

Referee 1

Many thanks for your detailed review and your feedback. All our responses or comments are written in green through the text.

General comments

Piel et al. describe the development of a cavity enhanced spectrometer for the simultaneous measurement of dioxygen concentration and its oxygen-18 isotopic composition. The instrument performances are excellent, reaching detection limits of 0.002 % for O₂ concentration and 0.06 ‰ for $\delta^{18}\text{O}(\text{O}_2)$ in 20 minutes. This development is of significant interest to the scientific community, with multiple environmental applications. However, although the manuscript is highly relevant to AMT's readership and overall well-written, it needs major improvement before being considered for publication.

Specific comments

- The instrument description should be rewritten including: a description of the instrument, showing all the elements currently used in the setup (pressure sensor, flow sensor, solenoid valve, mirror, photodiode) and specifying the product reference if commercial. A point-to-point comparison with the reference article should not be made. A technical scheme and an instrument picture should be provided.

The manuscript already provides detailed explanations (we added even more details and tried to improve their clarity) about the special modification of the instrument relative to previous realizations, for which we give references where the working principle of the technique and all details about the realization of different prototypes are fully explained. It would be completely redundant to provide an experimental scheme and description of the technique and of the working principle of the instrument which would not be different from what is presented in the cited references. In addition, since the presented instrument was realized by a private company, we cannot give details concerning the software and the control electronics or the model of pressure and temperature gauges which were selected by the company, which represent proprietary information and are not commercially available as individual parts (notably, the control electronics). In addition, we note that several works were published in this journal mentioning the exploitation, possibly with modifications, of industrial instruments (chromatographers, mass spectrometers, but also optical devices by Picarro or Aerodyne Research, eg. Berhanu et al., 2019; Kooijmans et al., 2016; Lebegue et al., 2016) without providing detailed descriptions of those instruments.

- In the results section, there is not enough explanation of how the tests were conducted and there are no data/figures to justify the conclusions given. The link between allan variance and

the time used to carry out the measurements is missing. The measurement strategy used should be explained in more detail.

An effort was made to clarify the result section.

- When results are given, they should be associated with uncertainties and the supplement should explain how they were obtained and the confidence interval chosen.

The Allan variance and response time (fig.5) plots are the results of a single, but highly repeatable measurement. An effort was made to clarify how the results were obtained.

- Overall, the manuscript is lacking details. It should be revised with additional data to support the development of the instrument.

While we added all specific details about this instrument that we could provide, we actually removed mention of the fact that we tested 2 different diode lasers since we realized this point of development was uninteresting to the readers of this journal.

- The overall structure of the manuscript should be revised. For some sections, the manuscript is written more in the form of a report than a scientific article. The authors should better guide the reader through their instrumental development methodology.

We kept the overall structure, but an effort was made to improve the manuscript and better guide the reader.

- There are numerous wordings that need to be revised.

A strong effort was made on this point.

- Greater attention should be paid to defining words and acronyms.

Indeed, we think we fixed all such problems.

- More references are needed throughout the manuscript.

References were added through the manuscript, in particular relative to past applications of the OF-CEAS technique, containing again descriptions of various similar realizations.

Technical comments

- All small deltas (δ) must be written in italics as “ δ ”. It has been done in all the manuscript

Title

- $\delta^{18}\text{O}$ should be defined: “High precision oxygen isotope ($\delta^{18}\text{O}$) measurements of ...” Done

Short summary

- Line 14: The temporal resolution and precision of measurements should be given. **Added**
- Line 15: $\delta^{18}\text{O}$ and O_2 should be defined. **Done**

Abstract

- Line 19: “(O_2)” should be placed after “Atmospheric dioxygen”. Then, only O_2 should be used throughout the manuscript. **Modified in all the manuscript**
- Line 20: CO_2 should be defined. **Done**
- Line 24: “isotopic” should be added between “oxygen” and “fractionation”. “occur” is missing an “s”. **Done**
- Line 26: Please add “isotopic” before “fractionation coefficient”. Of which isotopic fractionation coefficient are you talking about? **Done**
- Line 25: “($\delta^{18}\text{O}(\text{O}_2)$)” should be added after “ $\delta^{18}\text{O}$ of O_2 ” and then only $\delta^{18}\text{O}(\text{O}_2)$ should be used. **Modified in all the manuscript**
- Line 28: Please reverse “OF-CEAS” with “(Optical-Feedback Cavity-Enhanced Absorption Spectroscopy) as “optical-feedback cavity-enhanced absorption spectroscopy (OF-CEAS)”. Capital letters are not necessary. **Done**
- Line 33: “instrumental” should be added before “drift”. **Done**
- Line 33-35: need to be more quantitative on humidity and O_2 concentration effects. **We made it quantitative for O_2 concentration effect and decided to keep it this way for humidity effect. We explained in more details within section 3.2 the potential effect of water vapor and the solution we applied.**

Introduction

- Line 38: “ O_2 ” should be added after “Dioxygen” and then only O_2 should be used throughout the manuscript. **Modified in all the manuscript**
- Line 48: the $\delta^{18}\text{O}$ notation should be defined explicitly. “($\delta^{18}\text{O}(\text{O}_2)$)” should be added after “ $\delta^{18}\text{O}$ of O_2 ” and then only $\delta^{18}\text{O}(\text{O}_2)$ should be used throughout the manuscript. $\delta^{18}\text{O}_{\text{atm}}$ is useless as it is not used later in the manuscript. **Modified in all the manuscript**
- Line 57-59: $\delta^{17}\text{O}$, Ar, and $\Delta^{17}\text{O}$ should be defined. **Done**
- Line 78: Please reverse “CRDS” with “(Cavity-Ring-Down Spectroscopy) as “cavity ring-down spectroscopy (CRDS)”. **Done**
- Line 80: As isotopic ratios are expressed in per mill throughout the manuscript, the associated error should be expressed in the same unit. **Done**
- Line 83: same comment as for line 28. **Done**

Material and methods

- Line 95: the reference is not cited correctly, should be “described in Morville et al., (2005).” **Done**

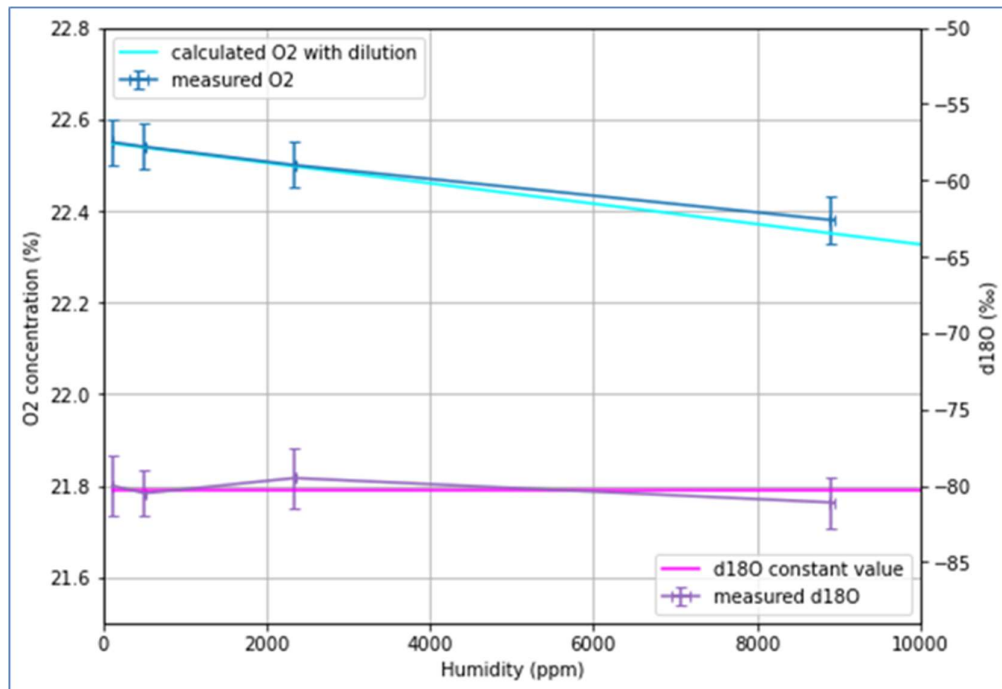
- Line 97 : What field applications? References should be given to provide examples. Added
- Line 98 : Simply providing a link to the company is not appropriate. More details should be given. We added there a paragraph to clearly explain that the presented instrument actually follows the principle of operation of standard OF-CEAS as described in previous cited works (of which some were added to provide more examples of specific applications as requested above). We hope that now it is clearer that all details, discussions, and schemes of the experimental setup were already provided in previous publications and that it would be completely redundant to add a figure and a couple of pages of explications about OF-CEAS as that are already available in the literature (as much as it is for Mass Spectrometry or, to give a closer case, Cavity Ring-Down spectroscopy for Picarro instruments).
- Line 99 : “Some specific demands”: which ones? Now explicitly written in the manuscript
- Line 100: “DFB” should be defined as “implementation using a distributed feed-back (DFB diode)”. Then only DFB should be used. Modified in all the manuscript
- Line 108: “the figure” should be replaced with “Figure 1”. Done
- Line 121-124: Data should be provided. Done
- Line 124 - 126 : “Instrumental drift, assessed by the Allan deviation as presented below, should then remain below the desired precision level over the measurement time for two samples of which one would be a reference”. This sentence does not bring necessary information. We actually think that this sentence is useful since it explains that we need to measure both sample and standard during a period of time not strongly affected by the drift. We have rewritten this sentence to be more precise:
“We then need to consider the instrumental drift (assessed by the Allan deviation as presented below) which should then remain below the desired precision level over the measurement time for two samples (~ 20 minutes) of which one would be a reference. “
- Line 142 : What is “working pressure”? Define here your cavity pressure. “as usual” should be removed. Working pressure is defined on the next line, we added it again after “working pressure”. “As usual” refers to OF-CEAS, we changed it to “As usual in OF-CEAS”
- Line 144-146 : “Another improvement is a more accurate, stable and fast control of the sample pressure inside the measurement cell, which is also important for low drift.” There is an important lack of information here. More elements should be provided. For example, a figure should be added and comparative values for any improvement of the instrument. This improvement, as specified, was implemented by the AP2E company. Apart from the fact that it is obvious that by improving system parameters stability decreases the measurement drifts in any such kind of spectroscopy-based instrument, we cannot provide additional proprietary information and a comparison of the performance before and after the stability improvement of AP2E instruments. Such tests were conducted at AP2E. We do provide however Allan plots of the presented instrument which illustrate well enough its rather long stability time.
- Line 154 : “Well known” should be removed and references for HITRAN spectral database should be given. HITRAN data base is indeed well-known, so we think we do not need to change the sentence. We add an online reference to the most used HITRAN web site (<https://hitran.iao.ru>) which is found as a first search result by typing “HITRAN” in Google.

- Line 156: “this point will be addressed below”. Without going into detail here, a few elements can be given here. It is “addressed below” because it cannot be addressed in short there. We maintain our text.
- Line 160: The delta notation should be defined when it is first used in the manuscript, i.e. on line 48. “permil” should be written as “per mill”. Done
- Line 162: “reference sample” of what? In order to be more precise, we changed it to “...reference gas sample relative to which the delta will be defined, ...”
- Line 163: “simple” should be removed and equation numbers should be added. Modified for all equation of the manuscript
- Line 164: Any equation given in the manuscript should have a number. Modified for all equation of the manuscript
- Line 170: How do you obtain the value 35 000? The cavity finesse is a standard optical cavity parameter which can be obtained from cavity length and measured ringdown time, as described in some of the cited papers. As we actually provide the cavity finesse for the 2 used system configurations in another section of the manuscript following this one, we remove “(35000)” from here as the exact value is actually not important for the discussion at this point.
- Line 188-189: what is the software used? “This works well”, please be more quantitative. The software, as mentioned in the added paragraph in a previous paper section, is proprietary of the AP2E instrument. We can’t give more details. We changed “well” with “perfectly”.
- Line 198 : What is the frequency dispersion of the cavity modes ? They are not absolutely fixed. Actually, as explained, the modes frequencies are determined by the cavity length (as also discussed in cited papers), and since the cavity is temperature stabilized by locking the position of the modes relative to the absorption lines, the modes have very well defined and stable frequencies.
- Line 200: References should be provided for the Rautian and Voigt profiles. References were added.
- Line 204: “Over the time span of presented results (18 months)”. It's not clear what the point of this information is. As mentioned, a few lines later: “In the following, we will specify which setup was used for which results, accounting which will account for somewhat varying performances”
- Line 208 : Add the cavity finesse value. For both cavity configurations, finesse and mirror reflectivity are now specified.
- Line 218-220: The symbol “®” should be added for any deposited trademark cited throughout the manuscript. PFA should be defined. Done
- N2 should be defined. Done
- Line 227: What is the difference between mode (2) line 221 and the routine mode? Why no longer use a trap with magnesium perchlorate? The term “routine mode” was indeed not appropriate. We needed to better precise “when measuring atmospheric air” and in this case, we indeed use magnesium perchlorate filter in the PFA tube. We have corrected it.
- Line 232: There is no need for a “-” between “Isotope” and “ratio”. Done

- Line 235: If this manuscript is to be published, the reference given must have been published previously. If this is not the case, further details will be required. It seems to us that sufficient details for the purpose of this paper are given in the rest of the paragraph.
- Line 241: A number should be given to the equation. Besides, the expression to calculate the O₂ concentration should be given explicitly. Equation number was added. However, the required expression is trivially derived from the given equation, which is more readable and meaningful than the derived one. We do not see the use of writing out the derived expression.
- Line 248: “is” should be “of”. More details are needed for the peak jumping sequences. Thanks! That was corrected.

Results and discussion

- Figure 2 and 3: A different color palette should be used. Black and green are not color-blind friendly. We checked using the Coblis Color Blindness Simulator: our figure is visible for all types of color blindness, except monochromacy.
- Line 252-253: This information can be provided earlier and not in the results section. Done, it now also appears in the introduction
- Line 254: What is allan deviation? A reference should be provided. Modified
- Line 260-261: The minimum of the allan deviation is not reached at the same time for the oxygen concentration and the isotopy. The time required to reach the minimum for each species must be given with the precision. Done
- Line 264 : The figure is complicated to understand because of the y-axes. The figure has been modified.
- Line 271: It should be clarified what is considered as a “moderate shift” and “regular measurement. Modified.
- Line 276: The time chosen for the measurement must be explained. A sentence was added: “This calibration frequency, higher than required by the Allan deviation discussed above, was chosen as a compromise towards obtaining measurements with high time resolution.”
- Line 277: How was the time interval between each injection of standard selected? See response to previous point.
- Line 284 : The concentration should be kept on the same side of both graphs of figures 2 and 3. Modified.
- Line 290: Any results from the secondary configuration should be provided in a supplement. We do not see any advantage in moving the results of the secondary configuration in a supplement. It seems to us that it takes very little place in the manuscript, and the difference between configuration is clearly stated within the results section.
- Line 293: Data should be provided to support this statement. Because we remove the water, we only characterize with a few data points the dependency of d₁₈O and O₂ on humidity. The graph is provided below and can be included in the revised version of the manuscript if needed. The dependence of the O₂ concentration is only due to the dilution of the O₂ signal by the added water vapor quantity.



- Line 294: This sentence needs rewording. **Done**
- Line 300: A reference should be provided. Overall, the structure of section 3.2 should be revised. **Actually, contrary to what was expected from knowledge on other molecules, a reference was found (and cited) showing that the effect of pressure broadening of O2 lines by water vapor is not much larger than by other atmospheric molecules (O2 itself and N2). Thus, the discussion in this section was modified accordingly.**
- Line 303: The section title should be revised. **Done**
- Line 304: This sentence needs rewording. **Done**
- Line 314: The linear regression data should be provided in the text. Besides, the given increasing rate of $\delta^{18}\text{O}$ with O2 concentration seems wrong based on Figure 4. **Modified**
- Figure 4: The overall figure display should be improved (e.g., add label ticks, regression equation, ...). The symbol for the per mill unity should be used. The errors on the slope and intercept of the linear regression should be provided. **Done**
- Line 321: the section number where the initial configuration is described should be added. **Done**
- Line 323: Too general, should be more precise. **Modified**
- Line 325: Why every 15 days?
We did it regularly (i.e. every 15 days) during experiments due to our lack of hindsight on the instrument, which could be considered at that time as a prototype. Clarified in the text.
- Line 326: The section title should be revised. **We propose “Memory effect and response time”**
- Line 329: The flow rate used for purging must be specified
The flow rate for purging is identical with the one used for measuring. Clarified in the text.
- Figure 5: there is a typo in the figure legend. **Corrected**
- Line 335: Any results from the secondary configuration should be provided in a supplement.

See our answer for your comment about line 290.

- Line 336: The overall structure of section 3.5 should be revised which is not appropriate for an article. Done.
- Line 353: “small 1σ ” should be quantify. Done
- Line 367: This section critically lacks details. We added more details.

Conclusion

- Line 390: The unity used throughout the manuscript should be homogenized. Corrected. We used “%” everywhere
- Line 400-402: Further details can be given on the instrument’s application. 3 examples were given.

References

Berhanu, T. A., Hoffnagle, J., Rella, C., Kimhak, D., Nyfeler, P., and Leuenberger, M.: High-precision atmospheric oxygen measurement comparisons between a newly built CRDS analyzer and existing measurement techniques, *Atmospheric Meas. Tech.*, 12, 6803–6826, <https://doi.org/10.5194/amt-12-6803-2019>, 2019.

Kooijmans, L. M. J., Uitslag, N. A. M., Zahniser, M. S., Nelson, D. D., Montzka, S. A., and Chen, H.: Continuous and high-precision atmospheric concentration measurements of CO_2 , CO and H_2O using a quantum cascade laser spectrometer (QCLS), *Atmospheric Meas. Tech.*, 9, 5293–5314, <https://doi.org/10.5194/amt-9-5293-2016>, 2016.

Lebague, B., Schmidt, M., Ramonet, M., Wastine, B., Yver Kwok, C., Laurent, O., Belviso, S., Guemri, A., Philippon, C., Smith, J., and Conil, S.: Comparison of nitrous oxide (N_2O) analyzers for high-precision measurements of atmospheric mole fractions, *Atmospheric Meas. Tech.*, 9, 1221–1238, <https://doi.org/10.5194/amt-9-1221-2016>, 2016.