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Atmospheric Chemistry and Physics Discussions

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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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 O3)'. 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 O3 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 subsection 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 burn-

ing 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 40The 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 (NOx) (Fig. 7c) are emitted leading to higher OH radicals in the extended biomass burning region (up to 20This 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 burn-

ing 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

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 4.5The 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 O3'.

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$_{SOA formation occurs over land. This figure is now added in the supplement (Figure S11}$

Figure S11: Annual mean percent changes in the apparent aerosol yield from isoprene as compouted 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 O3 (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-1cm3s-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.

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

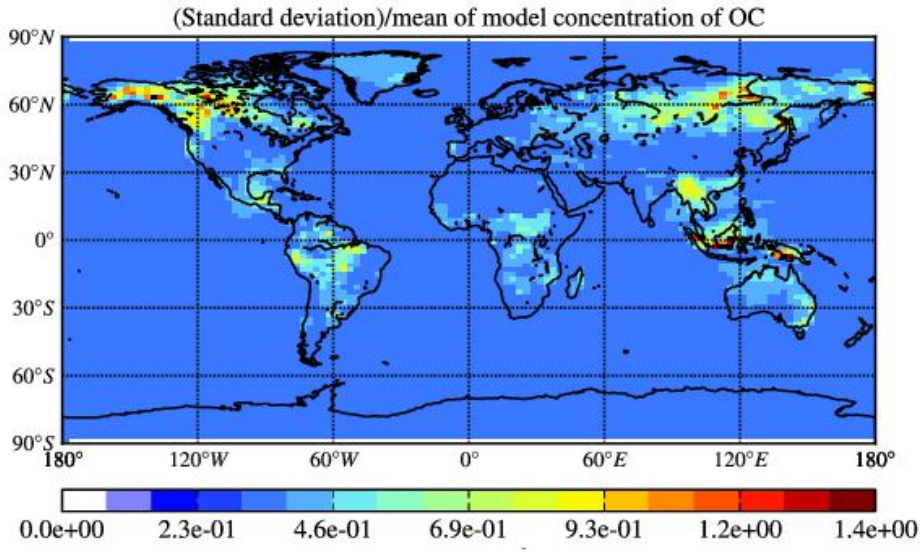

Fig. 1.

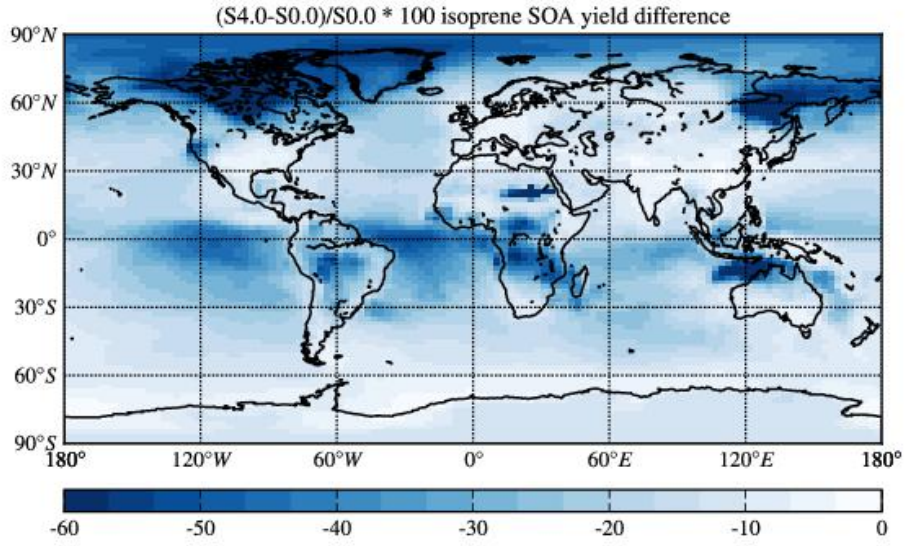
(S4.0-S0.0)/S0.0 * 100 isoprene SOA yield difference
90°N
60°N
30°N
0°
30°S
60°S
90°S
180°
120°W
60°W
0°
60°E
120°E
180°
-60
-50
-40
-30
-20
-10
0

Fig. 2.