

## ***Interactive comment on* “Sudden increases in the NO<sub>2</sub> column caused by thunderstorms: a case study in the northern subtropical region” by M. Gil et al.**

### **Anonymous Referee #1**

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I confess to still being somewhat confused after the response to my earlier review.

The author states..."The aim of the paper is to contribute to clarify the controversy related to whether or not NO<sub>x</sub> produced in thunderstorms remain in the atmosphere for long enough to be registered by DOAS technique in zenith mode at twilight (and/or satellite borne instruments)."

I'm not convinced that this is much of a controversy. Long-range transport of lightning NO<sub>x</sub> over the Atlantic (for multiple days) has been documented by in situ observations (for examples, see papers by Jeker et al., 2000 and Crawford et al., 2000). While long-range transport of lightning NO<sub>x</sub> is clearly possible, the more basic question is whether the DOAS technique can detect it? That is why I viewed this paper to be more focused

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on the SOTARC technique rather than the issue of lightning NO<sub>x</sub>. This paper is after all based on a single event.

The author states..."The radiation in two contiguous days is almost identical and the temperature profiles were very close on days 131 and 132. In such conditions the NO<sub>x</sub> partition due to photochemistry should be the same. As a matter of fact, the shape of the NO<sub>2</sub> slant column evolution versus sza has no change from one day to another, except on the spikes days."

This is precisely the point I tried to communicate in my first review. Things should behave the same "except on the spikes days." The partition of NO<sub>x</sub> is indeed the same (or at least similar) from day-to-day at a given altitude, but the ABUNDANCE is different. Thus, the abundance of NO<sub>x</sub> in the UT on day 132 (which was NOT present on the previous day) has a distinctly different evolution during twilight compared to the lower troposphere and stratosphere which normally dominate the NO<sub>2</sub> column amount. In the lower troposphere and stratosphere, NO<sub>2</sub> is ALWAYS the dominant species; thus increases in column amount during twilight are limited to a factor of 2 or less for these altitudes. In the upper troposphere, however, changes in NO<sub>2</sub> abundance can be factors of 5 to 10, but the impact of such large changes would only be visible when the NO<sub>x</sub> abundance in the upper troposphere is large enough to influence the total column amount (e.g., the spike days).

The author states... "While the upper troposphere suffers large changes in NO<sub>2</sub> during twilight, the contribution to the total column is very small. We have run the photochemical model that accompanies SLIMCAT for the site and season. For the 3 km layer centred at 12.77 km the contribution to the column is of only 0.5% at noon and 1% at midnight conditions (and in between at twilight) far below of what is observed during the spikes events. We think that the best way to analyze the differences is by comparing them with the previous day."

I agree that comparison with the previous day is best, but again I do not find these

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statements to be at odds with my comments. The fact that upper tropospheric NO<sub>2</sub> typically makes an insignificant contribution to the total column is precisely why you should expect a different trend during twilight on spike days when there is a large abundance of NO<sub>x</sub> in the upper troposphere. Without investigating this possibility and factoring in the possible effect, the premise that the column is changing "dynamically" remains to be a rationalization.

#### References:

Crawford, J., et al., Evolution and chemical consequences of lightning produced NO<sub>x</sub> observed in the North Atlantic upper troposphere, *J. Geophys. Res.*, 105, 19,795-19,809, August 16, 2000.

Jeker, D. P., et al., Measurements of nitrogen oxides at the tropopause: Attribution to convection and correlation with lightning, *J. Geophys. Res.*, 105, 3679-3700, February 16, 2000.

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Interactive comment on *Atmos. Chem. Phys. Discuss.*, 4, 2263, 2004.

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