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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Assessment of the interannual variability and impact of the QBO and upwelling on tracer-tracer distributions of N_2O and O_3 in the tropical lower stratosphere" *by* F. Khosrawi et al.

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We thank reviewer 1 for the constructive, helpful criticism. We followed the suggestions of reviewer 1 and revised the manuscript. However, the page numbers and lines given by reviewer 1 did not fit to any of the available manuscript versions (submitted draft or the two online versions). We hope that we nevertheless succeeded to provide the correct answers to the reviewer comments as well as to make the anticipated improvements on the text.

General comments:

The paper by Khosrawi et al. uses N_2O and Ozone data from different satellites for model evaluation and to assess the impact of the QBO on tracer-tracer distributions. The paper relies to a large part on N_2O and Ozone data by the Odin instrument. My main concern is that an analysis is performed which discusses subtle changes in a data set which shows rather obviously problems, especially with respect to the N_2O values of Odin, showing values above the tropospheric background at 25 km altitudes in the tropics. There have been a number of in-situ aircraft measurements (see e.g. Volk et al., 1996, 1997; Strahan et al., 1999) in this region deploying high precision in-situ N_2O measurements. None of these data sets have found enhanced N_2O even at 20 km altitude.

We apply Odin/SMR here as our main data set though the occurrence of too high N_2O mixing ratios is most pronounced in this data set. We do not dispute that these high values in Odin/SMR data are unrealistic. The occurrence of these high N_2O mixing ratios is caused by the instrument noise of Odin/SMR and become quite visible due to the method we apply. This is discussed in detail in section 4.2, p22646, l4ff. However, this does not conflict with the main objectives of this paper and the intention to show that the inter-annual variability in the monthly averages of N_2O and O_3 are low independent of which data set is used and that the method applied in this study can serve for both model evaluation and satellite data intercomparisons. Further, the objective of the method applied here is for evaluating models and performing satellite data intercomparison concerning O_3, not N_2O (e.g. Khosrawi et al., 2009). Thus the uncertainties in O_3 are much more important for the applicability of this method than the discrepancies in the N_2O measurements. We discuss the discrepancies in N_2O due to the fact that these were quite obvious in our previous model evaluation study (though not affecting the results, see Khosrawi et al.(2009)). This raised our curiosity on looking deeper into this issue. Since we found out that this is a matter not only affecting Odin/SMR but also other satellite instruments we decided to discuss this as it is done in the present study.

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N_2O values higher up are certainly lower due to the photolytical sink of N_2O. Further there is no plausible mechanism which could produce large amounts of N_2O or transport them to these altitudes. Looking at the in-situ climatology by Strahan et al (1999), I wonder what we can learn about variability of O_3 for N_2O larger than 300 ppb at 650 K pot. temperature? Most probably that there is a problem with the N_2O measurement. Even though the climatology by Strahan is somewhat older and N_2O values have increased by some 3-4% since, this climatology would suggest values on the order of 300 ppb as a maximum N_2O in the tropics at 500 K and certainly significantly less at 25 km altitude.

We agree that these high N_2O values in the satellite data sets are definitely due to biases or instrument noises, but the seasonal and inter-annual variability we see with which these values occur are caused by a physical process such as the QBO. Further, though we explain the O_3 variations for N_2O>300 ppb at 650 K as to be caused by the QBO the method is a valuable tool for model evaluation and satellite intercomparison where the entire N_2O/O_3 space and all altitudes are considered and not only the O_3 variation at N_2O/O_3 at 650 K for $N_2O > 300$ ppb. Nevertheless, when to such an evaluation/intercomparison is done one of course has to take into account the uncertainties of the satellite data sets. Nevertheless, model deficiencies are still clearly distinguishable from the differences caused by the uncertainties in the observations (see e.g. Fig. 2).

This is also supported by the correlation between N_2O and O_3 from ACE (Fig. 1) which goes to values on the order of 270 ppb at 650 K and by the correlations shown in Fig. 4. and 5. which do not show such high N_2O values (even though they are higher than from the in-situ climatologies). Therefore, I think that the N_2O axis chosen as a reference in this correlation study is highly uncertain, which puts the whole basis of this analysis on very shaky ground. My suggestion would be to exclude all tropical Odin N_2O data from this analysis, as it seems that the data base is not solid.

The fact, that the N_2O data derived from ACE-FTS as shown in Fig. 1 is significantly lower than from Odin/SMR is to a major part caused by the limited sampling of ACE-

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FTS. We find these values in the Odin/SMR data in the tropics and the tropics are very poorly covered by ACE-FTS. Further, also in the ACE-FTS data such high values occur as can be seen by the N_2O/O_3 correlation and N_2O/O_3 PDFs derived from ACE-FTS data over several years by Hegglin and Shepherd (2007). Although it seems that such high N_2O values are measured by all satellite instruments it does not contradict the three major purposes of our analysis. For showing that (1) the inter-annual variability in the monthly averages of N_2O/O_3 is low and differences can be easily be distinguished from model deficiencies, (2) the seasonal and inter-annual variability in the occurrence of higher/lower N_2O values is caused by the QBO (though we agree that the absolute values are incorrect as discussed above) and (3) the method can be applied for satellite intercomparisons, these uncertainties in the Odin/SMR measurements have no major influence. Thus, we do not think it is necessary to exclude the N_2O data from Odin/SMR measured in the tropics.

Specific comments:

p.4, l17: please specify which lifetime is meant (global, local etc.)

We refer to the global life time of N_2O. The sentence has been changed as follows: *The global mean lifetime of N_2O is in the order of 122 ± 24 years in the troposphere (Volk et al., 1997) and decreases with altitude from several years in the lower stratosphere to $\approx$8 months in the middle stratosphere (Stanford and Ziemke, 1991).*

p.5, l5ff: O_x is mainly destroyed in the high latitudes. The lifetime of ozone itself is shorter in the low latitudes.

We agree and changed the text as follows: *Ozone (O_3) is rather short lived in the troposphere (days to weeks), although it has a longer lifetime in the lower stratosphere. The lifetime of O_3 is about 1 month in the lowermost stratosphere and in the winter hemisphere high latitudes but decreases strongly with altitude in the stratosphere, particularly during summer months (Garcia and Solomon, 1985). [....] Since O_3 is not photochemically conserved it has limited application as tracer of transport.*

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p.14, l3.: I think that the decrease/increase of N_2O/O_3 is better visible in a simple vertical profile then in this view.
We agree, but our intention is to show the impact of this well known vertical increase/decrease of N_2O and O_3 on the monthly averages of O_3/N_2O.

p.14, l5: I have to admit that I do not understand this statement: what is meant by a set of curves lying in potential temperature bins? These are curves for separated potential temperature ranges.
We agree that this sentence is somewhat misleading. The set of curves are not lying in the potential temperature bins, but each curve derived for a potential temperature bin lies within this potential temperature bin. We have changed the sentence as follows: *Second, the N_2O and O_3 binned by potential temperature are averaged over 20 ppbv N_2O resulting in a set of curves. Therefore, each curve lies within its potential temperature bin (e.g. one curve at 400±25 K, 450±25K and so on).*

p.14, l10ff: I think that this is actually a bit a comparison of apples with pears. The ATMOS correlations are from one latitude and the variation in N_2O and O_3 comes from the different altitudes sampled. In the present study the variation in N_2O and O_3 is caused by the latitudinal variation in sampling. This is something entirely different.
We do not compare apples with pears when comparing the reference curves derived from ATMOS with observations from ACE-FTS. The reference curves from the ATMOS observations were derived for different latitude and altitude regions as described in Michelson et al. (1998a) and Michelson et al. (1998b). This is exactly the same as for the ACE-FTS data. These are also derived for different latitude and altitude regions. Though the data is treated differently afterward (fit of the O_3/N_2O correlation and separation in different altitude bins and then averaging in our method) they still serve the purpose of differentiating between air of polar, midlatitude and tropical character.

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p.16, l16ff: I do not understand some of the argumentation in this section.
We unfortunately cannot address this comment. It is not clear which paragraph is meant since the page and line numbers given do not fit to any of the manuscripts version available.

p.17, l11: I suppose that the reasonable agreement refers to the 650 K level? Or is the agreement between the models meant?
This sentence refers to 500 K as it is written in the text. However, we agree that this may be misleading and have dropped the first sentence. Our intention was to discuss the differences we found at 500 K between models and observations and not to grade the model performance. This has already been done in Khosrawi et al. (2009) and does not need to be repeated here.

p.19, l10ff: Further to the general remarks given above, it is not possible from Fig. 3. to distinguish in which year the climatology extends to higher N_2O values or has more ozone variability. In any case I wonder why for the discussion presented here a correlation is chosen, as this could be discussed straight forward using the latitudinal distributions observed (as the O_3 is not really used in the discussion). It is interesting to note that these extremely high N_2O mixing ratios occur at O_3 values on the order of 5 ppm, i.e. values typical of the middle stratosphere.
The O_3 or N_2O inter-annual variations are not that large and show that especially concerning O_3 these variations are negligible. The variations in N_2O are somewhat larger and we agree that the years during which year the climatology extends to higher N_2O values is not clearly visible from e.g. Fig 3. However, we have made this variation visible in Figure 7 as well as in the tables provided in the electronic supplement. References to both Figure 7 and the electronic supplement are given at several places in the text.

p.20, l4: I do not see why a higher number of observations should lead to a smaller

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standard deviation.
It is correct that a higher number of observations itself does not necessarily leads to a smaller standard deviation. However, a high number of measurements smoothes out gradients in the measurements of trace gas distributions. Further, the high vertical resolution of Odin/SMR is accompanied with a higher noise of the Odin/SMR data compared to other data sets. These differences in sampling and vertical resolution do influence the standard deviation of the monthly averages but not the monthly averages itself. Nevertheless, the main reason for the higher standard deviation is the higher noise of Odin/SMR.

p.21, l22: I would suggest to sort the data according to the QBO phase and then derive a QBO-east and a QBO-west correlation and see if they deviate in a statistically significant way from each other.
We appreciate this suggestion for sorting the data to check for an deviation in a statistically significant way. This would be worthwhile. However, this is beyond the scope of this study and this idea has to be kept for future studies.

p.22, l25ff: Isn't this quite trivial? I would expect the maxima of the averaged bins to occur when the maxima in the data occurs.
This relationship reviewer 1 is referring to is indeed trivial. However, what we meant here was that when we see maxima in the N_2O anomalies that are caused by the QBO we see also the maxima in our maximum N_2O mixing ratios of the averaged bins. We changed the text as follows and hope that what we meant becomes clearer now: *Further, the N_2O fields for the stratosphere (Fig. 8, third panel) show that due to a stronger upwelling N_2O was transported to greater altitudes in 2002, 2004, 2006, 2008 and 2010. This is in agreement with the maxima of the maximum N_2O mixing ratios of the averaged bins we found in these years (Fig. 7, as well as electronic supplement).*

Section 5.2.2.: I think that one should accept that the observation of large regions of

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N_2O well above the tropospheric background at 25 km altitude is just not a realistic feature. This occurs in no other data set and I find it very hard to imagine a process which produces N_2O in-situ in the stratosphere (see general remark above). With respect to the CRISTA N_2O data, I would like to point the authors to a paper by Kuell et al (JGR, 2005) in which the problem of trace gas retrieval from CRISTA data in the tropics is discussed, due to the high uncertainty in the temperature fields. These data are highly suspect and should not be used as a reference. There have been a number of high-precision in-situ measurement campaigns in the tropics, which all confirm that there are no N_2O values above tropospheric background in the stratosphere. I think the only valid approach is to exclude these data from any further analysis before there has been a thorough validation. I am convinced that these values are just an instrumental artifact.

We agree that the observations of high N_2O mixing ratios we found in the satellite data cannot be realistic. We have stated this several times in the text and are confident that this point comes across now in the paper. However, though the absolute values of high N_2O are definitely unrealistic the seasonal and inter-annual *variation* we see in all satellite data sets is caused by a physical process as the QBO as it is discussed in the paper. To emphasize this also in the previous section before discussing further satellite data sets we added the following text at the end of section 5.2.1: *Thus, though the absolute values are most likely caused by instrument noises and biases the winter/summer variation of the occurrence of higher/lower N_2O values can be attributed to the QBO.* CRISTA data is applied in our study since it is a data set with a very high spatial resolution. We are aware that there are high uncertainties in the absolute values of CRISTA N_2O data as represented by the large systematic error (e.g. 26 %, see Table 1). Since tropospheric measurements are well within the error bars, no additional mechanisms are needed to explain the "enhanced" CRISTA values, but the error bar has to be taken into account, when the data are interpreted. Therefore, we agree that our statement that Odin/SMR and CRISTA data are in good agreement concerning deriving monthly averages of N_2O and O_3 at $N_2O > 300$ ppbv is somewhat misleading. We therefore changed the sentence as follows: *Although these measurements were performed several*

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years earlier than the Odin/SMR measurements, here we also find N_2O mixing ratios up to 330 ppbv at 25 km (∼650 K). However, this does not mean that CRISTA-1 observations suggest N_2O values above the tropospheric background levels, since the systematic error of the CRISTA-1 observation is rather large in the tropical lower stratosphere (26 %, see Table 1). A more detailed discussion is given at the end of this section (page 22653, l15ff). We agree that it would be worth finding a reasonable way to exclude the unrealistically high N_2O mixing ratios from the satellite data sets. However, we will leave the decision on how the data quality can be improved to the respective satellite teams and their staff working on the data retrievals. In case of Odin/SMR, however, one cannot simply remove noisy data. E.g. if one would remove high values from a statistical distribution through filtering, one would introduce a negative bias of the mean which should be avoided. So one has to live with the noise in the data and consider all characteristics of a statistical distribution (here of single N_2O observations) simultaneously, which are mean, standard deviation, bias etc. Nevertheless, we show in our study that despite biases and noise of the N_2O data derived from satellites these data sets are suitable for scientific application.

p.27, l17ff.: I think that the MIPAS 2003 must be considered an outlier when looking at the entire MIPAS data set (see Fig. 5) and should not be used for intercomparison purposes. Otherwise the 2003 (especially April) data would not be in agreement with the statement that inter-annual variations are low. Therefore I doubt if they are best suited for a comparison.
It is correct that the MIPAS 2003 data deviates from the MIPAS data for the following years. However, it is well known that after the interruption of the MIPAS measurements in 2004 the MIPAS operation has been continued with low spectral resolution instead of the high spectral resolution applied before. The high bias of MIPAS data of about 23 ppbv between 6 and 25 km altitude (2002-2004) for the first, so-called full-resolution period of MIPAS observations has been identified earlier and is well-known (e.g. von Clarmann et al., 2009; Palazzi et al., 2011). Efforts were made to reduce the high bias for the later so-called reduced (or optimized)-resolution data set (2005 to 2012),

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and the production of a consistent mission-long bias-free data set is under way. The high bias of the first sub-set and the differences between the two MIPAS sub-sets are discussed on page 22652, l26 to p2653., l2. We changed the sentence as follows: *These low values are a result of the effort to reduce the well-known high bias in MIPAS N_2O observations of the period before.*

Section 5.3.: Taking into account that there are large uncertainties especially with respect to the ODIN N_2O data, I wonder if a validation using N_2O as one of the coordinates is a very sensible approach. One could actually find differences in ozone between different instruments which could turn out to be caused only by differences in the N_2O-axis. I suggest sustaining all findings with respect to ozone by a direct decompression, e.g. on eq. lat. - pot.temperature coordinates.

We agree that there are some uncertainties (biases/noise) in the N_2O data from Odin/SMR as well as in the data from the other satellites. However, this does not affect any model evaluation or satellite intercomparison study performed with this method. This method focuses on ozone and in case of an evaluation study differences between the data sets are only calculated for ozone (as done in Khosrawi et al. (2009)). We have not seen any indications that these uncertainties in N_2O affect the N_2O/O_3 monthly averages derived from measurements or model simulations. Note: These high values result in an extention of the N_2O/O_3O curves on the N_2O axis by 1 or 2 bins. In case of e.g. a model evaluation they won't be considered since these bins are not found in the model simulation data and differences are only calculated where bin values are available for both data sets. Thus these uncertainties do also not affect the evaluation/intercomparison. Of course, the method presented here is only one method that can be applied for model evaluation or satellite data intercomparisons. Other methods could be applied as well. We agree that an analysis in the equivalent latitude/potential temperature space could be an alternative to our analyses approach. However, potential temperature is already used in our analyses. Further, equivalent latitude and latitude are similar in the tropics. Using the N_2O/O_3 tracer space as done

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here is at least an alternative way to remove dynamical variability from the anlyses (e.g. Khosrawi et al., 2009).

p.31, l.2.: The Odin values at 25 km are 10 ppb higher than the tropospheric values. Other data indicate, that a substantial fraction of N_2O (typically about 15%) should already have been removed photochemically at this altitude. Therefore the difference is much larger than the stated 10 ppb, more in the region of 50-60 ppb.

We agree though the differences are not as large as suggested by reviewer 1. The decline of N_2O in the vertical profile is more pronounced at the midlatitudes due to the lower lying tropopause there. In the tropics N_2O is destroyed at altitudes above 25 km. The principal region of photolysis of N_2O in the tropics occurs at 30-35 km altitude (Warneck1988, Kuttipurath2010). Therefore, mixing ratios of around 300 ppbv can still be expected in the tropics at 25 km. We changed the sentence as follows to explain this in the paper: *However, the difference we found in our monthly averages between models and observations is much higher than the differences from the ground-based N_2O measurements. That is, We found a difference of 20-40 ppbv between model and satellite measurements compared to 10-20 ppbv between satellite and ground-based measurements. N_2O is destroyed in the tropical stratosphere at altitudes above 25 km, thus below 25 km we can expect an N_2O mixing ratio nearly unchanged from what is found in the troposphere (Warneck1988). Such a high positive bias between model and simulations (20-40 ppbv) was not found in validation studies applying Odin/SMR N_2O observations. We conclude that, this difference is probably caused by the combination of N_2O values derived from model simulations being too low, and the mixing ratios measured by Odin/SMR being too high.* Further, the simulation of lower N_2O mixing ratios than observed is a common feature of most models which arise due to difficulties in simulating atmospheric transport. We added additionally the following sentence: *In fact, the simulation of too low N_2O is a common feature of most models as it was discussed e.g. in Kuttipurath et al. (2010) and references therein.*

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References:

Garcia and Solomon (1985), The effect of breaking gravity waves on the dynamics and chemical composition of the mesosphere and lower thermosphere, J. Geophys. Res., 90, 3850-3868.

Hegglin and Shepherd (2007), O_3-N_2O correlations from the Atmospheric Chmistry Experiment: Revisiting a diagnostic of transport and chemistry in the stratosphere, J. Geophys. Res., 112, D19301, doi:10.1029/2006JD008281.

Khosrawi et al., (2009), Evaluation of CLaMS, KASIMA, and E5M1 simulations in the lower stratosphere using observations of Odin/SMR and ILAS/ILAS-II, Atmos. Chem. Phys., 9, 5759-5783.

Kuttipurath et al. (2010), Aircraft measurements and model simulations of stratospheric ozone and N_2O: implications for chemistry and transport processes in the models, J. Atmos. Chem., 66, 41-64.

Michelson et al. (1998), Correlation of stratospheric abundances of NO_2, O_3, N_2O and CH_4 derived from ATMOS measurements, J. Geophys. Res., 103, D21, 28.347-28,359.

Michelson et al. (1998), Correlations of stratospheric abundances of CH_4 and N_2O derived from ATMOS measurements, Geophys. Res. Lett., 25, No. 15, 2777-2780.

Volk et al. (1997), Evaluation of source gas lifetimes from stratospheric observations, J. Geophys. Res., 102, D21, 25,543-25,564.

Warneck (1988), Chemistry of the Natural Atmosphere, Academic Press, San Diego, USA.

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