

We are very grateful to both reviewers for their very careful and constructive reviews.

1. First review

(a) Role of PAN

- i. *Rapid PAN formation in fresh fire plumes. In fresh fire plumes, NO_x is rapidly converted to PAN in a few hours and then PAN becomes the dominant NO_y form (Mauzerall et al., 1998; Yokelson et al., 2009). NO_x in biomass burning plumes could be solely produced from the slow decomposition of PAN on a timescale of several days. I don't think this feature can be well represented in global models, unless NO_x is partly released as PAN (Hudman et al., 2007; Alvarado et al., 2010). This delay would largely change the interaction between biomass burning and biogenic emissions, as described here. Some discussion on this uncertainty is needed.*

The model does not include any emission of PAN from biomass burning though as pointed by reviewer 1 it may help capture subgrid processes. We will add the following sentence to section 4.2

“Injection of biomass burning emissions outside of the boundary layer ([9]) as well as the formation of PAN in biomass burning plume [5, 12, 1] may, however, extend the spatial impact of biomass burning on tropical ozone [7, 8].”

- ii. *PAN formation from isoprene oxidation. Numerous papers have addressed the important role of PAN on tropical ozone (e.g. Poisson et al., 2000; Roelofs and Lelieveld, 2000; Aghedo et al., 2007). PAN can act exactly as ING, consuming NO_x locally and releasing NO_x remotely. It seems to me that this is another pathway competing with ING chemistry. Relative importance of these two pathways must be addressed in order to understand the impact of isoprene oxidation on tropical ozone.*

In our simulations, we had calculated the sensitivity of ozone and NO_x to the decomposition of PAN. We will add it to the supplementary materials. We find that the effect is significantly smaller than that of the isoprene nitrate yield. The impact of this parameter is masked on the time and geographical scales considered here (>2 months, tropics).

As the reviewer points out, PAN is important for the large scale impact of isoprene. To characterize this impact, one needs to calculate the sensitivity to PAN precursor formation (e.g., peroxy acetyl radical from MVK ...). In this study, we chose to focus on isoprene nitrates. We intend to carry a more detailed study of the influence of PAN in the future.

(b) Other comments

- i. *A (Africa) is not mentioned here in the explanation. Also "ozone" refers to "mean tropospheric ozone"*

corrected as:

“ $S_{\text{A}}^{\text{OH}+\text{ING}_0^{\text{O}_3}}$ refers to the normalized sensitivity of Atlantic mean tropospheric ozone (in % per ° longitude) to the rate of oxidation of ING_0 by OH over Africa.”

- ii. *Page 25624 Line 5: $S_{\text{E}}^{\text{O}_3}(\text{ISOP})$ is minimum from April to May when $D(\text{ING})$ contributes most to LNO_x ”. I don't see this minimum from Fig 7.*

there is not much difference between December-February and April-May. In both periods biomass burning in South America is small and ING chemistry is a major sink of NO_x. We have corrected the sentence accordingly

“ $S_{\text{E}}^{\text{O}_3}(\text{ISOP})$ is minimum from December to May when \mathcal{D}_{ING} contributes most to $\mathcal{L}_{\text{NO}_x}$.”

- iii. *Page 25625 Line 20: “by Eq. (6)” : Is it really Eq. (6)?*

No this was a mistake and has been removed

- iv. *Page 25626 Line 20 are positive:” : Do you mean “ O_3^+ING_0 ” instead of “ $\text{OH}+\text{ING}_0$ ”? “ $\text{OH}+\text{ING}_0$ ” is always negative in Fig. S8.*

This is a mistake it should read $S_{\text{OH}+\text{ING}_0}^{\text{O}_3} + S_{\text{O}_3+\text{ING}_0}^{\text{O}_3}$

- v. *In Fig. 3, please make the minus sign more apparent in the legend.*

Corrected

2. Second review

- (a) *The paper includes no comparison with observed measurements. The paper uses a rather coarse version of GEOS-chem (4 x 5) in the base simulation and changes a number of reactions in this version of GEOS-chem. For the results in the paper to be valid the authors need to show the reference simulation compares reasonably well with measurements. For example Figure 1 shows the tropospheric column from satellite the authors could easily show the model comparison here. Can one tell if the reference simulation gives a better representation of relevant HOx measurements vis-à-vis the standard GEOS-chem version?*

The reviewer raises an important point. The changes in the isoprene mechanism results in an increase in HOx and NOx in tropical regions. This obviously results in more ozone production in tropical regions, thus reducing the bias highlighted by [13]. However, in northern midlatitudes where the standard geos chem simulation performs reasonably well, these changes tend to worsen the agreement with observations. We will include a comparison with observed ozone but this is definitely insufficient to validate (or invalidate) this mechanism. Before such a task can be undertaken, it is essential that the “transfer function” that relates ozone to different emissions and processes be properly characterized. This is one of the primary aim of this study. HOx is greater than with the GEOS-Chem standard model but the resolution of the model is too coarse to evaluate HOx (c.f. [2])

- (b) *The reader really gets little sense of the magnitude of the changes in tropical ozone to changes in the reaction mechanism. Are the actual changes large or small? This could be easily remedied by showing a “high and low” impact on tropical ozone from the sensitivity tests. Also, it would be interesting if the authors could give the overall sensitivity of tropical ozone to the reaction of OH and CH₄ versus that to changes in isoprene yield and isoprene nitrate recycling (e.g., integrate local changes in Figure 5 zonally and give the average sensitivity). Figure S5 gives a hint of this but the units are not particularly helpful.*

The sensitivity to the treatment of isoprene nitrate is fairly large. This has been pointed by many studies before and can be seen on Fig. 4 where changes in the treatment of isoprene nitrate lead to a +/- 20% change in the production of ozone. Similarly Fig. S5 shows that the standard deviation of the distribution of tropospheric ozone columns simulated with the different representation of isoprene nitrates is around 9%.

We have now included the integrated sensitivity of ozone to methane and isoprene nitrate yield in the caption of Fig. 5.

- (c) *The paper presents a complex subject – however I found it somewhat difficult to read. Below I include a number of suggestions of how the authors could help out the reviews to make the paper more readable. Other suggestions are given in the minor comments.*

We are very grateful to the reviewer for his constructive suggestions. We have attempted to follow his suggestions to improve the clarity of the manuscript:

- i. *The readability of the paper would be improved by providing more of a road-map of what the paper is about and where it is going*
 - A. *P25607, l 21-24. At the outset another sentence of two about how the paper goes about diagnosing the isoprene chemical cascade would be helpful (e.g., if the authors would say something about diagnosing the impact of isoprene nitrates as simulated in a chemistry-transport model).*

We have replaced

“This study aims to better characterize the mechanisms that control the spatial extent of the isoprene photochemical cascade, the complex suite of photochemical and physical transformations undergone by isoprene and its photochemical products, as well as its interplay with the Ox and NOx tropical budgets.”

by

“In this study, we first use forward sensitivity simulations to show that the impact of different representations of isoprene nitrate chemistry on the regional budget of ozone can be well understood by characterizing their overall impact on NO_x. We then use adjoint sensitivity simulations to trace the processes that contribute to the long-range influence of the isoprene photochemical cascade on tropical ozone.”

- B. *I think it would be much clearer if the oxidation mechanism given in the supplement is moved to the text. It is very difficult to understand the paper without referring to this supplementary table.*

The discussion in the introduction would also be much clear and more concrete if it referred to the reaction mechanism used. Thus I would recommend that the reactions and discussion given in the introduction be consistent with the set of reactions given in the supplement.

we have added the oxidation mechanism to the main text. We have added references to the reaction given the text and are now referring to specific reactions in Table 1 throughout the text.

- C. *The clarity could be improved by providing a road map at the end of the introduction as to what the various sections in the paper discuss.*

We have added the following paragraph:

In section 2, we briefly describe the model and its adjoint. In section 3, we detailed the treatment of isoprene chemistry used in our simulation that is used as a starting point for the sensitivity simulations and the adjoint simulations. In section 4, we describe how the interplay between ozone and isoprene chemistry varies depending on the chemical environment and the distance to the source. Finally, we conclude by presenting some avenues that may help tease out these different processes and improve their representations in chemical transport models.

(d) *Minor comments*

- i. *Please define NO_x and Ox in the abstract.*

we have defined NO_x and remove Ox since we are addressing tropical ozone

- ii. *The set of reactions given in the supplement refer to the reactions that differ from the standard GEOS-chem mechanism. What is the standard GEOS-chem mechanism? Please give an explicit reference to this mechanism or preferably publish the relevant parts of this mechanism in the paper.*

The standard geos chem mechanism is primarily derived from [4]. The most recent changes are highlighted on the GEOS-Chem wiki (http://acmg.seas.harvard.edu/geos/wiki_docs/chemistry/chemistry_updates_). We will add both references to the table describing the mechanism.

- iii. *Footnotes in the supplementary table of reactions have not been completed. Also please reference where the rates come from*
we added references for the reactions we have modified
- iv. *Nitric acid is not always a terminal sink. It would be more accurate to qualify this sentence somewhat, e.g.:“Unlike nitric acid, usually a terminal sink for NO_x in the tropical boundary layer”*

Corrected as follow:

Unlike nitric acid, usually a terminal sink of NO_x in the boundary layer, organic nitrates may not be a terminal sink of NO_x

- v. *A little more explanation is required as to what x and y are in equation (7) and the range of values. It looks like they are given specific values in the reactions in the supplement. Please explain the relation between this equation as given in text and that given in the reaction table in the supplement.*

We have removed the chemical reaction. and now refer directly to Table 1.

- vi. *The yield “Y” is not adequately defined in the text. Its definition is much clearer referring to the mechanism in the supplement. This is another reason for moving the supplementary reaction table in the main text.*

We have added a reference to the reaction describing the formation of organic nitrates in Table 1

- vii. *known-known:*

Corrected

- viii. *Should read “the RO₂ and HO₂ reaction”*

Corrected: The rate of the reaction of RO₂ with HO₂

ix. *Instead of isoprene do you mean isopo2?*

Corrected

x. *Page 25612. The model description is given (page 25612) before we even know what the model and its adjoint are used for. A brief description of the overall simulation plan would be helpful here.*

We added the following at the end of the previous section

In section 2, we briefly describe the model and its adjoint. In section 3.1, we detail the treatment of isoprene chemistry used in our reference simulation that is used as a starting point for the forward sensitivity simulations (section 3.2.1) and the adjoint sensitivity simulations (section 3.2.2). We then discuss the different processes that control the impact of the isoprene photochemical scale at the regional scale (section 4.1), far from isoprene sources (section 4.2) and immediately downwind of large isoprene sources (section 4.3). Finally, we conclude by presenting some avenues that may help tease out these different processes and improve their representations in chemical transport models (section 4.4).

xi. *In my opinion the emphasis in the text on describing the GEOS-chem model and the adjoint model should change. This reference to GEOS-chem is rather old (10 years). Does Bey et al. give a complete description of the version used in this study? If not give a summary of the changes. In particular convective and boundary schemes utilized may have large impacts on this study. Additional information that should be included is: what is the convective scheme used, the boundary layer scheme used, the scheme for lightning NO_x parameterization? What are the global lightning NO_x emissions? On the otherhand, in my opinion the adjoint sensitivity is described in too much detail. I don't think anything is particularly new in the description given here. I would suggest skipping the details. If someone already understands the mathematics the adjoint the description here will not be very informative. On the otherhand for someone not versed in the adjoint this section is not sufficiently detailed to be helpful. It seems important to emphasize the parameters you are taking the sensitivity to, the units of the sensitivity and the fact it is a linear sensitivity. I think the details of the adjoint equations should be skipped (or put in the supplement).*

A. We have added reference to the geos-chem website that describes the different modeling version in detail. Bey et al. still provide a good description of the main processes included in the model and we have referred to the papers describing the emissions currently used in our model.

B. Emissions and dry deposition are uniformly distributed in the PBL (added to the text)

C. The treatment of convection has been described in [11, 3]

D. tropical lightning NO_x emissions have been added to Fig. S1. In the tropics, there are ~ 61 Gmol/yr (over the time period considered here).

E. We have removed the section devoted to the adjoint and emphasized that the sensitivity calculated with the adjoint is only applicable to small changes in the different parameters investigated here

xii. *The rather coarse horizontal resolution may have an important impact on the results. The NO_x and Isoprene emissions are not properly segregated. The authors should comment on this. What will be the projected impact of the rather coarse resolution on the results?*

Ito et al. ([6]) have indeed shown that the grid resolution has important consequences for the impact of isoprene nitrate chemistry on the simulation on ozone because of the dependence of ozone production efficiency onto the grid resolution. A similar impact was recently highlighted for NO₂ [10]. Most of the impact of isoprene nitrate photochemistry is found outside of the biomass burning when NO_x emissions are low over most of isoprene source regions with an important contribution of soil NO_x. This can be expected to reduce the segregation between NO_x and isoprene source. The importance of the grid resolution is likely to be more significant during the biomass burning period. The different mechanisms identified in this study should however operate regardless of the resolution (e.g., contrast between OPE). but it would be interesting to assess whether their relative magnitude change.

- xiii. *p25616 line 8: “allows” → “allows us”* : Corrected
- xiv. *how is the recycling set? Presumably through alpha. Please be explicit.* : Corrected
- xv. *Page 25612, l 2: Figure 5 does not give the isoprene emissions.* : Removed
- xvi. *Page 25612, on a carbon basis is not necessary here. On any basis.* : Corrected
- xvii. *Page 25614, what are the lightning emissions* : added to Fig. S1
- xviii. *Page 25616, line 18: It would help if you would give a rough estimate in the text about how much is oxidized outside the boundary layer?* : ~ 20%. Added to the text
- xix. *Page 25617, line 5 what is HPALD?*

HPALD is the carbonyl produced by the isomerization of the isoprene peroxy radical. Only its hypothesized products are treated explicitly in our simulations. This acronym is now introduced when isomerization is introduced.

- xx. *Sensitivity simulations: Please explain (or recap) why these are the critical sensitivity simulations to explore.*

We have added the following sentence: This range of parameters captures the uncertainty in the representation of isoprene nitrates in chemical models.

- xxi. *Figure 2 – The dry-dep and wet-dep colors are hard to distinguish here. Also, it would be much more convenient if you put the abbreviations for locations in the figure captions.*

We have used a different colormap that differentiates better dry and wet deposition. We have also corrected the figure caption.

- xxii. *p 25617 lines 24-28. I don't think alpha has been explained at this point and Y is barely mentioned. You might want to repeat definitions of these parameters or emphasize to a greater extent in the preceding text. Again reference to a reaction mechanism within the text would be very useful.*

We have added references to the mechanism table

- xxiii. *Page 25618, line 16 and beyond. This discussion would be more concrete if appropriate reactions in supplementary table are explicitly referred to.*

We have added references to the reactions listed in Table 1

- xxiv. *Page 25622, line 9: it would help here and perhaps in other strategic locations to remind readers of the notation: e.g., to explicitly state “the sensitivity of tropical ozone to African variations in Y is.” – this would make the text less difficult read.*

Done

- xxv. *P 25622 line 10: Could you give more explicitly how the adjoint sensitivity is related to variations in Ding. Are you saying that the seasonal and regional variations in each follow the same pattern?*

Yes. We use D(ING) as a “coordinate” to interpret the adjoint results.

- xxvi. *p25622 Beginning line 11: Can you relate the overall average sensitivity of Y in comparison to that of methane oxidation? It would be informative if the authors gave a quantitative comparison of the two sensitivities averaged over the entire tropics (this also relates to point (ii) in the main points: what is the relative importance of the questions addressed in this paper?).*

We believe Fig. 5 illustrates the importance of the processes explored in this study. In particular it shows the heterogeneity of the sensitivity of ozone to isoprene chemistry that is related to the NO_x regime under which isoprene is oxidized.

- xxvii. *p25622 line 12: The notation here without a subscript to the left of S is initially confusing. While it becomes obvious eventually, it would be easier on the reader if the authors could clarify at the outset*

We have added the following sentence in section 3.2.2. We will use the notation ${}^{R_1}S_B^A$ to refer to zonal changes in the sensitivity of A in region R_1 to B.

- xxviii. *p25622 line 16. But OH is presumably not small throughout the column. Please comment.*

Right but outside of the boundary layer, OH can be expected to be relatively homogeneous. Hence zonal changes in the sensitivity of ozone to methane oxidation reflect changes in boundary layer OH.

xxix. Page 25624 line 19, Please remind readers of the timing of the biomass burning season over this part of Africa

Corrected

xxx. Page 25624 line 20: It is not clear how Figure 7 shows O_x is very low in the boundary layer

We are not sure what the reviewer is referring to.

xxxi. “this competition”. Please remind the reader what competition “this” refers to. Corrected as: The

competition between the local destruction of ozone and its downwind production

xxxii. Overall comment: What is the role of the reaction of isoprene and ozone?

It is fairly small as far as the column is concerned. We had calculated the sensitivity of ozone to ISOP+O₃ rate with the adjoint and we have added it to the supplementary material with the sensitivity of O₃ to PAN decomposition (cf. comment to reviewer 1). We have added the following sentence

${}^t\mathcal{S}_Y^{O_3}$ is significantly greater than ${}^t\mathcal{S}_{ISOP+O_3}^{O_3}$, suggesting that the segregation of NO_x by isoprene nitrate is more effective at reducing ozone than its direct destruction by ISOP + O₃

xxxiii. Figure 5. I believe the sensitivity is not to methane but to methane oxidation

In geos-chem, methane is kept constant. It is therefore equivalent. However, to avoid confusion, we have corrected the sentence.

xxxiv. Figure 5, Why do you use 1%? It should be .028% shouldn't it?

No, 0.028% is per degree longitude, so we need to multiply by 360 to get the overall sensitivity to methane oxidation in this region.

xxxv. What is small s in Figure 8? I thought small s referred to a region.

s designates the sensitivity to an individual grid cell (cf. equation 12). We have corrected it as follow ${}_{R_1}\mathcal{S}_\alpha^{O_3} = \sum_{p>p_i} {}_{R_1}\hat{s}_\alpha^{O_3}$

xxxvi. I find Figure 9 confusing. This figure seems to sum up a lot of the discussion in the paper. However, it is passed over with barely a mention in the text and not well described in the figure caption. Can you describe in more detail?

We have modified the caption as follow

Schematic representation of the spatial modulation of the O_x budget by the isoprene photochemical cascade. Isoprene chemistry alters ozone locally by altering the abundance of NO_x ; reduction in NO_x due to formation of isoprene nitrates ($\mathcal{D}_{ING}/\mathcal{L}_{NO_x}$, continent) leads to reductions in O_x . Downwind of continents, however, the release of NO_x by ING promotes ozone formation. The resulting net impact of isoprene photochemistry on regional ozone, thus, depends on the NO_x conditions near isoprene source regions. Under high- NO_x , an increase in the continental removal of NO_x (green filled arrow) by isoprene results in a net increase of ozone downwind of continents ($\Delta O_3 > 0$). This reflects the enhanced transport of nitrogen reservoirs from regions of low ozone production efficiency (continents), OPE, to regions of high ozone OPE (e.g., oceans) as illustrated by the black arrow. Conversely, under low- NO_x , a similar increase in NO_x removal can result in a decrease of ozone downwind of continents, as the contrast in OPE is not sufficient to overcome the decrease in continental ozone advected as well as the increased loss of NO_x during its transport (e.g., deposition of ING). The removal of NO_x by isoprene nitrate chemistry also increases the amount of BVOCs injected in the free troposphere, promoting the production of ozone. This indirect effect of isoprene chemistry on ozone is of opposite sign to the direct effect and impacts ozone away from isoprene sources.

xxxvii. Figure 10 is also barely mentioned in the text and the notation in the figure caption is confusing. What is (H-NO_x)? Please spend some more time explaining this figure or leave out of the text.

We have modified the figure caption figure as follow

Absolute changes in the adjoint sensitivity of tropospheric tropical ozone resulting from a large increase in anthropogenic emissions of NO_x (cf. text) ($\Delta^t \mathcal{S}_X^{\text{O}_3} = {}^t \mathcal{S}_X^{\text{O}_3}(\text{high tropical } \text{NO}_x \text{ emissions}) - {}^t \mathcal{S}_X^{\text{O}_3}(\text{reference})$, with ${}^t \mathcal{S}_X^{\text{O}_3}$ in $\%/o^2$) to changes in the emissions of isoprene (green), lightning NO_x (red), biomass burning NO_x (blue) and anthropogenic NO_x (black) resulting from a very large increase in anthropogenic NO_x emissions. Green-shaded areas denote regions where ${}^t \mathcal{S}_X^{\text{O}_3}$ become positive because of higher NO_x emissions. The grey shaded region denotes the regions over which the cost function are evaluated.

xxxviii. *This seems potentially like a good figure to show in the paper (not in the supplement), although I don't quite understand exactly what the normalized standard deviation refers to here. Do you mean spatially? Might I suggest showing the absolute changes in the ozone column in response to major point 2 above.*

Fig S5 summarizes the changes in tropical tropospheric ozone due to changes in the representation of isoprene nitrate chemistry. We show the standard deviation in the simulated ozone over the range of isoprene nitrate representation used in this study. As pointed by the reviewer previously, the manuscript already includes many figures. Because the primary focus of this study on processes rather than observations, we believe it is appropriate to leave Fig S5 in the supplementary materials.

3. Additional corrections

- (a) We are grateful to Ying Xie (EPA) who reported the following typos in the mechanism table
 The reaction of MRO2 with NO should have read $\text{MRO}_2 + \text{NO} \rightarrow \text{NO}_2 + \text{HO}_2 + \text{HAC} + \text{CH}_2\text{O}$
 The reaction INPN + OH should yield $0.3\text{INO}_2 + 0.7\text{OH} + 0.7\text{ING}_0$
- (b) We have become aware of an error in the tropopause height saved by the model. This bug affects the results plotted in Fig. 3 and Fig. S5. These figures have been corrected and (fortunately) the changes are small and do not affect our conclusions.

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