

## ***Interactive comment on “Estimates of lightning NO<sub>x</sub> production from GOME satellite observations” by K. F. Boersma et al.***

**K. F. Boersma et al.**

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We thank referee #1 for his or her critical remarks that have been helpful in improving the manuscript.

### GENERAL COMMENTS

In the eyes of ref.#1, section 6 would be "much too long considering all the other possible uncertainties before this stage of research" and therefore ref.#1 recommends shortening this section. We feel however that we cannot get around a thorough error analysis. Our paper gives a new (top-down) estimate of LNO<sub>x</sub> production, and many readers will only be convinced if the many steps from satellite measurement to the final NO<sub>x</sub> estimate are accompanied by motivated error estimates. In fact, ref.#1

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himself seems to agree by stating that "Although there are many uncertainties in the final calculation of the NO<sub>x</sub> production, these uncertainties are addressed in the paper, and even though there are many steps taken from the satellite data collection taken at 10:30am to the final NO<sub>x</sub> estimate, the results provide additional independent scientific estimates of this uncertain source of tropospheric NO<sub>x</sub>." Moreover, in perspective of recently published estimates (ranging from 1 to 13 Tg/N/yr) of LNO<sub>x</sub> production and their widely ranging uncertainties, we feel that an in-depth error analysis is required.

#### SPECIFIC COMMENTS

"Abstract: third line is written twice."

This has been corrected.

"Page 4, paragraph 1."

This has been corrected.

"Page 5, last line of section 2: This implies a 50% error!"

This is correct. The retrieval results for situations with small tropospheric NO<sub>2</sub> loading are subject to large noise-to-signal ratio. This is discussed in more detail in the responses to ref.#2.

"Page 6: Please explain what are "ghost column difficulties"."

We have updated the text as follows: "Ghost column difficulties occur in situations when clouds screen the lower part of the atmosphere and the retrieved total column comes to depend heavily on the assumptions on the lower, unseen part. To avoid these difficulties..."

"Page 6: How do you define ocean?"

The following sentence has been added: "Tropical oceans and continents are defined as the three oceanic/continent regions depicted in Figure 10."

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"Page 6: parameterization."

The misspelling has now been corrected.

"Page 6, last sentence: There are very large changes in cloud properties with altitude around 10km. Up until -40C you can have mixed phase clouds (ice and supercooled drops) while at temperatures below -40C you have ONLY ice. This is exactly the altitude (10 km) where these changes occur in the tropics."

This has been rephrased to better reflect the point brought up by ref.#1. We have added the point: "Also, there may be transitions from mixed phase (supercooled droplets and ice) clouds to ice-only clouds at the -40C level, so multiple scattering effects may change with cloud height." Furthermore we have changed the statement that we observe a sharp increase at 10km. Inspection of Figure 4 shows that the increase is smooth, continuous, and large for clouds higher than ~6.5km rather than sharp for clouds higher than 10km.

"Page 7, first paragraph."

This has been corrected.

"Figure 5: There are many more clouds over the oceans at 10:30. How did this influence your statistics in Figure 4?"

This can be seen in Figure 4 - bottom solid line, representing the standard deviation of the mean NO<sub>2</sub> column per bin. For cloud altitudes below 10.5 km, this standard deviation is smaller for oceans than for continents but since both are less than 10% the more frequent clouds over oceans are not expected to significantly influence the statistics. From a different perspective; we also calculated the power-law relationship for tropical oceans by selecting bins with clouds up to 10.5 km only (as in the continent case). This did not give significantly different results.

"Page 8: The CP relationship is only good over continents and not over the oceans. You have to be careful using continental relationships over the oceans, and vice-versa."

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We agree. The results of this study show that CP-to-lightning relationships obtained over land are not appropriate for oceans, even though they are corrected for an assumed 10 times weaker convective intensity. This is extensively discussed in section 4.4 with references to McCollum et al. [2000], Allen & Pickering [2002], etc.

"Page 8, section 4.1.1: How sensitive are your results to the assumption that "deep convection over the oceans is 10 times less efficient in generating lightning". If it was 5 times less, would the final results be twice as large?"

If we would have assumed in TM3 that deep convection is 5 times less efficient, this would mean that the fixed LNO<sub>x</sub> production of ~5 Tg/N/yr would be distributed differently, with more LNO<sub>x</sub> production over oceans and less over continents. Subsequently, the slopes (GOME:TM3 ratio) would increase over land and decrease over oceans, and the net effect would -via equation (5)- not be very different from the estimate reported now, since this is a global estimate. The modelled spatial distribution of LNO<sub>x</sub> IS of course very sensitive to the convective intensity ratio.

"Page 10, section 4.3: the correlations imply a good connection between the variability of the two estimates. What about the absolute values?"

Ref.#1 is right that the absolute values of GOME are ~2 times higher than the LNO<sub>2</sub> from TM3. However, as discussed in section 4.3.2, the GOME signal is not from lightning alone but contains a contribution from non-lightning NO<sub>x</sub> sources. For instance, the intercept in Figure 6 is interpreted as the background non-lightning NO<sub>2</sub> signal (with a pattern orthogonal to the LNO<sub>2</sub> pattern). Absolute values are also higher because of non-lightning NO<sub>2</sub> with a pattern similar to LNO<sub>2</sub>. In order to use the slopes to rescale TM3 modelled LNO<sub>2</sub>, we need to correct for both the orthogonal and non-orthogonal part. This is extensively discussed in section 4.3.2.

"Page 11: why were only 14 points used in the correlation? It would be more convincing if this number could be increased."

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Figure 7 is an example of a temporal correlation for one individual gridcell that happened to have 14 data points (cloud-free observations within the grid cell in the non-biomass burning period). Other grid cells have similar amounts of data points. We believe that the large number of grid cells used within a region (minimum ~100) ensures sufficient statistics.

"Page 12: middle: What about the influence of stratosphere-troposphere exchange, aircraft, etc.?"

All non-lightning sources are corrected to first order. STE is described in ECMWF meteo fields input to TM3 so this influence is also taken into account. We updated the sentence as follows: "The other sources include soil and biomass burning emissions, as well as inflow of anthropogenic emissions (including aircraft emissions)."

"Page 14, second paragraph: a CG:IC ratio of 1 would reduce the overestimation over the oceans, but would this not increase the error over the continents?"

Since initial TM3 total LNO<sub>x</sub> remains fixed at ~5 Tg/N/yr, A CG:IC ratio of 1 would reduce the overestimations over the oceans AND decrease the slopes over the continents. It is probably better to think here in terms of slopes -or scaling factors- rather than in terms of errors. With equal LNO<sub>x</sub> production, the slopes over oceans will be closer to 1.0, and they will have lower values over the continents. To prevent confusion we have adapted the text as follows: "...a CG:IC energy ratio of approximately 1.0 would effectively increase slopes over the oceans at the cost of slightly smaller slope values over the continents at constant initial LNO<sub>x</sub> emissions."

"Page 14, middle: Why was the TRMM data not used to investigate the diurnal cycle. In fact, the LIS/OTD website gives the mean diurnal plot for oceans and land which could be used in this study."

First, we would like to note that the concept of continent:ocean convective intensity ratio used in this paper means that for situations of equal convective precipitation or

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equal cloud height, continents are subject to the ratio value times more flashes than oceans. The ratio of 1.6 derived from GOME is valid for 10:30hrs local time from the fact that clouds of equal height produce 1.6 times more NO<sub>2</sub> (lightning) over continents than over oceans. The very interesting figure brought to our attention by ref.#1 however, shows the ratio of the continent:ocean flash FREQUENCY. This frequency ratio seems to be determined by both the convective intensity ratio AND the continent:ocean ratio of the number of storms occurring. In other words, our derived 10:30 hrs ratio of 1.6 and assumed 24-hour average ratio of 10 can not be directly compared to the plot ([http://thunder.nsstc.nasa.gov/bookshelf/docs/white\\_paper\\_driscoll.html](http://thunder.nsstc.nasa.gov/bookshelf/docs/white_paper_driscoll.html)). We can only compare our derived and assumed ratio's to the TRMM figure once this is corrected for the ratio of storms with lightning occurring over oceans vs. over continents. Moreover, we are not aware of any paper in refereed literature that discusses the plot. To conclude, the suggestion to use time-resolving capacities of the TRMM-instruments is certainly worthwhile, but would require extensive additional research, which is outside the scope of this paper.

"Page 16, Figure 10: I would not say that the agreement is very good. Africa in GOME analysis is a very strong source, and in the model quite weak."

We agree with the reviewer that it is not correct to claim that the agreement is very good. On page 16 of the manuscript we have not done so. Indeed, we mention the differences between modelled and observed NO<sub>2</sub> columns just as ref.#1 does.

"Page 21, last paragraph."

We changed "upper plot to "upper panel".

"Figure 4."

We changed "left" and "right" to "top" and "bottom".

"Figure 6: why only 14 data points when you have measured every day at 10:30am."

This figure contains much more data points than 14. The number 14 that ref.#1 is

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referring to concerns Figure 7, which illustrates the temporal correlation method for one single grid cell. The particular grid cell shown has only 14 data points since - as described in section 4.2.2, a masking method is applied that rejects data points measured in (1) situations of biomass burning, and (2) situations with cloud reflectance >50%. Furthermore, requirements were set with respect to the representativity of the observations within a grid cell, i.e. at least 3 GOME 320x40 km<sup>2</sup> observations should have been recorded in one 2.5x2.5 degree TM3 grid cell.

"Figure 8."

Left and right are now top and bottom.

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