Atmos. Chem. Phys. Discuss., 15, C12816–C12841, 2016
www.atmos-chem-phys-discuss.net/15/C12816/2016/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modeling lightning-NO_x chemistry at sub-grid scale in a global chemical transport model" *by* A. Gressent et al.

A. Gressent et al.

alicia.gressent@aero.obs-mip.fr

Received and published: 29 February 2016

The authors thank the referees for their comments on the manuscript. Their suggestions of correction and their questions on this work are very interesting and definitely help to improve this paper.

Referee #1:

Specific comments:

*P34100 L8-11: Is there a reason that the methods used to determine horizontal dispersion could not be used to find vertical dispersion as well? A greater understanding of vertical dispersion could be used to augment the profiles used to force the vertical distribution of LNOx given by Ott et al. If this is beyond the scope of this study as the

authors indicate, that is fine, and a sentence or two explaining why would suffice. Also, is the claim that vertical diffusion is less efficient than horizontal diffusion really true for the strong convective storms that create lightning?

We thank the referee#1 for his useful comment on the atmospheric diffusion issue. We agree that lightning triggering and NOx emissions from electrical discharges occur in the convective part of the cloud where the vertical diffusion (Dv) is stronger than the horizontal diffusion (Dh). Therefore, the vertical diffusion coefficient is a determining parameter for the LNOx distribution. However, this vertical distribution of LNOx is a priori estimated from Ott et al., 2010, as a reverse "C-shape" profile, prescribed in GEOS-Chem, and totally independent of our parameterization. It could be very interesting to estimate Dv in the cloud to improve and support the Ott et al., 2010 given LNOx vertical profile using similar methods than the one used in the present study. This is indeed beyond the scope of our study, as it would require taking over Ott et al., 2010 work. Moreover, the plume parameterization for LNOx chemistry is applied a posteriori after lightning NOx are vertically prescribed by Ott et al., 2010 and emitted in convective clouds. Our parameterization starts just after transport driven by the convective parameterization and will then mostly concerns convective outflow where the NOx are detrained in the troposphere. In this region of detrainment, the horizontal diffusion seems to be more efficient than the vertical one as mentioned by Cariolle et al., 2009. Also, future studies on the atmospheric diffusion such as in-situ measurements and meso-scale modeling calculations should be useful to better quantify Dv and Dh.

According to this explanation we have clarified this point in the text, section 3.1.1 as follows: "Note that the mean dispersion properties of the atmosphere were associated with the horizontal diffusion only. The lightning NOx emissions occur in the convective part of clouds where the vertical diffusion is strong. Therefore, the vertical diffusion coefficient is a determining parameter for the LNOx distribution in the cloud. As mentioned in section 2.1, the vertical distribution of the LNOx is a priori calculated from Ott et al. (2010) as a reverse C-shaped profile. The LNOx plume parameterization

is applied a posteriori after that lightning NOx are vertically prescribed and concerns convective outflow where the NOx are detrained in the troposphere. In this region of detrainment, the horizontal dispersion may be more efficient than the vertical one as discussed in Cariolle et al., 2009."

*P34109 L8: I wonder about choosing the 8-11 km range. A brief explanation as to why this range was chosen would be appreciated. Is it appropriate to use the same vertical range for tropical and midlatitude storms, given the differences in their convective depth? How will this affect the parameterization of lightning outside of this range (or, how sensitive are the results to altitude)?

Referee comment is legitimate. In a real atmosphere the detrainment region is commonly observed at higher altitude (up to 13-14km altitude, Folkins and Martin 2004, doi: http://dx.doi.org/10.1175/JAS3407.1) in the tropics than in the mid-latitudes (8-9 km altitude, DeCaria et al. 2000) In this study, the GEOS-Chem model uses GEOS-5 meteorological fields. The figure 1 illustrates the vertical distribution of the LNOx calculated by GEOS-Chem with GEOS-5 for the tropics and the mid-latitudes. This figure shows that the detrainment altitude is similar in both regions, i.e. around 8-11 km. GEOS-5 seems to underestimate the detrainment altitude in the tropics as shown in a previous work (Folkins et al., 2006, doi:10.1029/2006JD007325). Therefore, we decided to show vertical levels consistent with the GEOS-5 detrainment altitude level.

Outside this altitude range, our parameterization should mainly have influence between 6 and 12 km for which the calculated LNOx flux is still significant both in the tropics and in the mid-latitudes as showed by the following figure. However, the impact should be less important than between 8-10 km where the LNOx flux is the strongest.

We clarified this point in the manuscript, section 3.2.2 as follows: “The altitude range refers to the detrainment region estimated by GEOS-Chem using the GEOS-5 met fields (section 2.1) both in the tropics and in the mid-latitudes. Note that this range could vary depending on the met fields and the convection parameterization. In ad-

dition, the LNOx plume parameterization might have an impact outside of this altitude range mainly between 6 km and 12 km but in a lesser extent.”

*P34108 L16-: The authors indicate that 3-D turbulence is solved in their parameterization, and give a range of Dh values. More discussion of this would be interesting. How does Dh vary globally? Are there any trends or features in the Dh fields that are of interest? I, and I imagine other readers as well, would be interested in more details on the variability.

The 3-D turbulence is actually not solved online in the GEOS-Chem CTM because of the fine scale characterizing this process but prescribed by the met fields (GEOS-5). Therefore, the global variability of Dh is not calculated in the model and has to be deduced a priori. For our parameterization, in order to determine Dh in the outflow region (i.e. at small scale), we have used both meso-scale modeling and in-situ measurements in the cloud anvil. The three-estimated Dh values [0.1; 15; 100] m2.s-1 are very dispersed. There is a real gap to fill about the knowledge in this parameter in the upper troposphere and especially in the outflow region. In the future, further meso-scale modeling studies and in-situ measurements in the scientific community should help to better define Dh variability.

We clarified this point in the revised version of the paper, section 3.2.1 as follows: “It is important to note that the 3-D turbulence is not solved online in the GEOS-Chem model because of the fine scale characterizing this process but prescribed by the GEOS-5 met fields. Therefore, the global variability of Dh is not calculated by the CTM and it is beyond the scope of this study. ”.

*P34110 L25-: The ratio RLNOx is set to be consistent with GEOS-Chem. It’s good to be consistent, but I wonder if more can be said regarding this issue since this is a problem that is poorly understood. Is there anything in the parameters used in the new plume-in-grid parameterization that can shed some insight into what drives this difference between midlatitudes and tropics? This is related to my previous comment

regarding variability of Dh. More discussion on the spatial variability of the various model parameters may be enlightening.

As the referee pointed out, there are large differences in the LNOx production between the tropics and the mid-latitudes. The rate of flashes is higher in the tropics (Christian et al., 2003) but the amount of NOx molecules emitted per flash might be higher in the mid-latitudes. The amount of NOx molecules produced per flash may depend on different lightning parameters such as the number of flashes, but also the flash length, the stroke peak current and the stroke release height (Huntrieser, 2008, http://www.atmos-chem-phys.net/8/921/2008/acp-8-921-2008.pdf).

Our study aims to improve and represent the chemistry related to lightning NOx emissions in CTMs at global scale. The approach applied here is the implementation of a parameterization, which includes uncertainties regarding parameters estimate. The main parameters defined in this study could help us to get a more realistic idea of the NOx then O3 distribution at global scale. Also, the variability of these parameters may depend on latitude, as it is mainly the case for the plume lifetime. The horizontal diffusion coefficient doesn't appear directly in the plume parameterization in the CTM. It is estimated beforehand and used in the simple plume dispersion simulations. Therefore, we don't use and show the horizontal diffusion at global scale and the related variability.

The suggestion of the referee to use our results for understanding the tropics and the mid-latitudes LNOx emissions differences is interesting. However this is not easily feasible as our parameterization is linked with the convective and LNOx emission parameterizations used in the CTM, for which dependence on the tropics and mid-latitudes differences in terms of convective intensity and LNOx emissions are already considered (GEOS-Chem model, http://wiki.seas.harvard.edu/geos-chem/index.php/Lightning_NOx_emissions). Using similar LNOx emissions in the CTM (no a priori latitude dependence, RLNOx=1) with our sub-grid scales parameterizations, validated with in situ LNOx measurements, should indeed better help understand the RLNOx ratio, as suggested by referee#1. However, it would certainly require

more LNOx observations in the tropics, poorly documented, as already stated in the manuscript.

*Section 4.2: The comparisons between different model simulations (BC, P1, P2 experiments) do not include a direct comparison between the full model with and without the plume parameterization. Section 4.2.2 effectively does this by using the difference between P1 and BC experiments, but I found this to be unnecessarily confusing. A separately named direct comparison would be clearer to the reader.

As suggested, we have changed the (P1-BC) difference by a direct comparison name (P3) in order to make the comparison clearer to the reader.

*P34115 L5-6 "an approximate detrainment level (9 km altitude) where the LNOx are most concentrated": This sentence is a bit unclear. Is this the level where most LNOx is produced, or the level where the detrainment of LNOx from a cloud is the largest?

We thank the referee#1 for this good remark. Regarding the results in the entire vertical, the 9 km level corresponds to the altitude where the detrainment level of LNOx is the largest.

We have clarified this point in the text in section 4.2.1 as follows: "These results are shown for an approximate detrainment level (9km altitude) where the detrainment of LNOx is the largest."

*Section 4.2.1: The differences in plume lifetime for different seasons and locations are given without context. What drives these differences?

The plume lifetime (τ) depends directly on, first, the NOx content (rl) above which the LNOx plume exists. This critical value changes mainly with the latitude since its calculation is initialized by the background concentrations of species, which are different in the tropics and in the mid-latitudes regions (see Table 1). Then, τ varies depending on the amount of LNOx emitted by lightning (NOi, in the manuscript), which is higher in the tropics than in the mid-latitudes as mentioned previously. We do not show sea-

sonal variations of τ (because they are negligible). However, day and night conditions strongly influence the rl calculation. Therefore, we calculate different values of τ for daytime and nighttime (Table 2 in the manuscript). The seasonal variations are presented only for NOy and O3 distributions at global scale.

As suggested by the referee we clarified this point in the manuscript, section 4.2.1 as follows: "The plume lifetime depends on the latitude because of the different background chemical concentrations and the different amount of NOx emitted from lightning in the tropics and in the mid-latitudes. In addition, as mentioned before, we consider the influence of day and night conditions on the plume lifetime estimate."

*P34115 L27: Why are some emissions “less important”? Does this just mean there are fewer emissions, or is something making them less important somehow?

We agree with the referee that the expression “less important” is unclear to qualify the lightning NOx emissions. We mean fewer emissions and we have changed the sentence in the text, section 4.2.1, as follows: “So, the LNOx tracer is characterized by a shorter lifetime as a plume over North America than over Central Africa and around the Sahel while the model simulated fewer emissions over these regions especially in summer.”

*P34116 L5: what do you mean by “tracer is mainly reproduced”?

We agree with the referee that this sentence is unclear and we have removed the term “mainly”. Indeed, lightning NOx emissions are distributed from the surface to the cloud top height by the CTM according to the reverse “C-Shape” profile (Ott et al., 2010). Results show that the tracer mixing ratio is reproduced at the altitude where lightning emissions are calculated and detrained, mainly in the upper troposphere. This sentence aims to point out the consistency between lightning NOx emissions and the tracer distributions.

*P34121 L1 “Significant values of β”: How large does β have to be to be significant?

Or does this just mean non-zero?

We thank the referee#1 for this good comment. Our estimates of the β1 (10-4) and β2 (10-3) fractions are smaller than those from Cariolle et al., 2009 (with β1=0.06 and β2=0.2) in which the effects of the β fractions are non-negligible on the NOy distribution. Therefore, we assumed that if the β fractions would be higher than our estimates, their impact should be significant regarding Cariolle et al., 2009 results. Further modeling studies are needed (both box model and CTM) in order to determined what could be the significant values for the β fractions.

*Figure 9: The hatched areas are a bit hard to read since they overlap with multiple lines. The authors may want to consider revising this figure to make it clearer.

We agree with the referee#1 comment. The figure 9 has been modified to make it clearer.

*P34101 L5 “than the surrounding” should be “as the surrounding”

This has been changed in the revised version of the manuscript.

*P34108 L22 “cover all of horizontal” should be either “cover all horizontal” or “cover all of the horizontal”

This has been changed in the revised version of the manuscript.

*P34110 L16 L18 “the northern Colorado” and “the Ontario”. The “the” should be removed.

This has been changed in the revised version of the manuscript.

*P34111 L 24-25: This sentence is confusing and should probably be re-worded.

The sentence was re-worded as suggested by the referee#1 as follows: “Our Keff estimates are smaller than those calculated by Cariolle et al. (2009) for the plume chemistry related to aircraft exhausts. In this previous work, Keff varies from 1.0 to

4.2.10-18 molecules-1 s-1 cm-3 with a mean value close to 3.10-18 molecules-1 s-1 cm-3 depending on the NOx loading. The very low value for Keff point out that the plume parameterization implies a delay of the production of ozone at the large scale rather than its destruction within the plume."

*P34121 L8-11: The sentence beginning "That could be explained" is also confusing and needs to be re-worded.

The sentence was re-worded as suggested by the referee#1 as follows: "Indeed, our $\beta 1$ and $\beta 2$ estimates are smaller than those calculated by Cariolle et al., 2009 ($\beta 1$ = 0.06 and $\beta 2$ = 0.2) which showed a significant impact of this mechanism in the case of aircraft NOx emissions."

Referee #2:

*Lightning emissions are of NO, not NOx. It would be helpful if the paper discussed this from the beginning and not waiting until Section 4 to clarify this.

We agree with the referee#2 comment and we have clarified this point at the beginning of the Introduction as follows: "Lightning emissions are one of the most important sources of nitrogen oxides (NOx $\equiv$ NO + NO2) in the upper troposphere (WMO, 1999; Hudman et al., 2007). Lightning primarily produce NO and may also induce a negligible quantity of NO2 with a ratio NO2/NOx of 0.5 to 0.1 (Franzblau, 25 1991; Stark et al., 1996)."

*p. 34101: the Ox family is O + O3 + NO2 (not O2!)

This has been changed in the revised version of the manuscript.

*The English needs to be corrected throughout the paper - there are numerous mismatches of plural-singular, and extraneous or missing articles (the, a). Also, for example, on p. 34106, 'allows to form' should be 'allowing formation of'.

We thank the referee#2 for his/her comment. We have corrected the English through-

out the paper.

Referee #3:

General comments:

1. The plume parameterization uses both mixing ratios (molecules/molecules; all the variables that start with "r") and concentrations (molecules/cm3) in its equations, so it is important that the text correctly refer to each to avoid confusion. However, the original C09 equations have not had their units correctly translated to this paper. For example, Equation (2) labels rp as a concentration (molecules cm-3), but it needs to be a mixing ratio in order for its integration with air density to yield molecules. The subsequent continuity equations, especially the d(rO3)/dt equation, are also dependent on a careful distinction of concentration versus mixing ratio in its components. The authors will either need to maintain the original units from C09 in all their equations (a mix of both mixing ratios and concentrations), or reformulate them such that they are all concentrations as they are currently described (e.g., removing _ where necessary, changing rO3 to [O3], etc.). I also recommend removing the multiplication crosses in the equations, and think it would be easier for the reader if standard square brackets were used for the concentrations in Equations (5)-(7), e.g., d([O]+[O3])/dt = k2 [NO2] - k3[NO][O3] ...

We agree with the suggestion of the referee#3. The related changes have been made in the text for describing the equations with the appropriate terms according to Cariolle et al., 2009.

2. It is not clear to me what model is being used to estimate τ and Keff, which requires resolving chemistry and transport on spatiotemporal scales finer than the plume itself. I think the model section needs an additional part that describes the "simple plume dispersion" model referenced in Sections 3.2.3 and 3.2.4., in particular how the chemistry was included that was used to determine Keff .

We agree with the referee comment. We have added the section 2.4 to the manuscript in order to present the simple plume dispersion model for determining τ and Keff as follows:

"To model the dispersion of lightning NOx emissions we use a simple dispersion model similar to the plume model used for aircraft NOx emissions, except that the plume is supposed to be oriented along a vertical axis. The plume is represented as a cylinder that encompass horizontal diffusion with a constant coefficient Dh (section 3.2.1). This simple model is composed of 30 horizontal circles with spacing increasing progressively from the center axis. The discretization of the diffusion equation is mass conservative.

The chemistry scheme and associated reaction rate constants is adapted from the large-scale chemical model MOCAGE (Teyssedre2007). It includes the main reactions involved in the NOx-HOx system. Simple plume simulations were performed in order to estimate the physical and chemical characteristics of plumes related to lightning NOx emissions. "

3. The formulation of Keff in C09 assumes that only NOx is elevated in the plumes relative to the diluted background mean (whereas other species are assumed to match the background at the initialization of the plume). This is a decent assumption for aircraft and ships, but less so for lightning. Lightning NOx is released during active deep convection, by which sub-grid-scale processes rapidly mix air from non-local locations, yielding complex mixtures of ozone, VOC, HOx and H2O observed in convective outflows that would be atypical relative to the background grid cell. For example, the recent DC3 campaign saw complex mixtures of surface and stratospheric air masses alongside lightning NOx plumes (doi:10.1175/BAMS-D-13 00290.1). I think that the authors should briefly acknowledge these uncertainties and how they might affect their conclusions.

The comment about the Keff calculation from the referre#3 is very interesting. We

agree that in the case of lightning emissions other species like VOCs, HOx and H2O may be uplifted from the surface in the convective region. Nevertheless, Sauvage et al., 2007 showed that, in the upper troposphere, NOx rather than VOCs mainly influence O3 mixing ratio. In other words, the OPE in the UT is controlled by NOx. We assumed that our plume parameterization is a first estimate of the LNOx chemistry focused on NOx.

As suggested by the referee, we have clarified this point in the manuscript in section 3.2.4 as follows: "Note that in the case of lightning emissions other species like VOCs, HOx and H2O may be uplifted in the convective region that could bring uncertainties in our approach. However, we assumed that the OPE is mainly controlled by NOx in the upper troposphere as previously showed by Sauvage et al. (2007b). Therefore, Keff calculation is here mainly dependent on NOx content. Future studies should try to investigate this issue for lightning emissions mixed with strong surface emissions in order to sharpen our parameterization"

4. I would expect lightning plumes to be highly efficient PAN producers, since the convection in which lightning occurs would also loft short-lived peroxyacetyl radicals from the surface to react with the elevated NOx, and the temperatures in the cold free and upper troposphere will guarantee that PAN does not thermally decompose and it will outlast the plume. However, because the plume formulation does not allow NOx to be converted into PAN in the plume, it is released as NOx away from the regions of elevated peroxyacetyl radicals, and therefore global PAN decreases, as the authors correctly explain in Section 4. However, unless the ratio of PAN production to HNO3 production is relatively suppressed in the high-NOx plumes (which I would not expect), then this is likely an error in the PAN budget introduced by the plume parameterization. I think the authors should comment based on their DSMACC results whether relative PAN production is stable, enhanced or suppressed in the high-NOx conditions. Unless it is suppressed, then I think that a conclusion of this paper should be a recommendation that future studies with a similar lightning plume parameterization include an

additional β term that characterizes the conversion of NOx to PAN, and associated d([PAN]/dt) and d([CH3C(O)OO])/dt continuity equations alongside those for HNO3, O3, and NOx.

This is an excellent comment from the referee#3. As showed in the manuscript, our LNOx plume parameterization implies a decrease in PAN mixing ratio at global scale, which is mainly explained by the storage of NOx in the plume form along the transport.

The figures 2 and 3 show d[PAN]/dt and d[HNO3]/dt implied by high NO concentration (0.7ppb, 3.42ppb and 10ppbb) as results from DSMACC box model simulations (example for the mid-latitudes conditions at daytime). These figures show that high NO condition implies a significant increase in PAN, which is higher by a factor 105 than the HNO3 production.

Therefore, we agree with the referee#3 about the potential gap for PAN chemistry in our LNOx plume parameterization. It could be of great interest to add the PAN and CH3C(O)OO continuity equations and a new term to describe the NOx conversion to PAN within the plume.

A discussion about the PAN chemistry was added in the manuscript, in section 4.2.2, as follows: "Note that the production of PAN is limited by the supply of NOx or non-methane volatile organic compounds (NMVOCs). Above continental lightning sources regions, NMVOCs are uplifted by deep convection but with lower NOx due to the activation of the plume parameterization. That implies a less efficient PAN production in these regions. Downwind of lightning sources regions (oceanic regions), NOx increases because of the LNOx transport in the plume form but there is less NMVOCs available to produce PAN. Therefore, both in regions of LNOx emissions and downwind the PAN production is limited leading to overall lower PAN mixing ratios at large scale in P1 experiment. However, this may be nuanced by considering the PAN chemistry in future studies using similar LNOx plume parameterization by introducing the PAN and CH3C(O)OO continuity equations and a new term to consider the fraction of NOx

converted to PAN within the plume. This should allow the PAN production during the plume transport, which is inhibited in the current version."

Specific comments:

*P34093, L7-13: Only some lightning NOx is detrained in the cloud anvil, much is detrained at lower altitudes during the convective updrafts and downdrafts as seen in the Ott et al. (2010) profiles. I would rephrase this sentence to be "Most NOx produced by lightning is detrained into the free and upper troposphere, where ozone production efficiencies (OPE) per unit NOx emitted are 4 to 20 times higher than at the surface (refs), and therefore lightning exerts a disproportionately stronger effect on photochemistry than surface emissions (refs)."

We agree with the referee suggestion. This has been changed in the revised version of the manuscript.

*P34093, L21-23: Recommend changing the start of the sentence to "Therefore, lightning NOx production must be parameterized for inclusion..."

This has been changed in the revised version of the manuscript.

*P34093, L26-27: LIS and OTD are currently the only options, and OTD was not on the TRMM satellite.

We agree with the referee comment and we have change the sentence as follows: "Flashes simulated by CTMs are commonly constrained by satellite observations (Sauvage et al., 2007b; Murray et al., 2012) from the space-borne Lightning Imaging Sensor (LIS) on TRMM and the Optical Transient Detector (OTD) (Christian et al., 2003; Tost et al., 2007)."

*P34094, L1: Many models now use newer vertical distribution profiles from Ott et al. (2010), including GEOS-Chem, which are "reverse-C"-shaped, so please update the text here and elsewhere.

This has been changed in the revised version of the manuscript.

*P34094, L6: I would recast as “Despite the necessity of including lightning NOx emissions in global models, ...”

This has been changed in the revised version of the manuscript.

*P34094, L15: Please change “realistic” to “more realistic”, as the plume scheme is still a parameterization (and similarly in P34118, L18; P34121, L5; P34123, L27).

This has been changed in the revised version of the manuscript.

*P34096, L13-14: Heterogeneous reactions occur on all aerosol types in GEOSChem, not just sulfate and mineral dust.

This has been changed in the revised version of the manuscript.

*P34096, L18: Suggest replacing “specially estimate for” with “overwrite those for the”

This has been changed in the revised version of the manuscript.

*P34096, L20: “Fossil fuel” should be “Biofuel”

This has been changed in the revised version of the manuscript.

*P34096, L22: Lobert et al. is not a GFED reference (see doi:10.5194/acp-10-11707-2010)

We have changed the reference related to the GFED inventory in the text.

*P34096, L25-P34097, L2 - I recommend rephrasing the GEOS-Chem description to follow the order of the steps taken in the model to calculate NOx from lightning: (1) flash rates are calculated in active deep convection using the Price and Rind scheme, (2) flash rates are adjusted with local scaling factors to match the satellite climatology, (3) total column emissions are determined using NOx yields that differ in tropics and northern extratropics, and (4) total column is distributed vertically using the Ott profiles. I would also mention that the base lightning NOx scheme is described in detail

by Murray et al. (2012). Have the authors made any additional modifications to the standard GEOS-Chem implementation for their base case (BC) simulation, or does that simulation use GEOS-Chem as is publicly released?

We thank the referee#3 for his/her suggestion. In this present study, there is no additional change to the standard GEOS-Chem implementation related to the BC experiment. This has been changed in the revised version of the manuscript, in the section 2.1, as follows:

“In order to calculate the NOx from lightning, flash rates are first calculated in active deep convection using the Price and Rind Scheme based on the cloud-top-height (Price and Rind, 1992, 1994), then flash rates are adjusted with local scaling factors to match the satellite climatology (Sauvage et al., 2007b; Murray et al., 2012), and the total column emissions are determined using NOx yields that differ in tropics and northern extratropics. Finally, the total column is distributed vertically using the reverse C-shaped profile from Ott et al. (2010). Note that the base lightning NOx scheme is described in detail by Murray et al. (2012).”

*P34098, L22-23: Please give the mass of the LNOx tracer used, and specify whether or not it varies in space and time (as the mean mass of NOx does due to changes in NO/NO2 ratios).

The initial LNOx tracer mass corresponds to the NOx mass at the time of the beginning of the simulation within the GEOS-Chem model. However, the LNOx tracer is considered as a passive tracer within the model following the monotonic exponential decade applied by the plume parameterization. We have clarified this point in the manuscript as follows:

“Following Cariolle et al. (2009), a passive tracer (from the perspective of the usual model chemistry) is added to the CTM to represent NOx emitted by lightning. The LNOx tracer initial mass corresponds to the NOx mass at the start time of the simulation. Rather than increasing the concentration of NOx within the CTM, lightning

NOx emissions now increase the concentration of this new passive tracer, which is transported in the standard way by advection and turbulence. Plume chemistry is considered to be significant when the mixing ratio of the lightning NOx tracer is higher than a critical NOx content, hereafter denoted rl . Above this value the lightning NOx tracer is transferred to the normal NOx tracer at a rate described by a plume lifetime (τ), which is an exponential decay constant. This corresponds to an exchange time scale between the lightning NOx plume and the background NOx."

*P34099, L13: I should be the injection rate of "LNOx", and the units used here should be the same in P34113, L21.

This has been changed in the revised version of the manuscript.

*P34100, L17-20 - Equation (4) includes _NOx and EINOx. However, later in the text it is stated that these values are 1, which is non-physical based on what they are supposed to represent. However, the "fuel" tracer in this case is comprised only of NOx, so I believe the authors should just remove _NOx and EINOx everywhere from this work as superfluous (as long as the molecular mass of the LNOx tracer is specified). If they prefer to leave them in, please state what the g (N, NO, NO2, NOx?) and kg (air, LNOx?) are referring to in EINOx.

We agree with the suggestion of the referee#3. The EINOX variable has been removed from the equations.

*P34101, L4-5: Ozone is still an order of magnitude greater.

We thank the referee for his/her comment and the sentence has been change in the text as follows:

"In the case of large NOx injection by lightning, the NOx content (40 ppt in unpolluted atmosphere) becomes close (a few ppb, according to in-situ measurements, Dye et al. (2000); Huntrieser et al. (2002)) to the surrounding ozone (60$\pm$24 ppb) (Jaéglé et al., 1998)."

*P34101, L13-14: Recommend replacing "The sums of the concentrations as detailed by the Eqs. (5)-(7)" with "The rate of change of each chemical family are given by Eqs. (5)-(7)".

This has been changed in the revised version of the manuscript.

*P34102, L24-P34103, L2: It is given here that Keff is a function of the plume concentrations of NOx and O3, but the C09 derivation uses only in-plume NOx concentrations and background O3 concentrations in its definition of Keff. Please clarify what is used here and correct the integrals as necessary.

In the Keff equation, rNOxP and rO3P correspond to the mixing ratios of NOx and O3 respectively in the plume and the overlined rO3 term is the background ozone mixing ratio averaged in the model grid as described in Cariolle et al., 2009. We have clarified this point in the text.

*P34103, L17-P34104 L18: Please add "primarily" to the discussion of the day and night reactions, since both reactions occur both at day and night. Please clarify what is meant by "characterized by the coefficient β " (I assume molar fraction of NOx converted to HNO3?).

We have clarified this point in the text as follows: "In addition, we consider the conversion of NOx into HNO3 within the plume. This conversion takes place in two different ways depending on the day or night atmospheric conditions. During the day, NO2 reacts primarily with OH to give HNO3 directly and it is characterized by the coefficient β1. While at nighttime the conversion of NOx to HNO3 occurs mainly through N2O5 formation followed by a heterogeneous hydrolysis reaction, which corresponds to β2. In other words, the β coefficients are the molar fractions of NOx converted to HNO3 within the plume. These two fractions are unitless".

*P34104, L15: It is somewhat misleading to include PAN here. It is true that PAN is forming in the box model used to calculate β1 and β2, therefore reducing the magnitude

of those values by preventing some NOx from being converted to HNO3. However, the plume parameterization does not include a d[PAN]/dt equation, nor does d[NOx]/dt include any losses associated with PAN production in the plume, so this PAN remains as NOx as far as GEOS-Chem is concerned.

We agree with the referee#3 comment. PAN has been removed here.

*P34106, L23-24: Recommend changing to "related to highly elevated NOx concentrations relative to the background".

This has been changed in the revised version of the manuscript.

*Section 3.2.1: Dh was only estimated for outflow from deep convective anvils, but a lot of lightning NOx is released beneath the anvil. Please clarify if a constant Dh was used in all plumes, and didn't vary with altitude or latitude?

We agree with the referee, that LNOx may be released beneath the anvil. Three different values of Dh [0.1; 15; 100] m2.s-1 were estimated in our study. The first value Dh=0.1 m2.s-1 was obtained from in situ measurements outside the anvil but close to this region and could be related to the plume from beneath the anvil. The second value, Dh=15 m2.s-1 was obtained from measurements within the anvil region. Finally the last value, Dh=100 m2.s-1 corresponds to the horizontal diffusion in the anvil and was obtained from a convective cell simulation with Meso-NH.

From these estimates, we have a first guess of Dh in the anvil but also outside and close to the anvil from two different approaches. In this study, Dh is constant for each experiment performed with the GEOS-Chem model. Dh is used in the simple dispersion model (new section 2.4) in order to determine the plume lifetime and Keff but this parameter does not appear directly in our parameterization in the GEOS-Chem model. As discussed before in response to the referee#1, although it is very relevant, the global variability of Dh was not studied here because it is not solve at large scale by the GEOS-Chem model. We may expect that Dh varies mainly with altitude and also

latitude. Further modeling studies and in situ measurements should be performed in the future to better understand this parameter variability at large scale and improve the characterization of the LNOx plume dispersion.

We have clarified this point in the revised version of the manuscript in section 3.2.1, as follows: "In order to cover all horizontal diffusivity estimates discussed in this section the range of values 0.1, 15 and 100 m2.s$-$1 was used. The horizontal coefficient is constant for all lightning NOx plumes considered in the GEOS-Chem model. Hereafter, the results are detailed for the central value Dh = 15 m2.s$-$1."

*P34107, L5-7: "defined" would be better as "determined"? Please clarify what is meant by "mainly from previous in-situ measurement in thunderstorm anvil".

We agree with the referee comment. By the sentence "mainly from previous in-situ measurement in thunderstorm anvil", we would highlight that the Dh estimate from in-situ measurements may be the most accurate. The Dh estimate related to this method is more realistic (in agreement with Cariolle et al., 2009, Dh estimate) than the estimate from the modeling study. This has been removed in this paragraph but it is mentioned after in this section.

This has been change in the revised version of the manuscript in section 3.2.1, as follows: "The diffusion coefficient was determined by two different ways. A first estimate of the horizontal diffusion was performed by running the 3-D mesoscale Meso-NH model. Then, the Dh coefficient was calculated using in-situ measurements in thunderstorm anvil".

*P34107, L12: "performed" would be better than "made"

This has been changed in the revised version of the manuscript.

*P34107, L17: If e is not Euler's constant, please define.

The "e" is the exponential function here. It has been replaced by "exp" in the revised manuscript.

*P34109, L17-23: How are the tropics vs. midlatitudes defined when rl is applied in the model?

The critical content mixing ratio rl, above which the LNOx plume exists, is used in the plume dispersion model in order to estimate the plume lifetime (τ) and Keff. In these dispersion simulations, the tropics and the mid-latitudes conditions are distinguished using initial concentrations for chemical species relative to the two latitude regions (Table 1). From these runs, we got different values for τ (and Keff). Finally, in GEOS-Chen model, theτvalues are applied depending on the latitude region.

*P34110, L1: Please clarify what is meant by "an ensemble of spikes"

In our parameterization, each LNOx plume is associated with several electrical discharges at the convective cell scale. We have clarified this point in section 3.2.3, as follows: "Here we use a width of 500 m to refer to an ensemble of spikes at cloud scale (i.e. each plume is defined from several electrical discharges associated with a convective cell)."

*P34111, L2: Please replace "defined" with "estimated"

This has been changed in the revised version of the manuscript.

*P34111, L4: Is it appropriate to use separate rl for night and day, when some of the lifetimes are much longer than 12 hours? Why wouldn't we just use the smaller of the two values in both day and night?

There is some plume lifetimes longer than 12 hours but most are shorter (67 %). Also, there is a significant difference in the plume lifetime between day and night (Table 2). For example, for NOimax there is 3.6 hours and 13.6 hours difference in the mid-latitudes and tropics respectively. We cannot consider only the smallest value of rl because this could imply longer lifetime, which is less realistic than the use of the rl day and night distinguish values.

*P34111, L24-25: Please clarify the sentence so it is clear whether it is meant that Keff

is "very low" relative to the C09 values or the background K values?

This has been clarified in the revised version of the manuscript in section 3.2.4, as follows: "Our Keff estimates are smaller than those calculated by Cariolle et al. (2009) for the plume chemistry related to aircraft exhausts. In this previous work, Keff varies from 1.0 to 4.2.10$-$18 molecules-1.s-1.cm3 with a mean value close to 3.10-18 molecules-1.s-1.cm3 depending on the NOx loading."

*P34113, L23: I think "undiluted" should be "diluted" here

The use if the "undiluted" term is the good choice here. In fact, in our plume parameterization, the mixing ratio of the tracer (rLNOx) represents the NOx mixing ratio from lightning emissions. In other words, this is the NOx in the plume form not yet released to the model grid as a background concentration.

*P34113, L24: Lightning does produce some NO2 (as well as other species). I would change to "lightning produces negligible quantities of NO2 relative to NO and therefore E is effectively 0 in Eq. (15)."

This has been changed in the revised version of the manuscript.

*P34116, L5: "produced" (not reproduced). I would refer to the altitudes in Fig. 3c as "middle and upper troposphere" as the tropopause is $\sim$70 mb in the tropics.

This has been changed in the revised version of the manuscript as follows: "The lightning NOx tracer is produced at altitudes where lightning NOx are calculated and detrained (in the upper troposphere, between âĹij500 and 300 hPa) as shown in panels (c) in figure 3."

*P34117 L6: Should be "volatile"

This has been changed in the revised version of the manuscript.

*Sections 4.3.1-4.3.2: Here the word "variability" would be better replaced by "sensitivity", and all the various deltaO3 and deltaNOx values referred to as "ranges" or

“changes” associated with the uncertainty in the different explored parameters examined.

This has been changed in the revised version of the manuscript.

*P34121, L1: “significant” should probably be “large” here.

This has been changed in the revised version of the manuscript.

*Figure 1: This diagram needs a longer caption to describe what the lines, arrows, boxes, and colors represent. I would recommend removing the boxes from around the “[NOx] < rl” and “[NOx] > rl” to make it clear that those are a conditional statements (also, [NOx] should probably be [LNOx] there), and move the edge of the green “ERR” box to the other side of the conditional.

This has been changed in the revised version of the manuscript.

*Figures 3-7: I was originally confused because I interpreted these captions as that Dh and NOi were somehow applied in GEOS-Chem, not that the τ and Keff values were trained from the offline simple plume dispersion model using those Dh and NOi values. I would recommend rephrasing to make that distinction clearer.

This has been changed in the revised version of the manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 34091, 2015.

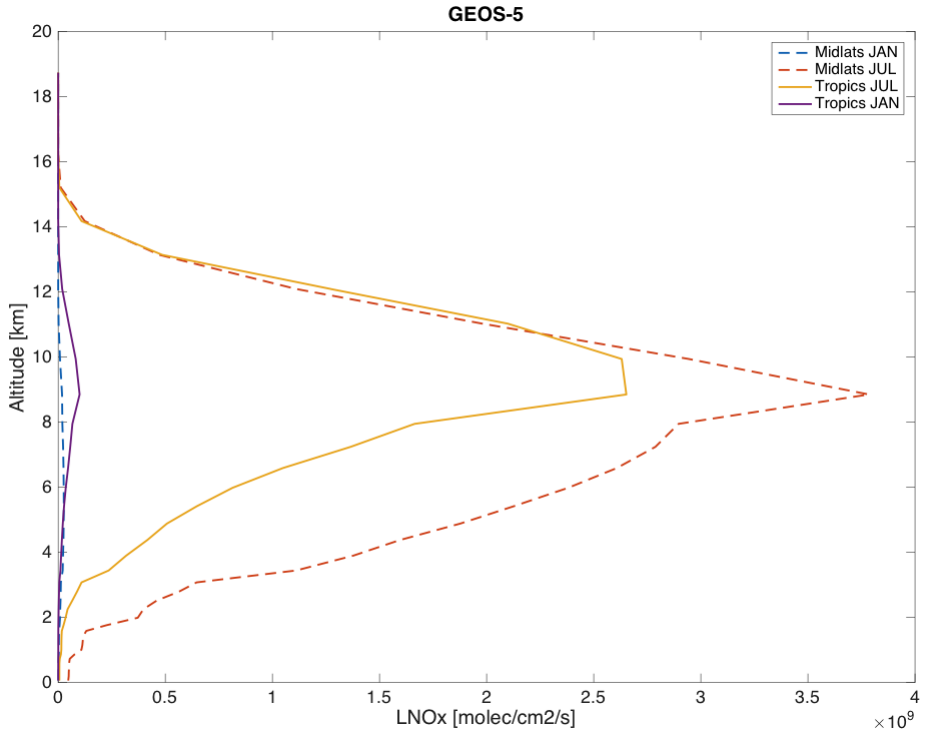

Fig. 1.

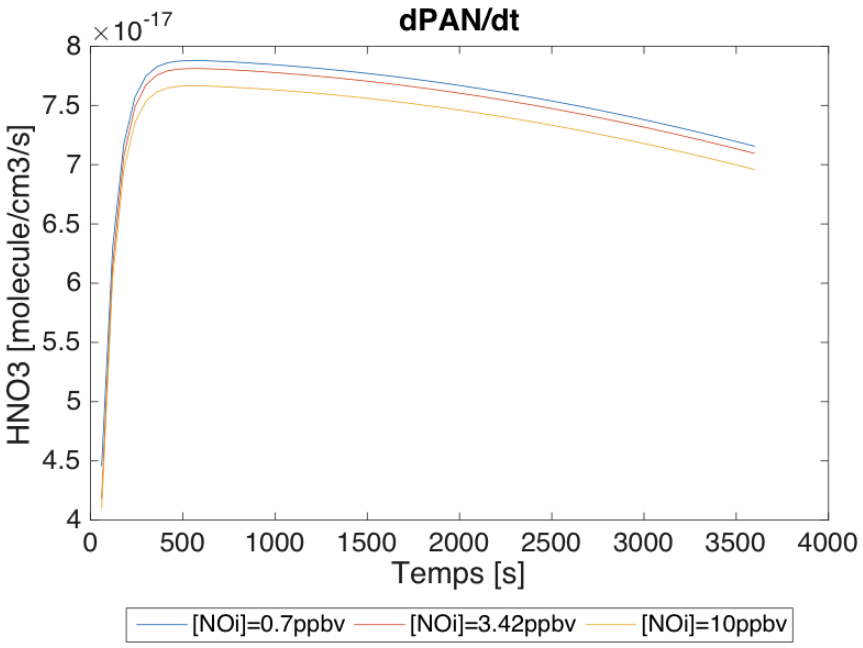
dPAN/dt
×10^-17
8
7.5
7
6.5
6
5.5
5
4.5
4
HNO3 [molecule/cm3/s]
0
500
1000
1500
2000
2500
3000
3500
4000
Temps [s]
[NOi]=0.7ppbv
[NOi]=3.42ppbv
[NOi]=10ppbv

Fig. 2.

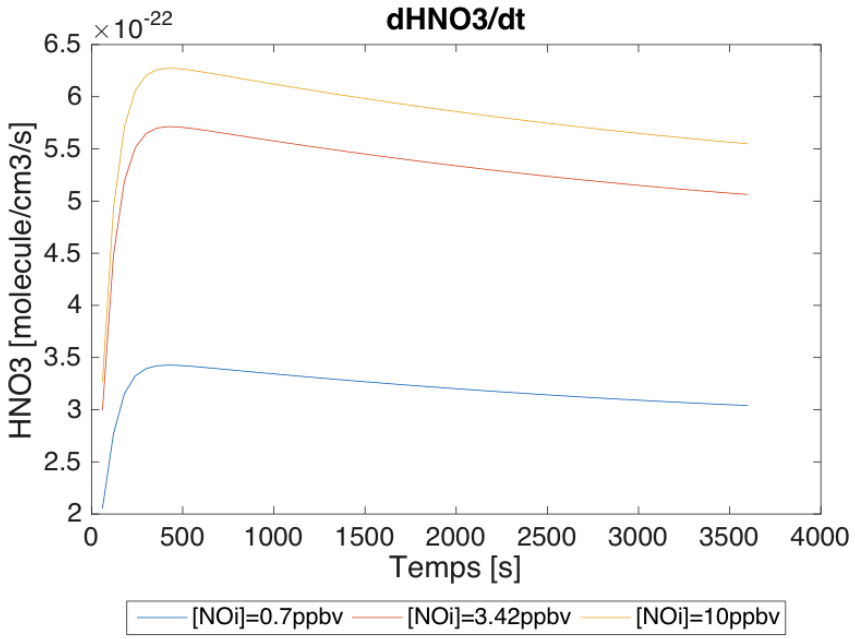
dHNO3/dt
×10^-22
6.5
6
5.5
5
4.5
4
3.5
3
2.5
2
HNO3 [molecule/cm3/s]
0
500
1000
1500
2000
2500
3000
3500
4000
Temps [s]
[NOi]=0.7ppbv
[NOi]=3.42ppbv
[NOi]=10ppbv

Fig. 3.