

## ***Interactive comment on “The use of O<sub>2</sub> 1.27 μm absorption band revisited for GHG monitoring from space and application to MicroCarb” by Jean-Loup Bertaux et al.***

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Interactive comment on “The use of O<sub>2</sub> 1.27 μm absorption band revisited for GHG monitoring from space and application to MicroCarb” by Jean-Loup Bertaux et al.

General answer: We are very grateful to referee #1 for his (her) careful reading of the manuscript, deep effort to reproduce some spectroscopic results, pointing out some errors, and one particularly interesting suggestion about the use of nadir viewing SCIAMACHY data that we have followed. As a result, we have made considerable rewriting

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of Section 2.3, and accepted most of the suggestions of referee #1. In the following, answers are in arial and blue in the following. Anonymous Referee #1 Received and published: 30 June 2019 The feasibility of using the 1.27 μm, or the “singlet Delta” O<sub>2</sub> band for satellite nadir remote sensing of greenhouse gases has been investigated previously (Sun et al. 2018). However, this manuscript, to my knowledge, firstly presents this observation strategy within the framework of a planned satellite mission (MicroCarb). Since such a satellite mission may cost hundreds of millions of euros/dollars and provide crucial data with global coverage for many years, this strategy may potentially have a far-reaching impact, and this work may become an import reference for future missions. Given the aforementioned significance, this manuscript at this stage may still be substantially improved in terms of both presentation quality and technical rigor. The following lists my general comments concerning the manuscript structure and specific comments concerning individual sentences, equations, and figures.

1 General comments

1.1 In general the manuscript has been clearly improved since the initial submission. That said, it still reads a bit fragmental with different sections apparently from different groups of authors. The O<sub>2</sub> molecules in a1\_g state, for example, was denoted as O<sub>2</sub>, O<sub>2</sub>(a1\_g), O<sub>2</sub> (a1\_), O<sub>2</sub>(1\_), and O<sub>2</sub>(a1\_) throughout the manuscript. As to the paper structure, Section 5 describes the MicroCarb mission and its physicsbased retrieval, 4ARTIC. Then it was deviated to a retrieval of SCIAMACHY nadir airglow using a different tool called LATMOS breadboard (sections 6.1–6.2) before 4ARTIC was applied to test different airglow mitigation approaches in section 6.3. The authors may consider moving sections 6.1–6.2 after section 3, because the SCIAMACHY nadir/limb airglow comparison naturally follows the onion-peeling VER retrieval, and combining section 5 with section 6.3. Some parts of section 7 seem to be out of the scope of this study (e.g., the discussion of methane and CO fluorescence), and the remaining parts may be combined with section 2.1.

1.2 Section 2.3 could be clarified and largely removed from the manuscript. First, the main conclusion from this section, equations 14 and 15 that will be used to convert absorption spectra simulated by LBLRTM to airglow emission spectra, seems to have already been demonstrated by equations 3–5 in Sun

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et al. (2018). Combining equations 4 and 5 in Sun et al. (2018): answer: the recommendation to clarify and to largely remove it from the manuscript is self-contradictory, and is anyway not acceptable for the following reasons: 1. The theoretical development (to obtain a theoretical spectrum of the O<sub>2</sub>\* airglow emission) that we present here was done in 2017, and completely independently from the work of Sun et al. (2018). If we were following the suggestion to just quote the equations of Sun et al. (2018) and not present our own analysis, it would give to the reader the false impression that we have followed blindly the developments of Sun et al. 2018, which is not true. The fact that both groups have developed the same kind of theory (form the same theoretical approach based on what can be found in Simeckova et al. (2006)) re-inforce the credibility of this approach, which is very important for “hundreds of millions dollars space projects”. 2. Our equations (14) and (15) give an original result: a formula giving the wavenumber variation over the whole band of the ratio  $\varepsilon/SS$  of airglow emission  $\varepsilon$  to the line strength SS found in Hitran table. This formula is NOT in the paper of Sun; the referee#1 had to check by some manipulation of equations (in black, below) that such a relation COULD be retrieved from formulas in Sun et al. (2018). Doing this, referee#1 shows that both groups are using consistent descriptions of the physics involved, a satisfactory piece of information for which we must thank him. 3. In fact, in the present study, we have used the formulation of this ratio in order to build very simply a synthetic spectrum of the airglow emission, by using the LBLRTM code computing the local absorption, and multiplying by the function  $\varepsilon(\nu)/SS(\nu)$  (14) and (15). This is a totally original method, and we wish that all the AMT readers to be able to reproduce it and use it. This is why all equations establishing  $\varepsilon(\nu)/SS(\nu)$  must be kept in the present paper. 4. The present manuscript was given to Iouli Gordon, one of the co-author of Sun et al (2018) and HITRAN expert and producer. It was also discussed by the first author(JLB) in a face-to-face meeting, and Iouli Gordon had no objection on the theoretical aspects. We consider that it re-inforces the validation of our approach.

The equation above is identical to equations 14–15 in the manuscript. As such, most equations/figures in section 2.3.2–2.3.6 can be replaced by a simple reference to equa-

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tions 3–5 in Sun et al. (2018). answer: no. See the answer above.

Second, the symbols are not consistently defined, making section 2.3 challenging to follow. For example, the upper state energy is E<sub>2</sub> in equation 9, E<sub>2i</sub> in equation 10, and E<sub>i</sub> in line 9, page 11. answer: some rewriting has been performed to clarify what is what.

The total partition sums for both absorption transition and airglow emission are denoted as Q<sub>tot</sub>(T), but separately as Q<sub>lotot</sub>(T) and Q<sub>up tot</sub>(T) in equation 15. answer: the text has been revised for more clarity, which was needed indeed. The summation index is i in equation 10, but appears to be J<sub>0</sub> in equation 11. The local emission rate spectrum is given by E<sub>m</sub>( $\nu$ ) (line 17, page 15) and E<sub>mn</sub>( $\nu$ ) (line 18, page 15) in section 2.3.7, but it is unclear what this quantity really is. answer: The additional letter n is not an index, but stands for “normalized”, since the values have been normalized. Finally, it will be very useful to discuss the improvements over previous studies. I did not find an equation that calculates Q<sub>up tot</sub>(T) in Sun et al. 2018, which is given by equation 10 in the manuscript. However, I cannot reproduce the results Q<sub>tot</sub>(T = 296K) = 147.196 (line 10, page 11) using equation 10 and HITRAN 2016 data because it is unclear what “index i refers to each value of E<sub>2</sub>” (line 36, page 10) exactly means. answer: index i refers to all values of the quantum number J' of each rotational level. This is now indicated in the text. One possible reason for a discrepancy on Q<sub>tot</sub> up may be that you may have taken the expression in (10), while we have (for convenience only) subtracted E<sub>0</sub> from all values of E<sub>2i</sub> in equation (10), to get a new definition of Q<sub>tot</sub> up, which probably coincides with the Q(T;airglow) from Sun et al. There is a factor 4.5x10<sup>-16</sup> of difference. The other possibility is that you took the head band value of Lischichina et al., (which is for J' = 0) while we took for E<sub>2i</sub> the energy of the first authorized level J' = 2. The relative distributions of levels are not affected by a particular choice of E<sub>2i</sub>. 1.3 One significant missing piece of this study is the fine-scale spatial variation of airglow, potentially due to gravity waves. The REPROBUS CTM (2Å~2<sup>-2</sup>) and SCIAMACHY limb data (400 Å~ 960 km) used in the study are too coarse to

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capture those fine-scale variations. The fine-scale airglow features may propagate to the retrieved XCO<sub>2</sub> as artefacts from the disturbed mesospheric temperature (spectral shape of both emission and re-absorption), excited O<sub>2</sub> molecule concentration (emission), and O<sub>2</sub> molecule concentration (re-absorption). The authors may have missed one opportunity as the SCIAMACHY nadir data over ocean have much higher spatial resolution and may reveal some gravity wave features. Larger scatter is seen from SCIA nadir ocean compared to SCIA limb (Figure 30). Is that just retrieval random errors or real spatial variation? How do they look on a map? Answer: We fully agree with your suggestion and made a new study. As a result, we have added at end of section 6.2 the following text: “Following an interesting suggestion of anonymous Referee#1, we have tried to estimate from nadir viewing SCIAMACHY data the small-scale horizontal variations of O<sub>2</sub>\* airglow that could be due to gravity waves and are not represented in REPROBUS CTM. This is not an easy task using the relatively low spectral resolution of SCIAMACHY data. At this resolution spectral features in airglow and O<sub>2</sub> absorption spectra are highly correlated and the estimation of airglow is accurate only for very low values of reflected solar flux as illustrated on Figure 29, where a large dispersion of airglow is observed for high values of reflected solar radiance. There are not enough observations reaching a low level of solar flux to plot maps of airglow. In spite of these limitations, we made an attempt to estimate at least an upper limit for the small-scale variations of airglow. We selected all pairs of nadir observations with reflected solar flux < 2 mW/m<sup>2</sup>/nm/sr, solar zenith angle < 60° and distance < 110 km. With these strong criteria only 1% of the observations were selected. The average difference in airglow intensity between the pairs of observations was equal to 1.0%. We consider this value as an upper limit of the impact of gravity wave perturbation in airglow intensity. At this level the impact on the retrieval of Psurf and XCO<sub>2</sub> will be very limited.”

We think that the absence of significant horizontal fluctuations of the O<sub>2</sub>\* airglow is due to the fact that the emission layer is rather thick, in contrast with the much thinner OH layer at night, where horizontal fluctuations gravity waves may be observed.

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2 Specific comments Line 31, page 1: the statement that the theoretical synthetic spectrum is from “a new approach” is contradictory to the following statement that this approach “is very similar (likely identical) to the approach of Sun et al. (2018)” (lines 7–8, page 2). The current work should be more clearly distinguished from previous before claiming it is “new”. Answer: for us, this approach was new, with respect to a first crude approximation. We have deleted the word “new” at this place. Note that our work was done independently , and before the publication of Sun. Line 32, page 2: may include OCO-3 and GOSAT-2. Done Line 40, page 4: why is pressure broadening proportional to the “square” of air density? Answer: Sorry for this mistaken statement and thank you for pointing it out. The number of collisions per cm<sup>3</sup> is proportional to the square of the density, but the number of collisions per O<sub>2</sub> molecule (which is relevant to pressure broadening) is proportional to the density. Sentence corrected. Figure 2 caption: how can “transmittance” be “larger” than emission? Did the authors mean “wider”? Answer: Yes, of course. We have rephrased the sentence and used “wider”: “Comparison at high spectral resolution of spectral shape of atmospheric O<sub>2</sub> transmission (transmittance) and spectral shape of O<sub>2</sub>\* emission. The FWHM of an individual O<sub>2</sub> line (red) is much wider than the FWHM of its counterpart in emission (black line), allowing in principle to disentangle absorption from emission at selected wavelengths.” Line 35, page 6: equation 3 looks the same as the second equation in the Appendix A1.1.1. They should be consistent. answer: They are consistent, because O atoms are in their fundamental state O(3P). This precision is now included in the text. Line 32, page 7: Dave Crisp et al. should be just Crisp et al. Done Line 30, page 9: quenching is significant below 50 km, and the airglow peak is also below 50 km. Are there any consequences if the quenching is ignored? Answer: The quenching becomes important at lower altitudes. If it were neglected in the airglow, it would result in a significant overestimate of the airglow. Line 15, page 14:  $E_0 = 7892.02 \text{ cm}^{-1}$  is inaccurate. Refer to Leshchishina et al. (2010). Answer: The work of Leschichina et al. is published in 2010. Co-authors are Gordon and Rothman, responsible for the HITRAN data base. We assume that the HITRAN2016 data base contains the best

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available constants. Probably Leschichina et al.(2010) and us are not talking of the same thing. We find in the HITRAN data base that the minimum energy level E1 for the upper state is 7892.01738, for V'=0 and J'=2, as clearly stated in our paper. We have changed our value of 7892.02 into the full digit value 7892.01738. Anyway, it is a multiplicative factor, and is not important in the relative distribution of the various rotational levels of the upper state. Line 40, page 16: " appears to be spectrally integrated here, but spectrally resolved in equation 13, and back to be spectrally resolved in line 28, page 18 again. answer: you are right. We have been more specific at line 40, page 16; while for line 28, page 18, there is the sentence: "In reality,  $\varepsilon$ ,  $\tau(s)$ ,  $Tr(s)$  and  $B$  all depend on wavelength  $\lambda$ .", which should be enough for the reader to understand. Line 5, page 18, equation 21, and line 4, page 19: a bold face " is used, but definition is unclear. answer: the classical notation (in english) for vectors and matrices is to write them in bold face, and this is what was intended here. We now have put  $B$  in bold face in the line just before equation (21).  $\varepsilon$  is already in bold face in this line. We have also change to bold face  $B(z)$  in the line 4, page 19 (now changed to line 32, p.19). Line 24, page 21: why does O2 absorption have to be neglected? It is important and all parameters should be readily available to include O2 absorption in the analysis. answer: The O2 absorption certainly does not need to be "neglected" and we did not say that. It just happens that at the time of this exercise, we did not include the absorption in the model, and we are not going to redo it for this paper. We were glad enough to see the improvement of similarities between SCIAMACHY observed spectra and our new model of the emission (compared to our "old" model) when the emission is properly computed (figure 13). We have rephrased here: "We tentatively assign this behaviour to the fact that we have not accounted for..." and also in the paragraph before figure 13: " we have not accounted for the O2 absorption..." Figure 11: this figure may be removed. The SCIAMACHY limb data are binned across track so it is unnecessary to show separate across-track positions. The globe distribution of SCIAMACHY orbits is well represented by Figure 15. answer: we think it is important to see the separation between the 4 FOV at the limb and the distribution of the data points along one single

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orbit. Figure 12b and the left panel in Figure 13: these should be removed, see the following argument. answer: we think that it is important to show that, at first glance (figures 12 a and 12 b), the observed spectra and the model spectra are quite similar, both for the crude approximation model and the "true" model. But when the ratios of observed/model are plotted, the better fit with the "true" model is obvious. So we wish to keep all these figures to show the importance of not using any longer the crude model.

Line 8, page 23: This is not a "simplified" model; the equation for absorption cross section is not simpler than equation for airglow emission. This is an "inaccurate" or "inappropriate" model. answer: we have replaced "simplified model" by "crude model", which is consistent with what is written in the first paragraph of Section 2.3.

Figure 16: SZArecomputed appears in the figure but is only explained at the end of page 31. answer: Figure 16 was redrawn without the word "recomputed". Lines 5–6, page 27: VER has been defined and should be only defined at first appearance. answer: Volume Emission Rate is now deleted here. Section 4.1.1, page 27: the authors are suggested to better articulate the usefulness of the REPROBUS model. Its low resolution is not sufficient to capture the fine-scale variation; its model top (0.01 hPa) does not fully capture the airglow profile (what is the error from neglecting airglows above 0.01 hPa?); the model is not used in the following MicroCarb retrieval tests. answer: We have added the following sentence: "It should be noted that, as a result of some discrepancies revealed by this comparison, the REPROBUS model will be modified in the future for a better representation of mesospheric ozone. Although the retrieval of O2 column does not need a model, it is likely that the output of the improved REPROBUS model (O2\* intensity) will be used as a prior information in the retrieval process." Lines 13–14, page 29: how much is the error due to neglecting the reabsorption, and what is its relative contribution to total error? answer: In the nadir viewing geometry, the absorption of the O2\* by O2 above 30 km would decrease the brightness by 1-2 % and was neglected. Figure 18: it's maybe useful to mark the

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subsolunar point in the plot. answer: we have added in the Figure 18a,b,c caption the sentence: "Because of the particular time and date, the subsolar point is at latitude 23.5° N and on the meridian 90° East, which is the one plotted at the center of each figure". Line 23–24, page 31: may change to "...an observer placed above the tangent points..." as there are multiple tangent points that are close to each other. Line 41, page 31: the SZA is defined at "one of the two points corresponding to the intersection between the line of sight and TOA". Which one? answer: we have rephrased the sentence: "We noted that the SZA value provided in the SCIAMACHY ESA products in limb viewing, as defined in the data product, is the SZA value of one of the two points (the nearest to ENVISAT) corresponding to the intersection between the LOS and TOA (Top of Atmosphere defined at 100 km altitude). But what we need is the SZA of the tangent point of the line of sight (LOS), which is different." Figure 19: removing the lines between markers may improve the figure clarity. answer: Figure 19 has been redrawn with the lines between points removed. Line 7, page 33: it could probably be delivered in a more rigorous way than asserting: "must have been". answer: yes, it could. But we are rather embarrassed, because a search on Google and ADS shows very little return on the topic of SCIAMACHY in-flight radiance calibration in the range around 1.27  $\mu\text{m}$  by comparison with MODIS or other well calibrated instruments. We note also that another referee who obviously knows very well both SCIAMACHY and english language (likely John Burrows) did not protest about our formulation, and we prefer to keep our wording. Figures 20–22: Figure 22 should be enough to present all results so Figures 20–21 can be removed. answer: we have deleted Figure 21 and kept Figure 20 for three examples of single profiles. Line 2, page 36: modified to "five orders of magnitude of variation of ozone with altitude". Done. Figure 25: what is the difference between the left and right? What are the observation time/SZA? answer: left and right are just two different examples of day side ozone profiles. The SZA are 38° (left) and 94°(right). During this second occultation, the tangent point is still solar illuminated. However, the chemical situation of ozone may be ambiguous at that SZA angle, and therefore we prefer to delete this figure and to keep only the occultation at

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SZA=38°. The exact date and SZA is now indicated in the caption.

Line 5, page 39: the claim that "the night side model bias is quite negligible below 60 km" contradicts the main conclusion of this section, i.e., the ozone deficit below 60 km of 10–20% in the REPROBUS model is the main reason of airglow underestimation. Moreover, is it possible to compare with other reanalysis/observation data, especially MERRA-2? answer: We agree that there was a little bit of confusion in the summary of the comparisons GOMOS/REPROBUS, in large part because we wrote "lower" when we meant "larger" in the following sentence, now corrected: "- GOMOS ozone concentration vertical profiles show quite similar values below 60 km between day and night, and larger lower values of O<sub>3</sub> at night above 60 km, a feature well understood from mesospheric chemistry. - there is a known shortcoming of the chemistry of REPROBUS model affecting strongly night side predictions above 60 km, quite apparent with GOMOS ozone night side comparisons (too much ozone in REPROBUS). - Because the model O<sub>3</sub> diurnal variation is small below 60 km (there, we are more confident in the model than in GOMOS dayside data to estimate the small ozone diurnal variation), the comparison GOMOS/REPROBUS on the night side showing a deficit (10-20 %) of the model versus GOMOS ozone below 60 km may be applied also to the day side. " The comparison with MERRA-2 output model is an interesting suggestion but beyond the scope of the present study. Line 8, page 41: change "maximum height Zaero" to something like "peak AOD height Zaero" to avoid confusion. answer: agreed! we changed to "the height Zaero of peak aerosol concentration" Lines 8–9, page 42: the water vapour lines are not present in this figure. answer: agreed. We changed the caption text. Figure 27: what are the vertical axes on the right? In addition, it is hard to see, but there is a red line very close to the horizontal axes. What is that? answer: The figure has been corrected and redrawn. Congratulation for your accurate viewing. Lines 12–19, page 42: those are repetition of lines 36–40, page 4, but not exactly the same. The same argument does not need to be repeated twice in the article. answer: in page 4, we illustrate the difference of width between absorption and emission, illustrated by figure 2; while in page 42, we show with the figure 27 the

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CIA. In addition, we write:” ..as noted before...” Figure 28: The “@1.27 $\mu\text{m}$ ” may be removed in the vertical axis label. answer: it could be removed but for practical reasons it will not be removed. Line 18, page 49: The REPROBUS model is coarse and cannot capture small-scale intensity variations, if they exist. answer: we have modified the sentence: “They should be very similar, if the characteristics of spatial lengths of intensity variations are as large as found by REPROBUS model, larger than the 2x2° REPROBUS resolution. This comparison would provide an important “sanity check” of the retrieval of Psurf (or O2 column). “ Line 4, page 50: what is the resolution of the “very high resolution” spectrum? used by 4AOP? answer: the spectral sampling used in 4AOP is adjustable by the user. In the exercise that were done, the sampling was 0.001 cm<sup>-1</sup>. Done. Lines 23–26, page 50: what are the a priori errors, especially for Psurf and airglow scaling factors? answer: On most inversion exercises, the a priori errors were large: H2O : 2145.16 ppm; Psurf : 5 hPa ; mean Albedo : 1 Scaling factor of Airglow : 1000 (totally relaxed) except in case#2 where it was taken at 0.2 (20%). What is the impact of a priori on posterior error? answer: we have not done studies of this type and it could be interesting to do them in the future.

Does the airglow impact retrieved XCO2? answer: if the airglow is not accounted for (ignored in the retrieval and direct model), then it induces an error of 70 hPa on Psurf and a correlated 7% error on XCO2.

Line 29, page 50: what is “Lmoy”? answer: the definition of Lmoy is given in the following line. We have modified the etx for better clarity: “We remind that the MicroCarb requirements on the Psurf retrieval for a median intensity luminance Lmoy scenario are 0.1 hPa in term of bias and 1 hPa in term of random error. This reference luminance value Lmoy corresponds to an observation with SZA=36° and albedo at 1.27 $\mu\text{m}$  = 0.2.” Lines 10–16, page 51: the cold/warm spectra here differ from what has been defined in equations 29–30 (spectra simulated at 217/270 K). Does this make any difference? answer: no, it does not make any significant difference. Line 31, page 56: change “dayglow” to airglow. Done. Line 39, page 56: may remove the exclamation mark after

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“contaminated”. Done.

Lines 13–15, page 57: the comparison with day-side GOMOS ozone profiles appears to be inconclusive according to section 4.3.2 and may not support the statement here.

Some pptrs of section 4.3.2 have been rephrased; nd here we have modified the sentence, in such a way that there is consistency between 4.3.2 and the sentence her: “For the time being, we assign this deficit to be due at least partially (but possibly not totally) to an ozone deficit in the REPROBUS model,...”

Lines 31–32, page 57: the first sentence may be inaccurate because a few factors stated previously in the manuscript. The absorption is still saturated while airglow emission grows linearly, the absorption is much more pressure-broadened, and only CIA is present in absorption. All those factors still hold even the airglow spectral shape were identical to absorption. answer: We still think that our sentence is correct: “...if the dayglow spectrum of O2\* were strictly identical to the O2 absorption spectrum,”: strictly identical is implying CIA, same pressure broadening at the same pressure, etc... Line 5, page 58: the O2 a1\_g band is significantly more complicated than just P, Q, and R branches. The same argument applies to discussion of transitions in sections 2.3.3–2.3.5, which is suggested to be removed. answer: The P,Q,R branches contains the most intense lines of the transition. One advantage to use the LBLRTM code based on the HITRAN absorption data base, and then multiply by a smooth function of wave number to get the emission, is that the HITRAN data base contains all the lines (within a certain wavelength interval) above a very small line intensity threshold, including for instance some of the lines V=1, V”=1, and some lines of branches N,O, S,T. Lines 31–32, page 58: I’ve found CIA being considered in Figure 3 and Eq. S5 in Sun et al. (2018), so it is inaccurate to state that “their whole analysis was done without accounting for the CIA O2 absorption”. answer: You are right. Therefore we have modified the text there: “Their whole analysis was done with already accounting for the CIA O2 absorption, whose broad size and smooth pattern is insensitive to spectral resolving power (Fig. 27). On the other hand, as can be seen in Fig. 27 with the same num-

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ber of spectels as MicroCarb and a coarser spectral resolution (and sampling), the whole O<sub>2</sub> band would be measured and would possibly allow to better constrain the CIA absorption and O<sub>2</sub> column retrieval.. The larger spectral sampling gives additional photons per spectel, which may be traded-off for an increased spatial resolution. However, the high resolving power of MicroCarb is an asset for the exact knowledge of the instrumental spectral function which is important for the retrieval accuracy.”

and further down, we modified also slightly: “We suggest though, on the basis of our analysis and the results of Sun et al. (2018), that when CIA is taken into account, a spectral resolving power of about 5,000 and a high SNR could possibly yield a sufficiently good accuracy...”

Please also note the supplement to this comment:

<https://www.atmos-meas-tech-discuss.net/amt-2019-54/amt-2019-54-AC3-supplement.pdf>

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