

Interactive comment on Hemispheric asymmetry in stratospheric NO₂ trends by M.

Yela et al.

Reply to Interactive comments from reviewer #1

We strongly appreciate the reviewer for his/her helpful and supportive comments. For clarity, our responses to the reviewer comments are in blue.

Anonymous Referee #1

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Review of “Hemispheric asymmetry in stratospheric NO₂ trends” by M. Yela et al. In this paper, the authors report on long-term ground based zenith-sky observations of stratospheric NO₂ columns at four stations and how they changed over the last 20 years. Interestingly, trends are of the order of 10% per decade at all stations but have opposite signs for the NH sub-tropical station and for the SH high latitude stations.

The stability of the trends with respect to time interval and parameters included in the trend model is evaluated, the difference between AM and PM trends discussed and the change in diurnal build-up of NO₂ evaluated. For Izana, DOAS NO₂-trends are also compared to other data sources from FTIR and satellite observations and qualitative but not quantitative agreement is found. In my opinion, this is an interesting and thorough analysis of stratospheric NO₂ trends which provides interesting data and results. While interpretation is mostly limited to qualitative discussion, and detailed comparison to dedicated model runs for the locations of the instruments is needed for a more quantitative evaluation, the study is relevant enough as it is to warrant publication. The manuscript is clearly structured, well written and the figures are clear and useful. As I have only few comments and suggestions, I recommend publication of this paper after minor revisions.

Major Comments

In the discussion of the difference in NO₂ trends between Marambio and Ushuaia, it is stated that the months with large differences are linked to the presence of the polar vortex. While this sounds plausible, it is not really supported by Fig. 8 which shows large differences already in April. To me this appears more like a latitudinal dependence which is more pronounced in winter than in summer. It is also stated that the trend in NO₂ columns could be linked to changes in vortex position and the resulting change in statistics.

This makes sense, and at least for Ushuaia, it would be relatively simple to check this assumption by repeating the analysis but excluding all measurements where the station was influenced by polar vortex air. I'd suggest adding this analysis to the paper.

Following the referee suggestion we have excluded the days inside the vortex from Ushuaia data series. Results show that, as expected, the trend decrease slightly (less negative) but the contribution to general trend is very small probably due to the fact that the days inside the vortex at Ushuaia, while increasing along time, are still very small compared with the total number of days. Even excluding these data the wavy pattern occurring in winter (reduction of the negative trend in July-August and increase in September-October) are mainly due to mid latitude dynamics. As a consequence we have reformulated the text in the following way:

Line 327 after "(Fig. 9)". Removed the sentence starting after (Fig.9).

Line 327 after "(Fig. 9)". Added the following paragraph: *"To test that the impact of the vortex position drifting is the main cause of the wavy structure in the trend observed in the Ushuaia winter (small trends in July-August large ones in September –October, see Fig. 8) we have repeated the monthly trend analysis excluding the days when the station was inside the vortex. Results show that while, as expected, negative trends are reduced, the magnitude is not enough to justify the observed trends. In fact, the changes are of only few tenths of a percent, probably due to the fact that the days inside the vortex at Ushuaia, while increasing along time, are still very small compared with the total number of days. In summary, when only extra-vortex data are used the winter wavy structure remains providing evidence that the evolution of the seasonal NO₂ trend is dominated by the mid-latitude dynamics along the year."*

In the comparison to satellite data, it would be good to add some information on collocation criteria used and the respective overpass times of the satellites. I think it would also be interesting to compare the satellite data to the PM DOAS measurements. Although the time difference between satellite observations and AM data is usually shorter, the AM measurements are strongly impacted by night-time chemistry whereas the satellite data at least over sub-tropical regions are more representative for daytime chemistry. In my experience, correlation of ground-based and satellite NO₂ data is better when using PM observations at least at low latitudes.

The purpose of considering all data available, including GB-FTIR and satellite for the Izaña station is to support, through additional and independent measurements, the genuine character of the trend. For that reason in this particular work no collocation has been applied since the absolute magnitude does not affect the trend. On the other hand, at Izaña, the ENVISAT overpassing time coincides with the effective SZA at which AM DOAS measurements are taken thus the photochemical correction has an almost negligible impact in the intercomparisons. OMI overpassing is later, and the NO₂ column is some 10% larger due to the diurnal build-up but, as mentioned, our interest lies in the trend rather than in absolute values. Recently a detailed intercomparison at Izaña considering collocation and "effective SZA" have been published (AMT Robles et al. 2016).

Minor comments

Line 38: I think the reactions listed do not lead to catalytic ozone destruction; for this reaction of NO₂ with O needs to be included.

We have improved the sentence: *Nitrogen oxides interact with ozone both directly and indirectly. Nitric oxide (NO) reacts with ozone, forming NO₂ and O₂. NO is recovered by NO₂ reaction with atomic oxygen and, in day time, by NO₂ photolysis. This catalytic reactions result in ozone reduction.*

Line 56: major source of NO₂ => major source of NO₂ in the stratosphere.

Done

Line 93: was installed Antarctica => was installed in Antarctica.

Done

Line 116: were derived from 6 typical individual measurements => were typically derived from 6 individual measurements (?).

We have modified the sentence: “...were derived from all available measurements between 89° and 91° SZA, typically 5-6 data per twilight”.

Line 134: effect on the cross-sections => effect on the effective cross-sections.

Done

Line 141: For monthly data – do you mean the fraction of months for which you have no data at all?

We clarified this point: “For monthly mean data, the rates of failures are 3.45 %, 0.40 % and 0.79 % of the total dataset for Izaña, Ushuaia and Marambio, respectively”.

Line 160: $\alpha > 0.1$ – α not defined.

α refers to the significance level. We simplified the sentence “to exclude the proxies exceeding $\alpha > 0.1$, corresponding to significant values of less than 90%”. Now it stands as “to exclude proxies with confidence intervals below 90%”

Line 160: significant values of less than 90% => significance values of less than 90%.

Changed. See above

Line 266: Both halogens should result => The observed changes in both halogens should result.

Done

Line 292: thus there was less N₂O₅ – while this is a reasonable explanation, I think other explanations cannot be completely excluded.

In lines 277-285 we argue that DBU is a good index to estimate changes in N₂O₅. However, as the reviewer points out, other explanations could be possible, particularly a change in the vertical distribution of the NO₂ column. Therefore we have modified the sentence as follows:

“In summary, all SH stations exhibit a negative decadal trend in their DBUs, ranging from -8 to -14 %, revealing either a reduction of N₂O₅ in the past few years at the middle and high latitudes of the Southern hemisphere or dynamically induced changes in the NO₂ vertical distribution”.

Line 364: N₂O oxidation is not the cause of the observed trend, nor of other global parameter changes. This sentence sounds odd, please rephrase.

Global NO₂ increase was expected at the beginning of the analysis due to the very long N₂O lifetime at a maintained increasing rate of 2.5%/decade. We have reformulated the sentence to make it clearer:

“The opposite sign in the NO₂ trends observed at the NH and SH stations shows that the NO_x distribution in the stratosphere does not directly reflect the increasing N₂O in the atmosphere, at least when individual stations are analyzed.”

Figure 10: Please add that these are AM DOAS measurements (see also my comment above).

Done

NOTE: Ushuaia data in figure 2, 4, 5, 6 and 8 are slightly different than in previous version due to the correction of few data from a recent quality control.