

Interactive comment on “2-D tomography of volcanic CO₂ from scanning hard target differential absorption LIDAR: The case of Solfatara, Campi Flegrei (Italy)” by Manuel Queißer et al.

Anonymous Referee #1

Received and published: 7 September 2016

Quaiser et al. 2016 amt-2016-166

The manuscript describes tomographic determination of the 2D distribution of the CO₂ mixing ratio (XCO₂) derived from a series of line integrated CO₂ measurements (or CO₂ column density measurements). While the overall structure of the paper is straightforward, there are many unclear points, missing information, and not very convincing statement, which make the paper rather hard to read and understand (see detailed list below). In particular the tomographic inversion is not convincing at all. It remains unclear why only such a small spatial resolution is reached. Also, no attempt is made to quantify the errors of the tomographic inversion and there are ample indications in the manuscript that they are huge. In summary, the manuscript describes an

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interesting approach, but the analysis of the data is poor and the errors of the derived fluxes are essentially unknown. I can not recommend publication of the manuscript in its present form.

In detail the following points need attention:

- 1) Page 1, line 17 (abstract): What are 1-D profiles?
- 2) Page 1, lines 18-19 (abstract): What is the meaning of the phrase "Acquisition was performed from a single half space only" ?
- 3) Page 1, lines 21-22 (abstract): "The method has important implications .." This is not treated in the text, other than saying that retrievals of this kind are difficult.
- 4) Page 2, lines 21-22: Why can "hard target DIAL" systems use continuous lasers and why is this an advantage?
- 5) Page 2, line 25: The term concentration (i.e. number density or mass/unit volume) is used as synonymous with mixing ratio (e.g. given in ppm)
- 6) Page 2, lines 34-35 (last Sentence): This statement is incorrect. Flux (through a particular plane) is obtained by integrating over the CO₂ concentration-distribution weighted by wind speed component perpendicular to the plane.
- 7) Page 3, lines 20, 25: Explanation of delta tau from line 25 should occur after line 19.
- 8) Page 3, line 30: The position of the glass wedge and the integrating sphere are not clear from Fig. 2, in fact it appears that the emitted beam passes through the sphere. It should be mentioned that the reference detector measures $P(\lambda_{\text{on}})_{\text{ref}}$ and $P(\lambda_{\text{off}})_{\text{ref}}$, respectively.
- 9) Page 4, lines 1-5: A lot more information needs to be given: What is the divergence of the emitted beam? What is the field of view of the receiving telescope? These quantities are required (a) to calculate the returned power (the quoted "few nW" can only be reached (at $R=1$ km) when the aperture angles of transmitting and receiving

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optics exactly match, is this the case? (b) The same quantities are needed to calculate delta Speckle in Eq. (5).

10) Page 4, line 9: What was the delta sigma used to convert delta tau to CO2 concentrations (and mixing ratios)?

11) Page 4, line 9: What could be the reason of the "instrumental offset"? Could it be an effect of differential reflectivity (different reflectivity for lambda on and lambda off) of the target? If this was the case then for each target (i.e. each measurement direction in a scan) there would be a different offset. Was this possibility checked?

12) Page 4, line 12: Why were mixing ratios calculated? For the flux determination (Eq. 8) the mixing ratios are re-converted to concentrations. While concentrations - as determined from the optical density - are independent of temperature and pressure, mixing ratios are not, so which temperature and pressure was assumed to calculate the values given in Figures 3ff?

13) Page 4, line 17: What were typical values of the three contributions to the error of the right-hand side of Eq. (3)?

14) Page 4, line 19: What were typical values of the SNR?

15) Page 4, lines 26-28: What are typical values of D, Xie, and the resulting delta Speckle (see comment 9, above).

16) Page 5, line 3: What is the meaning of the term "absolute Cartesian coordinates?" in this context?

17) Page 5, line 8: Do the authors here not just mean that the assumed CO2 concentration within a grid cell is constant (which would be the normal assumption when continuous space is divided into grid cells)?

18) Page 5, line 18: Here a comment about the relative magnitude of m, n is needed. If $m < n$ the system is underdetermined and can only be solved by making additional

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assumptions. If $m > n$ the system is overdetermined and a least squares solution can be found.

19) Page 5, line 19: What is the meaning of the double vertical lines, length of the vector ($m \times 1$ matrix)?

20) Page 5, lines 22,23: The CO₂ flux will be derived in kg/second?

21) Page 5, line 27: In 2016 the background CO₂ level reached 400 ppm, in industrialized areas probably even a bit more, why were 380 ppm used? what is the impact of this low bias on the derived fluxes?

22) Page 5, lines 27, 28: The statement about the "plume transport speed" is correct but misleading: since the scanned area is essentially the horizontal plane (I did not find information about a possible slope in the manuscript) the relevant component of u is the vertical component. Why not saying that?

23) Page 6, line 8, Eq. (10): What is the typical magnitude of the relative errors entering the Equation?

24) Page 6, line 21: It is confusing when the same letter (X) is used for two different quantities, the mixing ratio (X_{CO_2} , a_v , in ppm, i.e. dimensionless) and the column integrated mixing ratio (X_{CO_2col} in ppm, i.e. dimension of a length).

25) Page 6, lines 22,23: "Numerous wiggles indicate vigorous degassing activity ...". this appears to be in sharp contradiction to the "frozen plume" assumption (same page, line 20)?

26) Page 6, line 26: The meaning of the phrase "ranges and headings were converted to Cartesian coordinates" remains cryptic.

27) Page 6, line 29: What is the meaning of "associated coordinates" against which the X_{CO_2av} data are plotted. X_{CO_2av} are range averaged mixing ratios, the only coordinate available appears to be the scan angle (heading). So what is actually plotted

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in Fig 4?

27) Page 7, first three paragraphs: The discussion about the inversion of the CO₂ column data into gridded mixing ratio values is cryptic and not convincing in many aspects: i) Did the runs of the LSQR algorithm with test data also include noise added to the test data? ii) It is stated that for $n > 36$ there were oscillations in the retrieved XCO₂ values, did this behaviour occur for all test distributions? How large were the oscillations? What was the influence of (artificial) noise on the oscillations? What were the criteria for judging the retrieval unacceptable? iii) For real data even $n > 16$ were reported to be a problem, but what was the problem, "unreasonable high" XCO₂ or oscillations or both? What was the reason for the different behaviour of the algorithm for test data and real data? iv) There is no information given on the error of the tomographic inversion, which may be huge! The large chi square (norm) of the inversion (around 18%, page 8, line 26) is an indication that the inversion has problems. Moreover, what about the uniqueness of the inversion, which was apparently not studied at all?

28) Page 7, last paragraph: In the introduction of the manuscript it is stated that high resolution CCO₂ distributions (multiplied with associated plume transport velocities) would yield superior data compared to using average XCO₂. However, can a 4 x 4 grid of CO₂ data really called "high resolution" in particular when keeping in mind that the tomographic inversion (LSQR algorithm) likely adds significant errors, further diminishing the theoretical advantage of spatially resolved XCO₂ data?

28) Page 7, last paragraph: What is the plume transport speed? Is it the vertical component of u ?

28) Page 7, line 30: Do the stated errors include the errors of the tomographic inversion?

29) Page 8: line 2, figure 6: The Fig. is very confusing: How was the quadratic grid (Fig. 6a) mapped in the real shape of the area (Fig. 6c)? Were the squares distorted to match the real area? How? Also, the "smoothed" version of (Fig. 6a) shown in

(Fig. 6b) does not appear to match. smoothing should smear out the maxima, but they appear to be shifted in (Fig. 6b) relative to (Fig. 6a), why?

29) Page 8: line 11: What is a "symmetric increase"?

30) Page 8: lines 13, 14: The disagreement in peak positions is attributed to "physical" fluctuations in the CO₂ concentration. This appears to undermine the validity of the entire approach, effects like this will certainly give rise to large errors in the retrieved XCO₂, which of course, will propagate into the derived fluxes.

31) Page 8: lines 19, 20: Lack of spatial matching underscores the observation stated in comment 30.

32) Page 9: First two paragraphs: It appears that the large differences between the results of this study and other reported values are likely due to deficiencies of the method employed here. The attempt to bridge the gap is not convincing. For instance point sampling may miss spots of high CO₂ but equally likely miss spots of low CO₂, so it is not obvious that the data should be low-biased.

33) Page 9, lines 23, 24: Why should the inversion problem become non linear? After all tomographic inversions are linear transformations (see also Eq. (7), which is clearly a linear equation).

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2016-166, 2016.

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