

Interactive comment on “First multi-year occultation observations of CO₂ in the MLT by ACE satellite: observations and analysis using the extended CMAM” by S. R. Beagley et al.

S. R. Beagley et al.

beagley@nimbus.yorku.ca

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We wish to thank the reviewer(s) for their comments and have tried below to summarize the questions and answer them as shown in the following text.

General Comments:

Point 1

QU: 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.

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ANS: A number of the references reported by the reviewer have been added and are discussed in the paper. We were certainly remiss in leaving out some of these papers which have provided valuable additional information for the paper.

Point 2

QU: The paper lacks in thorough and deep description and discussion of all errors affecting the retrieved CO₂.

ANS: A new subsection has been added to the paper (section 2.3) which is dedicated to the discussion of the errors in the retrieval method and a new table of error estimate breakdowns are provided (Table 3).

Point 3

QU: Why the two step retrieval inversion is was performed?

ANS: The ACE observations section has been totally rewritten and considerably changed to describe the retrieval approach in far more detail as requested. See section 2.2 of the revised paper.

Point 4

QU: Significant artifacts due to sampling and averaging. More comments need to be made and how the sampling affects the shown distributions.

ANS: The text has been altered to describe earlier the issue of sampling impacts and the potential for artifacts when examining an comparing the 2D cross-sectional data of ACE with the CMAM. See also the answer to the specific question 22 below which addresses this aspect of the paper. February is not analysed in terms of the main focus of the paper which concentrate on April and August where the mid and low latitude data has the best coverage and resolution and thus minimizes the problems of sampling.

QU: Is the sharp altitude shift in February at about 15N caused by this artifact?

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ANS: We haven't tried to explain the CO2 behaviour near 15N in February, However, one may notice that this CO2 variation is within the ACE observational error bars presented in Figures 2 and 4.

Point 5

QU: Uncertainties in the dynamical processes may be responsible for data/model discrepancy.

ANS: We have explored parameter space including parameterized dynamical effects and found no signal that dynamical effects are the cause of the data/model discrepancy. Further dynamical variations can be explored by varying the model Brewer-Dobson overturning which may influence the upward flux of CO2 into the MLT, however this contradicts the results found in earlier papers which indicate that for other tracers such as methane and nitrous oxide that a stronger B-D overturning was essential for good simulation of the ACE observations, see Jin et al (2009). We found that uncertainties in parameterized turbulent flux of tracers alone cannot explain the discrepancy between the observed and modelled CO2 fields in a 3D GCM. It is important when considering the role of dynamics to look at any species and the impacts of changes rather than one or two species. The paper has been modified to reflect this statement and to expand on this issue by adding the following text:

"Given the uncertainty in our knowledge of the effects of diffusion generated by gravity wave breaking, a major contributor to the KZZ in this region, some concern over the role and strength of gravity wave induced motion is warranted. However, even if we neglect all diffusive transport associated with unresolved gravity wave breaking, the CO2 vertical profile does not begin to fall off in the model as low as the ACE observations indicate. Nevertheless we note that for other species in the MLT region, such as H2O, N2O, CH4, as well as CO2, for which their distributions are determined by vertical transport balanced by chemical loss, that knowledge of KZZ is important in the determination of their distributions (see also Jin et al., 2009). "

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Point 6

QU: The paper could be significantly shortened.

ANS: The text has been revised and where possible the redundant text has been removed, however the new material requested has increased the paper size too. Also in terms of figures we have, removed old figure 2, changed several, reduced the number of panels in some (new figures 8 and 9), and made panels bigger (figure 1) where feasible to increase the readability of the data presented.

Point 7

QU: Unreadable panels due to number of elements.

ANS: A number of figures have been altered in particular figure 1 to make the panels larger, however we have kept most of the figures since we feel they are important to the paper (also, see Point 6).

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Specific Comments: (in reference to page and line number queried).

1. P11553,I. 16-18:

QU: Lower knee shown much earlier than Kaufmann et al. (2002).

ANS: The Kaufman et al. paper was not the first to comment on the existence of the lower 'knee', but the data quoted and used (SAMS, ATMOS and ISAMS) in the earlier measurements were not global nor multi-year in extent and so were limited in its usefulness to define the issue. The CRISTA data provides therefore a more substantive measure of the issue. The paper has been modified to reflect the papers indicated by the reviewer and to indicate this is an expansion of the issue already worked on and reported in the literature for sometime. We now state in Introduction "The fact that CO2 starts to deviate from the well-mixed state lower in altitude than models predict has

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been reported by various researchers, and summarized in a review (Lopez-Puertas et al., 2000)."

2. P11554,I. 10-13:

QU: More clearly evidence that vibrationally excited hydroxyl affects the CO₂ asymmetric stretch mode is given in Lopez-Puertas et al. (2004). ANS: We thank the reviewer and has included this reference in the text.

3. P11554,I. 12-14: QU: Excitation of CO₂(001) from O(1D)

ANS: We discuss Kaufmann et al. (2002) (not Edwards et al., 1996) results here. In Kaufmann et al. the authors stated that the O(1D) excitation mechanism (along with the non-LTE parameters) constitutes the most important uncertainties of the CRISTA retrieval. The fact that Edwards et al. (1996) gave rather small constrains on this excitation mechanism is, in fact, an additional indication on how uncertain the non-LTE retrieval is. We thank the referee for the reference and has mentioned this contradiction between Kaufmann and Edwards results in the paper as follows: "The complexity of the non-LTE retrieval is illustrated by the fact that Kaufman et al. (2002) stated that along with the non-LTE model parameters, the O(1D) excitation mechanism constitutes the most important uncertainties of retrieved CO₂, whereas Edwards et al. (1996) gave rather small constrains on this excitation mechanism."

4. P11554,I. 18-20:

QU: NLTE is less important only if absorption from the ground state is considered. However, even in this case non-LTE can come through the vibrational partition function.

ANS: We agree with the referee that this assertion is true only if fundamental transitions are involved and that even in this case NLTE can be important above ~100 km through the vibrational partition function. However, as stated in Section 2, all microwindows used for the retrieval above 65 km contain spectral lines from transitions originating in the ground vibrational state. Also, since we are mainly concerned with altitudes be-

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low 100km, we didn't mentioned about the effect of the vibrational partition function. In a new subsection 2.3, the NLTE effect due to changes in the vibrational partition function is estimated. In the revised paper we clarified that we mean the absorption from the ground vibrational state and refer to subsection 2.3 for NLTE effect through the vibrational partition function. The text has been modified as follows: "However, for solar occultation measurements, when transitions from the ground vibrational state is considered, the absorption only depends on the CO₂ density, the kinetic temperature and the pressure and not on the vibrational excitation of the CO₂ molecule. Although some non-LTE effect in solar occultation measurement can occur through the vibrational partition function (Edwards et al., 1998), this effect is very small below about 100 km (errors related to the non-LTE vibrational partition function are discussed in Section 2.3)."

5. P11554,I. 25-27:

QU: The referee states he knows of no CO₂ measurements retrieved from the ATLAS mission and suggests to remove the statement.

ANS: There are some contradictions in the referee's comments here. In his comment on P11559,I. 17-19, the referee refers to the ATMOS measurements himself. Moreover, in the paper we refer to (Kaye and Miller, 1996) written in this comment? In Table 2 of the paper we refer to (Kaye and Miller, 1996), the ATLAS-3/ATMOS mission included CO₂ measured from 10-120km.

6. P11555,I. 20.

QU: Are there particular months for which the vertical resolution is bad.

ANS: The satellite observations produce a cyclic pattern in the resolution resulting from the orbit. The same patterns in latitude and beta angle repeat every year for the satellite. The worst spacing/resolution occurs for mid January, March, May, July, September and November and the best in February, April, June, August, October and December.

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The resolution varies from 2km at best to 6km at worst, effectively alternating in resolution every other month from best to worst. Hence in part our choice of the April and August data for analysis and discussion in the paper, when the resolution is ~2km. This point is clarified in the paper as follows: "The vertical resolution is generally ~3-4km, but varies (from 2km to 6km) with month, alternating from best to worst every other month and is best during both months analysed and shown in this paper, April and August."

7. P11555-6,I. 22-27 and 1-7:

a) QU: It is not clear how the retrieval was performed.

ANS: Section 2.1 has been written and a fuller retrieval description provided. see sections 2.1 and 2.2 in new text version.

b) QU: 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 used 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.

ANS: First, it should be pointed out that our retrievals do not employ optimal estimation. No a priori information is used as a constraint in the retrievals. However, we agree that the two sets of results (CO₂ VMR from the P/T retrievals and CO₂ VMR from the straight VMR retrievals) will not be statistically independent. If you average CO₂ VMR over several occultations, you expect essentially identical results. We would be quite sad if that were not the case, because that would mean something was wrong with the retrieval process. CO₂ VMR was not reported in version 2.2. The output CO₂ VMR from P/T retrieval was stored in a diagnostic file, but the result in that file did not take

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into account a small (typically the order of a couple of hundred meters) adjustment to the tangent heights in an altitude registration step at the end of the P/T retrieval process. To use these data, we would have needed to extract the data from the files and then do something (probably use interpolation, because we did not store the empirical parameters associated with CO₂ VMR in the P/T retrieval) to account for the small altitude shift from the altitude registration process. Alternatively, we had the option of using straight VMR retrievals to "reconstruct" the VMR profiles. This is what we did. The residuals from the straight VMR retrievals are smaller (thanks to the absence of the smoothing induced by the empirical function in the P/T retrievals), making results from the straight VMR retrievals more representative of the measurements, as is stated in the text. There is no scientific or philosophical problem with using these data. We do not imagine that we magically generate statistically independent information with the second retrieval step. That is not what we are suggesting.

8. P11556,I. 5-15:

a) QU: List the CO₂ transitions used in the retrievals.

ANS: A new table (Table 2) has been added to the paper.

b) QU: Does the temperature information come from the rotational distribution of the lines?

ANS: Yes, the absolute intensities of the lines provide information on CO₂ VMR, while temperature information is derived from the relative intensities of the various CO₂ lines with different lower state energies (i.e., different rotational lines have different lower state energies, and hence different population factors). This is clarified in the text as follows: "At high altitudes, CO₂ VMR and temperature are retrieved simultaneously. The relative transmittance strengths of lines with different lower state energies provide information on temperature, while the absolute values of the transmittances determine CO₂ VMR."

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9. P11556,I. 25-29:

a) QU: Is the indirect cell an accepted flow feature?

ANS: The meridional wind pattern seen in the extended CMAM with a thermally indirect cell (page 4, paragraph #21) lying above the direct Brewer-Dobson cell below 100km and then replaced again higher up by a further direct cell is a generally accepted and expected flow structure and this a basic response to GWD, all other extended models must/should get it. For example, Richter et al. (JGR, 2008) which discusses WACCM, shows in Figures 3 and 7 the zonal wind and the meridional advection of zonal momentum from which can be deduced that the WACCM model exhibits a thermally indirect cell at around 100km. The meridional flow is not shown explicitly in Richter et al., but using the U and U-V fields provided the reversed thermally indirect cell can be found, which sits between the middle atmospheric B-D cell below and the direct cell in the thermosphere. All higher lid GCMs should simulate these opposite circulation flow patterns if they model GWD correctly. Since this feature is well established and the reference to the extended CMAM is given, we do not discuss this point in detail in the current paper.

WACCM reference:- Richter J. H., F. Sassi, R. R. Garcia, K. Matthes, C. A. Fischer (2008), Dynamics of the middle atmosphere as simulated by the Whole Atmosphere Community Climate Model, version 3 (WACCM3), J. Geophys. Res., 113, D08101, doi:10.1029/2007JD009269.

b) QU: Explain what does a thermally indirect cell mean.

ANS: A thermally indirect circulation is a meridional circulation which moves heat, with warm air sinking and cold air rising, so as to be in the opposite sense of a normal thermally direct flow, like the tropospheric Hadley cell where heating gives rises to ascent and cool air descends. This thermally indirect cell is driven by the non-orographic gravity wave drag which occurs from gravity wave breaking in this region. The term indirect or thermally indirect is a term that refers to a circulation that has ascending motion in a

C5598

region of relatively low temperature and descending motion in a region of relatively high temperature. It therefore is sustained by dynamical processes rather than by thermal processes. Since this terminology is widely accepted and the reference is given, we do not discuss this point in detail in the current paper.

c) QU: Explain the difference between non-orographic GWD and resolved wave drag.

ANS: Non-orographic GWD is gravity wave drag generated by non-orographic sources such a convection, and ageostrophic motions. For the most part this form of drag has to be parameterized as the model cannot resolve the short wavelengths of these waves. Resolved wave drag is that portion of the wave spectra which the model can resolve and is principally the planetary waves forcing generated by the model. We have not modified the text as these facts are basic dynamical definitions and since the reference is given.

10. P11556 (bottom and 11557):

a) QU: It is difficult to picture the circulation from the figures. The figures are too small.

ANS: Figure 1 has been enlarged by putting 6 months on each page, splitting the figure into 2 parts.

b) QU: Limited latitude range.

ANS: The latitude range of the observations for a particular month is clearly a result of the satellite orbit character and observational capabilities. The two months analysed represent the 'best' data coverage and reliability as discussed in the paper.

c) QU: Sampling artifacts

ANS: The question of artifact features in the ACE data being mis-interpreted due due sampling etc., has been commented on in several questions. A full combined answer will be given below in answer to several questions which ask about the artifacts generated by the ACE sampling and averaging of the data. See reply to question 22 below.

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Figure 1 has been split into 2 and hence each panel is now larger to assist in allowing the reader to interpret the data.

d) QU: The error budget is incomplete. What of the temperature high bias reported in Sica et al.?

ANS: The error budget has been considerably expanded and a description of the reasoning for the various error estimates given including the temperature value used (see new subsection 2.3).

ANS: The error estimates now use temperature uncertainties consistent with the version 2.2 validation paper (2 K below 60 km, 5 K above 60 km). So in the mesosphere the error budget uses a 5K error estimate and this detail has been described in the text (new subsection 2.3).

e) QU: How well does the CO₂ values at the top altitudes compare with the errors of the measurements.

ANS: Figure 5 (Fig. 4 in the revised version) now contains the error bars and the errors and ACE measurements can thus be compared. The error bars cannot account for the CO₂ fall off.

f) QU: 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 difference from the measurements grid, then some kind of regularization should be applied which will impact the noise error and the vertical resolution (averaging kernel).

ANS: The quantity in question is the difference between the CO₂ VMR generated during P/T retrievals and the output from a straight VMR retrieval for CO₂. These differences arise in part from different approaches used to generate CO₂ VMR on the

C5600

1-km grid used in forward model calculations (an empirical function in P/T retrievals compared to piecewise quadratic interpolation from the "measurement grid" in VMR retrievals). You might also expect more variability in the profile from the VMR retrieval due to more degrees of freedom compared to the profile generated during the P/T retrieval (from noise effects). Note, however, that noise effects on the CO₂ profile determined by the straight VMR retrieval should be reduced by using the exact same set of microwindows (and therefore having the same associated spectral noise) for both retrievals. With CO₂ VMR smoothed in the P/T retrievals, the noise should induce errors in the T and P profiles instead. Ideally, with the noise errors transferred into the P and T profiles, this should allow the straight VMR retrieval to reproduce the smooth CO₂ profile from the P/T retrieval (again, when you have the same microwindows and associated noise). Its inability to do so is a measure of the limitations on internal consistency for the retrieval approach. These differences are random in nature (the differences are reduced through averaging profiles), so there is likely some influence from the noise (e.g., whether the piecewise quadratic interpolation gives higher or lower CO₂ than the corresponding values from the P/T retrievals between two measurements). However, it is mostly a smoothing/interpolation effect: the differences get larger as you move farther away in altitude from a measurement (i.e., is small near a measurement tangent height and peaks about halfway between two measurements). That being said, in the original version of the paper, the combined contributions for all systematic errors were estimated by looking at the effects in the straight CO₂ VMR retrievals when you introduced an assumed error in temperature. At the request of the reviewers, the current version of the paper estimates systematic errors in the CO₂ VMR generated during the P/T retrieval, from the "bottom up", considering each potential source of error separately (see new subsection 2.3). The difference between the P/T CO₂ VMR profile and that from the straight VMR retrieval does not factor into the discussion of errors in the P/T retrievals. Also, the quantity is small compared to the random errors and reduces in magnitude when averaging results from different occultations. Thus, we have removed this factor from the discussion in the paper.

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11. P11557,I. 23-24:

a) QU: If T is underestimated the T mapping in CO2 needs to be revised.

ANS: The retrievals reported here employ a preliminary version of the "next generation" P/T retrievals that are expected to improve on version 2.2 results. Hence the use of smaller temperature errors than were reported in the version 2.2 validation paper. However, because the new temperature results have not been officially validated, the error estimates now use temperature uncertainties consistent with the version 2.2 validation paper (2 K below 60 km, 5 K above 60 km). See new subsection 2.3.

b) QU: Quantification of ACE errors should be made.

ANS: We have extended the error discussion in the paper creating a new subsection 2.3 in the paper. See also Table 3 in the revised paper.

c) QU: A new process requires a good estimate of error to be made.

ANS: Yes good error assessment is required but since all observations except the rocket data seem to show the bias then either all are wrong or we have another mechanism acting on CO2. It seems since all observations considered in L-P et al (2000) and in this paper are lower than the model results that we are dealing with a bias rather than a random error.

12. P11559, I. 17-19 and ff:

a) QU: Why pick these measurement sets?

ANS: We choose data (except for the rocket data) which are almost global compared to the more limited earlier data. This point is now clarified in the text: "The rationale for choosing the CRISTA and SABER in the comparison is that alike the ACE both these datasets provide nearly global coverage. The rocket CO2 profile shown here is that used in the non-interactive model simulations."

b) QU: Why not include the SAMS,ISAMS and ATMOS.

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ANS: Firstly we are targeting an assessment of ACE, secondly we wished to concentrate on multi-year, global datasets of the same vintage (hence the use of SABER with the ACE).

c) QU: Why this rocket profile set used?

ANS: This rocket compilation set is used as it is that used in the non-interactive model radiation and is the climatology used in the Fomichev's radiation scheme used by various models around the world. We wished to compare the assumed climatology responsible for the background state and the modelled CO2 fields. We have modified the text to indicate our reasoning for use of the CRISTA, SABER and a given rocket datasets (see comment 12a above).

d) QU: SABER data v1.06 is very preliminary and is un-validated.

ANS: Yes SABER v1.06 is a preliminary dataset but these are a simultaneous modern dataset and hence add value to the ACE dataset usage by being consistent. No dataset has truly been validated except by analysis of simultaneous independent observation sets. We have amended the text to indicate to the reader the preliminary nature of the SABER data: "However it should be mentioned that the CO2 from SABER version 1.06 is very preliminary and not yet validated."

13. P11560,I. 1-10:

QU: Discussion about the altitude of the knee... and errors.

ANS: We have amended the text so as to have the reader keep in mind the error:"The various observations suggest an uncertainty in the location of the 'knee'."

14. P11560,I. 11-13:

QU: The referee dislikes the sentence.

ANS: The sentence has been modified to clarify its meaning:"ACE measures the ground state of CO2 and, hence fewer physical assumptions are required to obtain

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the CO2 abundance. The method is more straightforward and the physics involved simpler."

15. P11560,I. 15:

QU: The referee dislikes the sentence.

ANS: The sentence has been reworded:"Since CMAM is a climate model so does not produce a forecast and comparison with specific dates is inappropriate..."

16. P11560,I.18-20:

a) QU: How much of the model/observational discrepancy is due to artifacts.

ANS: The main area where artifacts will impact the observational data is in high latitudes. The main feature of the vertical profile which is dominated by the tropical mass of the atmosphere we feel will not be strongly influenced by the observational sampling issues. The manuscript has been altered and in section 2.2 where this issue is discussed.

b) QU: Use less zonal mean plots.

ANS: We do not agree with dropping the zonal data as they provide additional information. No difference plots are used as the plots show the various parameter space variations and the 'best fit' plot is not the "right answer for the right reason". Only by showing the different impacts can the analysis be meaningful.

c) QU: Have fewer panels and thus bigger plots.

ANS: The discussion paper is online and all figures can be enlarged by the viewer, however we have in some plots for the August analysis, (Figures 8 and 9), reduced the number of panels and hence the textual discussion too, where such a reduction does not detract from the analysis. Figure 1 was also enlarged to also aid in making the data in it more readable.

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d) QU: Why not use solstice data.

ANS: April and August were shown as they represent the months with best global coverage (as can be seen in Figure 1 which shows the data coverage for all months) from the ACE instrument. June-July and December-January (solstice) data do not have as good a coverage and so are more problematic to analyse.

e) QU: Stating the data and model are similar is in some contradiction with the text below.

ANS: We stated the data and model results were 'similar' not identical, as in "some common features, related in appearance or nature; alike though not identical."

17. P11560,I. 21:

QU: The referee claims the statement is not obvious in this plot.

ANS: The sentence has been reworded to direct the reader to the profile plots which more clearly show the issue in question:"... ACE measurements is seen to occur at lower altitudes (see also Figure 4) than for the model..."

18. P11561,I. 15:

a) QU: "...transporting CO2 up the vertical gradient"... transporting CO2 upwards?

ANS: The sentence has been rewritten to correct its meaning: "... impact, transporting CO2 upwards."

b) QU: Remove 3N panel

ANS: We agree that 3N and 30N are similar but we would argue that they characterize different atmospheric conditions and so the fact that they give the same conclusion broadens the results. Note also that 3N represents the major mass of the atmosphere and so is most representative of the profiles of the total atmosphere. So we wish to retain the panels.

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c) QU: Modify redundant text.

ANS: The text has been slightly modified but since the 3N and 30N represent different portions of the atmosphere we consider the text are not redundant.

d) QU: Add error bars to Figure 5.

ANS: The error bars are now shown in Figure 5 (Figure 4 in the revised version).

19. P11562,I.8:

QU: A reference to Lopez-Puertas et al, (2000) might be useful for the reader.

ANS: this and recognize the valuable addition this reference makes to the paper. The reference to Lopez-Puertas et al, (2000) has been added and the text changed to discuss their results: "Lopez-Puertas et al (2000) show in their Figure 7 that a reduced Kzz can using a 1D global model make a big difference in the CO₂/CO profiles lowering the 'knee' and altering the CO₂ profile significantly above 100km. However the lower Kzz results were not directly compared to observations or applied to a 3D GCM in their paper. Our results do not show as large a sensitivity to turbulent parameterized diffusion when undertaken within a full interactive 3D GCM suggesting that resolved waves and transport may be part of the extra transport seen in CMAM and represented as Kzz in the 1D tests from Lopez-Puertas et al (2000)."

20. P11562,I.9:

QU: Sentence rewrite.

ANS: We agree with the referee and have adjusted the text to be more readable: "The reason for CO₂ being diffused upwards to higher altitudes in the polar regions is that the impact of turbulent mixing generated by unresolved gravity wave breaking in the model is most important in this region compared to that in the tropics and mid-latitude regions (Figure 5)."

21. P11562,I. 19:

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QU: The referee claims our "reasonable representation" statement is incorrect due to features not modelled.

ANS: We have changed the text to indicate that the model produces reasonable representation of CO outside of regions of strong descent. As indicated in the text this is a climate state model and cannot reproduce specific year to year variations in descent and so will fail to reproduce such transient features. In this case the difference is not within 30% but we indicated in the discussion that it only generally was. The South polar ACE structure seems to imply ascent or weak descent but this is possibly affected by the ACE sampling issues at high latitudes (see also our reply re aliasing in reply to question 22). The text has been altered to discuss this: "... the CMAM simulation of CO provides a generally reasonable representation of CO (outside regions of strong high/mid latitude descent)."

22. P11562,I. 27-28:

QU: Are the features above also caused by these artifacts?

ANS: One issue which needs to be pointed out is that specific features in the CO₂ and CO datasets may not be accurately compared between the ACE and CMAM results. This is because at high latitudes the ACE experiment is subject to a sampling bias since data is less frequently observed and so strong shorter lived features may be mis-represented when the data is averaged into monthly bins. However the main analysis discusses the mid and low latitudes where this effect is not as prevalent and the observations are multi-year averages so as to reduce this problem. Though this means specific year gradients will be "smeared" and not well reproduced, but the climatological structure should be reasonably represented. Thus discussion of specific and transient features is not possible but the overall climatological can be assessed. So some of the specific feature discussion in the text has thus been removed or indicated as qualitative in their conclusions. The following discussion has been added: "... at high latitudes the ACE experiment is subject to a sampling bias since data is

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less frequently observed and so strong shorter lived features may be mis-represented when the data is averaged into monthly bins. However the main analysis discusses the mid and low latitudes where this effect is not as prevalent and the observations are multi-year averages so as to reduce this problem. Though this means specific year gradients will be "smeared" and not well reproduced, but the climatological structure should be reasonably represented. Thus discussion of specific and transient features will be more qualitative whereas the overall climatology can be better assessed."

23. P11563,I.5:

QU: Correction (minor) - insert comma after "turned off"

ANS: Done.

24. P11563,I. 5-8:

QU: CO obtained for case C for 30N seems in rather good agreement with measurements,

ANS: It looks like a comment not a question. However it seems to say that the $K_{zz}=0$ case is the best for CO and thus may well be the best scenario. The referee is correct that in Figure 8 (Fig 7 now) and hence in general a reduced K_{zz} improves the CO profile and for most of the atmosphere it appears to be the best scenario in simulating CO. However the impact of low K_{zz} on CO₂ is not enough and these two species are indelibly tied together in this altitude band so we are maybe getting the CO right for the wrong reasons as the CO₂ remains too high (and thus the local CO source is consequently too high). The text has been modified as follows: "... This highlights issues already brought out in Lopez-Puertas et al. (2000) and re-iterated in this paper that the uncertainties in the K_{zz} value needed in the MLT are important and our understanding of their temporal, vertical and latitudinal variations is lacking in order to correctly identify the role of the turbulence in the motion of species in this region."

25. P11563,I. 18:

C5608

a) QU: Remove 3N panel and modify text.

ANS: We use 3N as this represents a profile typical of the principal mass of the atmosphere. Thus we do not wish to remove these figures. The text discussion has been reduced.

b) QU: The referee claims CO₂ within error bars for this case if K_{zz} low is used.

ANS: As now seen in Figure 5 (Figure 4 in the revised paper) the $K_{zz}=0$ experiment does not fall within the ACE error bars above ~85km and even below 85km is on the high side at all levels indicating the discrepancy remains and low K_{zz} (which represents unresolved turbulent mixing) cannot in our model explain the observed CO₂ and CO fields. Also as shown in previous publications (Jin et al., 2008) the higher K_{zz} seemed necessary to explain other tracers in the MLT including methane and nitrous oxide. The text now includes the statement: "For Case C below 85-90km the CO₂ does lie within the ACE error bars but this is for an extreme sensitivity experiment where all GW turbulent diffusion is neglected, and above 85km the values do not fall within the error bars."

c) QU: No need to show zonal-mean for all simulations.

ANS: We agree for August that not all cross-sections are needed, but for April we wish to give a more overall picture including latitudinal structure. Thus Figures 9 and 10 (now, Figures 8 and 9) have been altered to reduce the number of panels.

26. P11564,I. 18-20:

QU: Discussion placed too far into text. Also an indication that ACE sampling is a cause of some of the discrepancies.

ANS: Our discussion on ACE sampling aliasing indicates polar regional data as being the primary region for possible impacts and this is partly why we have preferentially examined the low and mid latitude data where the main mass of the atmosphere resides and is less impacted both by year to year variations, strong high-latitude as-

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cent/descent and possible observational data sampling issues. See also our previous answer to question 22, above. We have moved this discussion forward to earlier in the paper text and have modified the text to expand on this issue.

27. P11565,I. 11-24:

a) QU: Missing reference and discussion of another model capable of simulating the 'knee'.

ANS: Figure 6 in Lopez-Puertas et al. (2000) shows a direct comparison of modelled and observed CO₂ and in this figure above 95km the model cannot represent the observed CO₂ though the position of the knee is comparable with the SL-3 mean profile but certainly does not reproduce the early decrease seen in ISAMS. In Figure 8 from Lopez-Puertas et al. (2000), which compares the ATMOS and TIME GCM only, shown using a log scale for CO₂ and so the model observational divergence above 95km is not small. Figure 7 however does argue that a reduced K_{zz} could produce the effect based on 1D simulations. Our model used a zero K_{zz} for the major unresolved turbulence process of the region, that of GW-breaking, inside a 3D model with resolved waves and transport and was not able to reproduce the desired change with a reduction in K_{zz} alone. The Lopez-Puertas et al. paper does not appear however to show a direct comparison of a "reasonable K_{zz}" simulation against the observations. However our assessment of their Figures 7 and 8 in Lopez-Puertas et al. (2000), indicate that the K_{zz} tests shown do not move the knee at 90km such as to reproduce the satellite observations and so even if the 1D results were applicable to the 3D simulation the figures do not conclusively and clearly show that a reduced K_{zz} can produce the observed CO₂ profile and location of the knee. We certainly agree with the argument that the unresolved and hence parameterized K_{zz} of this region is highly uncertain and most likely overestimated based on derivation from schemes such as the Hines GW drag scheme which tends to place a lot of wave breaking at high altitudes but in our comparison with ACE data the K_{zz} reduction not only cannot do enough to reduce the CO₂ profile to create the lower knee it also seems to be a needed element in the

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transport of other species such as CH₄ and N₂O which is contradictory to the idea of reduction for the CO₂ simulation. The text has been altered to discuss the results from Lopez-Puertas et al. (2000) and to expand on the issue of a representation of K_{zz} in GCM modelling. See answer to question 19 above.

b) QU: 2D Garcia and Solomon model.

ANS: Correction of molecular diffusion in Garcia and Solomon model did not result in reducing CO₂ similar to the ACE or CRISTA observation. But only brought the model results closer to the rocket observations (namely to the compilation presented in Fomichev et al., 1998) as shown in Chabrilat et al. (2002).

c) QU: WACCM claim via personal communication to get CO₂ correct.

ANS: Since the WACCM results the referee refers to are unpublished we cannot discuss their value in the paper even though it is of interest to know another model appears to solve the issue without the need for an additional CO₂ sink mechanism. As indicated in our text the GWD K_{zz} even when zero in our model was not able to reproduce the ACE observations and other tracers needed the stronger K_{zz} generated by the Hines for better simulation of ACE observations (e.g. CH₄ and N₂O) and so provides further evidence indicating the zero GWD driven turbulence K_{zz} scenario is inconsistent with the ACE data.

d) QU: Shorten this section

ANS: We do not feel that we have excessive repetition of the previous section here and since the discussion in this section is very important for the content of the paper, we have not shortened it, though as indicated above the section has been altered/ revised.

28. P11567,I. 14-17:

QU: The referee argues LTE valid for up to 100km so our discussion is invalid and asks to remove it.

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ANS: We are somewhat confused with this comment and do not want to remove this paragraph from the paper. First, we do not discuss here how the CO₂(010) state is close to LTE. The 15 μ m band is not the whole picture. Second, the referee is not right in his statement that the CO₂(010) is very close to LTE up to about 100 km. In fact, Fig. 6.4 of Lopez-Puertas and Taylor (2001) shows that vibrational temperature even for the main CO₂ isotope can be by about 60K higher than the kinetic temperature at 100 km. Third, even though only 1% of the total CO₂ molecules are in the excited states, it is not obvious how it can effect the temperature dependence of the CO₂ cross-sections. Cross-sections values for CO₂ molecules in excited states are not available. On the other hand, a highly excited CO₂ may have vibrational temperatures much larger than the kinetic temperature which means that these levels are strongly overpopulated compared to LTE conditions. In this paragraph we are estimating potential impact of non-LTE on the CO₂ cross-section by considering the upper limit of the effect. We have modified the text to clarify our point here: "...daytime vibrational temperatures are higher than the kinetic temperature (e.g. Lopez-Puertas and Taylor, 2001). We have estimated potential impact of non-LTE on the CO₂ cross-section by considering the upper limit of the effect and found out that non-LTE effects cannot likely lead to a considerable increase in the CO₂ photolysis rate."

29. P11568,I.9:

QU: Bad/Vague sentence.

ANS: The sentence objected to has been amended as it was badly written. The modified sentence is: "In the model control simulation, scenario A, CMAM CO is up to a factor of two too low above 0.01 hPa (80km)."

30. P11568:

QU: A lot of repetition of the previous section.

ANS: This section effectively discusses the previous section and hence refers to the

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results identified in the previous section. We do not consider it repetition. However the text has been reviewed and slightly modified in several places.

31. P11568,I.25:

a) QU: Is the "high" still beyond the error bars for this statement. ANS: Figure 5 (Figure 4 in the revised paper) now shows that above ~85km the model still cannot reproduce the ACE to within the observational error bars even with all GWD generated diffusion removed. Below ~85km the CO₂ does lie within the ACE error bars (on the high side) but this is for an extreme sensitivity experiment where all GW turbulent diffusion is neglected. The text has been modified to include:

"For Case C below 85-90km the CO₂ does lie within the ACE error bars but this is for an extreme sensitivity experiment where all GW turbulent diffusion is neglected, and above 85km the values do not fall within the error bars."

32. P11569,I.7-9

a) QU: The referee questions the "most reasonable scenario" statement. This needs the sentence to be re-phrased as the CO is clearly not reasonable only the CO₂ impact.

ANS: The sentence referred to the CO₂ data only and has been amended appropriately, to clarify the meaning we intended: "... for agreement between the ACE CO₂ observations and CMAM..."

33. P11569,I. 19-22:

a) QU: Can a sporadic process account for global and constant CO₂ depletion?

ANS: An additional process with a new effect of sequestering CO₂ can certainly diminish the CO₂ and produce a new equilibrium balance with lower CO₂ at the altitudes seen. A possible mechanism for the carbon removal on the meteoric dust is described in Plane (2004). This reference is now included. The process though variable would always be there (though of varying effectiveness) and hence can have a net loss/removal

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effect. The text has been modified to add clarity: "An interesting feature of such a phenomenon is that it will be spatially and temporally variable so that its effects will vary from season to season..."

b) QU: For April -v- August is there evidence of a secular change in dust consistent with CO2 impact? Would not a larger impact be expected in August and is this the correct sense to explain CO2 discrepancy?

ANS: The discussion is as indicated is speculative and further analysis is required to explore the details and hence it is beyond the scope of this current publication.

34. P11569,I. 23-27:

QU: Question over including temperature figures.

ANS: The temperatures are shown as we are illustrating how the basic modelled state compares with the ACE observations and that there is reasonable agreement with the ACE observations. This helps to suggest that the dynamical state is not subject to any anomalous temperature effects or major dynamical problems which could influence any interpretation of the observations and model comparison. We have moved temperature figure (Figure 10 in the revised manuscript) from the end to the middle of section 5 and include the following discussion: "In this context it might be of importance to consider how well the extended CMAM simulates temperatures. Figure 10 presents zonally and temporarily averaged latitude-pressure temperature for ACE and the CMAM control run for April and August. Bearing in mind that there are sampling limitations for ACE the agreement between temperatures is rather good. This suggests that the CMAM is capable to adequately model the large scale circulation and, hence, transport due to the resolved dynamics."

35. QU: Remove figure 2

ANS: Figure 2 has been removed after error bars were added to old Figure 5 (Figure 4 in the revised manuscript).

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36. QU: Minor change in Table 1 suggested.

ANS: Done.

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

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