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## ***Interactive comment on “Tunable diode laser measurements of hydrothermal/volcanic CO<sub>2</sub>, and implications for the global CO<sub>2</sub> budget” by M. Pedone et al.***

**M. Pedone et al.**

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General comments:

Dr Mather’s comment: Some more comments on the balance between diffuse and localised soil degassing at each of the locations is needed. One of the potentially interesting things about this technique would seem to be its potential to capture a cross section through whole areas of CO<sub>2</sub> degassing including diffuse degassing (where strong enough to be measurable) and visible localised sources such as fumaroles in the same cross-section. More discussion regarding this be very interesting and add to the impact of the paper. Is there any reason that it could not be used to measure main

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plumes also? Signal attenuation etc. perhaps.

Authors: We agree with Dr Mather that one of the advantages of using the tunable laser is the possibility to capture simultaneously the CO<sub>2</sub> contributions from both diffuse soil degassing and concentrated emissions (fumaroles). This topic can be easily covered with some additional sentences upon revising the manuscript. This technique can also be used also to measure main plumes provided the plume is not condensing and/or optically thick. Fog and/or others obstacles within the laser-mirror path reduce its functioning during field operations. We have for instance successfully used the laser in the plume of Mt. Etna (authors' unpublished results).

Dr Mather's comment: Would it be possible to include the Matlab script with the paper as a supplementary file? This would facilitate its use by others and increase the impact of this study.

Authors: the Matlab script is available to readers upon request (Addressed to the CA). The script was presented in an earlier paper on Bull Volc and has already been provided to some individuals upon their request.

Dr Mather's comment: Throughout more specific comparison of the spatial and temporal variation of CO<sub>2</sub> fluxes would be a very welcome addition to the paper and would further emphasise the usefulness of the technique. See specific comments regarding comparing the spatial maps with previous studies and the Vulcano flux through time below. Authors: The Spatial distribution of our CO<sub>2</sub> flux anomalies and overlapping with (and complementary to) the degassing anomalies seen via soil degassing surveys (see our specific replies below to the points made by the reviewer in the relation to the Santorini case). Unfortunately, we have only one single CO<sub>2</sub> flux available for Vulcano, so we are unable to explore temporal trends if not by comparing with earlier data from the literature (see specific point below on Vulcano).

Dr Mather's comment: Somewhere I would like to see the authors to explore how the TDL technique compares in terms of robustness, time to use, user-friendliness,

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cost etc. for application to a volcanic environment compared to other previously used techniques. These are very important considerations when working at volcanoes as well as the effectiveness of the technique.

Authors: We agree with Dr Mather that these are important aspects to consider for an effective use of the technique in routine volcano monitoring. We plan to cover these upon revising the manuscript. The TDL has both disadvantages and advantages compared other more consolidated techniques (e.g., FTIR). Only one species (CO<sub>2</sub> in our case) can be measured with the TDL (against multi-species simultaneous detection by FTIR) and no passive measurement is possible (FTIR uses passive sources such as the sun or hot rocks/magma). The TDL is however a factor 2-3 cheaper than FTIR, and more user-friendly (the instrument is simple to use in the field and post-processing is straightforward). The TDL is robust enough for its use in harsh/aggressive volcanic environments.

Major comments:

Dr Mather's comment: P2653, L17: I cannot see the red cells in Fig. 2a Authors: we meant the 16 cells separated by red lines

Dr Mather's comment: P2653, L 19/20 and P2655, L5: Do the errors in the ICA depend at all on where the high flux values fall? The authors state that they use synthetic data but do not state if it was tested for these sorts of issues. Errors in some kriging techniques are better assessed using a series of realisations with each specific dataset.

Authors: Yes, we used synthetic datasets exhibiting peak concentration in 1-3 cells to test the performance of the algorithm in conditions similar to those seen during field observations. The errors obtained are still <3%.

Dr Mather's comment: P2654: The shapes of the CO<sub>2</sub> concentration maps generated here should be compared with previous surveys for the 3 systems where previous measurements have been made to as great an extent as possible. This would help

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understand the balance between diffuse/fumarole degassing in terms of this technique compared to others.

Authors: We agree with Dr Mather that our measurements are complementary to results of previous studies focussing on the diffuse CO<sub>2</sub> degassing regime of the same areas. For the sake of illustration, and in the specific case of Santorini, we have prepared an additional figure that compares the spatial distribution of our CO<sub>2</sub> anomaly with that detected in the diffuse degassing flux map of Parks et al. 2013. The study of Parks et al. 2013 covered a wider exhaling area that contributes a diffuse CO<sub>2</sub> output of  $38 \pm 6$  t d<sup>-1</sup> (Parks et al. 2013), or  $\sim 60\%$  of our  $63 \pm 22$  t d<sup>-1</sup> fumarolic CO<sub>2</sub> output. From this comparison, we argue that our TDL measurements add new significant information to fully constrain the total CO<sub>2</sub> output at Nea Kameni, and the balance between concentrated (fumarolic) and diffuse (soil) contributions.

Dr Mather's comment: P2655: It would be really interesting to hear more about the time series of flux presented at Vulcano in Figure 6 and how it relates to other developments of this volcanic system.

Authors: The reviewer has probably misunderstood the content of this figure. In Figure 6, we are in fact comparing the results of the only individual TDL CO<sub>2</sub> flux campaign we have available with independent (earlier) results obtained with other techniques (CO<sub>2</sub>/SO<sub>2</sub> ratio + So<sub>2</sub> flux).

Dr Mather's comment: Section 4.5 is interesting but needs more clarification. For example, Vulcano is included in Table 1 and 2 and Nea Kameni, the Reykjanes volcanic system and Vulcano are all included in Table 3 of Burton et al. 2013 and see comments on Figure 7 below. I think that the argument in Burton et al. (2013) is that we only have a fraction of the strongly degassing 'main' plume fluxes of CO<sub>2</sub> determined. While I agree that saying we have only measured one fifth is probably too low, if the current authors agree that we are likely missing some significant fraction (e.g., PNG, more in Indonesia, the new CAVA data that is cited as Aiuppa et al. submitted etc.) this

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still has the potential to significantly change their distribution and regression shown in Figure 7. While I think that this section makes useful points a little more on the continuing uncertainties would be appropriate. This section is also quite long and if it were possible to cut it down then it would read better. Please rephrase the last sentence of the conclusions in line with my comments above.

Authors: We agree with the reviewer and Burton et al (2013) that a significant fraction of the large volcanic emitters is missing from current CO<sub>2</sub> datasets, and that these have the potential of impacting the shape of the CO<sub>2</sub> flux population. We agree this must be clarified in the text. The additional argument we make in this manuscript is that there is a very large number of small volcanic emitters that are missing too, and which combined output can be important (and possibly overlooked by previous work). Table 3 from Burton et al 2013 only counts the diffuse (soil) contribution to the CO<sub>2</sub> output for Nea Kameni, the Reykjanes; the fumarolic CO<sub>2</sub> contribution was undetermined until the present study.

Dr Mather's comment: Figure 1a and 2c: It is not very clear where exactly on Nea Kameni this is. How do these locations relate to those shown in the other degassing studies mentioned (Tassi et al., 2013; Parks et al., 2013)? This would aid the comparisons that I suggest above.

Authors: Our investigated area was located in the central part of the degassing structure investigated by Parks et al. (2013), right on-top the most actively degassing Nea Kameni summit crater. An ad-hoc figure has been prepared to show this.

Dr Mather's comment:Figure 7: It should be possible and would be helpful to indicate the new data points added beyond Burton et al. 2013 here on the Figure. There do not look to be enough data points to encompass the Burton et al. compilation as well as the new measurements they mention? More explanation of the red point in the caption would be helpful.

Authors: The new data points are in fact part of Figure 7. It is not obvious how new

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data can be distinguished from original data of Burton et al 2013.

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