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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.

Anonymous Referee #3

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1 Overview

The authors adapted a parameterization of high-NO_x plume chemistry developed for aircraft emissions by Cariolle et al. (2009) (hereafter, C09) into the GEOS-Chem global chemical transport model (CTM). Here, the parameterization has been modified to account for non-linear chemistry in plumes of highly concentrated NO_x from lightning.

It is a very interesting study, an important step toward a truer representation and understanding of how lightning affects chemistry in the real world, and worthy of publication. However, first there needs to be several corrections to the equations before it could be published. I also believe that there should be more clarification as to how τ and K_{eff} values are determined, and I believe a greater acknowledgement of some of the

uncertainties and biases introduced by the assumptions about the simplified in-plume chemistry, particularly those that are unique to lightning emissions, as outlined below.

2 General Comments

1. The plume parameterization uses both mixing ratios (molecules/molecules; all the variables that start with "r") and concentrations (molecules/cm^3) 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 r_p as a concentration (molecules cm^{-3}), but it needs to be a mixing ratio in order for its integration with air density (ρ) to yield molecules. The subsequent continuity equations, especially the d(r_{O_3})/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 ρ where necessary, changing r_{O_3} to [O_3], 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]+[O_3])/dt = k_2 [NO_2] - k_3[NO][O_3] ...

2. It is not clear to me what model is being used to estimate τ and K_{eff}, 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 K_{eff}.

3. The formulation of K_{eff} in C09 assumes that only NO_x 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 NO_x 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, HO_x and H_2O 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 NO_x 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.

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 NO_x, 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 NO_x to be converted into PAN in the plume, it is released as NO_x 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 HNO_3 production is relatively suppressed in the high-NO_x 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-NO_x 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 NO_x to PAN, and associated d([PAN]/dt) and d([$CH_3C(O)OO$])/dt continuity equations alongside those for HNO_3, O_3, and NO_x.

3 Specific Comments

- P34093, L7-13: Only some lightning NO_x 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 NO_x produced by lightning is detrained into the free and upper troposphere, where ozone production efficiencies (OPE) per unit NO_x 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)."
- P34093, L21-23: Recommend changing the start of the sentence to "Therefore, lightning NO_x production must be parameterized for inclusion..."
- P34093, L26-27: LIS and OTD are currently the only options, and OTD was not on the TRMM satellite.
- 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.
- P34094, L6: I would recast as "Despite the necessity of including lightning NO_x emissions in global models, ..."
- 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).
- P34096, L13-14: Heterogenous reactions occur on all aerosol types in GEOS-Chem, not just sulfate and mineral dust.
- P34096, L18: Suggest replacing "specially estimate for" with "overwrite those for the"

- P34096, L20: “Fossil fuel” should be “Biofuel”
- P34096, L22: Lobert et al. is not a GFED reference (see doi:10.5194/acp-10-11707-2010)
- P34096, L25-P34097, L2 - I recommend rephrasing the GEOS-Chem description to follow the order of the steps taken in the model to calculate NO_x 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 NO_x 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 NO_x 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?
- P34098, L22-23: Please give the mass of the LNO_x tracer used, and specify whether or not it varies in space and time (as the mean mass of NO_x does due to changes in NO/NO_2 ratios).
- P34099, L13: I should be the injection rate of “LNO_x”, and the units used here should be the same in P34113, L21.
- P34100, L17-20 - Equation (4) includes α_{NO_x} and EI_{NO_x}. 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 NO_x, so I believe the authors should just remove α_{NO_x} and EI_{NO_x} everywhere from this work as superfluous (as long as the molecular mass of the LNO_x tracer is specified). If they prefer to leave them in, please state what the g (N, NO, NO_2, NO_x?) and kg (air, LNO_x?) are referring to in EI_{NO_x}.

- P34101, L4-5: Ozone is still an order of magnitude greater.
- 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)”.
- P34102, L24-P34103, L2: It is given here that K_{eff} is a function of the plume concentrations of NO_x and O_3, but the C09 derivation uses only in-plume NO_x concentrations and background O_3 concentrations in its definition of K_{eff}. Please clarify what is used here and correct the integrals as necessary.
- 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 NO_x converted to HNO_3?).
- P34104, L15: It is somewhat misleading to include PAN here. It is true that PAN is forming in the box model used to calculate β_1 and β_2, therefore reducing the magnitude of those values by preventing some NO_x from being converted to HNO_3. However, the plume parameterization does not include a d[PAN]/dt equation, nor does d[NO_x]/dt include any losses associated with PAN production in the plume, so this PAN remains as NO_x as far as GEOS-Chem is concerned.
- P34106, L23-24: Recommend changing to “related to highly elevated NO_x concentrations relative to the background”.
- Section 3.2.1: D_h was only estimated for outflow from deep convective anvils, but a lot of lightning NO_x is released beneath the anvil. Please clarify if a constant D_h was used in all plumes, and didn’t vary with altitude or latitude?
- P34107, L5-7: “defined” would be better as “determined”? Please clarify what is meant by “mainly from previous in-situ measurement in thunderstorm anvil”.

- P34107, L12: “performed” would be better than “made”
- P34107, L17: If e is not Euler’s constant, please define.
- P34109, L17-23: How are the tropics vs. midlatitudes defined when r_l is applied in the model?
- P34110, L1: Please clarify what is meant by “an ensemble of spikes”
- P34111, L2: Please replace “defined” with “estimated”
- P34111, L4: Is it appropriate to use separate r_l 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?
- P34111, L24-25: Please clarify the sentence so it is clear whether it is meant that K_{eff} is “very low” relative to the C09 values or the background K values?
- P34113, L23: I think “undiluted” should be “diluted” here
- P34113, L24: Lightning does produce some NO_2 (as well as other species). I would change to “lightning produces negligible quantities of NO_2 relative to NO and therefore E is effectively 0 in Eq. (15).”
- 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
- P34117 L6: Should be “volatile”
- Sections 4.3.1-4.3.2: Here the word “variability” would be better replaced by “sensitivity”, and all the various ΔO_3 and ΔNO_x values referred to as “ranges” or “changes” associated with the uncertainty in the different explored parameters examined.

- P34121, L1: “significant” should probably be “large” here
- 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 “$[NO_x] < r_l$” and “$[NO_x] > r_l$” to make it clear that those are a conditional statements (also, $[NO_x]$ should probably be $[LNO_x]$ there), and move the edge of the green “ERR” box to the other side of the conditional.
- Figures 3-7: I was originally confused because I interpreted these captions as that D_h and NO_i were somehow applied in GEOS-Chem, not that the τ and K_{eff} values were trained from the offline simple plume dispersion model using those D_h and NO_i values. I would recommend rephrasing to make that distinction clearer.

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