Interactive comment on “Variability and trends in stratospheric NO$_2$ in Antarctic summer, and implications for the Brewer-Dobson circulation” by P. A. Cook and H. K. Roscoe

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Authors’ final response.

We must thank the referees for some useful comments, and particularly for encouraging us to do multiple regressions, to define the systematic errors, and to be quantitative about the implications for the Brewer-Dobson circulation. Each of these has significantly improved the paper.

Both referees suggested that Atmospheric Measurement Techniques might be a more appropriate journal than ACP. Two of the three products in the submitted paper (NO$_2$ at a standard solar zenith angle, and NOy) are calculated by a chemical model from the measured NO$_2$, and this would surely be stretching the definition of a measure-
ment. Setting that argument aside, the revised paper has important discussions about the significance for the Brewer-Dobson circulation that deserve to be seen by the more general audience of ACP readers. In particular, the fact that after multiple regression there remains a large unaccounted variability on a time scale of at least 17 years, goes well beyond the remit of AMT.

Below, comments by Referee 1 and Referee 3 are in square brackets, followed by our responses. A revised manuscript is prepared for submission.

[1A. I am a bit torn about this paper for the following reasons: The authors provide a very thorough and detailed description of the method used to interpret their NO2 slant column measurements. This is certainly very informative but at the same time highly technical. The actual scientific interpretation and discussion - as promised in the title - only happens at the very end. However, the authors also write that there will be a follow-up paper presenting a "quantitative interpretation of their NO2 and NOy trends in terms of changes to the Brewer-Dobson circulation". My suggestion would be to either tighten the technical part and extend the discussion a) to include a proper trend analysis and not just a straight line fit, b) to then also include whatever else is planned for the "more quantitative version"; and c) reflect this appropriately in the title OR to stick with the paper pretty much as is but submit it to a more technically orientated journal such as AMT (Atmospheric Measurement Techniques) and then submit the follow-up paper to ACP.]

When we prepared this work for submission to ACPD, our separate work with a model to quantify NOy trends caused by changes to the Brewer-Dobson circulation was incomplete. The paper describing the model and its results is now under submission to Geophys. Res. Lett., and is too lengthy to be included in full in this ACPD paper. But we can now quote in detail from its results, the most important of which is that a change in Brewer-Dobson speed leads to an almost equal change in NOy, of opposite sign. We have also included a proper trend analysis as part of the multiple regressions (see 1B). We feel the existing title reflects rather accurately the revised contents.
In any case, a significant part of the paper discusses NOy results that have been derived from the measurements via a model. This combination of model and measurements could hardly be described as an Atmospheric Measurement Technique.

[1B. If the paper should stay with ACP, then I would strongly suggest that the authors provide a trend analysis (multi-linear regression) which apart from the linear trend includes e.g. QBO, ENSO, solar cycle and possibly a volcanic term. This would be much more convincing given the aim of the paper (implications for the Brewer-Dobson circulation) and the high inter-annual variability clearly displayed by the data set.]

We have now done multiple regressions, which show that, contrary to our earlier impression, ENSO does not have strongest correlation with NO2 and NOy, although it is still significant at the 90% level. Instead, the solar cycle and QBO are better correlated, close to or above the 95% level. We have now included the results of these regressions in the extra Table 4 and a new Figure 10, and have made important modifications to the interpretation in Section 6. As before, the error bars on the linear components (trend) include zero. The revised paper now includes:

"The results in Table 4 show that the solar cycle is well correlated and the QBO anti-correlated, each of them close to or above the 95% level. ENSO is less well correlated although it is still significant at the 90% level. The reconstruction components from the regression results for NOy, shown in Figure 10, shows that the maximum in NOy in December 2000 is close to the solar maximum in 2001/2, and coincides with a strong negative QBO in December 2000. But although the regression accounts for 37% of the variance, much is unaccounted for. In particular, the residuals show that much of the increase up to 2000 and the reduction since is unexplained. This remains partially true even if the solar and QBO slopes are pushed to the limits of their error bars - the reconstructed value in 1990 then agrees, and the slope to 2000 almost disappears, but the slope since 2000 is almost doubled.

Importantly, the linear term from these regressions gives a much more rigorous trend
estimate than a simple straight-line fit to the data. This is because dependence on slowly-varying quantities such as the solar cycle can impart a false trend that varies with start and end time of the data set. The results still show little or no overall trend, that in NOy being 1.1 +/- 3.5 %/decade. Including the total systematic error in Table 2 would increase this error bar by less than 0.1 %/decade."

[1C. Also, if the paper stays in ACP, I really would expect to see more emphasis on the discussion of the trends and implications for the Brewer-Dobson circulation.]

We agree, and have now included:

"To investigate the relationship between trends in NOy and trends in speed of the Brewer-Dobson circulation, we constructed a simple multi-box model of the coupled troposphere and stratosphere (P.A. Cook & H.K. Roscoe, "Does an increase in the Brewer-Dobson circulation increase or reduce the amounts of stratospheric reactive gases?", submitted to Geophys. Res. Lett.). The results show that the trends are of opposite sign, and after subtracting the trend in tropospheric N2O they are of almost equal magnitude (e.g. a 40% increase in speed results in a 39% decrease in NOy). Tropospheric N2O has been increasing by about 2.5 %/decade (WMO 2007), so that our trend in NOy signifies a small and not significant increase in Brewer-Dobson circulation of 1.4 +/- 3.5 %/decade. The correlations with solar cycle and QBO in Table 4 imply equal correlations of opposite sign in Brewer-Dobson circulation: the speed of circulation follows the solar cycle with amplitude of about 20%, being slowest at solar maximum; and follows the QBO with amplitude of about 10%, being fastest at the positive phase of the QBO. There remains an unexplained reduction cycle of amplitude at least 15% and of period at least 17 years, with a minimum in speed in about 2000."
simply in the fourth paragraph of Section 4.

[1.2) Some of the figures are quite hard to read and I had to look at them on the screen quite strongly enlarged; would be helpful if they could be edited for easier reading (e.g. at least enlarge axis title).]

We agree. We have improved all the figures.

[1.3) Figures 1a+c should have a second y-axis (e.g. on the right side of the figure) with the actual SZA values rather than using one axis with SZA/15.]

We agree, and after the usual struggle with interpreting Excel 2007 we have succeeded.

[1.4) Page 839, lines 6-9: "... demonstrating that further processes are involved". These "further processes" have been discussed in McLinden et al., and that should be mentioned here; please add the ref: McLinden, C.A., S.C. Olsen, M.J. Prather, and J.B. Liley, Understanding trends in stratospheric NOy and NO2. J. Geophys. Res. 106(D21): 27787-27793, 2001. ]

We agree, and have revised this sentence. It was an oversight on our part not to have included the reference to this important work on the subject of NO2 trends.

[1.5) Page 840, line 11: should read something like that: "..., but the AMF also depends on the wavelength";]

We agree, and have revised this sentence as above.

[3A. Most useful conclusions of the paper is the demonstration that the many sources of error, which could impact the retrieval (Air Mass Factor correction, Langley plot, stratospheric temperature and vertical profiles of the species) have limited consequence on the NO2 vertical column during the summer, and therefore that the large inter-annual variability of about 12% peak to peak amplitude observed is fully meaningful.]

But in the absence of multi-regression analysis of influent parameters (i.e. QBO, ENSO,
geomagnetic activity, etc), the conclusions on a possible impact of changes in the B-D circulation and moreover on a link between these and ENSO, is very little convincing.]

We have now done multiple regressions, which show that, contrary to our earlier impression, ENSO does not have strongest correlation with NO2 and NOy, although it is still significant at the 90% level. Instead, the solar cycle and QBO are better correlated, close to or above the 95% level. We have now included the results of these regressions in the extra Table 4 and the new Figure 10, and have made important modifications to the interpretation in Section 6. As before, the error bars on the linear trend components include zero. See our response to 1C above for text of the revised paper.

[3 Summary. Since the most useful conclusion of the paper is the demonstration of the existence of a large NOx/NOy inter-annual variability exceeding the uncertainty, based on a thorough discussion of errors, it could be of great help for the UV-Vis community carrying similar studies. But given the limited audience, I feel that it would be more appropriate for publication in Atmospheric Measurement Techniques.]

As in our response to 1A above, we can now quote in detail from the results of our separate work with a model to quantify NOy trends caused by changes to the Brewer-Dobson circulation (under submission to Geophys. Res. Lett.). We have also included more emphasis on the implications for trends in the Brewer-Dobson circulation, including quantifying the upper limit from our measurements. This is important because climate models have predicted a significant increase in Brewer-Dobson circulation due to increased greenhouse gases, but recent work suggests that changes in stratospheric ozone have had an opposite effect. Our discussion deserves to be seen by the wider audience of ACP rather than the narrower technical audience of AMT.

[3.1. The influence of diurnal NOx / NOy photochemical changes, Langley plot intercepts, AMF calculations, stratospheric temperature, ozone and aerosol and species vertical profiles on the retrieval are described in great details with a number of figures, but what is missing is a quantification of each. Sentences such as "the sensitivity to re-
alistic changes is modest" for temperature, "has little effect on the overall NO2 column" for ozone, "effect is small" for aerosol, are very little informative. Most useful would be numbers in a table summarizing the error budget.]

We agree, and we have now included exact values for the change in NO2 at the end of the offending sentences listed above, plus a new Table 2 which details the resultant systematic errors in our trend in NOy.

[3.2. The figures are relatively poor (scale change between two comparable plots, scales like SZA/15 or SZA/30) and they are too many. Appropriate numbers, summarized in a table as suggested above, could easily replace them. ]

We agree about the quality of the figures, and have improved them. We also agree about the unorthodox scales, and have now replaced them in Figures 1a and 1c.

We disagree about the number of figures. There are 3 pairs of figures showing the pattern of changes (measured NO2, interpolated NO2, and calculated NOy respectively). It is an important conclusion of the paper that these patterns are almost identical, and it is impossible to show patterns in a table. The referee agrees with us that the patterns are almost identical, but would the referee or anyone else be convinced by a succession of statements followed by "not shown"?

The trends are already in a table (now Table 3), the new results of multiple regressions are in the extra Table 4, and the implication of the trend from multiple regressions is discussed in detail in the interpretation section.

[3.3. The discussion of implications for the Brewer-Dobson circulation is very crude and therefore little convincing. Indeed Randel et al (2006) have observed a fast reduction of stratospheric H2O, ozone, and tropopause temperature attributed to a change of upwelling of B-D circulation but, in 2001 and not in 2000 and in the opposite direction: increase and not decrease upwelling, which Rosenlof et al (2008) are correlating with SST changes. Is it the origin of the proposed relation between NO2 and La Nina? The
NO2 column at Rothera shows a significant drop in 2006 and 2007. Any idea of the origin of that? I am not aware of any significant change of source gases in 2006-2007, though a large number of measurements are available from ODIN, MIPAS, AURA.]

We have now significantly revised our interpretation, including the multiple regression results which account for a significant portion of the inter-annual variability discussed above by the referee. More importantly, we now quote results from our simple model of Brewer-Dobson circulation, to enable a quantitative interpretation of Brewer-Dobson speed given our NOy results (see response to 1B above).

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 837, 2009.