Manuscript: “Remote sensing of volcanic CO_2, HF, HCl, SO_2, and BrO in the downwind plume of Mt. Etna” by A. Butz et al.

Reply to interactive comment by M. Queisser

We thank Dr. Queisser for his comments on our manuscript. Please find our point-by-point reply below. We will consider modifications of the manuscript after all reviews will have become available.

General comments:

<<The paper presents results of a measurement survey, sensing volcanic gas concentrations (CO_2 , HF, HCl, SO_2 and BrO) a couple of km downwind Mt. Etna, Sicily (Italy), using passive remote sensing apparatus. In particular, a fourier transform spectrometer operating in the SWIR, and a DOAS operating in the UV were employed. I have not come across ground based passive remote sensing of volcanic gas from this long distance from the volcano mouth. While the methods used are established the data are new and an important result for climate related science and environmental monitoring in general. The paper is well written in a concise and straightforward manner.

I like the fact that the authors measured other volcanic species (HCl, HF etc.) in parallel as this allows a rather precise distinction of the volcanic CO_2 in space. The measurement precision, particularly that of CO_2 VCD is impressive, given that the observatory was moving on a road (even though CO_2 absorption spectra were recorded when the car stopped). How sensitive is the setup to shocks and vibrations? Is there an influence on the vibrations and have you quantified them or at least have a semi-quantitative measure?>>

The reason why we had to stop the truck for CO_2 measurements was that shocks and vibrations prevented the solar tracker from reliably tracking the sun over the 12 second integration time of the FTS. Heavy vibrations and shocks caused entire loss of the solar tracking. Lighter vibrations that caused brightness fluctuations could be monitored via the DC-part of the recorded interferograms. The later effect was used to discard such data.

The EM27/SUN FTS itself proved remarkably resistant against shocks and vibrations. It did not require any optical realignment during the entire campaign deployment. We monitored optical alignment through measurements of the instrument line shape (ILS) along a procedure for example described in Klappenbach et al., 2016. To this end, we operated an ordinary halogen lamp in a few meters distance from the spectrometer and coupled the light into the spectrometer via the solar tracker mirrors. Then, we fitted a model of the instrument line shape to water vapor absorption lines around wavenumber 7000-7400 cm^{-1} (1.4 micron wavelength) . The ILS did not show any significant changes during the campaign.

Thus, there is no direct influence of shocks and vibrations on the reported CO_2 records but recording was only possible when the truck was stopped.

<<The retrieval algorithm used remains mysterious, as well as the impact of model and fitting errors on the VCDs (see specific comments).>>

The retrieval algorithms PROFFIT and DOAS are standard methods in the atmospheric measurement community and our paper provides the relevant references. We would prefer not to expand on these technical aspects further.

Concerning the error estimates, we took the standard deviation of all background soundings as a measure for the retrieval error. This is conservative with respect to fitting errors, since our estimates thereby include instruments noise, residual fluctuations of solar tracking quality, as well as residual variability of the background gas. Systematic errors such as discussed for the determination of the CO_2 enhancement (removal of surface elevation effect and gas background) are not part of the estimated retrieval error. Our paper discusses these assumptions and caveats.

<<In my opinion the paper could be further improved by comparing the measured enhancements with plume dispersion models (e.g. Burton et al., 2013, p. 325), which are in line with your result. But this might be out of scope for an AMT paper.>>

We agree. But indeed, we find it out of scope for the present paper to run a plume dispersion model. Our goal here is to demonstrate the capabilities of our measurement technique in terms of detection accuracy for volcanic CO_2 and with respect to the number of volcanic gases that can be observed simultaneously.

Since we believe that the technique is of great interest for others working in the field of volcano remote sensing, we chose for a relatively quick and direct path to publication.

Specific comments:

<<P2, l 22 and 23: Lidar BILLY measures range resolved CO_2 concentrations. Whether or not it has to be close to the source depends on various parameters, including instrumental parameters such as the pulse energy, excess CO_2 concentration, aerosol density etc.. There is no fundamental reason why the LIDAR has to be close to the plume. The CO_2 plume of big cities is visible in airborne LIDAR signals from several km flight altitude.>>

We will change the manuscript taking this comment into account. In the manuscript, though, it says "Thus, they need to sample the plume in the proximity of the source." Our intention was to refer to the fact that techniques such as a LIDAR need to point toward the source and lines-of-sight must cross the plume close to source where path-integrated CO_2 enhancements are still much larger than several ppm. So, we did not directly refer to the location of the LIDAR which indeed can be deployed far away if the local environment allows for unobstructed view to the source and if a suitable reflection target is available. But, we understand that our statement might lead to misunderstanding.

<<P4, l31: How sensitive is the measurement precision of the gases, particularly CO_2 (i.e. the 3.7×10^18 molec/cm2) to errors of your atmospheric model. Do the assumptions of your model (e.g. horizontally homogeneous layers) cause a bias? Are you actually able to quantify that bias since you do not know the "true" atmospheric composition (e.g. transmittance at a given wavelength etc.).>>

The measurement precision (i.e. the random error component) is not affected by our retrieval assumptions.

However, our assumptions on plume geometry and atmospheric layering can introduce a systematic error if the reported VCDs were used to calculate the volcanic emission flux without considering plume geometry. The reported VCDs amount to the "true" volcanic enhancements if the assumption holds that the lines-of-sight cross the plume perpendicularly to its propagation direction and that the plume is homogeneous (page 6, line 1 ff). For inferring emission fluxes, one would need to run a plume model which provides plume geometry, propagation direction, and gas inhomogeneity. This could be used as "true" atmosphere in the retrieval or for translating the retrieved VCDs into a (slant) path-integrated enhancement. Since we argue that running a plume model is out-of-scope for this study and since it comes with its own uncertainties, we chose to report VCDs as defined in the paper. This approach should be sufficiently clear to enable follow-up studies.

We want to emphasize (as in the paper page 6, line 4), that ratios of gases are not affected by these assumptions if the gases share the same distribution in the plume.

<<P5, l9: I do not understand the phrase, seems like the subject is missing>>

"For CO_2, HCl, and HF, we only scaled the lower tropospheric part of the vertical profile and adopted the a priori for the upper tropospheric and stratospheric part."

->

For CO_2, HCl, and HF, we only scaled the lower tropospheric part of the vertical profile while the upper tropospheric and stratospheric part was taken from the a priori."

<<P5, l27: It would be interesting to know how high the fitting error is and how and if it propagates into VCD.>>

As outlined above, our error estimate is empirically derived and includes various error sources. In particular, the noise error calculated by PROFFIT and the fitting error calculated by DOAS are smaller than our top-down error estimate.

<<P6, l8: It is clear that the measured VCD vary with observer altitude, but it is not very clear why this is a challenge to obtain the volcanic enhancement. Isn't the VCD looking through the plume larger than when looking outside of the plume (at constant observer altitude)?>>

Indeed, at constant observer altitude, no O_2 correction was necessary to calculate CO_2 enhancements. But, intra-plume and background soundings were never taken at the same observer altitude. Mt. Etna's rough topography implies that lateral displacements of the observing platform virtually always go along with changes in observer altitude which cannot be neglected.

<<P7: Section 5 is largely a discussion rather than pure result section and as such it might be better placed in the discussion section.>>

We tried to separate the discussion of quantitative results (section 5) from more qualitative statements on our work and how it links to past and future efforts (section 6). We believe this separation supports a clear structure of the manuscript.

<<P9, l17: How did you estimate the measurement precision? P7, l29 does not really make it clear.>>

Page 7, Line 29: “We estimated the overall ΔCO_2 precision from the standard deviation (1-σ) of all the background measurements identified via the HF threshold.”

We divided our data record in background soundings and intra-plume soundings. The background measurements are those which show negligible HF concentrations assuming that detection of HF is a good indicator for our lines-of-sight crossing the volcanic plume. The intra-plume measurements are those with significant HF amounts. As described in section 4.3, in particular equation (2), we used the background soundings to subtract the XCO_2 background concentration and to calculate the CO_2 enhancement ΔXCO_2.

For estimating the precision, we simple calculated the standard deviation of all background soundings. We will consider adding some more text in the revised manuscript.

<<P9, l23: “The O_2 column was used to compensate CO_2 variations due to changes in observer altitude.” What means “CO_2 variations”? Variation in VCD?>>

Yes, “Variation in VCD” is correct.

<<P10, l10: Have you thought about measuring closer to the crater of Mt. Etna? Being _1 km away the enhancement would be greater. Being off-roads, you would not be constrained to roads. This might allow assessing some of your sources of uncertainty (negative enhancements, minimum integration time etc.).>>

Yes, we have considered this option for future work (page 10, line 23) and, indeed, being somewhat closer to the source would enhance our signal-to-noise ratio. But, of course, we aim at remote sensing in safe distance from the crater with small logistics overhead. Going off-road closer to the crater counteracts these goals. But, thinking of a permanent, stationary remote sensing network, distances of ~3 km instead of 5-10 km might be better suited.

<<Technical comments:
P4, l8: direct or directed?>>

“direct” since it is a general explanation how the setup works, not only for our study but for essentially all EM27/SUN work.

<<P13, l17, space before comma: FTIR measurements , J.>>

Yes.