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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The global lightning-induced nitrogen oxides source" *by* U. Schumann and H. Huntrieser

Anonymous Referee #1

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General comments:

This manuscript represents the first comprehensive review work in the field of lightning-produced nitrogen oxides in over a decade and is perhaps a much needed piece of work. It is well structured and covers the relevant aspects of research into lightning-produced nitrogen oxides and thorough manner. The authors review the importance of NOx for tropospheric chemistry as well as its concentrations, sources and sinks. The phenomenon of lightning is then considered before delving into the formation of NOx by lightning, touching upon the formation of other trace gases by lightning as well. The importance of lightning-produced nitrogen oxides in tropospheric chemistry is discussed as is its impacts on ozone and its probable climate implications. The importance of accurately determining the source of NOx from lightning is discussed in the light of the

importance to balance the budget of other NOx sources, such as that from aviation, proceeding then to derive specific lightning-NOx accuracy requirements. The authors then discuss the various methods for constraining the source of NOx from lightning in the light of the most recently-available set of satellite observations. The results of a number of studies using global models aimed at either assessing the importance of lightning-produced NOx in tropospheric chemistry or at determining its source, or both, is discussed in detail. From these studies a best source estimate is determined and guidelines from improving this estimate and reducing its associated error in future studies are given. It is striking that in the intervening years since the last major review paper in the area, source estimates from model studies still gravitate in the 1-20 Tg yr-1 range, and despite the fact that most studies suggest a range of 3-7 Tg yr-1, there is still no definitive evidence that allows discarding the upper and lower ends of the estimate range. Very interestingly, the best source estimate resulting from this study, in particular from the global model fit method, i.e., (5ś3) Tg yr-1, is not vastly different from that of last review paper in the area, while the uncertainty range remains, in essence, almost the same; the change results chiefly from a revised, higher estimate of the number of NOx molecules produced per flash. This manuscript represents a significant effort to compile more than 3 decades' worth of research effort in the area of lightning-produced NOx and not only arrive at a best estimate on the source magnitude based on it, but also lay out strategies for improving these estimates in the future. The methods used to derive the conclusions are sound and while no breakthrough conclusion is arrived at (through no fault of the authors), the manuscript does a very good job of putting the current state of research in the area in good perspective. While the manuscript is well structured and the main points are laid out in a clear manner, the authors should be more careful with spelling in future work. Once the minor spelling and grammar corrections suggested below are introduced, I would recommend this manuscript for publication in ACP.

Specific comments:

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Part of the reason why global atmospheric chemistry-transport model have problems accurately reproducing observed trace-gas distributions, other than the obvious shortcoming regarding the representation of vertical transport and other parameterized phenomena, have to do with the fact that, after every timestep, the tracers produced are evenly distributed within each gridcell. Many of field observation campaigns carried out, even those targeting lightning-produced NOx usually involve sub-gridscale-range flights measuring point-sources of trace gases. This naturally leads to inaccuracies in the simulation of both the observed vertical and horizontal distribution of the tracers (despite prescriptions of the vertical distributions of tracers in the models). While the problem with the vertical distribution is mentioned, brief mention should be made of the horizontal distribution problem in the section where model estimates are compared with field campaign data.

Technical corrections:

Page 2626, line 22: Change "..summarises the conclusions" for "presents" or "gives" the conclusions.

Page 2627, line 2: change "..for ultraviolet radiation.." to "of ultraviolet radiation"

Page 2629, line 19: change "..its speciation.." for "their speciation"

Page 2630, line 12: "..from an order of 1-10.."

„ „ , line 15: "..an order of 0.05.."

Page 2632, line 22: "..satellites suffer the effects of cosmic radiation"

Page 2633, line 4: "..molecules per unit surface area.."

Page 2633, line 5: "..clouds prevents the formation of NO2 below them" (redundancy)

Page 2634, lines 1-4; The start of the paragraph would read better if you were to switch the order of the first two sentences.

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Page 2634, line 1: “..Earth’s atmosphere..”

Page 2636, line 10: “..to reach the ground..”

Page 2636, line 24: “..of the order of”

Page 2638, line19: “..2.8 and 1.2 strokes, respectively.”

Page 2642, line 8: Spell out NLDN.

Page 2643, line 8: “..the formation of NOx by lightning..”

Page 2650, line 4: “..operation’s area..” or “area of operation”

Page 2653, line 29: “..of the order of..”

Page 2655, line16: “..it contributes strongly..” Redundancy.

Page 2661, line 1: The assumption of a constant mixing ratio in the vertical for the source of NOx from lightning does not come only from the notion of complete vertical mixing, but also from a further series of assumptions, i.e., that IC flashes are ten times as energetic as CG flashes (Price and Rind, 1994), but also ten times more frequent (Turman, 1978, Kowalczyk and Bauer, 1982), and the fact NOx production by lightning was computed to exhibit a strong dependence on ambient air, being less for lower densities (Goldenbaum and Dickerson. 1993). The first two assumption can be regarded as cancelling each other out, and the third results in the even mixing ratio. Please mention this.

Page 2662, line 20: “..have often been..”

Page 2665, line 12: “..in convection are missing, meaning that this approach is hard..”

Page 2669, line 19: this sentence would read better like this: “Over the Gulf of Mexico, during the 1997-1998 El Niño event, a 100-150% increase in lightning days is found, compared to the 1996-1997 and 1998-1999 winters, apparently...”

Page 2672, line 3: “..aviation-induced..”

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Page 2672, line 7: “..supersonic aircraft’s NOx emissions..”

Page 2672, line 17; Which implications?

Page 2673, line 26: “..global budget..”

Page 2674, line 9: “..per discharge unit energy..”

Page 2674, line19: “..energy-specific..”

Page 2675, line 26 “..factor of 2..”

Page 2676, line 4: “..(about quadratically)”

Page 2676, line 5: “..energy-specific..”

Page 2676, line 8: delete “/”

Page 2676, line 14: “..LNOx production per flash..”

Page 2677, line 2: “Flashes may even have..” or “A flash may even..”

Page 2677, line 3: Please define “tortuosity”, however briefly.

Page 2676, line 8: “..approaches resulting from..”

Page 2680, line 6: “nearly all”, please be more precise.

Page 2681, line 21: “..peak flash currents..”

Page 2681, line 24: “..OTD-derived”

Page 2682, line 24: “relative to the inflow air..”

Page 2682, line 27: “..to the global scale..”

Page 2685, line 9: “..source must be correspondingly smaller..”

Page 2685, line 16: “..combination of measurements, directly by emissions (NOx, NOy) and indirectly by photochemistry of trace gases affected by LNOx..”

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Page 2687, line 7: “..emission databases of tracers other than LNOx..”

Page 2688, line 20-24: is this mini-summary of the sections to come really necessary at this point?

Page 2689, line 18: “..has increased..”

Page 2699 line 24: “..is larger the larger ε is..”

Page 2702, line 20: “global scale”

Page 2703, line 20: “..rain unit mass..”

Page 2704, line 20: “..seem to simulate the variability..”

Page 2705, line 3: “..and this is important because..”

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 2623, 2007.