Reply to Anonymous Referee #1

The authors would like to thank the reviewer for those insightful comments, which greatly helped to improve the paper.

Major comments:

Presentation: - parts of the paper are too long and divert the reader from the main and strong elements brought by the paper. In particular, some rather general and introductory statements are given all along the manuscript (about the emissions, chemistry and dynamics...). It could be good if the authors try to shorten the paper and keep introductory elements to the introduction. Some examples of lengthy parts are mentioned in the detailed comments. - the paper is very often referencing to results from its accompanying paper which makes the reading and understanding somehow difficult. One example about O3 production regime is given below. - the same is true concerning the supplementary material which makes the paper a bit heavy to handle.

We agree that the presentation could be more focused and try to incorporate all corresponding recommendations. The final shape of the paper will depend on the outcome of this discussion, which might slightly redistribute the weighting between different aspects in the text. Therefore we are cautious singling out individual paragraphs for shortening at this stage, but we do aim to make the main text of the revised version more concise and shorter.

Each reference to the accompanying paper will be reconsidered to make the paper more stand-alone. Some references from the main sections can be moved to the introduction, some can be removed or reformulated to look less mandatory for the understanding of this paper, others can be substituted by existing or new analyses presented in the revised version (e.g. about the O_3 production regime).

We also reconsider each reference to the supplementary material to make sure the main text is understandable without looking in the supplement. We prefer to keep references to individual supplemental figures in the main text, but now state in the introduction that the supplementary material contains only figures that illustrate side aspects, or show some additional details not compulsory for the understanding of the main text.

LiNOx and O3 production: Fig. 3h displays higher LiNOx production from EMAC in the Tibetan part of the ASMA during spring than during summer as mentioned P10L4-5. Nevertheless, the net O3 production is larger in summer than in spring down to 200 hPa below the tropopause (Fig. 2h). The authors explanation is that (i) in spring lower COV are uplifted by convection resulting in COV limitation and reduced O3 production (ii) LiNOx are produced locally in spring and not in summer. The latest argument also appears in the Annexe about LiNOx (p20L14-16).
Concerning (i) 1/ LiNOx production is linked to deep convection, especially in the models where both parametrization are coupled (in EMAC flashes are linked to convective updraught velocity as mentioned P20L10-11). Therefore more LiNOx should be associated with larger uplift of pollutants. Why EMAC displays more LiNOx with less uplifted COV in spring?

Figs. 2 and 3 in the discussion paper show profiles that have been averaged over relatively large lateral regions. Therefore spatial co-location of convection and increased CO mixing ratios in the lower troposphere is not guaranteed. Panels c, f, i, m of Figs. C1 and C2 reflect the spatial and temporal match of deep convection and increased CO in different altitudes. The reviewer's comment mostly concerns the region marked "Tibetan" in Figs. C1 and C2. Convection is indeed stronger in spring (Figs. C1behk) than in

summer (Figs. C2behk), which is also supported by observations (Fig. C3). However, convection in April 2012 is very localized over the coastal regions of Western Bengal and Bangladesh. In contrast, during August 2012 convection is ubiquitous throughout the "Tibetan" region. It is most persistent at the south-western flank of the Himalayas and over the Tibetan plateau. This coincides with the highest CO mixing ratios, which are accumulated there by the prevalent south-westerly winds during the monsoon season. Consequently, more CO is transported through the troposphere in the "Tibetan" region during summer. However, the CO flux decreases towards higher altitudes (Figs. C2cfim), but profiles of the "Tibetan" region show almost constantly increased CO throughout the UT (Fig. 2d in the discussion paper). There is a different explanation for that: In the UT the ASMA is an –although leaky- transport barrier, allowing some accumulation of the uplifted pollutants. There is no such transport barrier in spring.
The manuscript will be revised to include the above reasoning.

2/ Over South and East Asia the season of largest deep convection takes place in summer during the monsoon rather than in spring. Why are there more LiNOx in spring in EMAC?

We compare EMAC-simulated lightning activity (intra-cloud + cloud-to-ground flash frequency) to the corresponding TRMM-LIS/OTD observations (Cecil, 2006) (Fig. C3). Convection is not explicitly resolved in the simulation, and the parameterizations for convection and lightning both introduce uncertainties to the simulation results for lightning. Uncertainties in the observations are due to a space- and time-dependent detection limit of 69% to 88%, and the application of a 3 month smoothing. Considering those uncertainties, the match between simulated and observed global distribution and the orders of magnitude of lightning activity is reasonable. In particular we note that also the observations over South Asia show stronger lightning activity during spring than during the monsoon season. The observed maximum of lightning activity over the coastal areas of Western Bengal and Bangladesh in April is well reproduced by the simulation. The above comparison will be made available with the revised manuscript.

Concerning (ii): Looking at Fig. A1 displaying monthly LiNOx at 168 hPa, we see that they are localized over NW and NE India and Pakistan in May while in August they are more over SE India and Himalaya/Tibet. Nevertheless, the source is much stronger in spring. In both cases, the LiNOx emissions are very "patchy" and localized with also in June a single large emission spot over Bangladesh and in July the LiNOx spot localized over northern central India. Therefore, it is difficult to attribute a lower O3 production to more localized LiNOX emissions in spring.

We agree that there is more lightning-produced NO_x (LiNOx) in the "Tibetan" region in April/May compared to the monsoon season (Fig. 3h of the discussion paper). However, deep convection in EMAC is more evenly distributed throughout the "Tibetan" region in August than in April 2012 (Figs. C1, C2). This is reflected in the corresponding LiNOx distributions (Fig. C4dh). The LiNOx distribution is also more homogeneous in July and September (Figs. C4gi), compared to April – June (Figs. C4def). The strong source of LiNOx in June is also rather limited in time (Fig. 3h).
Photochemical O_3 production (ProdO3) depends on a variety of parameters, e.g. ambient mixing ratios of H_2O, O_3, CO and NO_x (Ehhalt and Rohrer, 1994; Grooß et al., 1998; Jaeglé et al., 1998; Seinfeld and Pandis, 1998). We focus on NO_x and CO in the following (Fig. C5). Prod3 non-linearly depends on ambient NO_x mixing ratios: It increases proportional to NO_x in the NO_x-limited regime, but is almost independent of NO_x variations at higher NO_x mixing ratios. A further increase of NO_x even leads to decreasing ProdO3. Increasing CO has two effects: (i) It increases ProdO3; (ii) It shifts the point of maximum ProdO3 to higher NO_x. Increasing H_2O impacts ProdO3 qualitatively similar as increasing CO. Decreasing O_3 leads to higher ProdO3, but NO_x at the point of maximum ProdO3 is lowest for medium O_3 mixing ratios. Just to give some approximate numbers (Ehhalt and Rohrer, 1994; Jaeglé et al., 1998; Grooß et al., 1998): For UT

conditions at northern mid latitudes the point of maximum O_3 production may vary between 200 and 700 nmol/mol NO_x. Maximum net O_3 production may vary by a factor of about 4, depending on ambient conditions.

We attribute the lower ProdO3 simulated for spring conditions mainly to the following two effects. Firstly, locally very high NO_x in spring does not help ProdO3 – or even pushes the system into the NO_x-saturated regime. NO_x close to maximum ProdO3 conditions throughout the region in summer leads to higher ProdO3 in the lateral average (Fig. 2h). Secondly, more CO in the UTLS in summer (Fig. 2d) increases ProdO3 and the maximum possible O_3 production. We will clarify this in the manuscript accordingly. The above reasoning is also illustrated by an example, see the below discussion of Fig. C6.

Why are the LiNOx emissions so "patchy" on a monthly scale? The averaging should smooth horizontally the distributions because convection does not always occur at the same place. It could be interesting to compare LIS/OTD distributions of lightnings to EMAC LiNOx distributions.

The comparison is shown in Fig. C3 and discussed above. Smoothing applied to the observations certainly makes them less patchy. The parameterizations for convection (Tiedtke, 1989; Nordeng, 1994; Tost, 2006) and lightning (Grewe et al., 2001) used in our simulations have been tested in several studies (Tost et al., 2007; Grewe, 2009; Lopez, 2016) and appear to be state of the art. Simulated and observed NO along the HALO ESMVal flight track agree remarkably well within the ASMA region (Gottschaldt et al., 2017). The same is true for comparisons of CARIBIC (www.caribic-atmospheric.com) measurements of NO and our simulation's output along the CARIBIC flight tracks in the ASMA region for the period May 2005 – April 2014 (Fig. C7). The agreement is particularly noticeable for the monsoon season.

There is no proof that EMAC is right for the right reasons, but at least those comparisons provide some confidence that the ESMVal monsoon case has been captured well by the simulation.

We are nevertheless aware that parameterizations for convection and LiNOx in global models are a notorious source of uncertainty. We do not consider an in-depth discussion of this aspect a focus of this paper, and therefore put most of the discussion concerning LiNOx in an appendix.

Finally, in Barret et al. (2016) the LiNOx are not shown but a sensitivity test shows that O3 and NOx produced by LiNOx are the highest during the monsoon season which seems rather logical for the reasons discussed above. This discrepancy between the EMAC and GEOS-Chem models concerning LiNOx should be discussed.

Our simulations also show that net O_3 production is highest in the ASMA, as well as lightning being an important NO_x source in the UT. We argue that LiNOx alone can not explain higher O_3 production in the ASMA compared to spring. The main difference between the two seasons is the availability of CO (and related precursors) in the UT, resulting from different transport patterns and partial isolation of air in the ASMA circulation. More localized NO_x emissions in spring, combined with the lack of confinement and stirring in the ASMA also contribute to less ProdO3 in spring. Despite higher LiNOx emissions in spring, our simulation shows the highest net O_3 production in the ASMA.

EMAC sensitivity simulations also show a strong effect of LiNOx variations on ProdO3 (Figs. C6pq), so that aspect might not be too different to GEOS-Chem. The EMAC RC1SD-base-10a simulation is certainly not perfect regarding the temporal and spatial distribution of lightning, but at least the aspect of higher flash rates in South Asia during spring compared to summer is supported by observations (Fig. C3).

However, given the uncertainties related to LiNOx, a detailed comparison with other models is definitely warranted. Disentangling the various facets of ProdO3 beyond exemplary snapshots, and in different modelling systems would certainly contribute to a more robust understanding of the peculiar chemical conditions in the ASMA. Since this paper is considered to be too long already, we just add that as a

recommendation for future studies. This will also remind readers of the uncertainties associated with our results, derived from just one family of simulations.

The east-west O3 net production gradient is logical as explained in the manuscript (P10L11-12) and in agreement with previous comparable evaluations by e.g. Liu et al. (JGR,2009) and Barret et al. (ACP,2016). Furthermore, the values of Fig. 2 seems in rather good agreement with those of the above mentioned papers for the monsoon season. A comparison and discussion of the EMAC O3 production with these previous studies could be interesting to strengthen and put the results in perspective.

The agreement of the values for net O_3 production is indeed remarkable, given the uncertainties discussed above. Both studies have been cited already, but we will point out the agreement in the context of the different methodologies.

Referring to the accompanying paper, it is mentioned that O3 production at 168 hPa is rather limited by CO than by NOx (P15L12-13). The exact sentence in the accompanying paper is "Net O3 production seems to depend more on CO (and related precursors) than on Nox". The use of "seems" shows that the authors are rather uncertain. I do not really understand how is this possible because of the rather high CO concentrations within the ASMA (70-100 ppbv according to HALO and all references cited in the paper). This statement of a generally CO-limited regime in the ASMA at 168 hPa needs to be demonstrated and is not supported by the literature. For instance, according to Brune (IGAC Isuue 21, 2000), in the upper troposphere, the O3 regime is NOx limited for NOx concentrations lower than some hundreds pptv.

We illustrate the consequences of Fig. C5 (introduced above) by snapshots from a series of sensitivity simulations (Fig. C6). Going down from the tropopause in the ASMA, NO_x and O_3 generally decrease, while CO and H_2O increase (Figs. 2 and 3, H_2O not shown). Net O3 production (Fig. C6γ) in the UT is determined rather by ProdO3 (Fig. C6δ) than by LossO3 (Fig. C6ζ). Increasing CO increases ProdO3 and decreased CO results in decreased ProdO3 (compare Figs. C6ab with Figs. C6no). ProdO3 per NO_x shows a strong gradient in the altitude of maximum ProdO3 (Fig. C6ε), indicating the transition from the NO_x-limited to the NO_x-saturated regime. The maximum corresponds to about 300 pmol/mol NO_x (Fig. C6β). Variations of NO_x in the altitude region of increased NO_x (above max. ProdO3) have a relatively little effect on ProdO3 (compare Figs. C6gh with Figs. C6pq). ProdO3 even decreases in the region of the largest NO_x increase (Fig. C6m: 22°N, 16 km). CO sharply decreases in that altitude region (Fig. C6α), so the photochemical regime corresponds to the blue section ("3") in Fig. C5. More CO is available in the regions of maximum ProdO3, corresponding to the green section ("2") in Fig. C5. Towards lower altitudes, NO_x decreases and CO increases. Although NO_x increases/decreases less than in higher altitudes, ProdO3 increases/decreases more (~12 km in Figs. C6ghpq). This is the NO_x-limited regime ("1" in Fig. C5). ProdO3/NO_x also varies (Figs. C6tu), indicative of the non-linear region (grey in Fig. C5). The non-linear dependence of ProdO3 on ambient trace gas mixing ratios leads to the simulated maximum within the opposite gradients of those trace gases in the UT ASMA. In principle, all above mentioned operating modes of the chemical system ("1", "2", "3" in Fig. C5) could be in the NO_x-limited regime and still lead to a maximum of net O_3 production in the UT. It's a multi-dimensional problem. CO (and others) determine the curve in the NO_x-vs-ProdO3 diagram, and NO_x determines the operating point on the curve. The formulation in the paper is indeed misleading in that respect and will be revised.

Tracer-tracer relationships:

This part is very interesting because the 3 tracers document different transport and chemical processes. HCl in particular which is rarely used is good to trace stratospheric air because O3 is photochemically produced in the troposphere. The authors explain that "mixing line with negative CO/O3 slope dominate"

in Fig 4c (corresponding to positive slopes in Fig. 5c). They correspond to mixing stratospheric air with photochemically processed tropospheric air. Neverthelesse, in Fig. 5c, we also see some horizontal mixing lines (red and green). According to the discussion in p11 and 12 they correspond to mixing of fresh uplifted pollution (increasing CO, horizontal lines not so clear in Fig. 4c) and stratospheric air (increasing HCl in Fig 5c) with antagonist effects on O3. Another line with O3 decrease / HCl increase in Fig. 5c and O3 decrease / CO increase in Fig. 4c corresponding to mixing of fresh pollution in the UTLS can also be isolated. It is difficult to see whether these mixing lines correspond to important part of the sampled air masses but they could be mentioned.

Explanations for the different types of mixing lines are offered in the context of the hypothetical lines (L1-L5), shown in Figs. 4b and 5b. In the revised manuscript we refer more often to specific hypothetical lines when discussing Figs. 4c and 5c. The discussion of vertical lines in Fig. 5c has been corrected.
A detailed quantification of different processes' contributions to individual measurements would require more sophisticated analyses along back-trajectories.

Details:

Part 3: the description of the different species at beginning of §3.1; 3.2, 3.3 and 3.4 (origin, chemistry etc.) are too close to "textbook" descriptions and should be shorten for readability.

We feel that the main characteristics of each tracer should be mentioned here, at least as far as needed to understand the tracers' behavior in Figs. 2 and 3. We also see the need for shortening and will remove side aspects from the main text.

Part 5: this part tries to describe the different processes that control the ASMA composition one by one. It is interesting and well documented but rather lengthy and descriptive. For instance, the description of the evolution of the CO and HCL distributions P17L1-15 is very detailed and could be summarized.

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P17L1-14: the dynamics are very detailed with the evolution of the air masses but is it really necessary to give so much details?

Will be revised accordingly.

Fig2 and 3: the plots are shown in pressure coordinates that makes the region around the tropopause very compact. Readability would be better in logP coordinates

Will be changed.

(altitude plots are provided in the supplement but it makes the reading uncomfortable and could be simply removed).

Ok.

p5l13: why choose PV = 3.5 for the tropopause in the extratropics? Most studies choose 2 or 1.5.

This is the standard tropopause definition of EMAC in the extratropics, as introduced by Jöckel et al. (2006).

P5l27: Fig.1 is referenced after Fig 2 and 3.

Fig. 1 is referenced first on page 4.

p9l5: "indicates that relatively: : :"

Reformulated.

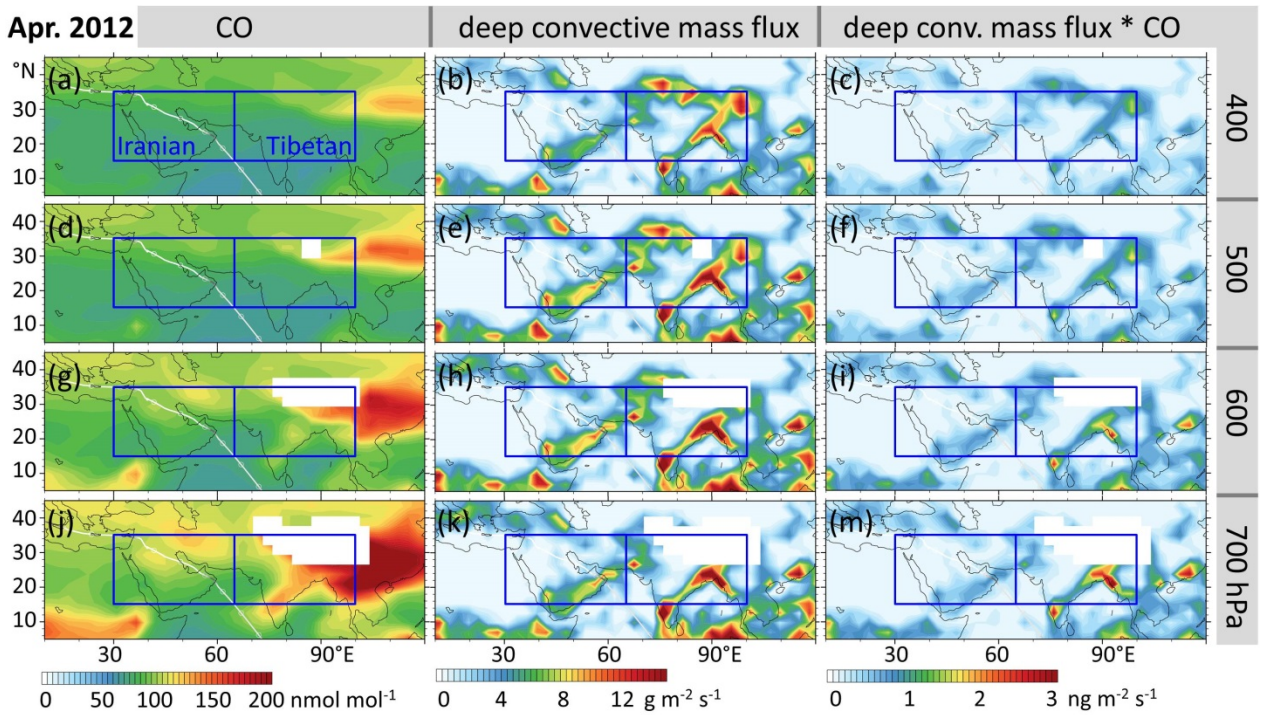

Fig. C1: EMAC-simulated monthly mean distributions of CO and deep convective mass flux in different pressure altitudes during spring (April 2012). The right column shows the deep convective mass flux of CO based on individual output steps. Blue rectangles mark the outline of the regions used to produce Figs. 2 and 3 of the discussion paper.

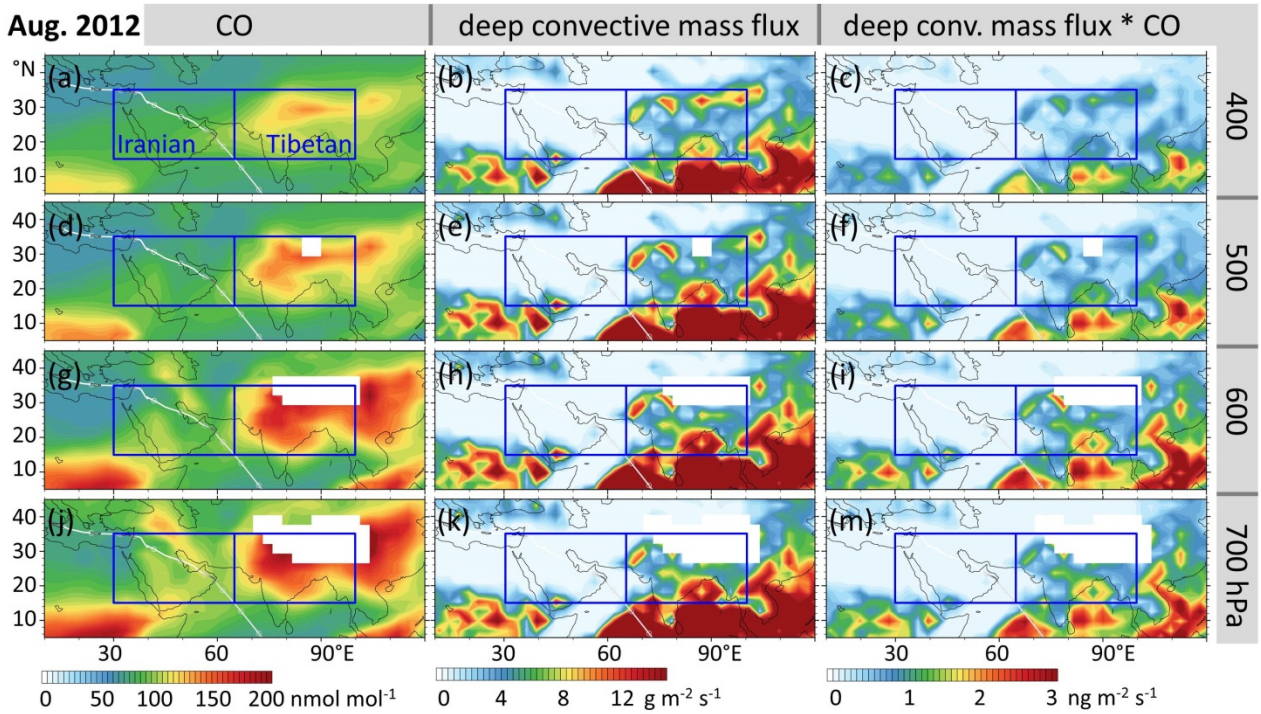

Fig. C2: As Fig. C1, but for August 2012, i.e. during the monsoon season.

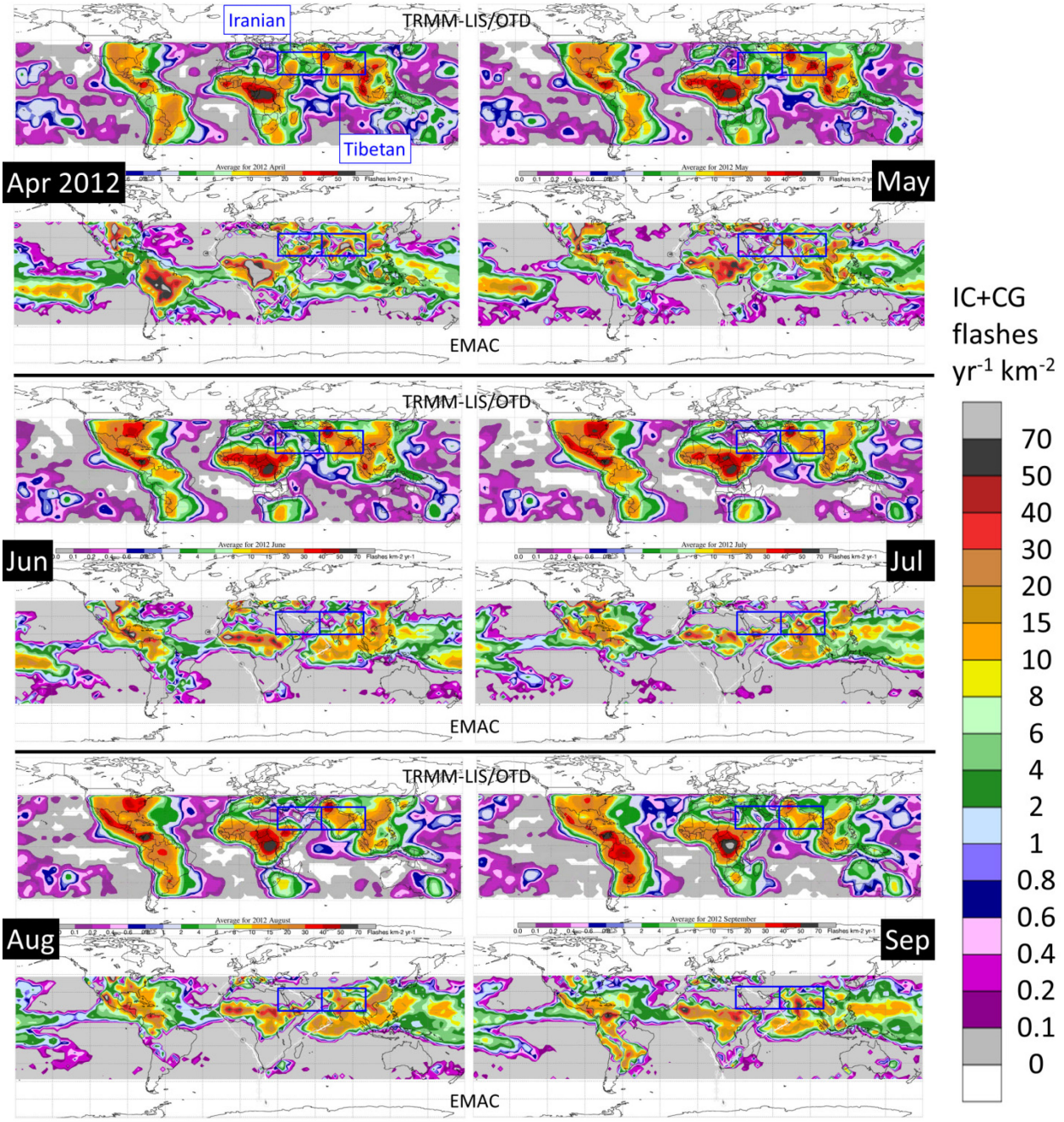

Fig. C3: EMAC-simulated monthly mean lightning activity compared to the corresponding TRMM-LIS/OTD observations. Note that a 3-month smoothing operation has been applied to the observations, which are also subject to a 69% to 88% detection efficiency. Data coverage and color scale are determined by the observations. Simulated lightning appears to be more localized, and thus exceeds the scale more often.

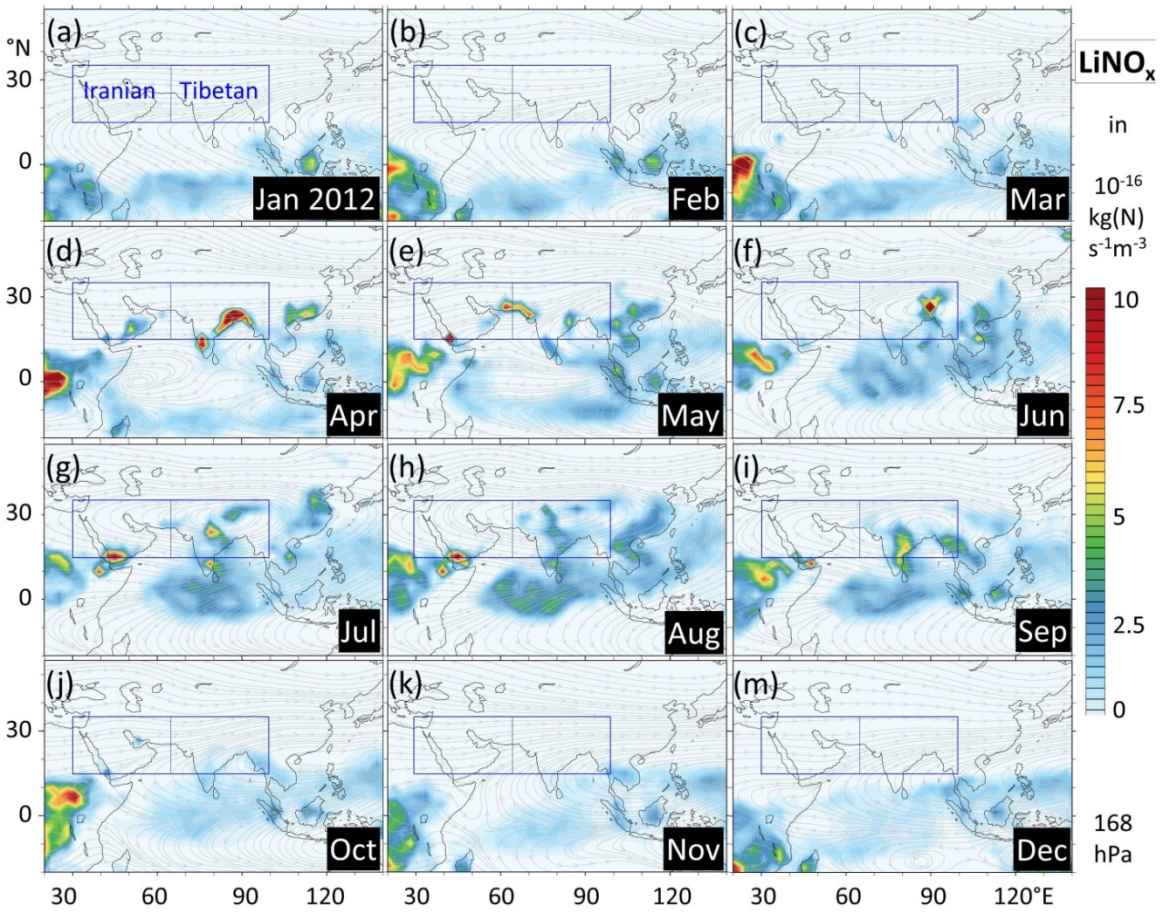

Fig. C4: Monthly mean lightning NO_x emissions in 2012 at 168 hPa, based an EMAC simulation RC1SD-base-10a. This figure is identical to Fig. A1 from the discussion paper, except of corrected outlines of Tibetan and Iranian regions, and added panel annotations.

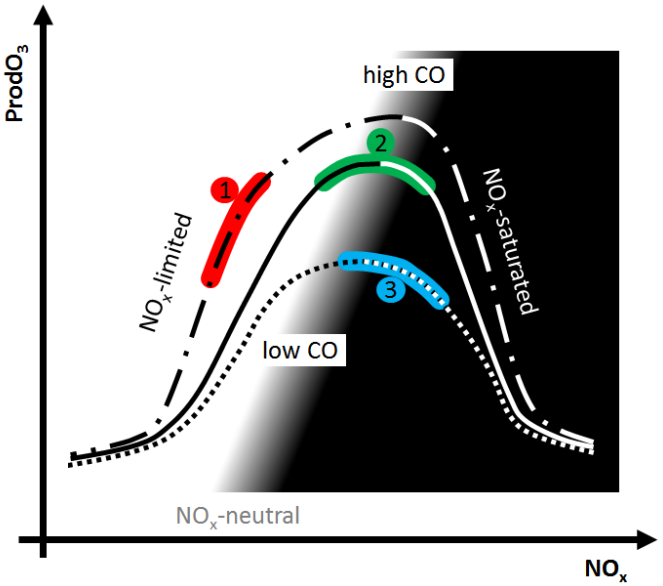

Fig. C5: Schematic of the dependence of photochemical O_3 production on NO_x and CO mixing ratios (after Grooß et al., 1998). Red, green and blue highlight photochemical conditions that are discussed in the text.

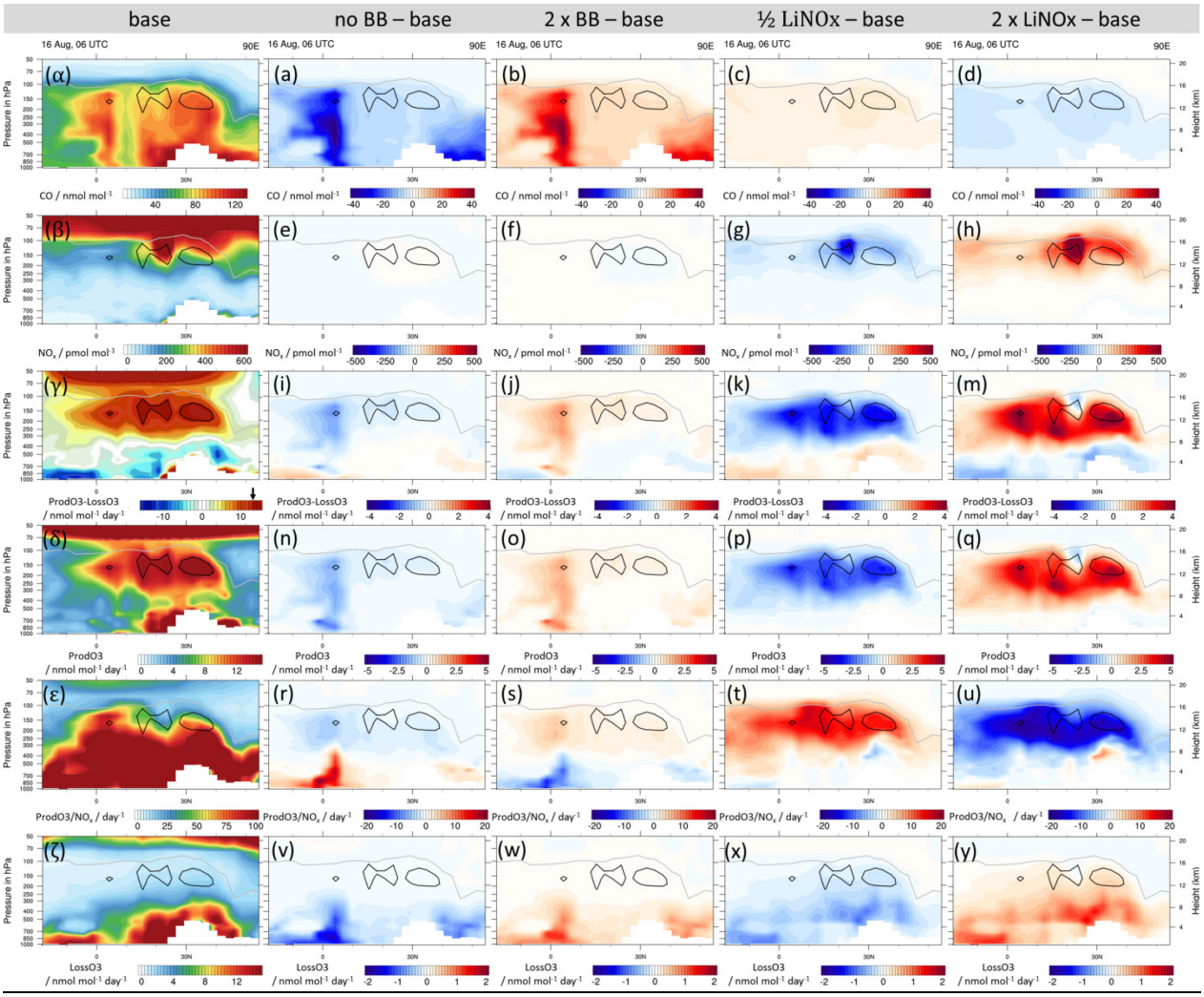

Fig. C6: Selection of parameters related to photochemical O_3 production in a meridional curtain through the Tibetan part of the ASMA in a snapshot taken mid August 2012. The left column shows the results of the EMAC QCTM simulation that has been introduced in appendix A of the discussion paper. The other columns show the difference of that reference to sensitivity simulations, which feature identical dynamics but differ in biomass burning (BB) and lightning NO_x (LiNOx) emissions. The black lines represent the 13 nmol mol^{-1} day^{-1} isocontour of net O_3 production (taken from panel γ), grey is the tropopause.

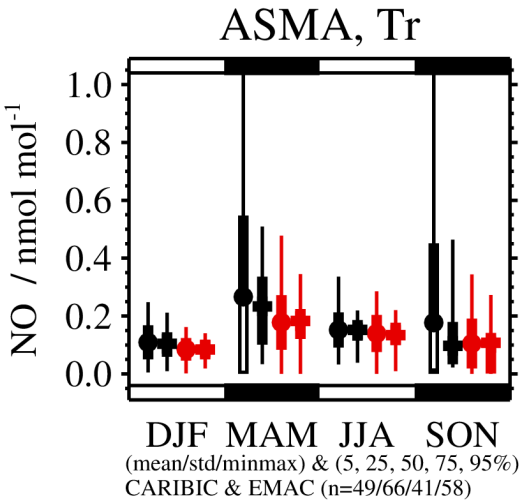

Fig. C7: Comparison of CARIBIC (black) NO measurements in the ASMA region (15-35°N, 30-100°E) with corresponding results of the EMAC RC1SD-base-10a simulation (red). Considered are data from between 300 hPa and the tropopause, from the period May 2005 to April 2014. The simulation was sampled along the CARIBIC flight tracks with a resolution of 12 min, and CARIBIC observations were subsequently interpolated (interval mean) to a resolution of 12 min. Number n below the plot shows the number of the remaining data pairs (after interpolation and filtering) available for the respective seasons. Dots represent mean values, rectangles the median. Whiskers indicate standard deviation, min & max values, and the percentiles given below the plot.

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