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Interactive Comment

# Interactive comment on "Regional lightning $\mathrm{NO}_{\mathrm{x}}$ sources during the TROCCINOX experiment" by C. Mari et al.

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Received and published: 20 July 2006

### **General Comments**

The manuscript by Mari et al. describes the implementation of a lightning  $NO_x$  (LiNOx) source in the deep convection scheme of the Meso-NH model. This approach is different from what has been done in several global and regional models. In these other models, the vertical placement of lightning produced  $NO_x$  was prescribed based on profiles from cloud-scale model runs by [Pickering et al.(1998)]. The major advantage of the approach by Mari et al., is that the vertical transport of the lightning produced  $NO_x$  is consistent with the (parameterized) deep convection in their model. The profiles by [Pickering et al.(1998)], on the other hand, have the merit of being calculated by a cloud-scale model, which is expected to provide a



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better description of transport than a parameterization. (Note that recent results by [Barth et al.(2005)] indicate good agreement for the transport of inert tracers between different cloud-scale models, while different parameterizations are known to show rather large differences (e.g. [Mahowald et al.(1995), Lawrence and Rasch(2005)]). The [Pickering et al.(1998)] profiles are, however, averaged results from model runs for a few specific events at certain mid-latitude continental, tropical continental, and tropical marine locations (mid-latitude marine locations have not been covered), and it is not clear, how representative the profiles are of the various "regimes". Furthermore, in order to fit the depth of the modeled deep convection the prescribed profiles are stretched or squeezed in the vertical (see [Labrador et al.(2004)]), possibly leading to large systematic errors. Thus, it remains to be seen which of the two methods yields the more realistic results.

A potentially large source of uncertainty (which is also inherent in the profiles by [Pickering et al.(1998)]), is the placement of the lightning NO<sub>x</sub> source (i.e. prior to transport), in particular the ratio of NO molecules per intracloud (IC) flash to NO molecules per cloud to ground (CG) flash (P(IC)/P(CG)). In both studies (i.e. Mari et al. and Pickering et al.), P(IC)/P(CG) is 0.1, while more recent studies indicate that intracloud flashes are likely to be as effective in producing NO as cloud-to-ground flashes, as stated in the manuscript. In addition, substantial uncertainty still exists about the magnitude of P(IC) and P(CG). Since the magnitude and the vertical placement of lightning NO<sub>x</sub> source can both have important impacts on global chemistry (as recently demonstrated by [Labrador et al.(2004)]), these uncertainties are important to address. Model sensitivity studies in association with extensive observations can certainly help to carefully explore and reduce these uncertainties. Unfortunately, different combinations of P(IC), P(CG), and flash rates for IC and CG flashes, can still lead to similar results in comparisons between model data and observations. The simulations by Mari et al., show a good agreement between simulated and satellite observed brightness temperatures, which indicates that the geographical location of the deep convection within the model domain is well represented by the model. This is an important prerequisite for 6, S1963–S1973, 2006

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such studies.

As noted in the manuscript, the number of NO molecules per CG flash adopted from [Price et al.(1997)] is one of the highest ever published (e.g. Table 1 of [Labrador et al.(2004)] showing estimates ranging from less than 0.1 to  $30.10^{26}$  molecules per flash). This is only partially offset by the low ratio of NO molecules per IC flash to NO molecules per CG flash (P(IC)/P(CG)=0.1). Unfortunately, no observed flash counts appear to be available for the episode studied by Mari et al.. Since the LiNOx source equals the number of flashes times the number of NO molecules produced per flash, this could together with the in-flight observations presented in the manuscript help to constrain the number of NO molecules produced per flash in the area.

Mari et al. derive an estimate for the global LiNOx production of 39-55Tg(N)/year, which is significantly higher than the upper bound of the range suggested by a number of recent studies. The days used for the extrapolation were, however, chosen for their high convective activity, as noted in the manuscript. By itself, this choice would lead to an over-estimation of the extrapolated global lightning NO<sub>x</sub> production. I would also like to point out a more general aspect of the extrapolation. I assume the global LiNOx production is calculated as follows (see Section 5, page 5211):

"lower bound": 1.8·10<sup>7</sup> kg(N)/day  $\cdot$  31 days  $\cdot$  (100/18)  $\cdot$  (100/8)  $\approx$  39 Tg(N)

and "upper bound": 2.5  $\cdot 10^7$  kg(N)/day  $\cdot$  31 days  $\cdot$  (100/18)  $\cdot$  (100/8)  $\approx$  54 Tg(N)

These calculations don't contain a factor which accounts for the ratio between the horizontal domain size used in the study  $(100.100.30 \text{ km}^2)$  to the size of continental south-

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ern America (about  $18 \cdot 10^6 \text{ km}^2$ ). If one would include such a factor, one would end up with several thousand Tg(N) of lightning NO<sub>x</sub> per year, which is definitively unrealistic. This unrealistic result reflects the strong variability in the occurrence of lightning. Mari et al. took into account the spatial and temporal variability by assuming that the number of flashes in their model domain equals the number of flashes in a much larger domain (90S-Eq, 180W-30W). This ad-hoc assumption could have a dramatic impact on the results. In order for the results of the estimate of the global LiNOx production to be credible, this assumption would have to be either sustained by some additional arguments or changed.

I would also encourage the authors to include results from additional methods of approximating the global LiNOx source based on the regional simulations in the paper. One possibility might be to scale the modeled LiNOx by the ratio r of the global average number of flashes (N<sub> $\sigma$ </sub>) to the number of flashes calculated by the model (N<sub>re $\sigma$ </sub>) during the 66 h period (r=N<sub> $\sigma$ </sub>(66h)/N<sub>re $\sigma$ </sub>(66h)). The average global flash rate could be adopted from [Christian et al.(2003)] ( $44\pm5$  lightning flashes per second). The total (time and domain) integrated number of model calculated flashes (IC and CG) is currently not stated in the manuscript, but should be included in the ACP paper, (and the caption of Fig. 6 should be expanded as suggested in the technical corrections section). The month-to-month variability of the global average flash rates could also be taken into account in this estimate (assuming 8% for March as was done in the manuscript), while the variability on the timescale of a few days would be neglected. Furthermore, this estimate could possibly suffer from too low model calculated flash rates. (Note, that the good agreement of simulated and observed NO<sub>x</sub> along the flight tracks together with a relatively high number of NO molecules per flash prescribed in the model hint towards an under- rather than an overestimation of the flash rates in the domain). The result of this calculation can directly be compared to the global estimate of Tg(N)/yearfrom lightning one gets via multiplying the global average number of flashes in a year based on [Christian et al.(2003)] to the molecules per flash by [Price et al.(1997)] (using an assumption about the IC/CG ratio). Using different approaches for estimating

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the same number may help to gain an impression of the uncertainty in these estimates, and show where refinements are necessary.

Finally, I would like to emphasize that I see the paper as being an important contribution to a field of ongoing research. The approach of calculating the transport of lightning  $NO_x$  by the deep convection scheme is especially likely to prove very valuable in future studies, regardless of the obvious need to better evaluate present day transport parameterizations. A similar approach will almost certainly be considered in other global and regional models in the future as well.

### **Specific comments**

 p. 5198, last sentence: I think, it would be important to point out the uncertainties involved in extrapolating the results of the regional model run to the global scale in the abstract (see comments above). I would suggest to for example change the last sentence of the abstract from:

"Extrapolation of these regional fluxes gives global LiNOx production ..."

to: "Extrapolation of these regional fluxes would yield a (hypothetical) global LiNOx production ..."

or, as you stated in the conclusions: "A first extrapolation of these regional fluxes yields a global LiNOx production ..." (although, this is still the abstract and the meaning of the word "first" only becomes clear later.)

- p. 5199, line 11 from bottom: Does the lightning NO<sub>x</sub> source dominate the southern hemispheric NO<sub>x</sub> budget, even if the amount of lightning NO<sub>x</sub> produced globally would be at the lower end of the current estimates?
- p. 5199, line 24–25: where it can recycle HNO<sub>3</sub> and PAN → where it can take part in the formation HNO<sub>3</sub>, HNO<sub>4</sub>, and PAN, which act as NO<sub>x</sub> reservoirs (optional!)
- p. 5200, line 16: Meteorological initial and boundary conditions?

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- p. 5202, line 18: Note: Rust and MacGorman, 2002 (it should be Rust and MacGorman, not Rust et al.) as well as e.g. [Rust and Marshall(1996), Stolzenburg et al.(2002)] show that charge structures are often more complex than a simple dipole or tripole.
- p.5208, line 12: In Fig. 8 NO mixing ratios reached very high values (>15 ppbV): The NO axis in Fig. 8 ranges from 0–4 ppbV (SI unit: nmol/mol) and I can't see any values as high as 15 ppbV!
- p. 5212, line 26: Did you state the reason why the vertical transport of high NO<sub>x</sub> air from the boundary layer as a source of elevated upper tropospheric NO<sub>x</sub> mixing ratios can be excluded for the flights studied in the manuscript? (Probably this was explained in the text, but I may have missed it).
- Please state the total number of IC and CG flashes
- Would you expect a difference if NO<sub>x</sub> instead of NO was transported in the deep convection parameterization?

### Suggestions for technical corrections

- Abstract, fist sentence: based on your usage of the abbreviation "LiNOx", it would be better to write "A lightning NO<sub>x</sub> (LiNOx) source".
- p. 5198, line 6: In this approach the vertical transport of NO inside clouds is calculated by the parameterization of deep convective transport, thus eliminating the need for a-priori LiNOx.: I would add the word profiles at the end of the previous sentence. There is still a need of assumptions regarding the location in which the NO molecules are produced (prior to transport).

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- p. 5198, line 15–16: number of IC lightning  $\rightarrow$  number of IC flashes
- p. 5199, line 20: Remove the colon before the brackets
- p. 5200, line 1: regional model  $\rightarrow$  regional models
- p. 5200, line 16: change to: 70 levels with grid spacings from 40m (bottom) to 600 m (top). At what altitude is the model top?
- p. 5200, line 25: NOx: change x into subscript
- p. 5202, line 19: Rust et al., 2002  $\rightarrow$  Rust and MacGorman, 2002
- p. 5202, line 24–25: Formula (7) is valid over land only, so just in order to prevent future misinterpretations maybe better add "over land" in line 24: the lightning frequency (over land), ...
- p. 5203, line 2: "icy levels": I think, Price and Rind call it "cold cloud"?. Perhaps, you could add the definition.
- p. 5204, line 5: Missing period before "The region ..."
- p. 5204, line 10: Overview  $\rightarrow$  overview
- p. 5205, line 17: is it really "pushed" northeastward by the cold front?
- p. 5207, line 11: ... on the diurnal cycle
- p. 5207, line 16: chemical species  $\rightarrow$  mixing ratios
- p. 5208, line 4: as was show by the satellite data  $\rightarrow$  as has been shown by satellite data
- p. 5209, line 16: initial  $\rightarrow$  diagnosed?

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- p. 5209, line 25: the max of convective  $\rightarrow$  the maximum of convective
- p. 5210, line 4: CG lightnings  $\rightarrow$  CG flashes
- p. 5210, line 8: was no appropriate measurements during the studied flights to confirm or not ... → were no appropriate measurements during the flights to show whether or not production of NO occurred below the cloud base.
- p. 5211, line 10 (and some other places): put the word "however" between two commas
- p. 5211, line 13: representative could also mean that the same flash density is expected over the whole area (90S–Eq, 180W–30W). If I understood it right, you assume that the number flashes in the model domain is equal to the number of flashes in the area (90S–Eq, 180W–30W), i.e. that most lightning during this episode takes place inside the domain?
- p. 5211, line 12–13: This hypothesis may thus contributes to overestimate ..  $\rightarrow$  This may contribute(no s here) to an overestimation of ....
- p. 5212, line 14: (90S–Eq, 180W–30W): make clear that this is an area from the Christian et al. paper (otherwise it's easy to stumble over this since continental southern America is obviously not bounded by the dateline and the South Pole).
- p. 5211, line 22: omit the word associated (it's really two different things)
- p. 5211, line 26: budget  $\rightarrow$  budgets
- p. 5212, line 9–10: These flights operated by aircraft measurements ... Why "operated by"?
- p. 5212, line 12: brightness temperature  $\rightarrow$  brightness temperatures

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- p. 5212, line 12: observed from GOES-12 satellite  $\rightarrow$  observed from the GOES-12 satellite
- p. 5212, line 19: .. on the diurnal cycle.., next line: The comparison...
- p. 5212, line 19: ... flight 10 and which horizontal and temporal resolutions are much higher than the model ones. → ??... flight 10. The horizontal and temporal scales of these anvils are much smaller than the smallest scales that can be resolved by the model??
- p. 5212, line 25: which can be  $\rightarrow$  which could be
- p. 5212, line 27: Why back?
- p. 5212, line 27: from lightning source  $\rightarrow$  from the lightning source
- p. 5215, line 4: Goldembaum  $\rightarrow$  Goldenbaum
- Fig 3: Two lines under (a) are completely unreadable (same in Figs. 4,5). Caption: The black line..
- Fig 6, caption: Please state the unit of the "total" (better write domain integrated) number of simulated IC flashes? It must be flashes per some time interval.
- Fig 10, caption: over the whole domain of the simulation (add the second "the")
- Fig. 11: I think, "Top:", "Middle:" and "Bottom:" would be easier to read, but there may also be an ACP standard for this.

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# References

- [Barth et al.(2005)] Barth, M. C., S.-W, K., Wang, C., Fridlind, A., Ackerman, A., Pinty, J.-P., Mari, C., Leriche, M., Cautenet, S., Spiridonov, V., Pickering, K., Ott, L., and Stenchikov, G.: Summary of the chemistry transport in deep convection cloud modeling workshop intercomparison, Geophys. Res. Abstr., 7, SRef–ID: 1607– 7962/gta/EGU05–A–05 419, 2005.
- [Christian et al.(2003)] Christian, H. J., Blakeslee, R. J., Boccippio, D. J., Boeck, W. L., Buechler, D. E., Driscoll, K. T., Goodman, S. J., Hall, J. M., Koshak, W. J., Mach, D. M., and Stewart, M. F.: Global frequency and distribution of lightning as observed from space by the Optical Transient Detector, J. Geophys. Res., 108, 4005, doi:10.1029/2002JD002347, 2003.
- [Labrador et al.(2004)] Labrador, L. J., von Kuhlmann, R., and Lawrence, M. G.: Strong sensitivity of the global mean OH concentration and the tropospheric oxidizing efficiency to the source of  $NO_x$  from lightning, Geophys. Res. Lett., 31, L06102, doi:10.1029/2003GL019229, 2004.
- [Lawrence and Rasch(2005)] Lawrence, M. G. and Rasch, P. J.: Tracer transport in deep convective updrafts: Plume ensemble versus bulk formulations, J. Atmos. Sci., pp. *2880–2894*, 2005.
- [Mahowald et al.(1995)] Mahowald, N. M., Rasch, P. J., and Prinn, R. G.: Cumulus parameterizations in chemical transport models, J. Geophys. Res., 100, 26173–26189, 1995.
- [Pickering et al.(1998)] Pickering, K. E., Wang, Y., Tao, W.-K., Price, C., and Müller, J.-F.: Vertical distributions of lightning NO<sub>x</sub> for use in regional and global chemical transport models, J. Geophys. Res., 103, 31 203–31 216, 1998.

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[Price et al.(1997)] Price, C., Penner, J., and Prather, M.: NO<sub>x</sub> from lightning: 1. Global distribution based on lightning physics, J. Geophys. Res., 102, 5929–5941, 1997.

[Rust and Marshall(1996)] Rust, W. D. and Marshall, T. C.: On abandoning the thunderstorm tripole-charge paradigm, J. Geophys. Res., 101, 23499–2350, 1996.

[Stolzenburg et al.(2002)] Stolzenburg, M., Marshall, T. C., Rust, W. D., and Bartels, D. L.: Two simultaneous charge structures in thunderstorm convection, J. Geophys. Res., 107, 4352, doi:10.1029/2001JD000904, 2002.

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