Response to Reviewer 1

We would like to thank the referee for the helpful review and the constructive comments. His/her comments are given below in black and our responses in blue.

General comments:

1) In my opinion, the manuscript tries to cover too many topics despite the fact that the title states that this is an update to existing parameterization. Well, it doesn't hurt to recall the main milestones of the previous work, but the manuscript in its current form is "unfocused". If it is intended to be a text-book chapter on non-LTE in the CO2 bands, then the same information could be found in the first authors' book (Lopez-Puertas and Taylor, 2001). If it is supposed to be a technical paper then it should put more stress on technical aspects like the accuracy, interfacing, and so on. In addition, the text is overloaded with figures. I counted more than a hundred (!) of individual panels in the main part that consists of 41 pages. This is definitely an overkill and one can tell the same story in a much more concise way. I suggest the authors to shorten the manuscript and to keep only the essential parts. For example, all the inputs can be gathered on one two-panel plot, where the left-hand side would be occupied by the reference temperatures and the right-hand side would show the profiles of CO2 (only one is needed) and atomic oxygen, which is crucial for the estimate of the cooling rate. The first figure is not informative at all and can be easily omitted. The large number of panels in figures like Fig. 5 does not add much to the understanding of the principle of the radiative cooling or its calculation, and so on. Overall, I would reduce the number of non-essential figures to a minimum and put more efforts to a description of the routine itself, its implementation to a GCM and its limitations.

Reply: The point is well taken. We propose the following actions.
- About the text, we will retain only the major points driving the CO2 coolings, e.g their dependence on the pT structure, on the CO2 and O vmrs, the different bands contributing to them and how these contributions and their deviation from LTE depend on the p-T and CO2 and O(3P) vmr profiles. These parts are essential to understand the difficulty and the rationale of the parameterization. Further, note that we are extending the CO2 vmr range and this implies changes in the LTE to non-LTE switching regions, e.g. in the boundaries of the previous version of the parameterization. We think these changes have to be justified and explained to the reader. The part of the paper which is somehow redundant with Lopez-Puertas and Taylor (2001) is not that much, only part of Sect. 4.2 where we describe the reference non-LTE cooling rates. Here we focus more on the details of the input parameters (collisional rates, etc.) for the non-LTE model, which we think are important, for example, if compared to other parameterizations. In any case, the point is taken and we will try to reduce as much as possible the text of Sec. 4.2.

- About the figures, following your suggestions, we propose the following actions:

a) To move all figures of the Appendices to Supplementary information.
b) To remove Fig. 1.
c) To combine Figs. 2 and 3 in one 2-panel fig. The referee also proposes to include one Fig for O. How can it be done for the 3 parameters and only 2 panels? Further, combining CO2 and O in one figure is difficult because of the different (linear and log) scales. We propose a 3-panel 1-col fig. for p-T, CO2 and O.
d) Fig. 5. Sorry but we do not agree with the referee on this point. This is an essential figure to discuss the points mentioned above as a reference for the line-by-line accurate cooling rates. Nevertheless, it has been reduced to keep only 4 essential panels.
e) Fig. 6 Contributions of the different bands. It has been reduced to only 4 panels (as Fig. 5).

f) Fig. 7 Contributions of bands in the lower thermosphere (1 panel). It will be moved to the Appendix

g) Figs. 8 and 9. Comparison of non-LTE and LTE cooling rates. Fig. 8 will be reduced to four panels, but not moved to the appendix because the other referee suggested that we should describe the non-LTE-LTE differences in order to show the relevance of the non-LTE errors of the parameterization.
Fig. 9, however, will be moved to the Appendix.

h) Fig. 10. The effects of the K(CO2-O) collisional rate. We will show only 4 panels, consistent with previous figures.

i) Fig. 12 (escape probability functions). It will be moved to the Supplement.

j) Figs. 13, 14 and 15 compare the previous and current par. They are essential and are kept in the main text.

k) Figs. 16 and 17, and 18 and 19 show the accuracy of the parameterization for intermediate CO2 vmr profiles and for the K(CO2-O)/2. We will keep in the main text only Figs. 17 and 19 (the summaries) and move Figs. 16 and 18 to the Appendix.

l) Fig. 20: MIPAS temperature profiles. It will be kept and likely modified (see below).

m) Fig. 21. MIPAS zonal mean temperature maps for 4 days. Will be moved to the Appendix.

n) Fig. 22. Zonal mean of the differences between the previous and new par. for the MIPAS temperatures (8 panels). We suggest keeping in the main body only 4 panels (solstice (January) and equinox (March) and moving the other 2 panels to the Appendix.

o) Figs. 25, 26, and 27 show the performance of the parameterizations for the elevated stratopause conditions. Fig. 25 will be moved to the Appendix. The others will be kept.

Overall, the number of Figs. in the main text will be reduced from 28 to 19 (and some of them with a reduced number of panels); and the number of Figs. in the appendix will be reduced from 20 to 8. 21 figures will be moved to a Supplement.

About "... put more efforts on the description of the routine itself, its implementation to a GCM and its limitations", the description and its limitations are already discussed in detail. Its implementation is described in the Readme file of its distribution and, as suggested by the other referee, we will add a paragraph about the altitude region where it should be used. That is, the parameterisation is specifically developed for the CO2 15 µm **non-LTE region**, and, although it also works for the LTE region, it should be used with caution in that region. We recommend that the users utilize their radiation scheme in the LTE region and this parameterization in the non-LTE region (above~50, 60 km).

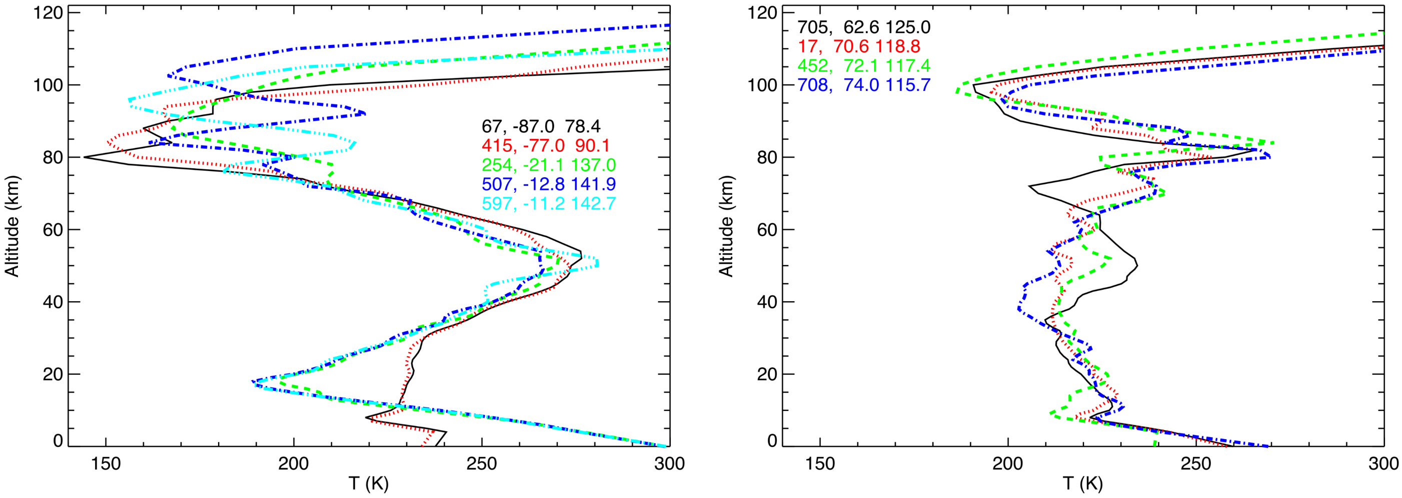
2) Regarding the routine, I did not see any reference or repository for it provided along with the manuscript. As far as I understand, this is now a requirement for EGUsphere journals, so I wonder when and how did the authors plan to publish the routine. Besides a general conformity with the journal's rules, it would be advisable to get a self-consistent compilable code with the cooling rate parameterization routine called from the main code with some standard atmospheric profile, which could be replaced with a test profile by the user. In the next paragraph, I explain the current problem I see with the accuracy of the updated parameterization, and I guess the GCM-modelers would have liked to test it themselves prior to implementing it to the model. Summarizing this point, the authors must provide a ready-to-use code and provide an instruction for its compiling and running both as a standalone routine and as a part of the test code, that might be useful not only for GCM-modelers, but for other researchers wanting to get a quick estimate of non-LTE cooling profile for a given atmosphere.

Reply: The referee is fully correct. We were not aware of that and thought of providing it during the review process. We will provide it in its final version as a Fortran 90 code jointly with the revised version of the manuscript. In the meantime, the parameterization is now available provisionally as a Python routine, please see the reply to the Editor comment 1 (https://zenodo.org/doi/10.5281/zenodo.10547026)

We thank the referee for the advice and recommendations. We have already taken them into account and are providing input and output files for the 6 reference atmospheres, for two CO2 concentrations (#3 (current) and #8 (10x preindustrial values)).
We have a modeller in our team and we will incorporate all of his comments about its implementation in the WACCM model in the documentation of the code. Further, the referee made a very good point to provide a version that could be used by any researcher and not only in GCMs. We will facilitate its use for this purpose.

3) The authors show that the routine works fairly well for the multitude of atmospheric profiles they use in the tests. This is quite impressive given a large variability of CO2 concentrations used in the study. Still, I cannot consider these tests to be fully representative because the real atmospheric temperature profiles are far from those shown in Fig. 2 of the manuscript in terms of vertical variability. Moreover, they are even worse than the ones shown in Fig. 20 because the averaging kernels of MIPAS instrument in the middle atmosphere are broad (e.g. Dinelli et al., 2021) and, therefore, this instrument cannot capture any wave shorter than 5-7 km. In addition, MIPAS is a limb measuring instrument, so it washes out horizontal inhomogeneities and this further reduces its vertical resolution. At the same time, the gravity waves (GWs) are known to be composed of several waves of different wavelengths and their amplitude increases with height up to the top of the “wave-turbopause” layer at 90-110km (e.g. Offerman et al., 2007; Ge et al., 2022), which overlaps with the non-LTE-affected area. I believe, the MIPAS profiles used in the study do not represent a full picture of a gravity-wave perturbed temperature profiles, so the tests performed with them show only a partial truth. Moreover, large RMS values presented in Fig. 24 even for these, partially smoothed, profiles make me think that the real accuracy of the parameterization is far from 1−2 K/day as it is stated in the abstract (I do not consider the specific case of elevated stratopause, which is addressed; I’m talking about the accuracy of “regular” profiles). If the tests I suggest below do not disprove my suspicions, this wouldn’t cancel the suggested parameterization. But, the GCM modelers will have a clear picture of what they will use and act accordingly if their models produce lifelike GW-perturbed temperature profiles.

Reply: Overall we agree with the referee in testing a few more cases, although we are quite confident that the results will not change significantly (see below). However, let us first clarify a couple of points about the MIPAS temperatures used. It is true that they have a limited vertical resolution and do not fully recover the actual pT altitude structure. However, they are also affected by the noise of the instrument, which frequently maps in the profiles as a vertical variability larger than the real one. The given reference about MIPAS (Dinelli et al., 2021) describes the retrieved temperatures from the MIPAS **nominal** mode. The MIPAS data we used in the manuscript are those retrieved from the middle atmosphere (MA) mode, which has a better vertical sampling and finer AKs than the nominal mode ones (see García-Comas et al., 2023).
Hence MIPAS profiles (eg Fig. 20) are not that far away from real ones, we think.
We are including below a couple of Figs. showing some profiles extracted from MIPAS data for 15 February 2009 (Fig. 25). They clearly show wave structures and the very unusual pT profiles during the elevated stratopause.



We concur with the referee that the values of 1-2 K/day given for the accuracy are for the reference p-T profiles. When dealing with individual p-T profiles the errors are larger, as demonstrated in its application to MIPAS data, particularly for polar summer and polar winter conditions. For that reason, we did the test with the MIPAS individual temperature profiles and even showed the case of elevated stratopause conditions. However, those errors for MIPAS can be considered representative.
In general, we think the most proper way of expressing its accuracy is by a mean error value (bias), which will inform us about its accuracy in a global sense, and by the RMS (not the standard deviation), as an appropriate estimate of its error for individual profiles as assessed from a significant sample.
We will clarify in the abstract that the values now given for the accuracy are for the reference atmospheres, and will include the RMSs as an estimate of the individual profiles for the different atmospheric conditions. Further, we will also assess similarly its accuracy for the specific lidar cases described below.

4) Here is the test I believe is necessary to conduct in order to properly estimate the accuracy of the suggested parameterization. Instead of using the MIPAS profiles and showing the averaged effects, I ask the authors to use the individual temperature profile coming from a ground-based lidar (e.g. Spitsbergen (78°N, 15°E), ALOMAR at Andøya (69°N, 16°E), Kühlungsborn (54°N, 12°E), Boulder (40°N, 105°W), Fort Collins (41°N, 105°W), Logan (42°N, 112°W), Arecibo (18°N, 293°E), Cerro Pachon (30°S, 71°W), or McMurdo (78°S, 167°E).) Since these lidars do not cover the whole atmospheric profile, in the lower atmosphere one can take a corresponding smooth profile from the reanalysis or from any other model. Among the profiles provided by the lidar teams, one has to pick up the ones, which clearly show the GW-signature (at least, the wave of +/- 10 K amplitude at 100km), and perform the exact and parameterized calculations of radiative cooling rate for these profiles. It would be also interesting to see how different wavelengths affect the accuracy, because the non-LTE effects are different in the case of a short-and large-scale perturbations. I have to stress here that due to a nonlinear nature of temperature perturbation effects on non-LTE cooling rates, these results cannot be replaced with the ones obtained for averages.

Reply: We will check several of the lidar profiles suggested by the referee. We do not know now (we will check) how different they are from the variability shown above for MIPAS. If they are significantly different, we will assess the accuracy of the parameterization for a statistically representative ensemble of lidar p-T profiles in addition to the MIPAS profiles. We will assess its accuracy for the specific lidar cases in a similar way as described above.

Minor comments and technical corrections

Lines 4 and 41: this statement is too general. In fact, there are ways of including the non-LTE calculations to GCMs. For example, this was done for Martian GCM (Hartogh et al., 2005) and I don't see why this can't be done for Earth. The corresponding manuscript is still in discussion in the same journal, so it can't be referenced, but the authors claim that the same approach works for the Earth's atmosphere.
Reply: We agree that there are different ways of including non-LTE in GCMs but generally they are all very CPU time consuming. "The corresponding manuscript … " Does the referee mean the manuscript submitted recently by Kutepov et al.? We will include the mentioned reference and a citation to the manuscript recently submitted to this journal in the revised version.

Line 11: since the atomic oxygen is a variable component and the reaction 1c is directly linked to it, it would be good to have it as a variable parameter, see the second general comment
Reply: The user will have the option of including both, the atomic oxygen concentration profile and the collisional rate K1c. This is specified in the documentation of the code provided now.

Line 17: the measured profiles themselves is not a ground truth because of the vertical resolution, see the third general comment. Moreover, one cannot average the results for individual runs and present this difference as an error, because the GCMs accumulate the error due to non-linearity of the processes.
Reply: First point: We agree it is not the ground truth but we think it is not thus far (see plots above). We will also analyse lidar pT profiles and test the parameterization for them.
2nd point: We agree. Still, that average gives us a measure of the "error" (systematic) in a global sense (eg the cooling at high altitudes in the previous version is always about 2K/Day larger than in the line-by-line calculations). But will also provide the RMS as an estimate of its error for individual profiles as assessed from a significant sample.

We do not understand what the referee means by "... because the GCMs accumulate the error due to non-linearity of the processes." Does he/she refer to the fact that the cooling is proportional to exp(-kE/T) and not to T? We can only assess cooling rate errors induced by the parameterization, but not non-linear effects on modelled quantities such as temperature and winds.

Line 58: Figure 1 is not informative, see the first general comment
Reply: OK. It will be removed.

Lines 120-150: I guess, the readers will be confused here. It looks like GRANADA non-LTE code cannot work in LTE mode whereas normally it requires one line or one flag to calculate LTE populations. Could you, please, explain?
Reply: GRANADA computes non-LTE populations and non-LTE cooling/heating rates. We do not need a code for computing LTE populations and GRANADA has not been specifically designed/coded for computing LTE cooling rates. We already clarified in the manuscript that we use KOPRA for LTE cooling rates using a Curtis Matrix approach.

Line 125: I didn't find any mention of line mixing effects here. Are they not important?
Reply: KOPRA does include line mixing and it was used in these calculations. We will include a sentence stating it.

Line 135: I didn't get the phrase about the "oscillations in RFM results". Could you, please, clarify?
Reply: We refer to the minor oscillations in the RFM cooling rate profile, e.g. around 50 km, 60 km, 65 km, and 85 km.

Line 145: And what about the results without this additional iteration?
Reply: In short, the cooling rates would be less accurate**.** Without that final iteration (for specified accuracies for the vibrational temperatures), the non-LTE populations, e.g. the vibrational temperatures do not change significantly (see Funke et al. 2012). However, we did not describe in that work the non-LTE cooling rates. To properly calculate the cooling rates, in addition to the specific procedure described in that work, a final iteration to compute the radiative field in all bands is required in order to properly account for the overlapping between the different bands. The effect of not including it is to have slightly less accurate cooling rates.

Line 171: I recall that the difference is about 2-3K, and I wouldn't call this a small change, so one should not neglect the daytime vs nighttime differences. This is addressed in Sect. 8, so the wording has to be changed here
Reply: It is not that large, about 1-2 K at most (see e.g. Fig. 3 in López-Puertas et al., 1990) which in relative terms (the day/night differences are larger above about 90 km) are very small.
Our more recent calculations of day/night differences of the CO2 15 µm cooling rates are smaller than 1K/day for all pT profiles, at any altitude and for CO2 vmrs up to 5 times the pre-industrial

value, except for MLS and SAS pT profiles near 105 km and CO2 vmr 4x and 5x the pre-industrial value.
We already state that this point is addressed in Sec. 8 (line 175).

Lines 200-215: all these descriptions are correct, but I doubt that they add something to the understanding of the routine or its accuracy, see the first general comment on the scope and focus of the manuscript.
Reply: We already state the reason why including this discussion, lines 201-202: *"This comparison is useful from a physical point of view and it is required to establish the boundaries of the different atmospheric regions of the parameterization"*. We have moved Figs. 8 and 9 to the Appendix and will try to lighten the text in that section.

Lines 220-225 (see also the comment to lines 437-462): if we assume that the k(CO2-O) used in the models is equal to 3 x 1e-12 cm3 s-1, then this experiment shows the effects of quadrupling the atomic oxygen mixing ratio. In addition, the reader might be misled by switching from atomic oxygen to k(CO2-O) and back. It would be enough to present this numerical experiment as the one performed for doubled (or quadrupled) atomic oxygen.
Reply: That is what we did, just to half the k(CO2-O) nominal value of ~6e-12 cm3 s-1. We will clarify this in the text and change "perturbed the kCO2−O collisional rate by a factor of two" to "divided the kCO2−O collisional rate by a factor of two".

Lines 248-395: in fact, this deserves a general comment, but I cannot suggest to modify the general approach, it is what it is, and it works for smooth profiles for a current atmosphere. But what if the atmosphere changes some day and the LTE/NLTE1,2,3,4 regions become different? Let's imagine that a modeler wants to calculate cooling rates for an Earth-like atmosphere, but with a different vertical temperature structure. How can he/she tell the limits of applicability of this parameterization?
Reply: Very likely, it will not work with high accuracy for those situations. This parameterization has not been designed from its origin for any planetary or p-T temperature profile. It has been specifically tailored for the current-like Earth's atmosphere and projected high CO2 vmr in the future.

Line 304: this is just one of the examples of the problem outlined in the previous comment. The error introduced by this "implicit assumption" depends on the atmospheric profile and is hidden deep in this module. It is small for the current atmospheres, but it is not guaranteed that it will remain small for some unusual temperature profile.
Reply: That particular aspect is not an issue because the atmosphere is in the optically thin regime at those altitudes and the parameterization is very accurate there. Look at all the tests performed and see how the errors tend to vanish at high altitudes.

Lines 437-462: this is another potential candidate for a general comment. The problem is that there are three values of this rate coefficient, k(CO2-O): the first one is measured in the lab and is about 1.5−2.0 x 1e-12 cm3 s-1 (e.g. Castle et al., 2012), the second one is about 6.0-9.0 x 1e-12 cm3 s-1 retrieved from space-borne observations of 15-micron CO2 emissions, and the third one, 3.0 x 1e-12 cm3 s-1 was suggested for using in the GCMs. This paradox has not been resolved over the decades of its study, but it shouldn't matter for the parameterization itself, because it is supposed to calculate the cooling rate with any given k(CO2-O).
Reply: Exactly, we fully agree with the referee. The point we want to stress is that the parameterization has been optimized for k~6.0e-12 cm3 s-1, and we would like to assess the errors when it is used for half of this value. This is important to tell the reader that if using a rate half of the value for which it has been optimized, it does not lose significant accuracy.

Line 455: how do you explain that the errors obtained for a smaller k(CO2-O) are larger? Usually, the larger the quenching rate, the larger the cooling rate, and the magnitude of this error is linked to cooling, as in the case of enhanced CO2.
Reply: The very likely reason is that the coefficient has been calculated, and it is therefore optimized, for the larger k(CO2-O) rate. In any case, the differences are very small.

Line 462: Fig. 20 is barely readable. Please, provide only a couple of profiles coming from a different source (see general comment 4) and show the errors for the corresponding cooling rate profile on the right-hand side panel.
Reply: The intention of showing this figure is not to distinguish between the different pT profiles but to show that the parameterization has been tested for an ample range of temperature structures, ranging from polar-summer-like profiles to polar-winter-like profiles and for elevated p-T profiles. That is, it is a very demanding test.
We have shown above a few profiles extracted from Fig. 20. We will show a few pT profiles like those in the Figures above together with the cooling rates estimated by the “exact” model and by the parameterization. We will make clear that they are examples but should not be taken as representative of the performance of the parameterization.

References

García-Comas, M., Funke, B., López-Puertas, M., Glatthor, N., Grabowski, U., Kellmann, S., Kiefer, M., Linden, A., Martínez-Mondéjar, B., Stiller, G. P., & von Clarmann, T. (2023). Version 8 IMK–IAA MIPAS temperatures from 12–15 µm spectra: Middle and Upper Atmosphere modes.*Atmospheric Measurement Techniques*, *16*(21), 5357–5386. https://doi.org/10.5194/amt-16-5357-2023