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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Sudden increases in the NO_2 column caused by thunderstorms: a case study in the northern subtropical region" *by* M. Gil et al.

M. Gil

gilm@inta.es

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We appreciate and thanks the anonymous referee for the careful reading and comments.

General comments:

The aim of the paper is to contribute to clarify the controversy related to whether or not NOx 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). We have demonstrated for a case study, that a short duration well defined spike observed in the NO2 column record is due to very large concentration located in the upper troposphere in airmasses coming from thunderstorm, and that these increases in a relative narrow layer can be observed by DOAS. While we have used the SOTARC technique for NO2 altitude estimation, the final purpose is to identify the last origin of the ŞanomalousŤ increase. We believe that the title focus on the objectives of the paper.

The extension of the procedure to treat it statistically is no as straightforward as it may appear, since isolation of the different potential contributions to the column require clear skies and very low aerosol content (also the need of large spectrometer signal to noise ratio), which is generally not the case. Low pressure systems in the vicinity of the Archipelago result in a break up of the trade wind inversion layer and vertical mixing. We see that in most of the spikes the storm is close enough to perturb the observations through convective clouds. Under these situations the solar spectrum is slightly modified by unknown optical processes (absorption by pollutants, Ring effect not due to Raman scattering, etc) and the error in the retrieval increases making difficult the use of the technique.

We admit that, mainly in the abstract, it can be interpreted that we claim that all spikes are produced by thunderstorms. We will clarify this point in the text.

The referee suggests the need to address the NOx changing partition during twilight. Since we are mainly interested in finding the origin of the spike, we have decided to compare the evolution of the column versus the sza with the previous day. 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 NOx partition due to photochemistry should be the same. As a matter of fact, the shape of the NO2 slant column evolution versus sza has no change from one day to another, except on the spikes days.

While the upper troposphere suffers large changes in NO2 during twilight, the contribution to the total column is very small. We have run the photochemical model that accompanies SLIMCAT for the site and the season. For the 3 km layer centred at

ACPD

4, S833–S836, 2004

Interactive Comment

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 analyse the differences is by comparing them with the previous day.

Specific comments

Page 2264, line 18. The referee is right. The sentence should state, ŤChemical steady state model calculations underestimate the NOx/NOy ratioŤ.

Page 2264, line 23. We will follow the suggestion of the referee and concentrate in global budgets. The sentence will be reformulated

Discussion (page 2268, line5): By the sentence Şthe NO2 column is increasing dynamically during the eveningŤ we mean that the column is increasing due to transport. If we assume that the partition NO/NO2 is only dependent on the sza and reservoirs do not change from one day to the next one, the contribution of the changes versus sza due to photochemistry are eliminated by ratioing the curve (slant NO2 column versus sza) from that of previous day. We have tried to explain in this section how if the increase takes place in the stratosphere at the same altitude where the maximum is located, then AMFs are the same for both days (AMFs are no dependent on the magnitude but on the shape of the vertical distribution) and a constant value versus sza should be seen in figure 6. Increases in NO2 column ratio toward larger sza can be related to either increase in the column due to transport to the station or increase in the parameter p(sza). If we increase the amount of NO2 in the upper troposphere, the AMFs decrease, and therefore decrease the ratio p. The only possible interpretation for the increase of the 132/131 ratio of the columns towards sza is that air masses rich in NO2 are arriving the station during evening of day 132.

Discussion (Page 2268, line 17). We refer to the previous point. It is clear that the NO2 increases abruptly close to sunset. That is the main reason to use the ratio between both days for the same solar zenith angles. We are searching for ŞexcessŤ of NO2

ACPD

4, S833–S836, 2004

Interactive Comment

apart for the one produced due to reduction in the photolysis. If unclear in the text we will add a sentence to clarify this point.

Discussion (Page 2269, line 18). We agree that the sentence does not add anything. We will skip it

Figure 1. We will add the following sentence in the caption: ŞLarger values in the pm data result from N2O5 photodissociation during the dayŤ

Figure 2. It was wrongly plotted. It is corrected now. As concern to the more spikes in 2000 than statistically expected, we believe that it is simply due to natural variability. The number of low pressure systems is highly variable form year to year depending on the position of the subtropical jet. Generally spikes appear when the MBL inversion breaks up. According to the suggestion of the referee a sentence clarifying this point will be added.

Technical corrections

Figure 3. It will be clarified in the text and in the caption. As the spike occur in the pm data, the am adds nothing.

Figure 5. Corrected

Figure 7. Deleted Legend

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