A Review of: “New infrared spectroscopy instrument for reliable low humidity water vapour isotopic composition measurements” (2023-2457), submitted by Mathieu Casado et al. to Atmospheric Measurement Techniques (via EGUsphere)

This manuscript puts forward a new measurement setup for measuring stable water isotopes in extremely dry air. Though the core of the measurement technique is based off existing technology, it combines it with additional technology in a novel way. Additionally, the ever-present issue to combat instrument drift in CRDS instruments is addressed in an innovative and creative manner, via frequency referencing with a physical constant.

In this review, I will focus mainly on the use of CRDS technology to measure stable water isotopes in low humidity environments as this is closest to my area of expertise. However, some of the finer details of the spectroscopy are beyond my expertise, specifically Sect 2.3. Nevertheless, I will do my best to fairly assess all sections of the manuscript.

Overall, I very much enjoyed reading and learning about this new development in stable water isotope measurement. The science presented certainly warrants publication, and the manuscript is nearly there. With only a few revisions and better management of the readers' expectations, this work has the potential to become a foundational piece of literature in the further advancement of CRDS technology and the stable water isotope research community.

Major comments

1. My most pressing critique concerns the assertion that the instrument accurately measures isotopic signal down to 1 ppmv. The abstract and introduction mention this capability multiple times, which would be an outstanding achievement and got me very excited to read more of the manuscript. However, in Sect. 3 (Results), the authors show that the lowest experimental measurements are at 25 ppmv and that the precision at 1 ppmv was extrapolated. This undercuts the very good work by the authors to make accurate measurements at 25 ppmv, which is a formidable achievement in itself. Therefore, this claim needs to be rephrased here and elsewhere to comply with the actual measurement results that are shown. For example, I would suggest modifying the initial claims of “1 ppmv” to “10 ppmv”. I believe that this order of magnitude better reflects the experiments conducted by the authors with the calibration unit available to them. I also suggest moving the sentences discussing extrapolation down to 1 ppmv to Sect. 4 (Discussion), with the context of vapour generation limitations. To be clear, I don’t disbelieve the authors that the instrument could be capable of accurate measurements at 1 ppmv if given the chance, however if it remains a “*could*” it should be presented as such.

This should also be reflected in Fig. 8, with some differentiation needed in the bottom section of the OFFS-CRDS grey bar, below 10 ppmv (perhaps a lighter shade of grey or dashed outlining). With this measurement expectation established early in the manuscript, I believe readers will still find the work exciting, while knowing what to anticipate from the instrument.

2. I think that the authors should re-consider how they establish the comparison between the new instrument and the commercial CRDS analyzer. As it stands now, the manuscript portrays the new instrument as an alternative to a commercial device, using a Picarro L2130i as a benchmark. As this is an instrument that many in the field would be familiar with, it serves as a relatable reference for readers. However, unlike the Picarro, the new instrument cannot be deployed to the field in its current form and is much larger and more sensitive in regard to handling than the simple benchtop analyzers. This does not detract from the quality of the scientific advance put forward by the authors but establishing the reality of the instrument in Sect. 1 (Introduction) with a short remark (big, bulky, and fragile) would better manage the expectations of readers.

Minor comments

1. Introduction

The authors do a satisfactory job of conveying the utility of stable water isotopes and the gaps present with their observations. Additionally, an overview of the technology used to detect stable water isotopes is well covered. Numerous, relevant references are made to past bodies of work. Aside from the two issues mentioned in the Major comments, the authors do well to situate readers for the work that follows.

2. Methods

2.1 The authors detail the 3 main components and their workings in a logical manner, paralleling the structure of Fig. 1. While I compliment the authors on the appealing design of the figure, I'm left wondering why the authors didn't try to connect it more to the text, as they only generally refer to it once (L66). I'd suggest re-considering the level of detail included in Fig. 1, making it even more of a bare schematic for the main text. Then the more detailed version could be included in the Appendix.

2.2 Fig. 2 is very nicely designed, with a helpful connection between subfigures a) and b).

2.3 Fig. 3 suffers from the specific placement of the "Lamb-dip" label. At first, I thought I had misunderstood what the Lamb-dip was, and that the sharp decrease and subsequent

recovery in the red line at 90-140 hours was somehow evidence of the Lamb-dip. I believe this misunderstanding should be fixed by changing the location of the label to around the (40,-0.05) area.

3. Results

3.1 The authors separate the findings of their experiments into precision and accuracy, with an additional highlight on the frequency auto-referencing. Aside from the extrapolation item mentioned in my Major comments, the methodology for determining instrument precision and stability is well documented.

3.2 Though it also naturally fits in the explanation of the experimental results, the influence of the vapour generator (L172) and the difficulty of disentangling its impact should be mentioned in Sect. 4 (Discussion), including the lower limits of vapour generation.

3.3 Fig. 4 is also well-designed, with creative linkages drawn between the subpanels, which help to orient the reader. However, the coloured surface in b) is a bit distracting for this purpose. Perhaps a greyscale colourmap would better show the lines drawn for the connections to subfigures a) and c).

3.4 It may also be worth mentioning that the exact isotope-humidity response is unique to each individual analyzer (Weng et al., 2020), so an instrument without any such a response is even more valuable.

3.5 Though complex, Fig. 7 nicely visualizes the concepts presented in the text, especially b), d), and f). I would only suggest keeping the labelling consistent with Fig. 3, since the red "Self referenced" is the Lamb dip method and the blue "External reference" is the optical comb method (right?).

4. Discussion

4.1 The authors do well to put forward how the current work would fit into previous work. The speculated benefit that the instrument would bring to Antarctic field research is very exciting.

4.2 I appreciate that the authors also connect the potential benefit of these low humidity measurements to a concrete scientific aim, which is the fractionation between gas and solid phase at such extremely low temperatures. If it is actually feasible, I suggest adding a brief comment on the possibility that the new instrument could measure this fractionation in lab experiments, inside which it seems the instrument would be most comfortable in its current iteration. Such an application would be a further demonstration of its value and utility.

4.3 As the instrument is being compared to a commercially available Picarro, consider a qualitative comparison of the financial cost, even if this comparison is as simple as “much more/less” or similar.

5. Conclusion

5.1 The authors concisely present the key findings of the work, though the term “cheap telecom” or similar was absent from the rest of the manuscript. As mentioned above, including some indication of cost in Sect. 4 (Discussion) would support this conclusion.

5.2 The viability of the frequency reference technique is deservedly highlighted in its own right. By finishing with a statement focusing on the lower measurement limit of the instrument, the authors nicely echo the title of the work, and end on a definite.

6. Appendices & References

6.1 As mentioned above, the influence that the vapour generator might have on the experiment should be touched upon in Sect, 4 (Discussion), but the authors made good use of Appendix A to contain the more specific details.

6.2 A similar point can be made for Appendix B, which concisely justifies why the authors focused on $\delta^{18}O$ and not δD. However, I would suggest renaming the current appendices to B and C, and making a new Appendix A, which contains the detailed breakdown of the current Fig. 1.

Detailed comments

- L21: Consider “stable water isotopes” as this word order is used in L159.
- L39: This sentence is rather long, and the knowledge gap (no instrument able) is buried in the middle. Consider separating it into two. For example: “Ritter et al., 2016). This is despite attempts to develop a new generation”
- L41: Consistent spacing between value and unit “20 ppmv”. Not necessary for % or ‰
- L54: 0.06 Pa
- L55: -90 °C
- L56: “or on other planets.”
- L71: Include abbreviation expansion in the figure caption (e.g. MZM).
- L81: “which enables us to tune" or “which enables tuning of the frequency”
- L85: 0.9 mW
- L94: 0.01 mbar

- L100: Consider explicitly connecting the mode established in the previous paragraph end sentence to the opening of the new paragraph. For example, “In full spectrum mode, the instrument has a high spectral resolution and can be used”
- L101: Does the word “Here” refer to the high spectral resolution mode mentioned in the previous sentence, or is it being used to establish the other high pace mode used in this work? Again, consider using explicit reference to the previously established mode names introduced at the start of Sect. 2.2 (full spectrum vs. high pace)
- L101: The phrase “water vapour isotopic,” doesn’t really fit here. Maybe “isotopic water vapour,” or “water vapour isotopes,” fits better.
- L104: Remove the word of: “This is done by “jumping” exactly one FSR”
- L104: Expand FSR abbreviation
- L105: Consider re-phrasing to “the spectral resolution is only multiples of the FSR”
- L108: “which leads to additional”
- L125: I would personally recommend minimizing the use of latin phrases unless absolutely necessary or further explained in the text. Consider instead “tackled by an empirically established drift correction” or similar
- L126: “frequency comb which is itself locked on the”
- L130: “by very small linewidth”
- L140: “and then measure the Lamb dip”
- L143: Fig. 3 would be even better understood if there would be clarification that the figure is the result of a 6 day experiment somewhere near this sentence. For example, “Across a 140 hour experiment, we measure the frequency deviation obtained with the Lamb dip method…”
- L153: “in Leroy-Dos Santos et al. (2021).”
- L161: Is there an upper limit to the humidity that the instrument can measure? In other words, is the upper measurement limit comparable to the Picarro, or does the special configuration of the instrument impose a limitation? A brief sentence in Sect. 4 (Discussion) would do.
- L170: Appendix B
- L185: “from Fig. 3 of Leroy-Dos Santos et al. (2021),”
- L186: “from Casado et al. (2016),”
- L190: “While we were not able”
- L200: “response until humidities around 200 ppmv”. Consider instead “flat humidity response for humidities above 200 ppmv.”
- L209: Remove “as detailed by”. Otherwise, “as detailed by Casado et al. (2016) and…”
- L210: 500 ppmv
- L210: 100 ppmv

- L214: 100 ppmv
- L220: “in Leroy-Dos Santos et al. (2021)
- L232: 50 MHz
- L232: 1 MHz
- L232: 10 MHz
- L243: “measured Lamb dip features every hour.”
- L257: “Lamb dip measurements”
- L263: “would mitigate a large part”
- L273: -40 °C
- L276: Consider “In inland Antarctica, some campaigns monitored isotopic water vapour composition but were”
- L280: “use of the high sensitivity”
- L280: “to an Antarctic field station”
- L283: As mentioned in my opening comments, I think “10 times lower” is more accurate. But this would be a good point to speculate on the potential precision down to 1 ppmv.
- L284: -80 °C
- L286: Remove one of the “monitoring”. Consider “all the hurdles that limit water vapour isotopic composition monitoring in the coldest…” or “all the hurdles that limit the monitoring of water vapour isotopic composition in the coldest…”
- L293: 230 kg
- L301: Remove either “Indeed” or “currently”
- L301: “gaseous solid phase”
- L302: -30 °C
- L305: -30 °C
- L307: I’m not sure what is meant by “hyped”. Would maybe “enhanced” or “complimented” fit better?
- L309: durations
- L314: -80 °C
- L318: 1 hour
- L330: Specific humidity
- L387: spectroscopy of H_2S
- L445: “Available from: %3CGo”. What is %3CGo?
- L452: “$\delta^{18}O$ and δD”