

Interactive comment on “LISA: a lightweight stratospheric air sampler” by Joram J. D. Hooghiem et al.

Anonymous Referee #2

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General comments

The authors present a new stratospheric sampling system that can fill a niche between current techniques, as it provides larger air samples with better vertical resolution than AirCore while operating with much lower cost and payload weight than typical cryogenic whole air systems. The manuscript is suitable for AMT and well-written and should be published after addressing the following comments and requests. In particular, a possible error in the calculation of uncertainty for isotopic analysis should be resolved.

Major issue: Section 6.4

I agree with Reviewer 1 that the underlying assumptions used to estimate fraction of sample from contamination (fc) are incorrect. The ultimate source of the error seems

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to derive from the expression used to define $f_c = \text{bias}/([\text{X}] + \text{bias})$. Here I presume that $[\text{X}]$, defined by the authors as “typical mole fraction,” is therefore meant to be $\approx [\text{X}]_m$, the measured mixing ratio.

If we rearrange equation (7) to $\delta m = \delta s - \Delta\delta$ and rewrite for mixing ratio as $[\text{X}]_m = [\text{X}]_s - \Delta\text{X}$, where $[\text{X}]_s$ is the stratospheric mixing ratio and ΔX is the bias, then substitute this into the equation above, we have $f_c = \Delta\text{X} / (\Delta\text{X} - [\text{X}]_s + \Delta\text{X}) = \Delta\text{X} / [\text{X}]_s$. But since the authors also define $f_c = \Delta\text{X} / ([\text{X}]_s - [\text{X}]_c)$ in equation (8), this implies that the authors are assuming the mixing ratio of the contamination is 0 when they calculate the f_c on pg 14, line 28. Instead, they should use measured or typical mixing ratios, nominal 400 ppm and 1800 ppb for CO₂ and CH₄, respectively. This would result in roughly the same estimate as Reviewer 1 has calculated.

An alternative method to estimate f_c would be to solve equation (8) for both CO₂ and CH₄ simultaneously. Since the authors already have estimates for $\Delta\delta$ and δs stated (pg 14, lines 26-28), this results in two equations with two unknowns, which can then be solved trivially. This would be a worthwhile exercise to confirm the estimate of f_c provided by Reviewer 1.

Finally, it is not clear to me why the authors don't use the storage test results, in addition to the ambient data, to estimate f_c . This should either be performed, or the authors should explain why this analysis is not valid.

Additional general comments

I agree with Reviewer 1 that the introduction is a bit unfocused and broad. I think a more focused discussion of the literature with regards to the particular challenges of making sufficiently accurate and precise mixing ratio measurements in the stratosphere would be more useful. See the introduction offered by Membrive et al., 2017 for an example.

The manuscript presents multiple tables of information (e.g. Table 2 is derived fit coefficients, Table 3 is instrument operational settings) that would be better suited in

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supplemental materials, as this information is not critical to main discussion here). Meanwhile, there is no table provided that summarizes the instrument specifications (e.g. weight, power requirements, sample resolution at stated altitude) and comparison to the AirCore and other systems. I would be appreciative of such a summary table.

Additional comments

p. 1, line 11 - The abstract discusses the stability tests, which include H₂O mole fractions. However, the rest of the abstract only discuss CO₂, CH₄ and CO mole fractions. A phrase noting the significance of H₂O (interferent?) would be appreciated.

p. 1, line 29 – “Stratospheric changes in ozone and water vapour levels” is awkwardly phrased. How about “Changes in stratospheric ozone and water vapour levels”?

p. 2, line 1 – “stratospheric air up to 35 km” Please define this value (e.g. ASL).

p. 2, line 13 – “remarkable scientific efforts” I would avoid using words like “remarkable” without a substantial defense of this term.

p. 2, line 15 – Introducing mean age of air is unnecessary here unless it is discussed later in the text

p. 3, line 8 – “AirCore does not provide large sample amount” Please quantify this statement.

p. 4, line 1 – “a diaphragm pump (KNF, product no. NMP 850.1.2 KNDC B)” Please provide a description of wetted surfaces for this pump.

p. 5, line 20 – Storage test results. Can the authors take advantage of the results of these tests to separate leak and permeability effects for all species? The discussion of the water vapor experiment is nicely done and touches on observed changes in CO; can this be extended?

p. 6, line 11 – “we observed an offset” Please describe which direction these offsets

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were (increase or decrease). As a reader, I wondered if this effect could have been a cause of the CO₂ offset observed during the April flight discussed later

p. 6, line 24 – “We do not include any sampling error in the presented evaluation, although it might be significant for high-precision measurements, especially for CO in the stratosphere when the ozone concentration is high.” This statement was ambiguous to me. Please explain exactly what is meant by “sampling error” and the significance of ozone concentration.

p. 6, line 28 – “However, detailed information about storage conditions are required to correct for the drift, which is usually unavailable in the field.” Why wouldn’t this be available? The detector used for analysis was in the field (see p. 9, line 27) so this data may have been collected for the samples presented here. Please explain if the data is available and whether it should be collected in future work.

p. 7, line 12 – “The performance of the small diaphragm pump is to our best knowledge not previously investigated under the atmospheric conditions in the stratosphere, e.g. at low-temperature and low-pressure conditions.” This is a nice presentation of this experiment. Was the pump subject to stratospheric temperatures? The manufacturers specification is for ambient temperature between +5°C and +40°C. Any insight that can be provided about the pump performance at stratospheric temperatures would be appreciated.

p. 8, line 6 – “19.7 seconds.” How precise is this value? I’m a bit surprised that the pumping speed would be so consistent at varying pressure. There are no data points shown in Figure 3 prior to 20 sec, which makes this statement a bit hard to evaluate. The uncertainty of this value under operating conditions should be discussed with respect to the modeled behavior that follows.

p. 8, line 25 – “The sampled volume at STP can be modelled for all pressure levels ranging from 200 to 0 hPa” It seems inappropriate to model to 0 hPa, given that the data collected ends at 30 hPa.

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p. 9, line 5 – “To be on the safe side,” should be rephrased “Conservatively,”

p. 11, line 10 – “the vertical resolution increases” The vertical resolution decreases since the vertical height increases. There are several significant deviations from the relationship between altitude and vertical resolution, as shown in Figure 4b: 26-Apr at 150 hPa, 4-Sep at 100 hPa and 5-Sep at 30 hPa. Could these data points be explained in the context of the discussion of the variability in vertical resolution starting at p. 11, line 14?

p. 11, line 25 – While the vertical profiles of the mixing ratios are shown in Figure 5, I don’t see the actual values presented. Could this be added to Table 3 or as a Supplemental Table (along with individual uncertainties of both mixing ratio and vertical height)?

p. 12, line 8 - While there are significant differences between the LISA and AirCore retrieved mixing ratios for CO₂, the CH₄ and CO mixing ratios show good agreement. Can this allow the authors to make a statement about the validity of their assumptions for estimating altitudes and pressures for the failed data loggers? And does this indicate that the offset CO₂ measurement is a result of the measurement of mixing ratio rather than altitude? This comment is also for p. 12 line 30 to p. 13, line 2.

p. 12, line 17 – “0.5 ppm or 0.13%” I’m not sure that the percentage reported here is appropriate, as it implies greater analytical precision that is achieved. If the CO₂ mixing ratio varies only 20 ppm or less between the troposphere and the top of the sampling region, then 0.5 ppm is a more significant deviation. I would suggest simply removing these percentages.

p. 13, line 13 – “decreases fast.” Should be “decreases quickly.” In the following sentence, “the gain in sample amount, for example adding 10 extra seconds of sampling time, does not increase” is not correct, but rather the gain in sample amount is very small in comparison to the decrease in vertical resolution. This should be restated.

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p. 13, line 16 - As an aside, I would suggest additional bags as an alternative to larger bags, as the altitude profiles presented in Figure 5 indicate that there are periods during the ascent when no sample is collected, i.e. the system is sitting idle.

p. 13, line 28 – It is not clear why the uncertainty in the sample amount is important. Could the authors explain the importance of this?

p. 23 – Figure 3b. I found the reuse of marker shapes between 3a and 3b confusing (I was looking for a relationship between the same marker shape in both figures). Could the authors change the marker shapes in Figure 3b to avoid this? For Fig 3c, it is not clear if the units on the vertical axis use volumetric liters or standard liters. This should be standard liters for consistency. Also, in Fig 3d please extend the vertical line at 0.76 L for the full vertical height of the figure, as was done in Fig 3b.

p. 25 – Figure 4 and Figure 5. As a color-blind reader I cannot distinguish between the colors used for the 26-Apr and 4-Sep flight data. Could the 26-Apr color be changed to a different color (e.g. blue)?

p. 27 – Table 6. Please define the acronyms VPDB and VSMOW.

Reference: Membrive, O., Crevoisier, C., Sweeney, C., Danis, F., Hertzog, A., Engel, A., Bönisch, H., and Picon, L.: AirCore-HR: a high-resolution column sampling to enhance the vertical description of CH₄ and CO₂, *Atmos. Meas. Tech.*, 10, 2163-2181, <https://doi.org/10.5194/amt-10-2163-2017>, 2017.

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