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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

Interactive comment on "Equatorial transport as diagnosed from nitrous oxide variability" *by* P. Ricaud et al.

P. Ricaud et al.

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Reply to the Reviewers' comments on the manuscript "Equatorial transport as diagnosed from nitrous oxide variability" by P. Ricaud et al. (Atmos. Chem. Phys. Discuss., 9, 4899-4930, 2009)

REVIEWER 1 (REV1) & REPLY (REP)

REV1: This paper uses 5 years of ODIN satellite measurements to study spatial and temporal variability of N2O in the tropics, and makes comparisons with results from 2 chemistry transport model simulations (SLIMCAT and MOCAGE). This is the first analysis of the ODIN N2O data that I have seen, and these results are novel and interesting. The data and models all show the well-known seasonal and QBO variations over ˜20-50 km, with some noted differences between the observations and model results. However, the most interesting results and comparisons are in the altitude region near the tropopause, where neither model captures the observed seasonal or spatial variability. The authors document the behavior of the observations in this region, and make arguments about the importance of overshooting convection and transport for the lowermost stratosphere. Overall the paper is novel and interesting, and these new data will make an important contribution to understanding of the TTL region. However, I am not in agreement with all of the inferences regarding these observations, and I suggest the authors address the following points in revision.

REP: We thank the reviewer for these comments. We fully understand his questions regarding some of the inferences. Several features shown by ODIN were very new for us and not readily understandable. We have addressed at best the reviewer's remarks below.

REV1: 1) The lack of seasonal variation in the models at 100 hPa is very interesting, but this could be due to a number of problems (such as N2O seasonality in the model upper troposphere and/or problems in model transport in the TTL).

REP: These are exactly our arguments developed in the present paper.

The SLIMCAT model does not have any seasonal variation in the troposphere. A gradually increasing surface vmr is imposed and this is well-mixed up to the level below the tropopause. The seasonal cycle seen at 100 hPa in SLIMCAT is presumably because SLIMCAT uses theta levels and so, while N2O is constant on the model levels up to the tropopause, there is this variation on p levels in the very low stratosphere. In contrast, as shown in Fig. 10, MOCAGE has variable sources. The N2O concentration is not homogeneous in the troposphere, but the 1.5 ppbv amplitude of variation at 100 hPa is rather small compared to the 12-15 ppbv shown by ODIN.

REV1: I agree with the logic that the vertical gradient in N2O is small, so that seasonal variations in vertical velocity probably do not completely explain the overall TTL seasonality (although the seasonal N2O maximum in NH winter is consistent with stronger

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upwelling in this season). However, I am not convinced that overshooting convection is a main contributor to the observed variability at 100 hPa.

REP: We agree that the seasonal variation of N2O in the upper troposphere is rather hypothetic. MOCAGE, which includes geographic and seasonal changes of sources at the surface, does not show more than 1.5 ppbv variation at 100 hPa and the recent IASI N2O total column measurements (Ricaud et al., ACP, 2009) does not show more than 1.6% zonal contrast in the troposphere neither.

This is why, since the first submission, we have explored other possibilities for explaining the N2O seasonal cycle in the lower stratosphere: a) meridional exchange with the mid-latitudes known to be larger in the summer, particularly in the NH summer (however not in May-June but later). But since the models are known to well represent horizontal transport, most of the observed N2O seasonal cycle has to be attributed to something else. b) sinking regions in the lower stratosphere, like above Indonesia as shown by Sherwood (GRL, 2000), required for closing the energy budget following the overshooting irreversible injection of cold and heavy air at great height. Though still tentative, the advantage of this process is that it could provide an explanation for the larger reduction over the Pacific than over Africa, and moreover, the stronger difference in May coinciding with the TRMM maximum overshooting volume.

The discussion of possible contributors has been deeply revised.

REV1: A key point is that the seasonality of the overshooting convection (above 14 km) in Fig. 7 (maximum in March-May) does not match the observed N2O minimum during May-July (Fig. 4), and results are not shown for the amount of overshooting for higher altitudes (the text suggests overshooting up to 500 K or 21 km; can TRMM results for these levels be shown?).

REP: The measurements of the TRMM precipitation radar are showing extreme events of 20 dBz at altitude $>$ 18.5 km (0.1%) or even at 19.5-19.8 km (0.01%) (Liu and Zipser, JAS, 2007).

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But overshoots are expected to increase the N2O concentration at and above the tropopause and not the opposite, unless there is a large annual cycle of concentration of the species in the UT in contradiction with current understanding of N2O sources at the surface. Moreover, we agree that the minimum N2O does not match the seasonality of overshooting volume, even if calculated within 20°S-20°N instead of 10°S-10°N, which shows larger overshooting volume in Sept-Nov.

This is why we propose the idea of a contribution of local drain associated to intense convective overshooting, maximum in May. Another advantage of this mechanism would be an explanation of the propagation of the minimum in May up to 500K.

REV1: I am unconvinced that the vertical profiles in Fig. 9 demonstrate injection of tropospheric air up to 500 K, but rather I see the largest gradient changes during May-July below 450 K (most pronounced in the Western Pacific); this is difficult to interpret given the poor knowledge of seasonal variability of N2O in the upper troposphere. Overall I do not think that there is convincing evidence that overshooting convection is a dominant process influencing seasonal or longitudinal structure in the TTL.

REP: Indeed the largest gradient changes occurring between April and May at 400 and 450 K cannot be explained directly by an injection of tropospheric air which should result in an N2O increase and not the opposite. If convection was involved (the event coincides with the month of max TRMM overshooting), the mechanism is certainly more complex.

The description of Fig. 9 has been revised as well as the discussion of possible mechanisms and conclusions.

REV1: 2) I would like to see a comparison of the vertical velocities used in the two model simulations (some time series at a couple of pressure levels). I am surprised at the substantial differences in the vertical structure of N2O between the two models in Fig. 9, and wonder if this is due to the imposed vertical velocity or some large differences in N2O photochemistry between the models. It would also be helpful to

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understand better the behavior of N2O in the upper troposphere in both of the models. Does SLIMCAT incorporate stratospheric ozone variations in its radiative calculations?

REP: It is indeed possible to get heating rates from SLIMCAT, but then they are difficult to compare with MOCAGE. As the two models are using the same analysis after 2002, numerous studies have been published regarding vertical transport. The SLIMCAT model has an option to use a similar setup to MOCAGE (i.e. the TOMCAT model with stratospheric pressure levels and vertical motion from the divergence). Previous studies have intercompared the tracer transport obtained with the SLIMCAT/TOMCAT model in the stratosphere (Chipperfield, 2006; Monge-Sanz et al., 2007). They have shown the more rapid vertical tracer transport in the pressure-coordinate model than in the sigma-theta coordinate model. This is explained in the text.

SLIMCAT has no N2O seasonal variation in the UT. It uses a slowly increasing global surface mixing ratio and assumes this vmr is mixed to just below the tropopause. This is now explained in Section 2.2.

SLIMCAT does incorporate stratospheric ozone variations in its radiative calculations. This has been added in Section 2.2

"The modelled ozone field is used in the calculation of diabatic heating."

REV1: 3) I think there is some confusion regarding seasonal upwelling in the TTL. The seasonal cycle in upwelling is a dynamically forced phenomenon, with a resulting seasonal cycle in temperature and radiative heating (temperature responds to the upwelling, and the radiative heating is a response to the temperatures being out of equilibrium). Radiative heating is a response to the dynamically-forced upwelling; radiation is not a driver of the seasonal cycle. Similarly, the corresponding variations in TTL isentropes are a (diabatic) response to the dynamical upwelling, so that isentrope movements are not a useful explanation of constituent seasonal variability.

REP: The reviewer is right and we have carefully reconsidered the explanations relative

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to this point. We are using diabatic heating as a diagnosis of the dynamical change, not as the forcing. That is why theta level / heating rate models work. We do not mention anymore that variations are forced by changes in diabatic heating. We rephrased the appropriate sentences.

REVIEWER 2 (REV2) & REPLY (REP)

General Comments

REV2: This paper discusses transport mechanisms in the equatorial region as diagnosed from the occurrence of semi-annual, annual, and quasi-biennial oscillations in the N2O field observed by the ODIN. The findings for the SAO and QBO are largely confirmation of previously reported work, but the results presented for the annual oscillation in the UTLS, the main focus of the paper, are new and interesting. The mechanism advanced to explain the observations, convective overshooting coupled with an unobserved variation of tropospheric mixing ratio, is somewhat speculative and not entirely compelling, but serves as a starting point for further thought on the subject. Ideally, it would be nice to see the observations reproduced in a model which, unlike the two used here, incorporates the invoked processes. The arguments in the paper are generally well presented, but the text contains a number of minor grammatical errors or unusual phrasings which would benefit from tidying up. Also, some more detail is required throughout the paper on the data, models, and methods used.

REP: We agree that the evoked seasonal variation of N2O concentration in the upper troposphere is rather speculative and certainly very little consistent from what is known about the sources and lifetime of the species. Moreover, it is in contradiction with the zonal contrast between oceanic and land convection over the West Pacific and Africa. But since the N2O and dynamical upwelling are out of phase by 2 months, another process is required. Since the first submission we have found an alternative tentative explanation better matching the observations with the idea of "stratospheric drain" proposed by Sherwood (GRL, 2000) from wind measurements over the Indonesian re-

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gion, tentatively explained as resulting from the sinking required to close the energy budget after injection of cold and heavy air in the lower stratosphere by convective overshooting.

The description of possible explanations for the N2O seasonal variation and zonal contrast, together with the discussion and the conclusions have been deeply revised.

Specific Comments

P.4900,L. 20, "...peaking in May and absent in the models"; "absent in the models" is slightly ambiguous. It's not clear whether or not the models even attempt to simulate overshooting. For the abstract the phrase could probably be omitted altogether.

REP: We removed the term "absent in the model" from the abstract.

P.4901, L.7-9, Some more specific information on the N2O lifetime in the stratosphere and its variation with height would be helpful. We are told the stratospheric lifetime is "less than one year", but it is then referred to as a "long-lived species".

REP: We clarified this point by modifying the sentence into:

"where its lifetime decreases from ˜1 century in the lower stratosphere to ˜1 month in the upper stratosphere."

P.4902, L. 7, Reference for the TRMM data?

REP: We indeed refer to the TRMM data and study presented in Liu and Zipser (2005). The missing reference has been added.

P.4902, L.9, Definition of TTL, and how it relates to UTLS?

REP: The definition of the TTL adopted here is that of the transition region between the level of zero net radiative heating (LZH) (˜14 km, ˜150 hPa) and the top level of tropospheric influence or the bottom level of stratospheric lapse rate (˜18.5 km, ˜70 hPa), as suggested by Fueglistaler et al. (Review of Geophysics, 2009). The UTLS is the conventional layer covering the upper troposphere (including the altitude of convective outflow) at about 12 km up to the lower stratosphere at about 23 km (550 K).

We have thus modified the sentence into:

"As an extension of the previous study, here we investigate how N2O behaves over a five-year period in the entire equatorial stratosphere including the Tropical Tropopause Layer (TTL), the region of intermediate lapse rate extending from the level of zero net radiative heating (LZH) (˜14 km, ˜150 hPa) to the level of stratospheric lapse rate (˜18.5 km, ˜70 hPa). We have combined ODIN/SMR measurements from 2001 to 2006 and long-term runs of the three-dimensional (3D) chemical transport models (CTMs) SLIMCAT and MOCAGE."

P.4903, L.14, How many measurements are made within each bin? Is the averaging sufficient to effectively remove the measurement noise (precision error)?

REP: Within single bins, the number of measurements can vary between 100 and 500 depending on time, pressure, and latitude-longitude bin within the equatorial band 10°S-10°N from November 2001 to June 2005. Typically, precision errors are ranging 0.8-7.0 ppbv at 100 hPa, 0.8-2.0 ppbv at 70 hPa and 0.35-1.00 ppbv at 10 hPa per bin. If we consider the equatorial evolution of N2O wrt to time or wrt longitude, the precision errors are ˜0.2-1.2, 0.2-0.4 and 0.07-0.20 ppbv at 100, 70 and 10 hPa, respectively. These figures are indeed much less than the calculated amplitudes of AO, SAO and QBO, and the amplitudes of the longitudinal gradients.

We added a new sentence.

"Typically, precision errors per bin in the latitude band 10°S-10°N are ranging 0.8-7.0 ppbv at 100 hPa, 0.8-2.0 ppbv at 70 hPa and 0.35-1.00 ppbv at 10 hPa. If we now consider the equatorial (10°S-10°N) evolution of N2O wrt to time or wrt longitude, the precision errors are then ˜0.2-1.2, ˜0.2-0.4 and ˜0.07-0.20 ppbv at 100, 70 and 10 hPa, respectively."

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P.4904, L.8, Why were the SLIMCAT results zonally averaged instead of being placed into lat-lon bins like the ODIN and MOCAGE data?

REP: Only monthly- and zonally-averaged SLIMCAT results were actually available for the present study. We added a new sentence.

"Note that only ODIN and MOCAGE are represented in Fig. 5 since only monthly zonally-averaged fields are available for SLIMCAT."

P.4904, L.14, State specifically that exactly the same ECMWF files were used for forcing MOCAGE and SLIMCAT, or explain the difference if there is any.

REP: We indeed added a new sentence to clarify this point.

"Therefore, MOCAGE uses the same analyses as SLIMCAT from January 2002 onwards but before this date the models use different ECMWF products (ERA-40 for SLIMCAT)."

P.4904, L.24, Could perhaps mention how the surface mixing ratios of N2O compare in SLIMCAT and MOCAGE.

REP: We again clarified this point by adding the following sentence.

"The mean tropospheric N2O is constrained in MOCAGE to a value of 316 ppbv over the 2000-2005 period whilst in SLIMCAT it increases from 315.6 ppbv in January 2000 to 320.5 ppbv in December 2005."

P.4904, L.25, The implication from later in the paper is that the Betchold convection scheme does not allow overshooting and transport of material into the stratosphere. This should be stated here.

REP: In fact, the Betchold convection scheme used in the MOCAGE model does allow overshooting and transport of material into the stratosphere. Nevertheless, these overshootings across the tropopause are highly improbable (as it was already reported in Ricaud et al., 2007) because of the weak horizontal resolution used in the MOCAGE

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runs (5.6°x5.6°). We modified the sentence into:

"Regarding convection, the present run was performed using the scheme of Betchold et al. (2001) that allows overshooting and transport of material into the stratosphere. However, given its coarse (5.6°x5.6°) resolution, MOCAGE can hardly capture overshooting across the tropopause. Indeed, as shown by Ricaud et al. (2007) for the period of MAM 2002-2004, the probability density function of altitude reached by convection in MOCAGE is negligible above 150 hPa within 10°S-10°N whatever the area considered: Africa, South America or Western Pacific."

P.4905, L.6, A sentence indicating how the various oscillation amplitudes were extracted from the data (Fourier decomposition?) should be included. What period was used for the QBO?

REP: The oscillation amplitudes were estimated by using a gradient-expansion algorithm to compute a non-linear least-square fit of the non-linear function representing the AO, SAO and QBO. The period under consideration was covering the domain 2001-2005 and we fixed the QBO phase to be 28.2 months. We added a new sentence.

"To estimate the different amplitudes, we used a gradient-expansion algorithm to compute a non-linear least-square fit of the non-linear function representing the AO, SAO and QBO. The period under consideration was from November 2001 to June 2005 and we fixed the QBO phase to be 28.2 months."

P.4905, L.9, Clarify if these vertical winds are identical to those used by MOCAGE.

REP: Yes they are. We added the following sentence.

"The vertical winds presented in Table 2 are identical to those used by MOCAGE."

Section 3.1, It would be helpful to give the reader some more indication of which figure is being discussed at various points in the text.

REP: This has been done.

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P.4907, L.5, As I understand it, N2O from the two models is here being correlated with the vertical winds used by MOCAGE (or at least winds calculated in a similar manner). Thus it is hardly surprising that the correlation is greater in MOCAGE than in SLIMCAT.

REP: Yes indeed, but the N2O-omega correlation in the vertical domain 2.1-10 hPa from the MOCAGE data sets is very consistent with the ODIN data sets compared to the SLIMCAT data sets. We nevertheless added a new sentence.

"Since N2O from the two models is correlated with the vertical winds used by MOCAGE, we can expect that the correlation is greater in MOCAGE than in SLIMCAT."

P.4907, L.8-11, In addition to the vertical velocities themselves, how the N2O field responds to the velocities will depend on the vertical N2O gradient, controlled by the chemistry. How do the gradients throughout the stratosphere compare in the observations and the two models? Could this play any role in the differences in amplitude of the oscillations?

REP: We have indeed detailed the impact of the N2O gradients in the lower stratospheric N2O evolutions in the section 4.2 highlighted by Figure 9. We have not considered this issue throughout the stratosphere in our manuscript.

P.4908, L.9, Some explanation should be given for why the difference between the Western Pacific and Africa is not shown for SLIMCAT in Figure 5.

REP: Again, the SLIMCAT data set available for the present analysis was corresponding to monthly- and zonally-averaged data. We add a new sentence:

"Note that only ODIN and MOCAGE are represented in Fig 5 since only monthly zonally-averaged fields are available for SLIMCAT."

P.4908, L.17, Make clear you are referring to Figure 3.

REP: Yes indeed, we referred to Figs. 3 and 4. We clarified this point in the text.

P.4909, L.16, Where does the figure of 600-800 m month-1 for the ascent rate come

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from? And how does it relate to the 0.2-0.3 km month-1 given a few lines earlier?

REP: The apparent contradiction between the two ascent rates needs clarification. Firstly, the ascent rate of 0.2-0.3 km month-1 is related to the Brewer-Dobson circulation in the stratosphere (20-25 km) and cannot explain the upward propagation of the minimum in June up to 32 hPa. Secondly, the ascent rate of 0.6-0.8 km month-1 is the vertical speed for summer season of the northern hemisphere at 17.5 km as calculated by Randel et al. (2007) in agreement with Yang et al. (2008). We slightly rephrased the sentence and added the new reference Yang et al. (2008).

"Yang Q., Q. Fu, J. Austin, A. Gettelman, F. Li, H. Vömel (2008), Observationally derived and general circulation model simulated tropical stratospheric upward mass fluxes, J. Geophys. Res., 113, D00B07, doi:10.1029/2008JD009945."

P.4910,L.27, "The 2-month chemical lifetime of CO implies that its concentration decreases rapidly with time". That is not strictly true since in the absence of any change in the sources or sinks a short lived species will be in steady state. This argument should be expressed more clearly.

REP: Indeed this sentence is confusing. We modified the sentence into:

"Since a slow vertical displacement will have limited impact on the concentration of the species given the small vertical gradient, the observations will be better compatible with overshooting during the convective season in February-April, followed by a 40% photochemical reduction during the following 5 months, because of the 2-month lifetime of the species."

P.4911.L.12, "The fourth and last potential contributor". Are the four factors discussed really the only potential contributors to an AO? I doubt it. Better just to say "A fourth potential contributor".

REP: OK, modified.

P.4912,L.1, "A vertical displacement of 250m...". This sentence is repeated almost verbatim from P.4910,L.20. One or other of the occurrences should be removed.

REP: Yes indeed we modified the sentence into:

"As already mentioned in the previous section, given the large gradient, a small vertical displacement would be enough to explain the change."

P.4912,L.7, "...peaking in July" peaking is not the appropriate word for the point where the values are a minimum.

REP: We removed the term "peaking".

P.4912,L.21, As we are discussing the tropics, what months are meant by "the winter"? Are the overshooting episodes reported by Ricaud 2007 being advanced as an explanation of the fast increase in CO in November-December mentioned above. If so, this should be made clearer. If not, the fast November-December increase should be attributed to another cause. The likely impact on the CO cycle in the lower stratosphere of the large seasonal variations in tropospheric CO due to biomass burning should also be discussed.

REP: Indeed, we completely rephrased the section related to CO variations in order to take into account the time evolution of different mechanisms: biomass burning activities, uplift, overshootings, ... The new paragraph is presented below and includes new references to the study presented in Schoeberl et al. (2006).

"The MLS CO shows two maxima in the tropical troposphere related to biomass burning activity, in October-November above South America and in February-March above Africa (Schoeberl et al., 2006; Randel et al., 2007). However, the CO seasonal cycle at 68 hPa (19 km) only displays one single maximum in February-April like if the large November CO tropospheric burden was not lofted above the tropopause, in contrast to the February-April maximum reaching higher altitude. Since a slow vertical displacement, either radiative ascent or isentropic surface lift, will have limited impact on the concentration of the species, the observations will be better compatible with overshoot-

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ing during the convective season in February-April, followed by a 40% photochemical reduction during the following 5 months. Overshoot episodes, such as those reported by Ricaud et al. (2007) over Africa, could also explain the fast anti-correlated CO-O3 changes at 68 hPa shown by MLS (Randel et al., 2007), which would require vertical velocities exceeding by far that of radiative ascent (Folkins et al., 2006; Corti et al., 2005)."

P.4914,L.18, "The above findings fully confirm the hypothesis of the convective origin..." "Fully confirm" is a little strong based on the evidence presented. "Support" is a more appropriate word.

REP: Modified.

Figure 6, The caption should state the bottom panel is the potential temperature anomaly.

REP: Modified.

Technical Comments

"O3" and "ozone" are used interchangeably throughout the text. Best stick to one or the other.

REP: We have used the term "O3" within the manuscript.

P.4900, L.14, "...the variations are shown..." change to "...the OBSERVED variations..."

REP: Done.

P.4901, L.11, "straightforward diagnostic". Perhaps omit "straightforward"?

REP: Done.

P.4906, L.2, "diabatic thermal processES"

REP: Done.

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P.4906, L.21, "differences with the observations", differences FROM the...

REP: Done.

P.4907,L.6, "...is rather poor in SLIMCAT..", rather LOW...

REP: Done.

P.4908,L.9, "...interpolated at 400 ...", interpolated TO

REP: Done.

P.4908,L.28, "...such has seen in the HALOE...", such AS

REP: Done.

P.4910,L.23, "...a change of 3.5 ppbv only at 17.5 km..", "a change of only 3.5 ppbv at 17.5 km" is slightly less ambiguous

REP: Done.

P.4911,L.4, "In the case ozone...", In the case OF ozone

REP: Done.

P.4914,L.27 "confronting the measurements to", "COMPARING the measurements WITH";

REP: Done.

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