burnt areas from satellite, while in that same region also large scale burning could be detected. Finally, the AWB sector is arguably one of the most uncertain ones. Some uncertainly analysis is warranted: how do the assumptions on correcting for AWB affect the final answer.

We understand that the presentation of AWB emissions and of the various scenarios used in this study has been rather confusing for the reader. Therefore we have done a number of modifications in the presentation of the inventories used here and the simulations performed to avoid misunderstanding and improve the clarity of the work performed:

Table 2 has been modified by showing biomass burning emission estimates that do not include AWB.

Table 3 has been modified to provide the emissions from AWB in the three different emission inventories used in the present study.

Table 4 that summarizes the simulations performed, now explicitly states the biomass burning and the AWB emission inventories used for each one of the simulations. These simulations do not make any assumption on the AWB emissions other than using the specific inventories.

The text has been accordingly modified in section 2.2 it is now written: '*Since AWB is either included in the anthropogenic emissions or in the biomass burning emissions, caution was taken to avoid double counting of the emissions. For this, the AWB emissions (Table 3) are*

considered separately for the simulations that have been performed for this study (Table 4). The AWB in the ECLIPSE database (approximately 34.5 Tg a^{-1}) amounts to 4.5% of the total anthropogenic pollutants emissions for the year 2008'

The first part of section 2.3 now reads: '*For the present study a number of sensitivity simulations have been performed (Table 4) using different biomass burning emissions (Table 2) and AWB emissions (Table 3), all for the year 2008. For the base simulation (S0.0), the biomass burning emissions from the Global Fire Emission Database v 3.1 (GFEDv3; van der Werf et al. (2010)) are used, excluding the AWB sector, hereafter called GFEDv3-ECLIPSE biomass burning emissions (S0.X), while AWB emissions are taken from the ECLIPSE anthropogenic emissions developed in the framework of the ECLIPSE project. Additional simulations have been performed (Table 4) using both biomass burning and AWB emissions from the GFEDv3 (van der Werf et al., 2010) (S1.X), as well as AWB from ECLIPSE and biomass burning emissions from the Atmospheric Chemistry and Climate Model Intercomparison Project's (ACCMIP; Lamarque et al. (2013); http://ecaad.sedoo.fr) (S2.X) or from the Fire INventory from NCAR (FINN; Wiedinmyer et al. (2011)) (S3.X) and finally a simulation where no biomass burning emissions were taken into account (S4.0).'*

Regarding the comments of the reviewer on possible double counting of emissions in a biomass burning emission inventory, while this can be an important issue when constructing emission inventories, it is out of the scope of the present work that does not construct but uses such inventories to evaluate uncertainties associated with their use in global models.

l. 22646 l.2 Describe where the ACCMIP biomass burning emissions are coming from. If I remember well it was GFED2 for the year 2000. Do all emission datasets refer to the same year or years?

As indicated at the eccad.sedoo.fr web site, ACCMIP biomass burning is a combination of GICC, RETRO and GFEDv2 inventories and the inventory provided at this web site is year specific. The website information is now provided in the text. For the present study we use the emissions for the year 2008 as stated in the captions of Tables 2 and 3 and information is now also added in the first sentence of section 2.3.

22646 l. 7 what assumptions are made in the Dentener 2006 paper? I am wondering if no 'newer' studies are available.

This work has been done for the first AEROCOM model intercomparison exercise and is now commonly used for global modelling. In that paper it is mentioned "Large-scale wildland fire emissions are released distributed over six altitude regimes: 0–100 m, 100–500 m, 500–1 km, 1–2 km, 2–3 km, 3–6 km according to wild-land fire location and type based on detailed work by D. Lavoue (2003, personal communication). Emissions are distributed evenly within each altitude layer. Contributions assigned to heights below the actual surface altitude are moved into the lowest applicable height range while contributions assigned to the 0–100m altitude are always emitted in the lowest model layer.' This information is available in the Dentener et al 2006 paper, so it is not repeated here. More recent studies (Sofiev et al., 2012; Wiedinmyer et al., 2011) are now discussed in p. 22642 second paragraph.

22646 l. 12 What can lead to different seasonality across components?

This discussion has been rephrased based on the corrected emissions databases (aerosol emissions in FINN and NMVOC emissions in ECLIPSE).

22646 l. 16 In line with earlier remarks; why removed AWB from one inventory and not from others?

See our earlier detailed reply regarding AWB and biomass burning emissions.

22647 l. 20 What is the criterion to qualify as 'characteristic': more specific.

This sentence has been rephrased: '*While all available data have been used for model evaluation, only comparisons at stations that have been selected to make evident differences between the simulations using different biomass burning emission inventories are shown for OC, CO and O_3*'.

22648 l 15 . . . Tsigaridis. What was the outcome of this discussion, and to what extent contradicting or confirming discussion here. What is the difference of that paper and this one?

The Tsigaridis et al. (2014) is a completely different paper. We have now clarified this by adding the following text in section 4.1: '*Tsigaridis et al. (2014) OC global model intercomparison exercise has indicated that among the thirty-one models contributing to that study, some models emit all biomass burning aerosols at the surface, while most models distribute them to a number of layers above the surface, typically within the boundary layer. Most models are using GFEDv3 and ACCMIP inventories and all models appear to have similar seasonality in primary OC emissions with increased emissions during Northern Hemisphere summer due to the enhanced contribution of Northern Hemisphere biomass burning emissions from temperate and boreal forests to the total OC fluxes. Kaiser et al. (2012) found systematic model underestimation of smoke aerosol optical depth (AOD) observed by MODIS that can be as high as a factor of 3 on the global scale when emissions from bottom-up inventories like GFED are used. Petrenko et al. (2012) have demonstrated that such underestimate strongly varies by region.*'

22649 The conclusion is that the sparse observation of CO and Particulate do not constrain the inventories. This is perhaps not a really novel conclusion.

We agree with the reviewer that the current observational network does not provide sufficient information to constrain the emission inventories. This is why we suggested in section 4.1 to densify air quality monitoring close to the major biomass burning sources in the tropics, which are virtually absent. Furthermore following also suggestions by the other reviewers, in order to provide specific recommendations for measurement sites, we have calculated the ratio of the standard deviation to the mean of all model simulations to identify locations where biomass burning emission inventories produce the largest model divergence. We consider that these are locations where additional observations can help to better constrain the biomass burning emission inventories.

The figure below (new Figure 5) show these ratios for organic carbon and indicate that systematic observations over boreal regions, Alaska, South Asia and Indonesia can help constraining the used biomass burning emission inventories.
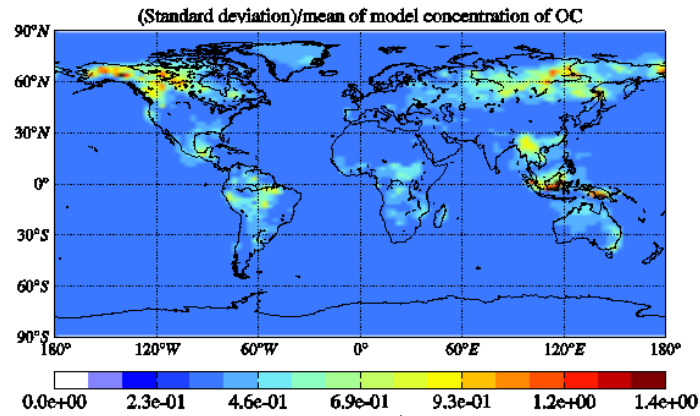

Figure 5 Spatial distribution of the ratio of the standard deviation to the mean of all model simulations, based on annual mean surface concentrations.

This is now discussed at the end of section 4.1 and in the conclusions

22650 l 1 There must be more studies on biomass burning source contributions. I recall the work of Marufu et al, there must be more. An adequate literature survey is relevant in view of evaluating the models sensitivity to biomass burning emissions in general and the effect of using different inventory assumptions. The two together can give some uncertainty range.

The discussion of our results (section 4) has been improved by comparison to relevant results from earlier published studies).

The following references have been added: (Crounse et al. (2009); Duan et al. (2004); Galanter et al. (2000); Palmer et al. (2013); Parrington et al. (2013); Ziemke et al. (2009))

22650 l 26 result in or lead to.

Done

22651 section 4.2.3 is an important section, which could be explored somewhat better, since it is perhaps the most novel analysis of this paper. Specifically I would suggest to analyse what is know in the literature (measurements) about co-occurance of biomass burning and isoprene emissions- the role of grid resolution. Is it possible to analyse a feedback factor (i.e. with and without the feedback process included).

See earlier reply

22651 As I understand it, aerosol yields from isoprene are still quite uncertain. Can the authors discuss an uncertainty range- and how this sensitive to biomass burning emissions. Where are the regions where these isoprene aerosols are becoming relevant (there will be a lot of direct biomass burning aerosol).

As earlier discussed and quantified the emission of primary organic aerosol from biomass burning is increasing the partitioning of semi-volatile products of isoprene oxidation to the aerosol phase. Furthermore, the uncertainties associated with the model spatial resolution and

uncertainties in the yield of the semi-volatile products of isoprene oxidation are discussed in the section 4.3.3 (new section number) as explained in our earlier replies.

The supplementary figure S11 (new figure) shows the spatial distribution of the percent changes in the apparent aerosol yield from isoprene as computed comparing simulations S4.0 and S0.0. This figure points to the areas where the impact of biomass burning emissions (in percent) on the apparent SOA yield from isoprene is calculated by our model to be significant. These changes are most important over the high latitude zone of North America and Asia as well as in the tropical regions over land as well as at the outflow from biomass burning regions. Note however that most isoprene_SOA formation occurs over land.

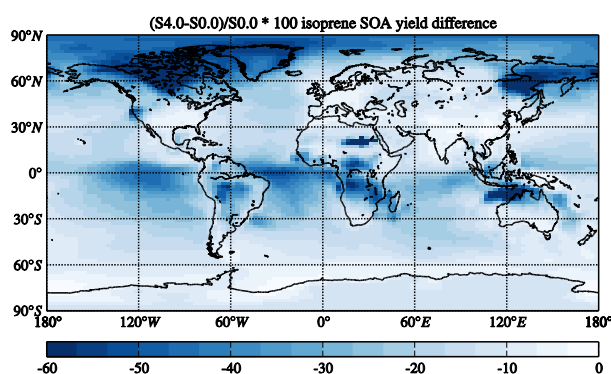This figure is now added in the supplement (Figure S11) and discussed in section 4.3.3.

Figure S11: Annual mean percent changes in the apparent aerosol yield from isoprene as computed comparing simulations S4 and S0. Apparent aerosol yield is calculated as the ratio of the annual mean tropospheric load of isoprene_SOA to the annual mean tropospheric load of isoprene.

Tables 22662 There are a couple of combinations of inventories/components standing out as 'unusual'. E.g. FINN BC/OC 5 to 8 lower than others, GFED-ECLIPSE NMVOC. It would be good to repeat discuss the reasons for such different estimates, as they will determine much of the answer.

22663 see discussion before. I do not understand why NMVOC fraction attributed is so much higher than for other components? A bug?

We thank the reviewer for pointing us these inconsistences due to the interpretation of the databases. These have been now corrected as shown in Tables 2 and 3 and above explained. All simulations affected by these corrections have been performed and analyzed again. The major outcome of the paper remains unchanged.

22664 Table would read easier when just having two columns for varying and surface.

Table 4 has been modified as suggested.

22669/70 Figure 3 and 4 Obviously these are a snap shot of available CO and O3 measurements. How was the selection made?

As earlier explained, while all available data have been used for the comparisons between model results and observations at surface stations (locations provided in supplementary figure S1) as mentioned in the manuscript (first paragraph of section 4.1), *only comparisons at stations that have been selected to make evident differences between the simulations using different biomass burning emission inventories are shown for OC, CO and O₃.* We further clarify this in the first paragraph of section 4.1.

22672 The color scheme of the figures is not very helpful.

For clarity, we have modified the colorbar scale of OC panel.

22673 The numbers below colorbar are not sufficiently describing the scale. Only one plot would be sufficient- they are almost the same.

Following reviewer's recommendation we have removed the OC panel and kept only the BC panel. In addition, we explain in the figure caption that the numbers below the colorbar refer to minimum and maximum values.

22674 Figure 8 In the main text should be some summary of what are the current insights in the ageing of OC and BC; the changes in lifetime displayed here are of course a function of these assumption- which are too my knowledge rather uncertain. Is lifetime applying to the column/burden?

Section 2 (model description) has been complemented to provide information on the parameterization of the ageing of BC and SOA (that for primary OA was already included in the initial version):

"Chemical aging of organic aerosol (OA) is also taken into account. For primary organic aerosol (POA) and black carbon (BC) chemical ageing is considered to occur by oxidation of organic material that coats the particles and is driven by O_3 (Tsigaridis and Kanakidou, 2003); while for SOA chemical ageing to non-volatile SOA (Tsigaridis and Kanakidou, 2003) is considered to occur by reaction with OH at the rate of 4.10^{-12} $molec^{-1}cm^3s^{-1}$, very close to that of the H-abstraction reaction of pinonic acid with OH (Praplan et al., 2012)."

22675 how is lifetime defined in Figure 9/10; tropospheric column?

All figures (either load or lifetime) are for tropospheric columns. This is now added in the captions of Figures 6 (now Fig. 7) and 9 (now Fig. 10), where it was missing.

Reviewer #2

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 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 %) are totally irrelevant, but this should be discussed in the paper.

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 O_3 reductions when fire emissions are omitted, the computed hydroxyl radical (OH) load (Fig. 6e) is significantly reduced (5-10%) over the same regions; while larger percent reductions are computed at high northern latitudes where OH loads are generally very low due to the very weak photochemistry there'*.

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 reviewer#1

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 (NO_x) (Fig. 7c) are emitted leading to higher OH radicals in the extended biomass burning region (up to 20% regionally) and slightly lower over northern hemisphere regions with intensive anthropogenic NOx emissions and their outflow. Thus isoprene ambient levels are reduced with the highest reduction over and downwind tropical forested areas. Isoprene global tropospheric column is calculated to be lower by 15% in S0.0 than in S4.0 (fig. 7f)."

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 O_3 losses under high NO_x 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 HO_x and O_3 reaction cycle for O_3 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 O_3 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 isoprene_SOA and isoprene itself are affected by the model resolution, the percent change in the apparent aerosol yield is almost unchanged. This is discussed in the manuscript and the conclusion where it is now stated *"These computed percent increases do not seem to be affected by the resolution of the model."*

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?
This is tropospheric column, now stated in the caption.

Reviewer #3

Section 2.1. Please include a table that shows the temporal and spatial resolutions of all emission inventories. Natural emissions such as those of isoprene are a function of meteorological variables. Biomass burning also has large temporal variations. These variations seem to have been omitted in this study. Please state what is omitted, give the reasons for doing so, and indicate potential effects on the model results.

Information on the temporal and spatial resolution of the inventories used in the present study has been added in Table 2. We have chosen to account for monthly mean emissions since not all inventories have higher temporal resolution. This is the reason we have also chosen to validate the model results comparing to monthly mean observations. A relevant comment has been added at the end of section 3.

(2) P. 22644, Line 9. How is chemical aging of OA computed in the model? How is BC hygroscopicity computed?

For chemical aging of OA, see our reply to reviewer #1. In our model, BC emissions are by 20% soluble while terrestrial POA emissions are by 50% soluble. For both BC and POA the insoluble fraction is converted to soluble during aging. This is now explained in section 2.

(3) P. 22644, Line 24-25. Why are MEGAN estimated emissions scaled to the PEGASOS estimate? What are the changes?

The biogenic emission inventory used is described in Sindelarova et al. (2014). Changes have been made in the manuscript to avoid confusion.

(4) Table 2, why is AWB NMVOC emission total twice as large as the global non-AWB biomass burning emission (compare GFEDv3 to GFEDv3-ECLIPSE)? The total (mass) amount of AWB burning is much less than non-AWB burning globally.

Rephrasing and corrections have been where necessary, as explained in our replies to reviewer 1.

(5) P. 22646, Line 7. How do the assumed plume heights compared to the observations by Val Martin et al. (2010)? A figure will be very useful.

A figure would give no clear information, since the Dentener et al. (2006) vertical distribution of fire emissions that we use here, assigns a specific percentage of the emitted compounds in pre-specified layer from 0 to 6000m, while Val Martin et al. (2010) has plume retrievals from satellite (MISR) data providing mean and maximum height values.

Taking into account the regions defined by Dentener et al. (2006) (3 regions for North America, Boreal, Temperate and Tropical) and the regions used in Val Martin et al. (2010) (6 areas, Boreal Forests, Boreal Shrublands, Boreal Grasslands, Temperate Forests, Non-Boreal Shrublands and Tropical Forests), we only qualitatively compare the two studies. Thus, according to Dentener et al. (2006) approach boreal fires emit higher than what MISR sees over North America, since there 50% of fire emissions are injected in altitudes from 2000 to 6000 meters while MISR detects a mean fire plume height of about 500 – 1500 meters. Furthermore, according to Dentener et al. (2006) temperate forest fires emit mostly up to 500-2000 meters altitude, in agreement with MISR which detects fire plumes at 500 – 1000 meters mean height. Similar agreement is found for tropical fires with emissions injected between surface and 1000 meters, while the mean plume height detected by MISR is about 700 meters. As short comments has been added in the introduction: '*A plume height climatology over North America has been also derived by analysis of 5-year satellite observations by MISR (Val Martin et al., 2010) which compared to the Dentener et al. (2006) vertical distribution of fires (2000-6000 meters) shows lower mean injection heights (500-1500 meters) for boreal fires but is in agreement for temperate and tropical fires*'.

(6) Figure 1 does not give enough information on spatial differences of the inventories. Please add a figure.

A new figure with spatial information of BC emissions has been added in the supplementary material (new Fig S2).

(7) P. 22647, Line 9-11. This is a very good point. It would be good to provide some suggestions in the conclusions.

Suggestions are provided in the conclusions as earlier explained.

(8) Section 4.1. This is a section that I am mostly concerned. The observations used are obviously incomplete. With a few exceptions, the comparisons show that the model cannot simulate the observed OC and that the observations cannot be used to evaluate if biomass burning emissions or the assumed plume heights are correct. I would have concluded that there is essentially no observational constraint on model simulation results by looking at Figs. 2-4. I would suggest that the writing of this section focuses on if the observations are useful to evaluate model, not on how model results differ among themselves. Model result differences are clear in the sections that followed. At the end of this section, some comments on what if any observations can be used to evaluate biomass burning simulations would be useful.

In addition to the suggested changes in section 4.1 by the other reviewer, and our reply to the previous comment of the reviewer, we have rephrased parts of this section to point out that no

simulation and thus no emission database stands out for its performance in reproducing the observations.

(9) P. 22651, Line 16-19. I am also very concerned with this result. It is obviously an important result, emphasized in the abstract and conclusions. I think it is necessary to provide more detailed explanation on why a relatively small change (5-10%) of OH (Fig. 6e) would lead to a 40% change of isoprene SOA. The formation mechanism of SOA is still very uncertain under low NOx and high isoprene conditions. It is difficult to verify this result using the observations but the model mechanism for this large change can be clearly understood. It may even be possible to point out what future observations should look for.

This section has been further developed as explained in our replies to reviewer #1.

(10) P. 22653 Line 13-14. The NOx lifetime difference seems to be much larger than that of OH (Fig. 6e?). I would have guessed that they are on the same order.
Figure 6e (new figure 7e) shows load differences and not lifetime differences, Hydroxyl radical lifetime (that we know it is extremely short) is not calculated here. Lifetimes are provided in Table 6 and differences in lifetimes in Figures 9-11 that do not include OH. Furthermore, in Figure 11 we refer to NOy lifetime as reported in the figure caption and not to NOx, as erroneously reported in the text. This is now corrected.

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