

Interactive comment on “Tracking isotopic signatures of CO₂ at Jungfrauoch with laser spectroscopy: analytical improvements and exemplary results” by P. Sturm et al.

Anonymous Referee #1

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The paper by Sturm et al. is generally well written. The paper reports on issues related to the instrument long term operation and in addition attempts to apply and correlate the experimental data with the FLEXPART model. Isotope measurements of ¹³C and ¹⁸O of CO₂ are one of the most challenging in laser spectroscopy and associated calibration methods. This paper builds upon a number of previous papers of the same group and associated partners (Nelson 2008, Tuszon 2008a and b, Tuszon 2011). To a some extent, significant overlap exists with previously published paper by the same group, in particular in the instrument discussion. Associated techniques, changes, and improvements are generally well documented and cross referenced. In some instances however, referenced as well as this paper lack details or lead to some confusion.

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1) In addition to stating the precision (0.02 per mil) in the abstract, the authors should include the replicate precision or accuracy of their measurement. A 10 min averaging time is stated to obtain 0.02 per mil precision. Given the series of drift compensation described in the paper, the authors should clarify the complete measurement time and duty cycle of measurement. Although this performance is outstanding it is easily misunderstood that such performance is obtained continuously (100% duty cycle). The statement of being able to acquire with a high time resolution (10 Hz) and stating a precision that is obtained for long averaging times is inappropriate and misleading. Further, the authors mention that the instrument has been deployed since 2008 measuring at 1 Hz. No data is shown for 1 Hz and it is not helpful to state in such way as a measurement cycle is much longer. Mixing in statements of time and precision out of appropriate context or with more specificity should be omitted. 2) It would be helpful to state how long it takes for a flask to be filled. 3) Clarify the temperature stability of the cell and associated limit of precision 4) Data processing and Performance: When mentioning the spectroscopic performance the authors state an improvement to the previous arrangement and papers. This is hard to follow given that different averaging times from this with previous papers are compared. Looking at the Allan variance plots and precisions mentioned in Nelson 2008, Tuszon 2008a, b, and Tuszon 2011, it seems that only the 1 s performance has actually degraded from ~0.2 to 0.4 per mil (¹³C). This paper, shows 0.02 (0.04) per mil for 600 s (400 s) of averaging, while Nelson 2008 shows 0.02 per mil for 400 s, Tuszon 2008a 0.035 per mil for 400 s, while Tuszon 2011 only states the performance of 0.046 per mil at 50 s. This is in contrast to the significant improvement shown in Fig. 3. – This reviewer suggests to clarify this by comparing only Allan variance derived precisions for identical averaging times (1s and 400 s for example) and better distinguish the precision for a complete measurement cycle involving drift etc. corrections (Fig.3). Also, the authors mention a degradation of performance due to the long term operation, but do not offer details. Clearly, the 1 s performance has diminished by a factor of ~2, but overall performance has increased (Fig.3). For laser based systems, spectroscopic degradation can oc-

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cur by long term mis-alignment, decrease of transmission through the multi-pass cells (and associated increase of scattering, although pulsed systems may be less affected by this), decrease of spectral purity and power of the QC laser, or degradation of the detectors. Speculating that the QC laser may be emitting with a broader line-width over time may significantly affect the absorption. Based upon the details from Nelson (2008), only 10 % of absorption is yielded in 7.3 m pathlength, which is perhaps a factor of 7 less than with a narrow linewidth laser and suggesting a relatively broad QC laser linewidth. In reference to these comments, it would be helpful for the authors to better tabulate the affects and dependence of precision, drift, and shifts from temperature changes, pressure changes, cal standard drifts, concentration correction and associated determination of CO₂ mole fraction. Some details are provided, but are hard to follow. Specifically, it lacks details on how the data is processed (including the many steps of corrections). In particular the 1st and 2nd drift correction is difficult to follow and understand without details. Nomenclature also changes from this to previous papers (cylinder 1, 2, 3 to working, drift, reference, primary etc.) makes it hard to follow if previous papers are referenced for details. A data processing chart in addition to the text description would greatly enhance and add clarity. The correction for the concentration dependence is also difficult to follow. Although previous papers from the same group are referenced, Tuzson 2008 (Fig 4 a. and b.) are not well understandable (13CO₂ / 12CO₂ ratios of ~ 0.8 are what ? absorption of respective lines ?). 5) The FLEXPART section of the paper is somewhat isolated to the instrument performance and improvements discussion. This perhaps could be a separate paper (with fig. 8, 9, 10, 11), yet complements this paper by offering clues on how the acquired dataset can be utilized.

In summary, the paper is by in large well written. However, in current form offers relatively minor value (instrument section) due to a large overlap of content presented in previous papers, and as discussed above lack of clarity, and detail of the discussed advances that would be most useful to understand (without having to read in parallel 4 core reference papers, but then to find many gaps in the actual descriptions therein).

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The first half of the paper therefore will benefit from incorporating clarification and expansion to minimize the dependence of having to read prior publications.

Interactive comment on Atmos. Meas. Tech. Discuss., 6, 423, 2013.