

## ***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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#### General Comments

This paper seems to be less about lightning NO<sub>x</sub> than the paper's title would suggest. Instead, the primary aim seems to be demonstrating the SOTARC technique. The authors might even consider changing the title of the paper to reflect this. Something like, "A technique for estimating the altitude of NO<sub>x</sub> enhancements responsible for sudden increases in NO<sub>2</sub> column over the Izana Observatory." The analysis determining that lightning was important for this particular case is still relevant, but not the main focus of the paper.

If the authors are seriously interested in attributing these "spikes" in NO<sub>2</sub> to lightning, they should expand their analysis to the other "spikes". Simple trajectory analyses

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for the other spikes and an examination of satellite imagery similar to that shown in Figure 8 would go a long way in establishing that these "spikes" are typically caused by lightning. As it is, I believe that the authors have convincingly established that lightning is the cause of the "spike" highlighted in the case study, but their results do not say anything substantial about the importance of lightning or the yield of NO<sub>x</sub> from lightning or even the frequency of lightning impacts on the upper troposphere over the Izaña Observatory.

Finally, the paper needs to more fully address and explore the changing partition of NO<sub>x</sub> during the twilight period. Diurnal changes in NO<sub>2</sub> are greater in the upper troposphere than anywhere else in the atmosphere. At these altitudes, the low temperatures and modest ozone concentrations create a daytime NO<sub>x</sub> partition that favors NO, thus the fraction of NO<sub>2</sub> is typically 20% or less. As the sun sets, the NO<sub>x</sub> partition rapidly shifts in favor of NO<sub>2</sub>, thus there is a dramatic change in upper tropospheric NO<sub>2</sub> during this period.

#### Specific Comments:

Introduction (Page 2264, Line 18): The statement attributed to Jaegle et al. is not correct. Jaegle et al. address the inability of point models to reproduce the NO<sub>x</sub>/NO<sub>y</sub> ratio. This problem is not related to NO<sub>x</sub> abundance, but rather the expected partitioning between NO<sub>x</sub> and its reservoir species (e.g., HNO<sub>3</sub> and PAN).

Introduction (Page 2264, Line 23): The statements contrasting the findings of Lamarque et al. and Ehhalt et al. regarding the importance of lightning NO<sub>x</sub> are a mischaracterization. Lamarque et al. were focused on the global NO<sub>x</sub> budget while Ehhalt et al. was much more narrowly focused on the upper troposphere at latitudes of 40–50N. It would be better here to reference estimates for the global NO<sub>x</sub> budget (e.g., Lee et al., *Atmospheric Environment*, 1997 and Bradshaw et al., *Reviews of Geophysics*, 2000) accompanied by some statements about the relative importance of surface versus upper tropospheric NO<sub>x</sub> sources.

Discussion (Page 2268, Line 5): What do you mean by "the NO<sub>2</sub> column is increasing dynamically during the evening"? If you mean that the NO<sub>2</sub> column is increasing due to transport, this is difficult to prove or disprove. On the other hand, if you are talking about increases in NO<sub>2</sub> due to changes in the partitioning of NO<sub>x</sub>, this is not only a possibility, but a fact that must be elaborated upon. Increases in NO<sub>2</sub> during the evening would have their largest impact in the upper troposphere where the daylight partitioning of NO<sub>x</sub> heavily favors NO rather than NO<sub>2</sub>. While NO<sub>2</sub> typically constitutes less than 20% of total NO<sub>x</sub> in the daytime upper troposphere, NO<sub>2</sub> rapidly becomes the dominant fraction of NO<sub>x</sub> as the sun sets. The trend in the ratio in Figure 6 is certainly influenced by this rapid change in the partitioning of NO<sub>x</sub> and therefore the abundance of upper tropospheric NO<sub>2</sub>.

Discussion (Page 2268, Line 17): The determination of altitude is based on correlations (sza = 85 to 92 deg) between the actual ratio in shown Figure 6 and the curves shown in Figure 7. A simple box model calculation, however, indicates that NO<sub>2</sub> at 10-12 km will increase by more than a factor of two between these two sza's. Some assessment of how this change in NO<sub>2</sub> abundance would affect the correlation should be explored.

Discussion (Page 2269, Line 18): I'm not sure that this statement is necessary. Variations in NO<sub>x</sub> are always related to ozone photochemistry. However, given a NO<sub>x</sub> enhancement of a few hundred pptv, the change in O<sub>3</sub> should be only a few ppbv at most. Such a change should fall well below a detectable change in the total column.

Figure 1: Why the systematic difference in am versus pm values for NO<sub>2</sub> column? A very short explanation or appropriate reference would be fine.

Figure 2: This is a bad figure...only 11 bars to represent 12 months? Also, there are only 50 events over 8 years, but 13 events in 2000 alone. Why does 2000 fall outside the norm? Even if there is no clear answer, it should be acknowledged that 2000 exhibits more "spikes" than than the average year. There are also some pm events in late 2000 (see figure 1) that do not show up in Figure 2.

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Technical corrections:

Page 2268, Line 2:  $VCD_{132}=VCD_{131}.f$  (your notation here is unclear to me, do you mean  $VCD_{131}$  multiplied by  $f$ ?)

Figure 3: This figure shows pm data only. This should be made clear.

Figure 5: The figure caption states that there are two wet layers while the text notes that there are three. Fix this for consistency.

Figure 7: The legend is unnecessary.

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

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