Atmos. Chem. Phys. Discuss., 9, C1644–C1656, 2009
www.atmos-chem-phys-discuss.net/9/C1644/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "First multi-year occultation observations of CO_2 in the MLT by ACE satellite: observations and analysis using the extended CMAM" *by* S. R. Beagley et al.

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Received and published: 10 June 2009

General comments

The paper presents observations of CO2 (and CO) in the mesosphere and lower thermosphere (MLT) as measured by ACE (using the solar occultation technique) and analysed with the CMAM model. This represents a new dataset of CO2 measurements in the upper atmosphere which is very important given the scarcity of global CO2 measurements in this atmospheric region. The paper also includes a sensitivity study of CO2 abundances to several model parameters which is very illustrative for understanding the CO2 distribution. The conclusions reached about the processes responsible for

the CO2 distribution are, however, rather speculative and probably the dynamics aspects should be further checked before those speculations are put forward.

The CO2 dataset presented represents a significance advance to our knowledge of the MLT region and therefore worth to be published. However, there are some major points which need to be addressed before the paper can be accepted for publication.

1. The paper does not discuss completely all important work previously done on this subject. Some important references are missing and the discussion of previous work is not fully accurate at some passages.

2. The paper lacks in a thorough and deep Âădescription and discussion of ALL errors affecting the retrieved CO2 field. Sentences as " .. and such error has not been taken account" should not be acceptable and the 2K perturbation temperature error does not seem to be realistic. This is very important since when reaching conclusions about the processes controlling CO2 in the MLT region from data/model comparisons, the errors in the former have to be taken into account and, depending on their magnitude, the conclusions might differ.

3. The retrieval inversion of pressure/temperature and CO2 performed as a joint retrieval in a first step but carried out in a second step retrieving only CO2, poses some problems from the information content point of view. Since the same spectral windows are used for retrieving temperature and CO2, both parameters should be retrieved simultaneously (using an adequate constraint) and I cannot see the point of why a second step retrieval, retrieving only CO2, is necessary.

4. The CO2 distributions shown in Fig. 1 seems to have significant artifacts due to the sampling and averaging (over one month). This fact is mentioned in p. 11562, l. 26 and ff. This point should be included when describing the distributions in this figure, and discuss in more detail how it affects the shown distributions. The fact that such plots have several discontinuities in latitude probably makes this point even more important. For example, is the sharp altitude shift in February at about 15°N caused by

this artifact?

5. I am not fully convinced that the uncertainties in the dynamical processes are not large enough for explaining the data/model discrepancy and that new chemical loss processes for CO2 are needed to be invoked.

6. About the presentation of the paper. I think that it could be significantly shortened. There are many figures which do not add new information and whose discussion make the paper lengthly and repetitive. I do not see any point in presenting the sensitivity study for all cases as zonal mean as well as profiles for several latitudes, and then for different months (April and August), e.g., figures 4, 7, 9 and 10). Also, much of what is said in the "Discussion" section is repeated from the previous section. I suggest to merge the "Results" and "discussion" sections or omit several paragraphs in the "Discussion" section already described before.

7. Including so many panels in each figure make them, at least in the printed version, really unreadable. This technical problem has to be solved, otherwise the features discussed in the text are hardly discernible in the figures.

Specific Comments

In the specific comment below I normally quote the paragraph in question from the manuscript and then comment on it.

INTRODUCTION

P. 11553, l. 16-18. "(Kaufmann et al., 2002). They found that the CO2 volume mixing ratio (VMR) deviated from a well mixed state, which we will call the “knee”, around 70 km."

The fact that CO2 deviates from the well mixed value much lower, at about 80 km, that models have predicted before, was shown much earlier than Kaufmann et al. by the SAMS, ATMOS and ISAMS measurements (See Lopez-Puertas and Taylor, 1989; Rinsland et al., 1992; Lopez-Puertas et al., 1998b; and Zaragoza et al., 2000).

See also the discussion on this topic in Lopez-Puertas et al., 2000, A review of co2 and CO abundances in the middle atmosphere, in Atmospheric Science Across the Stratopause, Amer. Geophys. Union, Geophys. Monograph 123, 83-100, 2000. This paper reviews all CO2 and CO measurements at that time and might be useful for the reader to mention it in the introduction.

P. 11554, l. 10-13. "There is also some evidence that highly vibrationally excited hydroxyl molecules affect the CO2 asymmetric stretch mode (Kumer et al., 1978)."

This has been evidenced more clearly from SABER measurements very recently, showing that the efficiency in transferring vibrational energy from OH to ÂăN2 is about a factor of 3 larger than previously thought, see, e.g., López-Puertas et al., JGR, 109, 09307, 2004, 10.1029/2003JD004383. The authors might want to include this more updated reference.

P. 11554, l. 12-14. "As stated by Kaufmann et al. (2002) the O(1D) excitation mechanism and the non-LTE model parameters constitute the most important uncertainties of retrieved CO."

Edwards et al., JGR, 101, 26577, 1996, 10.1029/96JD02133 were able, from CLAES CO2 10 μm measurements, to give rather small constraints on the excitation of CO2(001) from O(1D).

P. 11554, l. 18-20. "However, for solar occultation measurements the absorption only depends on the CO2 density, the kinetic temperature and the pressure and not on the vibrational excitation of the CO2 molecules".

This assertion is generally true only if fundamental transitions (e.g. those with the ground vibrational state as the lower level of the transition) are involved. For hot transitions, NLTE can be significant and actually this has been used for retrieving non-LTE populations, see, e.g., Rinsland et al., JGR, 1992. Being even more rigorous, non-LTE can be important in occultation measurements even for fundamental transitions, through the vibrational partition function, see, Edwards et al., J. Quant. Spectros. Radiat. Transfer, 59, 423, 1998. This situation does actually take place for ALL CO2 transitions at altitudes above around 100 km in our atmosphere. An extended discussion of this topic is given in Lopez-Puertas and Taylor, Non-LTE Radiative Transfer in the Atmosphere. World Scientific, Singapore/New Jersey/London/Hong Kong, Sec. 8.10, 2001.

P. 11554, l. 25-27. "... and in the Atmospheric Laboratory for Applications and Science (ATLAS) 1, 2 and 3 missions (Kaye and Miller, 1996)."

I have not seen any CO2 measurements retrieved from the ATLAS missions spectra in that reference. Has CO2 been actually retrieved from those missions? To my knowledge no retrieved CO2 have been published from these missions. I suggest to remove that sentence and the reference.

P. 11555, l. 20. "The vertical resolution is âĹij3–4 km" In line 20 (P. 11557) is mentioned that the vertical sampling can be as much as 6 km. Maybe some more details should be given here. Are there particular months for which the vertical resolution is best/worst?

P. 11555-6, l. 22-27 and 1-7. the p/T and CO2 retrieval.

It is not clear for me how the retrieval was performed. I have understood that p/T and CO2 were retrieved jointly in a first step but with a strong regularization on CO2. Then, in a second step, the retrieved p/T from the first step was use to retrieve CO2. In the two steps the same micro-windows were used for p/T and CO2. If this is what has been done, I think the method is not appropriate. You cannot use the result of the first retrieval in a MW as a priori for the second retrieval from the same MW of the same measurement, because then, the a priori and the measurement are no longer statistically independent, and the usual retrieval formalism does no longer hold. Maybe I did not understand correctly. I think this should be described in more detail and/or clarified.

P. 11556, l. 5-15. It would be very useful to list the CO2 transitions used in the retrievals. Does the temperature information comes from the rotational distribution of the lines? Are there enough lines to cover the range from optically thin to moderate for the whole altitude range (50-120 km)? No need to include hot bands? It would be useful to roughly indicate which bands were used for different altitudes.

P. 11556, l. 25-29. "As reported by McLandress et al. (2006), the meridional wind in the CMAM is characterized by summer-to-winter ïňĆow in the mesosphere and winter-to-summer ïňĆow in the lower thermosphere, between 100 and 120 km. The former is a feature of the thermally indirect circulation driven primary by non-orographic gravity wave drag (GWD), whereas the meridional wind reversal in the lower thermosphere is a direct result of the resolved wave drag."

Is this meridional wind patter, with two opposite circulations, below and about 100 km, a generally accepted result of 2D and 3D models or is it present only in the CMAM model?

Could the authors explain in some more details for the non-dynamicist readers the origin of these two circulation branches: What does "thermally indirect" circulation mean? Could they explain the differences between "non-orographic gravity wave drag" and "resolved wave drag"?

P. 11556 (bottom and 11557) "The meridional CO2 distribution for the solstice months appears to be consistent with the large-scale circulation exhibited by the extended Canadian Middle Atmosphere Model (CMAM)"

It is really difficult to get the picture of the circulation predicted by the model from the maps in Fig. 1. First, because they are very small. Second, because of the limited latitude range. Third, because of the presence of the sampling artifacts which which cannot be easily distinguished from actual features. For example, the authors mention, l. 5, "... the January CO2 data for the austral subpolar region appear to indicate the up-welling cell up to about 85 km (âĹij5.10−3hPa) and downwelling cell in the region above," The upwelling cell is clear, although it should be mentioned that it is about 50N (at higher latitudes, ~70N, it is not so clear and confusing, artifact?) but with respect to the descending cell, do the authors mean the only point at 3e-3 mb? It is really hard to see it. The authors should use larger figures, or maybe a zoom of the figures, or draw circles around the features or refer to the latitudes more specifically to show the features more clearly.

It is really hard to see these features at June/July. How does the CO2 values at the top altitudes compare with the errors of the measurements?

The evaluation of the error budget is incomplete and seems rather optimistic. The propagation of the temperature error in the CO2 retrieval, estimated by just shifting the temperature profile by 2K, is underestimated. The abstract of the ACE validation paper by Sica et al, ACP, 2008 state that "There is evidence of a systematic high bias (roughly 3–6 K) in the ACE-FTS temperatures in the mesosphere ..."

Further, I cannot see any scientific justification for estimating the CO2 error as the differences between the CO2 retrieved with a strong regularization and the CO2 derived including the temperature from the first step (If I did understand right). I suggest that only one p/T CO2 joint retrieval should be done and the errors (noise) will come up from the retrieval. If it is used a retrieval grid different from the measurements grid, then some kind of regularization should be applied which will impact the noise error and the vertical resolution (averaging kernel).

P. 11557, l. 23-24 "Above the mesopause, temperature changes rapidly with altitude, and a temperature uncertainty of 2 K is possibly an underestimate."

According to the ACE temperature validation paper, the temperature is underestimated at the mesopause and below. The temperature mapping in CO2 should be revised and quantified at all altitudes where the CO2 is presented. Spectroscopic errors and other systematic instrumental and forward model errors should also be quantified.

It is not clear which is the noise error of a single CO2 profile. This should be given and then an estimate of the values shown in the zonal mean, when some averaging are performed. I suggest to include a table listing the errors from the different sources, random and systematic at several altitudes covering the CO2 range.

As new processes occurring in the atmosphere are proposed, this requires a good estimate of the CO2 error to be considered Âăwhen comparing with models results.

P. 11559, l. 17-19 and ff. Comparison of CO2 is done for a particular set of CO2 profiles, e.g. CRISTA, SABER and rockets. Is there any rationale of why selecting those measurements? There are other measurements, e.g., SAMS, ISAMS and ATMOS, see e.g. Lopez-Puertas et al., 2000 cited above. Why have not them been included? About the rocket profile, this differs from that compiled by Wintersteiner et al. (see Wintersteiner et al. 1992, and Lopez-Puertas et al, 2000). It might be useful for the reader to know about distinct those rocket profiles are. Also, the CO2 retrieval from SABER, version 1.06, is very preliminary and was not validated. This should also be stated in the manuscript to warn the reader.

P. 11560, l. 1-10. Discussion about the altitude of the knee.

It might be useful to have in mind the errors in CO2 when comparing among different measurements. E.g. SABER being about 20 ppmv larger than ACE above the knee. This is 6%, which is within ACE errors (and probably SABER errors too). The rocket profile compiled by Wintersteiner et al. (1992) has the "knee" around 90 km, higher than the rocket profile shown here.

P. 11560, l. 11-13. "ACE measures the ground state of CO2 and, hence, provides more reliable information on the CO2 abundance."

I'm not in favor of this kind of sentences. It is true that ACE CO2 is less affected by non-LTE processes but the "goodness" of the CO2 profile should be judged on the overall error budget. I suggest that the authors state the errors and, if they want to compare, mention that is smaller/larger(?) than in other measurements.

P. 11560, l. 15. "Since CMAM is a climate model we cannot compare with the same dates .." You can, but it does not make sense.

P. 11560, l. 18-20. "The ACE-FTS gives a reasonable latitudinal coverage in April and from Fig. 4 we see that the overall structure exhibited by the model for the standard scenario, A, is similar to that of the observations. However, the measurements appear to have more structure with latitude."

It is mentioned (see above) that because of averaging over one month, the data includes some artifacts, shown as latitudinal structure. How much of the model/data discrepancy is due to these artifacts? This should be commented. Also, the best way to see if the model reproduce the measurements is to make a difference plot. I suggest that, instead of showing so many model zonal means for different conditions in Fig. 4, they authors include only the measurement plot, the model plot (whatever run with probably best fit), and a difference plot. The behavior of the different model runs are better shown along with profiles (as in Figs. 5). Having less panels might also help in showing larger figures.

Also, probably it would be best for showing any agreement/disagreement between data/model to compare for a solstice month (not April which is near equinox and exhibit less latitudinal gradients).

Stating that model and data give "SIMILAR" results is somehow in contradiction with the line 23 below where it is discussed about the disagreement between both. This is confusing and needs some rewriting.

P. 11560, l. 21 "fall-oïňĂ of CO2 mixing ratio with height for the ACE measurements is clearly seen to occur at lower altitudes than for the model results in the control run, scenario A."

This difference is seen more clearly in profile plots (not in zonal means). Certainly this

is NOT seen CLEARLY in this figure.

P. 11561, l. 15 "... transporting CO2 up the vertical gradient." ... transporting CO2 upwards.?

Figure 5. I suggest to remove the panel for 3°N. It does not add any new information to that for 30°N. This would help in enlarging the figure and reducing redundant text, e.g., (p. 11562, l. 4-6). Also, I suggest to include in the measured profile the CO2 error bars (and remove Fig. 2).

P. 11562, l. 8. The role of Kzz on the CO2 and CO distributions was already studied by Lopez-Puertas et al. (2000). A reference here might be useful for the reader.

P. 11562, l. 9 and ff. I would write these sentences in a easier way. Replace: "Viewed as an experiment on the role of GWD this suggests ... mid-latitude regions" by: "The reason of CO2 being diffused upwards to higher altitudes in the polar regions is that the impact of turbulence generated by unresolved gravity wave breaking in the model mixing is most important in this region than in the tropics and mid-latitude regions (see Fig. 6)."

P. 11562, l. 19. "... that the CMAM Âăsimulation of CO provides a reasonable representation of CO."

- At 30-40 N there is a significantly larger descent in the data than in the model - The increase at 90°N is not in the model. Are these features within 30% errors?

P. 11562, l. 27-28. "... descent over the pole. However, this is an artifact due to the sampling limitations of the ACE experiment during a time of strong descent."

Are the features above also caused by these artifacts? This should be discussed earlier in the description of the data and the description of figures 1, 4 and 7.

P. 11563, l. 5. Insert a "," after "turned off"

P. 11563, l. 5-8 The CO obtained for case C for 30°N and 80°S (Figs. 8) seems in rather good agreement with measurements; except for 80°S below 65 km, but in this case there is no other better model simulation.

P. 11563, l. 18. I suggest to remove also the panel for 3°N in Fig. 8 and the corresponding discussion (which is very similar than for panel 8a) in the text.

Could not be both CO2 and CO measurements reasonably explained using a low Kzz? It is true that CO2 is overestimated but it might be within the measured CO2 error bars. If so, there it will be no need to invoke any new mechanism for sequestering CO2.

Fig. 7. As for Fig. 5, I think there is no need to show zonal mean for all simulations. The differences of the sensitivity study are seen more clearly in profiles figures, as Fig. 8. The same applies to Figs. 9 and 10, which are similar to figures 5 and 7 except for August, instead of April.

P. 11564, l. 18-20. "We note that the slopes of the ACE contours in the polar region are affected by sampling as for April as can also Âăbe seen in Jin et al. (2008)."

This should be discussed much earlier in the paper (see comments above).

P. 11565, l. 11-24. Lopez-Puertas et al. (2000) were able to reproduce such a low "knee" in the CO2 profiles by using reasonable Kzz. The problem with overestimation of CO2 above about 80 km in the previous version of the 2D Garcia and Solomon model was that molecular diffusion was not properly taken into account. Probably this was the same reason for TIME-GCM not being able to reproduce such CO2 profiles. In any case, the low CO2 profiles of ATMOS, ISAMS, and SABER (similar to that of ACE) can be well reproduced, with no overestimation above around 75-80 km by the Whole Atmosphere Community Climate model (WACCM) (R. Garcia and D. Marsh, personal communication).

Part of the discussion in this section has already been described in the previous section, when describing the sensitivity study. I suggest to shorten this section in that sense.

P. 11567, l. 14-17 "However, there is some justification for higher temperatures being used. This is because the vibrational levels of CO2 are non-thermally excited in the mesosphere so that daytime vibrational temperatures are higher than the kinetic temperature (e.g., Lopez-Puertas and Taylor, 2001)".

The CO2 15 μm levels, mainly the CO2(010), is very close to LTE up to about 100 km. Furthermore, the number of CO2 molecules in this level (and in higher levels) represents only about 1% of the total number of CO2 molecules, most of which are in the ground state. I suggest to remove that paragraph. It does make sense to consider the temperature as low at 200 K in that region.

P. 11568. l.9. "In the control model simulation, scenario A, CMAM CO is up to a factor of two too low above 0.01 hPa (80 km) but it is not clear how serious a disagreement this is."

I do not understand this sentence. Is it not clear that a factor of 2 is a large disagreement? (Unless the measurements have a similar error).

P. 11568. As before, there is quite a lot of repetition from previous section.

P. 11568, l.25 "We note that the agreement between model and measurements is improved somewhat but that the model CO2 still remains too high. "

How high in comparison with ACE CO2 error?

P. 11569, l.7-9 "As is clear from above results the most reasonable scenario for agreement between the ACE observations and CMAM simulations is with an increased J value ..."

This is questionable. One could argue that this gives a very large overestimation of CO (see Figs. 8 a and c).

P. 11569, l. 19-22 "An interesting feature of such a phenomenon is that it will be sporadic, and its effect and its effects will vary from season to season with varying dust amounts which might account for the variation required in the “enhanced” J (CO_2) to account for the observations in April and August."

If sporadic, how can it account for a global and constant CO_2 depletion? Is there any hint that the dust amounts changed from April to August and in the correct sense? Wouldn’t we expect a larger meteoritic ablation in August? Ins this is the right sense?

P. 11569, l. 23-27 and Fig. 11 I can’t see the need for including the figures for temperatures. Possible temperature differences are not invoked to explain the CO_2/CO measurements/model differences or any comparison is made among modeled and measured temperatures. I suggest to remove this paragraph and Fig. 11.

The error bars of Fig. 2 are already shown in Fig. 3. I suggest to remove Fig. 2.

Table 1, scenario B. Insert a "," after "factor of 5"

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 11551, 2009.