

Heraklion, 24 February 2015

Dear Editor:

We would like to thank the reviewer for the useful comments that helped further improve our manuscript.

We have addressed both of them as explained below in a new revised version of our manuscript that we hope it is now suitable for publication in Atmos. Chem. Phys.

We thank you for your time and we are looking forward to hearing from you,

Sincerely yours,

Maria Kanakidou

Point-by-point replies to the reviewer comments:

(1) *“This feedback in the presence of biomass burning emissions increases by about 40% the global mean apparent 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.” I would think that the aerosol yield should be defined as a function of total isoprene emissions or oxidation rates. Tropospheric load of isoprene is not the proper term to use. It is roughly emission/isoprene loss rate constant, whereas SOA load = yield*isoprene load*isoprene loss rate constant/SOA loss rate constant. To use the definition in the paper, one has to assume isoprene loss rate constant = SOA loss rate constant, which is obviously incorrect.*

To satisfy the reviewer we have modified the manuscript to refer to the yield of SOA from isoprene oxidation as a function of total isoprene emissions. The discussion and the associated numbers have modified as follows:

In the abstract the last sentence now reads: ‘ This feedback is shown to be able to increase the global annual secondary aerosol yield from isoprene emissions, defined as the ratio of tropospheric loads of secondary aerosol from isoprene oxidation to isoprene emissions, by up to 18%.’

In section 4.3.3 (page 18) the second paragraph from the end has been modified as follows and Figure S11 has been changed to fit the discussion:

‘This feedback in the presence of biomass burning emissions increases by about 18% the global mean aerosol yield from isoprene emissions that is defined as the ratio of the tropospheric load of secondary organic aerosol from isoprene oxidation to the emissions of isoprene, while locally this difference can exceed 40%. Impacts on the tropospheric loads of the first generation gaseous products of isoprene are smaller, i.e. about 10%. The supplementary figure S11 shows the spatial distribution of the percent changes in the aerosol yield from isoprene emissions as computed comparing simulations S4.0 and S0.0.’

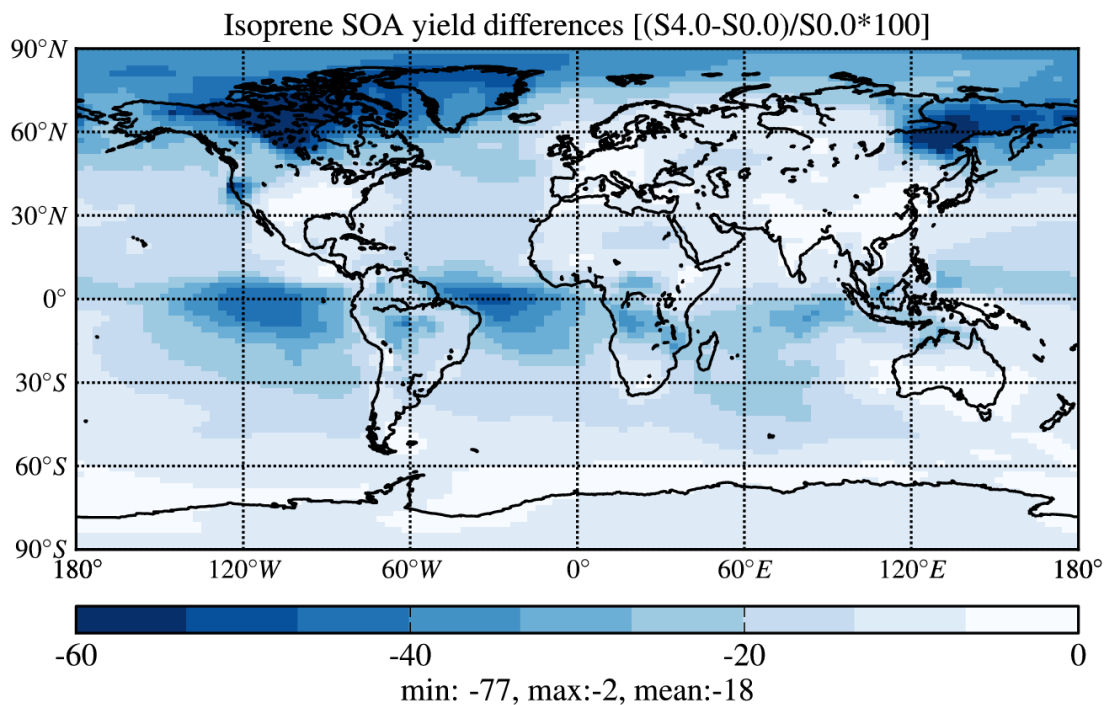


Fig. S 11 Spatial distribution of annual mean percent changes in the aerosol yield from isoprene emissions as computed comparing simulations S4 and S0 $((S4-S0)/S0 \times 100)$. The aerosol yield is calculated as the ratio of the annual mean tropospheric load of isoprene-SOA to the annual isoprene emissions.

(2) I still do not understand Figure 11. “The tropospheric NO_y lifetime ($NO_y = \text{sum of } NO_x, HNO_3, \text{ peroxyacetyl nitrate and organic nitrates}$) strongly responds to the wild fire emissions used in the model, with differences between about -40% and 60%.” Why would biomass burning affect NO_y lifetime by this much? Some mechanistic explanation and quantification should be given. Is it increased aerosol deposition of NO_y due to biomass burning? But I do not understand why the lifetime would increase by that much

The relevant discussion in the last paragraph of section 4.4 has been modified as follows and figure S12 has been added in the supplement.

The tropospheric NO_y lifetime ($NO_y = \text{sum of } NO_x, HNO_3, \text{ peroxyacetyl nitrate and organic nitrates}$) due to depositional losses strongly responds to the wild fire emissions used in the model, with differences between about -40% and 70% when taking into account all simulations that have been performed for this study. Focusing on central Canada and north eastern Asia, the S2.0 simulation results in a large increase in NO_y lifetime compared to S0.0 (Fig. 11b) that is weaker for the S1.0 (Fig. 11a). These differences are mainly attributed to the spatial distribution of the emissions favoring different chemistry pathways and resulting in different fractional composition of NO_y and thus different dry and wet removal fluxes. Note that high differences in NO_y lifetime due to deposition are calculated over the tropical regions with both positive and

negative differences over and off-shore tropical Africa. There, both NO_y burden and deposition losses are increasing due to biomass burning emissions and the corresponding NO_y lifetime in S0.0 varies roughly between 4 to 50 days (Figure S12a). When wild fire emissions are omitted in the model, the absolute differences in NO_y lifetime between simulations S4.0 and S0.0 in the tropics vary from -6.5 to +5 days (Figure S12b). Thus, the NO_y lifetime is increased by up to 67% locally (Fig. 11d), although on global scale a small lifetime change (about -2%) is computed (Table 6).

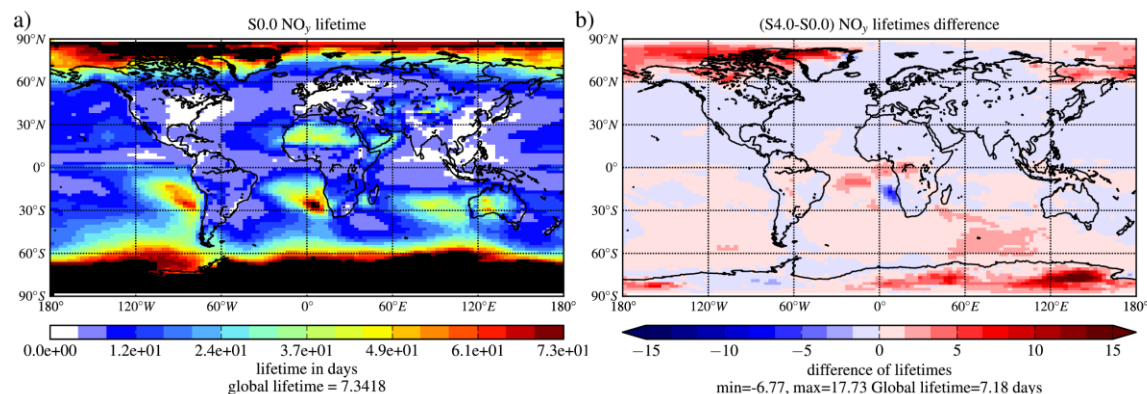


Fig. S 12: (a) Tropospheric lifetime of NO_y due to deposition losses for the S0.0 simulation and (b) absolute difference in NO_y lifetime between S4.0 and S0.0, units are days