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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric Band Fitting Coefficients Derived from Self-Consistent Rocket-Borne Experiment" *by* Mykhaylo Grygalashvyly et al.

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Dear Reviewer.

Thank you for taking a time to review our manuscript. We were trying to follow your suggestions.

In the following we address the comments of the reviewer point by point.

Reviewer write: "First, I do not consider necessary many of the analysis devoted to atomic oxygen direct recombination process. So Figures 2 and 3, and many of the discussion refereed to them, should not be here. Once the authors obtain from direct

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recombination analysis that an efficiency of 0.07-0.13 is required for O2(b) production means that this process, alone, can not explain O2(b) production ...."

McDade et al. (1986) made the conclusion that one-step mechanism is not working not because their efficiency to high, but because it depends on altitude, that is impossible by the essence of ε. They are writing: "The altitude dependence of ε suggests that the observations are not consistent with the direct excitation mechanism unless the efficiency for formation is strongly temperature dependent." Because our experiment is essentially differ (common volume observations) from former one, we should repeat the steps of McDade et al., (1986). Hence, figures 2, as well as short corresponding discussion should be saved. Please, approach with understanding. Nevertheless, in order to satisfy the reviewer suggestions, we delete figure 3 and radically reduce the discussion of one-step mechanism and made it as short as it is possible.

Reviewer write:" ...then, all additional the work of considering only direct recombination process and possible dependences of the efficiency with temperature and pressure is very speculative, first because of the large value of this efficiency and second because in the altitude range of the atmospheric region considered ($\sim$10km) there is not large variation of temperature and pressure condition (although they could reach +-50K and/or a few micro bars)."

The temperature enters into Eq. (2) via coefficients , , under the exponential and power functions, hence, even small fluctuations of temperature produce essential deviations.

Reviewer write: "Figure 4, 5 and 7 need some improvements: 1) Plot some subintervals in the vertical and horizontal axes. 2) There are points (data) only at about each 3 km. Points at each 1 km should be shown, although the error bars be shown only each 3 km. 3) Figure 3 shows very good fit between about 97 and 98 km (as I can guess in figure 3 without any subdivision!). It will be interesting to show in the same figure the temperature profile (with and appropriate temperature scale in the upper horizontal axis), and the number density to see how the structure of temperature and number

density can affect the fitting."

We modify figures 4, 5 and 7 according your suggestions: 1) we plot some subintervals in the vertical and horizontal axes; 2) we prefer to use 3 km step at figures 4 and 5 (ex. Fig.5 and Fig. 7) because it is more appropriate to the volume emission observations (see Hedin et al., 2009); 3) we think that you mean figure 4, because the fit shown on figure 4, but not on figure 3. We bring our apologies that do not show in the same figure number density and temperature. This does not give any information how the structure of temperature and number density affects the fitting, because: there are three parameters O, T, M; eq. (4) non-linear, and the effects of these parameters not obvious; thus, we may not distinguish influences of different parameters. Moreover, this is not the subject of our paper and it led to defocusing.

Specific suggestions of the reviewer.

Abstract.

Reviewer write:" Line 19- 20: ",we derived the empirical fitting coefficients,....(0,0) in terms of the atomic oxygen concentrations." Delete "in terms of the atomic oxygen concentrations" To read: ",we derived the empirical fitting coefficients,....(0,0)."""

Lines 19- 20 are changed according to Reviewer suggestion.

Line 25: Reviewer write:" Simultaneous and true common volume measurements of all the parameters used in this derivation, i.e...." Change to: Simultaneous measurements of all the parameters involved in the theoretical calculation of the observed O2(b) emission, i.e...."

"Common volume measurements" is a special term well known and generally accepted for in-situ observations. Hence, we should save this term. The rest of the sentence is modified according with Reviewer claim.

1. Introduction.

Reviewer write:" Line 33-35 Change: "particularly, by emissions in the Atmospheric Band that form the excited state of molecular oxygen O2(b)..." To read " particularly, by the emission of the Atmospheric Band which is produced by the emission of the excited state of molecular oxygen O2(b)..." Lines39-40 Change "Lopez-Gonzales" to "Lopez-Gonzalez" Line 66 Delete "O or" Line 67 Change "by known" To read "and" Line 67 add "values" to read " volume emission values" Line 70 change "leads to the loss of self-consistency (e.g Murtagh et al., 1990)." To read "leads to some degree of uncertainty (e.g Murtagh et al., 1990)." Line 70-71. Delete "and, consequently, to essential biases." To read "(e.g Murtagh et al., 1990)." Line 73-74 Delete " real common volume in-situ measurements of these..." To read: "simultaneous measurements of these..." Line 75-79 Change "chapter" To read "section""

Lines 33-35 are changed according to Reviewer suggestion. Lines 39-40 are corrected as Reviewer suggest. Lines: 66-67. The sentence is rewritten more clearly. Line: 67. The word "values" is added after the words "volume emission" as Reviewer suggest. Line 70 is modified according with Reviewer suggestion. Lines 70-71 are changed deleted by Reviewer suggestion. Line: 74. We save "common volume" as special term (see answer above), but the rest the sentence is modified according with Reviewer wish. Lines: 75-79. The word "chapter" is changed to "section" through the entire manuscript.

2. Rocket experiment description.

Reviewer write: "Here I have a question, it is described that FIPEX use two types of solid electrolyte sensors platinum electrodes sensitive to both molecular and atomic oxygen, whilst gold electrodes show a selective sensitivity to atomic oxygen. Is this mean that it can measure molecular oxygen too? If it is possible, It would be useful this O2 profile were shown in the plot."

We use the CONE number density measurements and partial partitioning from NRLMSISE-00 reference atmosphere (Picone et al., 2002). The FIPEX O2 density

measurements are not used: for reasoning and discussion see Eberhart et al. (2015). To avoid such confusion we rewrote the FIPEX experiment description.

3. Theory.

Reviewer write: “All this section is just a very detailed recompilation of the known mathematical expressions used in O2b calculations. There are too many details that should be reduced. I think expression (2) should be deleted is already said in line 135-136 (same that equation 1). Then expression (4) and (5) should be deleted and put only expression (6). Then delete expression (7), it is not needed, and write expression (8). Then (1),(3), (6) and (8) expressions could be written, to easy understand the "nomenclature".”

All of Reviewer suggestions are applied. Equations (2), (4), (5), and (7) are deleted.

4. Results and Discussion.

Reviewer write: “Comment: Figure 1, shows atmospheric concentration, [N], temperature, T, atomic oxygen concentration, [O], and Volume emission rate. Here Figures 1b) and 1c) should have additional subintervals in the logarithmic x axis. Additionally molecular oxygen concentration is used in the analysis performed. I have a question what values of O2 are used? (is there some additional measurement from FIPEX?, or are derived from the measurements of [N] (density) and [O]?)”

We add subintervals in Figure 1. Molecular oxygen is derived from CONE atmospheric number density measurements and partitioning from NRLMSISE-00 reference atmosphere (Picone et al., 2002). We add the notation into the section 2.

Reviewer write: “Question: Lines 174-177. It said: "Our rocket experiment shows an essential difference of emissions between ascending and descending flights (see Strelnikov et al., 2018). It also demonstrates a significant variability in other measured parameters, including neutral temperature and density as well as atomic oxygen density." How large is this difference? The time between ascending and descending flights

should be a very few minutes. Perhaps it could be interesting to show the profiles of the different parameters obtained in both, the ascending and descending, flights to see these differences."

We add the references where such difference is shown and discussed. Following Reviewer advice that the paper should have better focus we do not repeat this discussion in our manuscript.

4.1 One-step mechanism.

Reviewer write: "This subsection has to be very simplified. There is a lot of speculative exercise that leads to any point."

Situations in which the science allows a formulation of several reasonable alternatives, and it is impossible to show convincingly that only some one of them is right, are characteristic of all fields of scientific research. In this case researcher should discuss all available alternatives.

Reviewer write: "For example: The efficiency calculated by using direct atomic oxygen recombination for the production of O2b to explain the observed emission, is in the range of about 0.07-0.13, this value is too large compare with the obtained by laboratory and theoretical investigation (Wraight, 1982; Ogryzlo et al., 1984; Bates, 1988). Then, direct atomic oxygen recombination alone can not explain the observed emission in agreement with earlier works (McDade et al., 1986; Bates, 1988;...) Any other exercise is not necessary. So figure 2 y 3 should be deleted.".

As we discuss above, the figure 2 is saved but the section essentially simplified and figure 3 is deleted.

4.2 Two-step mechanism.

Reviewer write:" Figure 4. Please, put some subintervals specially in the y axis. Here I see a very good fitted region of about 97-98 km (as I guess because of the lack of subintervals!), and important deviation of the fit above and below this region. I

think it would be interesting to show, simultaneously in the figure, T profile (with a horizontal scale from 150K to 210K, in the upper x axis) and the N profile (the same scale as RHS Eq.8, in the lower x axis, is appropriated) to easy see how temperature and atmospheric density profiles affect to the features observed in RHS equation 8."

To address the Reviewer's point we put subintervals in figure 4. T and M on figure 4 does not give an ability to see how the temperature and number density affects the RHS, because beside T and M there is third parameter (O) and one non-linear eq. (4) from which we may not distinguish influences of different parameters. Moreover, this is not the subject of our paper and it led to defocusing.

Reviewer write: "Line 259: Delete "s" to read " and molecular oxygen" Line 261: Change "are 3.1 and 2.9" to read "of 3.1 and 2.9" Line 282-283 Change "we show Figure 5 with atomic oxygen concentration from... " To read "we compare in Figure 5 the atomic oxygen concentration from ..." ((Figure 5 and Figure 7 need to show subintervals in both axes (vertical and horizontal), also the data points should be plotted each 1 km.))"

Line: 259. "s" is deleted. Line: 261. Changed. Line: 282-283. Changed. The subintervals in both axes at Figure 5 and Figure 7 (now Fig. 4 and Fig. 5) are added. We save resolutions 3 km as it is more relevant to volume emission observations.

4.3 Combined mechanism

Reviewer write:" Figure 6 is not necessary, all the information is in table 3."

This contradicts to (*). Nevertheless, in order to satisfy the Reviewer suggestion we delete Figure 6.

Reviewer write: "I do not think to put the equation (9) is needed. Neither do I consider an appendix necessary. So I will deleted the appendix and will reduce the equation 9 and the explanations to: "... we have investigated a combined mechanism of direct and indirect atomic oxygen recombination, the fitting coefficients for the transfer energy

process were calculated for.... The results for the best-fit in each case are listed in Table 3." (Now in Table 3 only K3o/k3o2 and total efficiency would shown (delete D1 and D2)) “

We need this equation because it used to calculate fitting coefficients, hence, we have to show how it was derived, consequently, we save an appendix.

Reviewer write: “Line 305-310 Delete all.. "They are listed in Table 3. The altitude profile of the RHS of equation (9) and calculated fit-function are plotted in Figure 6. The deviations of fit function between limits and averaged values are negligible, hence, we only show the averaged case. Thus, we can recommend for future investigations the values of averaged case (last column of Tab. 3). Analogously to the two-step mechanism....<1.Taking into..." To read: "The results for the best-fit in each case are listed in Table 3. Taking into..." “

This contradicts to (*) and (**). Nevertheless, in order to satisfy the suggestion of the Reviewer we delete lines 305-310 and add “The results for the best-fit in each case are listed in Table 3.”

Reviewer write:” LIne 311. Add (see Table 3) to read "0.073 (see Table 3).”"

We add “(see Tab. 3)”.

Reviewer write:” Line 327-330. Delete this sentence "Note that... two point, respectively." It is not needed.”

This contradicts to (*). Nevertheless, in order to satisfy the suggestion of the Reviewer the sentences at lines 327-330 have been deleted.

Reviewer write:” Line 330-332. Rewrite this sentence. For example: The total efficiency of production of O2b through an energy transfer process and new coefficients derived in this work provide a valuable information about the chemistry of O2b. Moreover, the importance of make studies with the possibility of using simultaneous measurements is strongly pointed out.”

This contradicts to (*) and (**). Nevertheless, in order to satisfy the Reviewer the sentences were rewritten.

Conclusion.

Reviewer write: “This section has to be very summarised. There is a long text, and here the results has to be clearly established.”

Following by you suggestion the text of conclusion has been reduced.

Reviewer write: ”Line 336: Change "true common volume observations" to read "simultaneous observations". ”

“Common volume” is special term (see answer above). Trying to follow your suggestion we change "true common volume observations" to "common volume simultaneous observations".

Reviewer write: “Line 337-338 "delete one-step. two step and combined" to read "the mechanism of o2b formation were analysed".” We delete “one-step. two step and combined” to “the mechanisms of formation were analysed.”

Reviewer write: “The following discussion for one-step and two-step should be deleted. These are from line 339 to 356. Only the proposed mechanism should be mentioned.”

This contradicts to (*) and (**). In this paper we discus three mechanisms, hence, in conclusion should be highlighted all of them. Nevertheless, in order to satisfy the suggestion of the Reviewer we essentially reduce the conclusion owing to one-step mechanism.

Reviewer write: “Line 357-360 are when the main result are reflected. This can be rewritten as: Based on simultaneous observations of atomic oxygen, atmospheric band emission (762 nm), and density and temperature of the background atmosphere and all the information available up to now about reaction rates coefficients, branching ratios, quenching rates and spontaneous emission coefficients the mechanism of o2b

formation were analysed. A direct and indirect atomic oxygen recombination process to explain the production of O2(b) is the one chosen as responsible of the atmospheric emission observed. The total efficiency of production of O2b in the indirect recombination process is of 0.08 and the ratio of quenching coefficient of the precursor state is 0.231, when an efficiency of 0.02 in direct recombination is chosen. The analysis of the values of the total production indicates that O2A' or O2Pi may be possible precursors for the two-step mechanism."

We do not include the reviewer text instead of our conclusions because this contradicts to (*).

Reviewer write: "The lines 361-366 reflect the final thoughts, so here Lines 361-366 ramble on about these mechanisms again in a confused way. Instead, it should show the need to make simultaneous measurements to confirm and improve these results."

Note, lines 361-366 were written by authors. The Reviewer term "ramble on about" unacceptable. This contradicts to (*) and (**), hence, we save this formulation as it is. Nevertheless, in order to satisfy the suggestion of the Reviewer we include at the end of the conclusion the statement of the Reviewer about more simultaneous measurements in future to confirm and improve these results.

Thank you again.

With respect, M. Grygalashvyly, M. Eberhart, J. Hedin, B. Strelnikov, F.-J. Lübken, M. Rapp, S. Löhle, S. Fasoulas, M. Khaplanov, J. Gumbel, and E. Vorobeva.

(*) 3. A referee of a manuscript should judge objectively the quality of the manuscript and respect the intellectual independence of the authors. In no case is personal criticism appropriate. (**) 7. Referees should explain and support their judgements adequately so that editors and authors may understand the basis of their comments.

(General obligations for referees, ACP web page, https://www.atmospheric-chemistry-and-physics.net/for_reviewers/obligations_for_referees.html)

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