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Interactive comment

Interactive comment on "Global distributions of CO₂ volume mixing ratio in the middle and upper atmosphere from MIPAS high resolution spectra" by Á. A. Jurado-Navarro et al.

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Carbone dioxide is the main radiative cooler of the mesosphere and lower thermosphere (MLT), taking energy from the heat reservoir in the course of collisions with atomic oxygen and emitting it to space. Correspondingly, knowing its concentration, trends, and variability is of great importance for the atmospheric physics community. The present manuscript seeks to provide high-quality data retrieved from spectrally resolved radiances measured by MIPAS instrument in 4.3um CO2 band in limb observation mode, but its main purpose is to explain and justify the retrieval methodology and to demonstrate its high accuracy. The latter is stressed several times in the manuscript and this sets the aim high, but it also makes the requirements to quality tougher. In my

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review, I try to explain why I believe that currently presented methodology does not fit these requirements and what should be done to convince the community in robustness of the algorithm and, therefore, quality of the retrieved data.

Major comments

Most points of my concern have already been addressed in the detailed analysis of the first reviewer and they have to be addressed in the revised version of the manuscript. Here, I would like to draw more attention to three issues, which I believe are crucial not only for the atmospheric researchers and potential users of the CO2 product, but also for the ones who are just learning atmospheric retrieval since overlooking these issues in the future might be costly for them and for the community.

For me, the major issue in the suggested retrieval methodology is the regularization approach the authors have chosen. This method is known to give stable results, which correlate well with currently accepted model of this or that parameter. On the other hand, this method is also known as the one, using which one can easily "throw the baby with the bathwater", so it should be applied with great caution and the reasons for using a regularization approach should be explained along with regularization parameters (strength, matrix form, L-curve, residuals). We should not fool ourselves with the results of self-consistency study performed with very mild conditions, which is used in the manuscript as a proof of concept. First of all, as the authors know, the problem is non-local and non-linear, and the perturbation of the atmospheric profile should be performed in a more sophisticated manner than in the single-channel selfconsistency tests. In 2008, two of the authors of the manuscript participated in a joint work dedicated to H2O retrieval in the MLT [Feofilov et al., 2009], where two research and one operational codes were compared, and the robustness of the retrieval scheme was tested. Even in this single-channel retrieval, the self-consistency test was performed for the conditions much harsher than those used in the manuscript, which is supposed to address much more complex scheme. In real atmosphere, the temperature and trace gas profiles in the MLT are perturbed by a mixture of gravity waves

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with amplitudes in temperature reaching 20-30 K. But, in the case of rapidly increasing (or decreasing) volume mixing ratios (VMRs) of trace gases, the perturbations of their vertical distribution should be also taken into account. Last, but not least is the modification of pressure in accordance with hydrostatic law. Correspondingly, a correct setup for the self-consistency test for the retrieval scheme suggested by authors should look as follows (see also two-channel self-consistency test in [Rezac et al., 2015]):

(1) atmospheric profile is taken from the model and is perturbed by some realistic gravity wave above certain altitude (i.e. 80 km). The new profile is pressure-adjusted and the vertical VMR changes are taken into account properly;

(2) This profile is called a "reference" one and is used in forward radiance calculations with the help of non-LTE model described by the authors;

(3) either another perturbation is applied to the initial profile or a smooth profile is taken and is sent to the input of the retrieval algorithm along with simulated radiances obtained at the second step;

(4) retrieved CO2 profile is compared with the one obtained at the first step and relative values of average radiance discrepancy are shown for each height.

The discrepancies obtained in this test will give an idea of the robustness of the method itself and about the errors introduced by applying the regularization scheme. To address the uncertainty of the final product, the same test has to be repeated for the cases, when the atmospheric profile is perturbed below 80 km, down to the ground. At this stage, the uncertainties of surface pressure need to be taken into account.

The second point of my concern is the 90km feature reported in [Rezac, 2015], which is not present in the smooth vertical distributions shown in the manuscript. This requires special attention since SABER overlapped with MIPAS and the aforementioned work considered all possible mechanisms, which could lead to an enhancement of 4.3 um radiance. With the information at hand, one can suggest several explanations for this

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mismatch:

- (a) experimental artifacts in SABER;
- (b) experimental artifacts in MIPAS;
- (c) smoothing of MIPAS radiance profiles;

(d) using large regularization parameter in the MIPAS retrieval.

This is where real physics starts into play and this is what I meant under "baby and bathwater" example above. If we exclude (a) and (b) and assume that modern non-LTE models are not capable of explaining this feature then one has to concentrate on the missing pumping mechanism. If the authors prove that neither (c) nor (d) is responsible for a lack of this feature in their data then one has to understand the fundamental difference between integrated 4.3 um band signal and spectrally resolved radiances of the main isotopologue picked by the authors for the retrieval, taking into account that the main contribution to broadband SABER channel still comes from the main isotopologue. To compare apples to apples, it would be interesting to see the comparison of overlapping SABER vertical profiles of 4.3 um radiance and MIPAS spectra convolved with the SABER instrumental function. For the same reason, it would be also good to have an access to spectral information from MIPAS.

Summarizing this part, I would say that the manuscript addressing CO2 concentration in the MLT and not addressing in details the 90km enhancement is not complete.

The third ideological issue I see in this manuscript is an attempt of retrieving N variables from less than N equations. In the previous work [Jurado-Navarro et al., 2015], an optimized set of rate coefficients was obtained. I call it "optimized" and not the "true" one because the authors themselves write (p8, lines 4-8) that "the retrieved rates are expected to correlate with the errors caused by model parameter uncertainties". In principle, this "error compensation" approach is widely used in this field (the most obvious example is kVT(CO2-O) rate coefficient, which is estimated from atmospheric

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observations and is used to interpret atmospheric observations, but its laboratory value is 2-3 times lower than the "atmospheric" one). The only problem I see here is that kVT estimates have been performed with other profiles fixed, while in the present case we are dealing with simultaneous fitting of the radiance and CO2 profile, so one has to choose, what is known and what is unknown. If both the rates and CO2 profile are treated as unknowns, I doubt that the solution is unique since minimization of radiance discrepancy can have several minima. Moreover, the minimum of a multi-parametric function might be washed out. In the aforementioned joint work, it was just on the edge of detectability with the trace gas profile constrained by occultation measurements, and here we have another degree of freedom. In addition, the set of rate coefficients obtained in tropical region in this approach may be inconsistent with the one obtained in the polar region. In my opinion, the non-LTE rate coefficient retrieval should be based on a same approach as was used in previous works with maximum number of parameters is fixed and for CO2 I would use vertical profiles coming from ACE-FTS colocated with MIPAS since occultation measurements are not affected by non-LTE. In this case, the set of rates would still be an "optimized" one, but at least the solution would be more reliable and there will be more confidence in CO2 retrieved with these rate coefficients.

Minor comments

- The LOS approach needs to be described in more details – in its current form, the main idea is not explained.

- Horizontal inhomogeneities affecting the retrievals are mentioned and the methods of their reducing is suggested, but the explanation of "excluding opaque spectral lines" is not convincing. The tests show that the neighboring layers perturbed by gravity waves can affect tangent point retrievals even in transparent media. A sensitivity study is needed here, which would show the upper estimate of the horizontal inhomogeneity effects on the retrieval uncertainty.

- Neither hot oxygen pumping CO2 nor electron pumping of N2 is discussed. Do the

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authors believe that these mechanisms can be neglected at thermospheric heights?

- Vertical resolution of 5-7 km and up to 10-20 km is stated. At the same time, CO2 falls off rapidly in the MLT and the stated uncertainty of its retrieval is much smaller than CO2 change at 5 km distance not speaking about 20km offset. How do the authors explain this mismatch and what are the real estimates of the retrieval uncertainty associated with vertical resolution?

Reference

Feofilov, A.G., Kutepov, A.A., Pesnell, W.D., Goldberg, R.A., Marshall, B.T., Gordley, L.L., Garcia-Comas, M., Lopez-Puertas, M., Manuilova, R.O., Yankovsky, V.A., Petelina, S.V., and Russell III, J. M.: "Daytime SABER/TIMED observations of water vapor in the mesosphere: retrieval approach and first results", Atmos. Chem. Phys., 9, 8,139-8,158, (2009).

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