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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Equatorial total column of nitrous oxide as measured by IASI on MetOp-A: implications for transport processes" *by* P. Ricaud et al.

**P. Ricaud et al.**

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Reply to the Reviewers' comments on the manuscript "Equatorial total column of nitrous oxide as measured by IASI on MetOp-A: implications for transport processes" by P. Ricaud et al. (Atmos. Chem. Phys. Discuss., 9, 3243-3264, 2009)

4 May 2009

REVIEWER 1

This is a nice paper that shows how IASI trace gas data for the long-lived N2O molecule can be used to study atmospheric dynamics. In particular, operational total column data for N2O are sufficiently precise and accurate (after seasonal averaging on a 5°

grid) to show equatorial maxima over Africa and Indonesia, and a minimum over South America. The MOCAGE model helps to explain the observations by showing a convergence of airmasses over Africa. I have just a few questions and corrections:

1. I found repeated use of the term "factor 1.25%" when discussing the correction to N2O to be a minor irritation because the authors do not mean multiply by 0.0125, but multiply by 1.0125. I suggest simply replacing 1.25% by 1.0125 everywhere including in two figures.

The term "1.25%" has been changed into "1.0125" everywhere including in the Figs. 4 and 5.

2a. I very much liked Figures 1 and 2, but a few more comments on pg. 3249 would be helpful. I was puzzled by the residuals for what I assume to be CO on the left of Figure 1 and CO2 on the right. The CO2 residuals are attributed to NLTE effects and assuming that the lines are from hot bands (Is this true?) seems to be a plausible explanation. However, I am surprised that NLTE effects are significant in these mainly tropospheric spectra. No comment is made about the CO residuals. Are they hot band lines or is there some other problem?

In the region dominated by CO2 (i.e beyond 2240 cm-1) we see larger residuals and it is plausible that these larger values are associated to NLTE effects not modelled in RTIASI (it is a general limitation of RTIASI-5.3). The order of magnitude is consistent with results obtained with simulations of the NLTE effect. But the NLTE effect is only one of the factors that could affect the residuals. One should also consider spectroscopic errors and the specification of the temperature and absorber amount profile. This is also true for the spectral regions dominated by the other absorbing species. For instance, in the spectral region dominated by CO (namely below 2180 cm-1), the larger residuals are seen in correspondence of strong CO lines. Consequently, the residuals could be affected by the specification of the CO profile and by errors in the specification of the CO line parameters.



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We thus modified the two last sentences on pg. 3249 into:

"NLTE effects are not accounted for in the RTIASI-5.3. Although they may have an impact at 2240 cm-1 up to 2250 cm-1 ($CO_2$ bands) where the residuals reach ~2 K, one should also consider spectroscopic errors and the specification of the temperature and absorber amount profile. This is also true for the spectral regions dominated by the other absorbing species. For instance, in the spectral region dominated by CO (namely below 2180 cm-1), the larger residuals are seen in correspondence of strong CO lines. Consequently, the residuals could be affected by the specification of the CO profile and by errors in the specification of the CO line parameters."

2b. I found Figure 2 to be quite striking with the horizontal peaks at 700 and 250 hPa. My main complaint is that I find it hard to think in terms of hPa and perhaps an approximate scale in km on the right would be helpful (and giving a few more approximate heights in km in parentheses in the text would be nice). What is the tropopause height for this example (hPa and km please!).

We explicitly mention the approximate height on the right-hand side of the Figure 2 (lower panel) and in the text. We also added the following sentence in the Fig. 2 caption:

"Note that the tropopause pressure is about 93 hPa (~17 km) at this particular location and time."

3. In view of the fairly good agreement between the observations and MOCAGE over Africa and South America, I wonder why the agreement is so poor over Indonesia (and outside the equatorial regions) as shown in Figure 4. No comments are given although the discrepancies are noted in the text. Any suggestions would be welcome.

See the reply to the last comment from reviewer 2.

4. An extra A appears after MetOp-A on pg. 3257, line 22.

We corrected the above mentioned typo.



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REVIEWER 2

The manuscript of Ricaud et al. covers an important scientific topic, namely the interpretation of satellite-derived pattern of the greenhouse gas N2O. The topic is well suited for ACP. The manuscript is well-written and I recommend its publication after the comments listed below have been carefully considered by the authors.

General

The retrieval of tropospheric N2O from space and the interpretation of the results is a relatively new area. Previous papers, which are cited by the authors (mainly Chedin et al., 2002, and, Lubrano et al., 2004), basically show some first results exhibiting significant differences with reference data used for comparisons without any detailed interpretation on what may cause the discrepancies. The results discussed in Lubrano et al., 2004, which are based on high spectral resolution satellite data, are limited to only five real IMG spectra. It is therefore early days for N2O retrieval from space. The topic covered by this manuscript is therefore relatively new. Because of this, this paper is potentially quite important. On the other hand care is needed with the interpretation of the results. The authors aim at giving a clear interpretation of what the retrieved pattern mean in terms of, for example, transport processes. Based on the results shown, I have strong doubts that at this stage it is possible to draw the conclusions, which have been drawn by the authors. In the following I will give more details about this aspect.

Abstract

Based on Fig. 4, which shows large differences between IASI and MOCAGE N2O, I think one cannot say that "very good agreement" exists between the two data sets. I recommend to replace "very good" by "reasonable" (at best).

Corrected as suggested by the reviewer and in the conclusions.

"N2O measured by IASI": IASI measures interferograms, which are converted to spec-



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tra. N2O is retrieved from the spectra. N2O is therefore, strictly speaking, not measured, but obtained from an interpretation of the spectra (which implies many subjective choices how to do this). I recommend to replace "NO2 measured by IASI" with "N2O retrieved from IASI".

Corrected as suggested by the reviewer.

Last sentence: Based on the results shown (Fig. 4) this statement is too strong. I recommend to replace "we provide measurement evidence ..." with something like "our results are consistent with ..." or equivalent.

We modified the sentence into: "... our results are consistent with the fact that..."

This has also been modified in the conclusions.

In the last paragraph of the section 3, we changed "Furthermore, there is measurement evidence that confirms the theoretical study..." into:

"Furthermore, our results are consistent with the theoretical study...".

Section 2.1, page 3248:

As this is (one of) the first papers on IASI N2O more details about the retrieval algorithm are needed as part of this paper. Please add a more detailed description of the retrieval algorithm and the assumptions and input data the algorithm is based on. It needs to be explained how the algorithm works, but also how sensitive the retrieval is to errors of the temperature profile and how exactly the temperature profile has been obtained. Has the temperature profile been obtained assuming that CO2 is known? If yes what is the impact of this for N2O retrieval? N2O is due to its long lifetime (potentially) even less variable (more constant) than CO2 implying that the N2O results may depend critically on the assumptions made about CO2. How are varying surface properties dealt with (e.g., surface pressure / altitude variations, emissivity)?

We completely modified the paragraph related to the retrieval process into:



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"The retrieval algorithm is designed as described by Turquety et al. (2004), as feed-forward artificial neural network. The spectral samples entering the network basically include 30 $N_2O$ channels positioned in lines of the v3 band between 2200.75 and 2243.25 cm-1, and three micro-window channels at 2136.25, 2142.75, and 2150.00 cm-1, providing a "baseline" in the water-vapour absorption continuum. Additionally, the inclusion of four lines from the v1 band between 1278.75 and 1291.50 cm-1, carefully chosen to avoid the $CH_4$ fundamentals, proved to be advantageous in the training of the network. Surface temperature and a coarse temperature profile, as derived from an Empirical Orthogonal Function (EOF) regression retrieval enter the network as additional information. The temperature profile is derived assuming a constant $CO_2$ concentration."

We also added a new reference.

"Turquety, S., Hadji-Lazaro, J., Clerbaux, C., Hauglustaine, D. A., Clough, S. A., Cassé, V., Schlüssel, P., and Mégie, G.: Operational trace gas retrieval algorithm, for the Infrared Atmospheric Sounding Interferometer, J. Geophys. Res., 109, D21301, doi:10.1029/2004JD004821, 2004"

See also comment 2 from the Reviewer 1.

Of critical importance for this manuscript is also if averaging kernels are available from the retrieval algorithm? As IASI's $N_2O$ altitude sensitivity is far from constant, IASI cannot provide "true vertical columns". It is therefore mandatory to apply averaging kernels (which depend on the instrument and on the retrieval algorithm) to the model profiles before any reasonable comparison can be made. Has this been considered?

We indeed clarified this point since averaging kernels can only be calculated off-line, as the retrieval algorithm is just a non-linear regression, using an artificial neural network. The relation between the true vertical columns and the integrated profile applying the averaging kernel is implicit in the retrieval method and only represented statistically.



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We added the following sentences and a new reference.

"IASI does not provide true vertical N2O columns, as the sensitivity of the measurements is not homogeneous over the vertical profile. Usually, an averaging kernel is applied in the vertical integration of the retrieved profile (e.g. Rodgers and Connor, 2003). Here, the relation between the true vertical columns and the integrated profile applying the averaging kernel is implicit in the retrieval scheme and only represented statistically."

"Rodgers, C. D., and Connor, B. J.: Intercomparison of remote sounding instruments, J. Geophysical Res., 108(D3), 4116, doi:10.1029/2002JD002299, 2003."

The numbers given for the estimated accuracy and precision are much larger than the approx. 1 percent variations shown in the top panel of Fig. 5 on which most of the conclusions of this paper are based on. Important for this paper would be the relative accuracy (systematic differences between different regions and time periods). If I understand correctly, the accuracy (4 percent) is mainly supposed to be a general bias (offset). But what about the relative accuracy? Is the relative accuracy (e.g., between regions) supposed to be better that 1 percent? Or is it possible that the longitudinal variations shown in Fig 5., which are only about 1 percent, are on the same order as the estimated relative accuracy ?

There is no relative accuracy estimate available, as the method has not been validated so far.

Section 2.1, page 3249:

As good spectral fit does not necessarily imply a high accuracy of the state vector parameters.

We indeed agree with the reviewer. This is why we simply mentioned in the text the term "suggesting" a high accuracy of the state vector parameters, nothing more.

Section 3.2, page 3263:



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The main conclusions of the paper are drawn from the top panel of Fig. 5, showing good agreement between IASI and the model. The curves have been obtained by averaging the results shown in Fig. 4, where large differences are visible. The results shown in Fig. 5 are likely not very stable. I recommend to add two additional figures analog to Fig. 5 but for latitude intervals shifted by 10 degrees. They would probably show large differences. I don't think that strong conclusions can be drawn from Fig. 5 if good agreement is only present in one latitude band but large (unexplained ?) differences exist if neighbouring latitude bands are used.

Regarding the IASI retrievals outside of the 10°S-10°N band, the high values of N2O above desert regions (and particularly above Sahara) might have been linked to the inappropriate surface emissivity values either used in the retrieval process or actually retrieved by the scientific ground-segment. Indeed, the emissivity for that period was underestimated by IASI in desert regions when compared to the emissivity derived from the space-borne Moderate Resolution Imaging Spectroradiometer (MODIS) instrument (Seemann et al., 2008). Inside the equatorial belt, instabilities in the retrieval process cannot definitively be ruled out (molecule and cloud contaminations, etc.)

Regarding the MOCAGE results, we have again to bear in mind that the horizontal resolution was rather crude (5.6°x5.6°) to reproduce intense convective systems. Indonesia is an active convective region whatever the season considered (warm pool area) but the CTM has some difficulties to reproduce this phenomenon over that particular area. Indeed, for transport evaluation simulations, MOCAGE does produce a maximum of Radon 222 for instance, in the upper troposphere above America and Africa but a minimum above Indonesia, consistently with other CTM outputs (Jacob et al, JGR, 1997; Josse et al., Tellus, 2004). Another source of discrepancy between IASI and MOCAGE might also be linked to the N2O sources used in MOCAGE that could differ significantly from reality, and particularly in the Indonesian sector in 2008. And finally, since averaging kernels are not provided by the ANN, this might also impact on comparisons between measured and modelled total columns of N2O.



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Thus, at this stage of the analysis, considering satellite non validated products and CTM outputs with their own caveats, it is beyond the scope of the paper to assess the quality of the N2O fields outside of the equatorial belt.

We have thus added a new paragraph and three new references.

"Outside of the Equatorial belt (10°S-10°N), some large discrepancies between MOCAGE and IASI N2O fields are depicted (e.g. over Sahara) that might be linked to inappropriate surface emissivity values either used in the retrieval process or actually retrieved by the scientific ground-segment. Indeed, the emissivity for that period was underestimated by IASI in desert regions when compared to MODIS derived emissivity (Seemann et al., 2008). Within the Equatorial belt, the N2O field above Indonesia is more intense in MOCAGE compared to IASI. Indonesia is an active convective region whatever the season considered (warm pool area) but the model has some difficulties to reproduce this phenomenon over that particular area. Indeed, for transport evaluation simulations, MOCAGE does produce a maximum of Radon 222 for instance, in the upper troposphere above America and Africa but a minimum above Indonesia, consistently with other CTM outputs (Jacob et al., 1997; Josse et al., 2004). Another source of discrepancy between IASI and MOCAGE might also be linked to the N2O sources used in MOCAGE that could differ significantly from reality, and particularly in the Indonesian sector in 2008. And finally, since averaging kernels are not provided by the ANN, this might also impact on comparisons between measured and modelled total columns of N2O."

"Jacob, D. J., et al.: Evaluation and intercomparison of global atmospheric transport models using 222Rn and other short-lived tracers, J. Geophys. Res., 102(D5), 5953-5970, 1997."

"Josse, B., Simon P. and V.-H. Peuch, Rn-222 global simulations with the multiscale CTM MOCAGE, Tellus, 56B, 339-356, 2004."

"Seemann, S. W., Borbas, E. E., Knuteson, R. O., Stephenson, G. R., and Huang, H.-



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L.: Development of a Global Infrared Land Surface Emissivity Database for Application to Clear Sky Sounding Retrievals from Multispectral Satellite Radiance Measurements, J. Appl. Meteorol. Clim., 47, 1, 108-123, DOI: 10.1175/2007JAMC1590.1, 2008."

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