Dear Editor,

With regard to the manuscript:

MS-NR: acp-2012-201
Title: Analysis of stratospheric NO2 trends above Jungfraujoch using ground-based UV-visible, FTIR, and satellite nadir observations

Author(s): F. Hendrick, et al.

Please find below the replies to Anonymous Referee #1's comments.

Sincerely yours,

F. Hendrick (**franch@oma.be**)

Anonymous Referee #1

First, we would like to thank Anonymous Referee #1 for his/her helpful comments.

In this manuscript, Hendrick et al. report on trends in stratospheric NO2 columns observed at the station of Jungfraujoch by three independent measurement systems: FTIR, SAOZ, and UV/vis nadir satellite observations. The three time series are in very good agreement and all show a downward trend in NO2 column, in apparent disagreement with the increasing abundances of N2O. The observations are compared with previous results reported in the literature and possible reasons for the NO2 decline are discussed. The paper is well structured and clearly written. The topic of the paper (observations of temporal changes in stratospheric NO2 columns) is interesting and fits well into the scope of ACP. The main result of the study – a downward trend in mid-latitude NO2, consistently observed by three independent measurement systems in spite of the increase in N2O - is well supported and relevant for stratospheric research. I therefore recommend publication of the paper after minor revisions.

Major comment
The only major concern I have is the discussion of the possible reasons for the NO2 decline which I find much less convincing than the presentation of the measurement results. The discussion is completely neglecting the vertical distribution of the different species, and is thus based on the assumption that the vertical profiles are simply scaled over time. This implies that the change in NO2 column which is dominated by the bulk of the NO2 profile at higher altitudes can be linked to, for example, the change of HNO3 column which has a profile with a much lower maximum in the

atmosphere. In my opinion, this is not necessarily true. The same problem arises when discussing ClONO2. Also, possible changes in temperature can have complex effects on stratospheric chemistry and in my opinion a model is needed to evaluate such effects with any confidence. I therefore recommend either to shorten this section and emphasize that these are only plausibility arguments, or to add model based sensitivity studies supporting the arguments made.

In response to this major comment, we would like to clarify the scope of the paper: we present a trend analysis of stratospheric NO2 at Jungfraujoch based on three independent observational data sets and we give possible reasons/arguments explaining the negative trend values consistently derived from the SAOZ, FTIR, and satellite time-series of stratospheric NO2 columns. We agree with Anonymous Referee #1 on the fact that model-based sensitivity tests and analyses would help to give a firmer interpretation of the results. However, this would require a lot of efforts and it constitutes for us the subject of a separate study. So, we think that our paper and the way the results are presented in it (i.e. based on observational data only) is self-consistent and merit publication in ACP. Therefore we decided to follow the first suggestion made by Anonymous Referee #1 for the revised manuscript, i.e. emphasizing that the discussion is based on plausibility arguments only. Since we already proceeded like this in the first version of the paper (see lines 9-13 on page 12371, lines 20-21 page 12374, and more generally the way the discussion is written), we only made the three following changes in the revised manuscript:

Line 11, page 12371: '…suggest a change…' is replaced by '…suggest a possible change…'

Line 12, page 12371: '…we discuss possible explanations…' is replaced by '…we discuss plausible explanations…'

Line 4-8, page 12375 (last paragraph of the conclusions): 'Similar trend analyses are needed at other locations using FTIR, UV-visible, and satellite observations to augment the interpretation of our findings by improving the statistics of such a trend study. This also suggests that more effort should be put into consolidating the different ground-based and satellite observational data sets, which is one of the major tasks of the NDACC.'

is replaced by:

'Model-based sensitivity studies would certainly augment the interpretation of our findings, as well as similar trend analyses at other locations using FTIR, UV-visible, and satellite observations. However, these are beyond the scope of the present paper. This work also suggests that more effort should be put into consolidating the different ground-based and satellite observational data sets, which is one of the major tasks of the NDACC'

Minor comments

P 12362, l 15: For the satellite data, stratospheric temperature is taken into account but not for the SAOZ ground-based observations. Please comment on possible effects of this difference

We have estimated the impact on the SAOZ columns of using the NO2 cross sections at 220 K instead of taking into account the variation of the stratospheric temperature. Sensitivity tests in the 425-490 nm range using the Vandaele et al (1998) NO2 cross-sections at 220 and 294 K show an increase of the NO2 slant column density of 20% from 220 to 294 K, i.e. an increase of 0.3%/K assuming in a first approximation a linear temperature dependence. The NO2 effective temperature at Jungfraujoch, defined as the mean temperature of the stratosphere weighted by the NO2 concentration profile (Gil et al., 2008), is estimated using ERA-Interim temperature profiles and NO2 profiles from the SLIMCAT/PSCBOX model. It ranges in average from 212 K in winter to 232 K in summer. This means that the correction for the difference between 220K and the NO2 effective temperature to be applied to the SAOZ NO2 columns is of -2.4% in winter and +3.6% in summer. On average over the year, we can therefore expect slightly larger SAOZ NO2 columns when applying a correction for the variation of the stratospheric temperature. Since the relative difference satellite-SAOZ is about +2% on average (satellite larger than SAOZ, see Figure 3), this would improve a little bit the agreement of the SAOZ data with satellite observations.

P12366, l10: Why was a simple geometric AMF used? Isn't the model vertical column used here? This sentence is not clear to me.

The output of the assimilation process is assimilated stratospheric NO2 slant columns. They have to be divided by an AMF (in this case, a simple geometric AMF) in order to get vertical columns.

P12368, statistical model: Why are there seasonal terms for QBO and aerosols, and why are 2 seasonal terms used for the QBO and one for the aerosols?

According to the Bodeker et al. (1998) paper on which the statistical model is based, 2 seasonal terms are used for QBO because the extratropical influence of the equatorial QBO is stronger in winter-spring and weaker in summer. 1 seasonal term is used for aerosols to allow for seasonally dependent effects as expected from observational and modeling studies.

P12370, l11: If QBO and solar circle do not contribute significantly, wouldn't a trend model be more robust without these terms?

The way we proceeded was to use the more complete statistical model. Afterwards, we have seen that the QBO and solar circle terms did not contribute significantly. For the 1990-2009 and 1996-2009 SAOZ time-series, we have

performed some sensitivity tests by removing both QBO and solar terms. The trend values obtained with both versions of the statistical model were negative and within their combined error bars.

P12371: formatting problem for N2O

Corrected.

P12371: Why is hydrolysis of N2O5 not considered as HNO3 source?

We have only considered the main source of HNO3, which is the three-body gas-phase reaction OH+NO2+*M* (see e.g. Urban et al., 2009).

P12374, l22: if you consider stratospheric cooling, what about the possible impact of the temperature dependence of the NO2 cross-section in the SAOZ retrieval?

According to the above calculations, a stratospheric cooling will cause a decrease of the SAOZ NO2 columns due to the temperature dependence of the NO2 cross-sections, going into the direction of a negative trend.

References:

Bodeker, G. E., et al.: Trends and variability in vertical ozone and temperature profiles measured by ozonesondes at Lauder, New Zealand: 1986-1996, J. Geophys. Res., 103, 28,661-28,681, 1998

Gil, M., et al.: NO_2 climatology in the northern subtropical region: diurnal, seasonal and interannual variability, Atmos. Chem. Phys., 8, 1635-1648, 2008

Urban, J., et al.: Nitric acid in the stratosphere based on Odin observations from 2001 to 2009 – Part 1: A global climatology, Atmos. Chem. Phys., 9, 7031-7044, 2009

Vandaele, A. C., et al.: Measurements of the NO_2 absorption cross section from 42000 cm^{-1} to 10000 cm^{-1} (238-1000 nm) at 220 K and 294 K, J. Quant. Spectrosc. Radiat. Transfer, 59, 171-184, 1998