RC 1

General Comments:

In this manuscript, the authors describe a data-informed numerical evaluation and instrument model for volcanic gas analysis using scattered near-infrared light. They provide an approach to estimating signal-to-noise ratio under varied conditions and thereby measurement precision for target gas species. While the text is clear in its derivation of SNR and concise in the outcome for three gas species (CO2, HCl, and HF), the overall presentation would benefit from a stronger emphasis on the novelty of the work and impact on future directions for atmospheric measurements.

We thank the reviewer for this important and constructive feedback. In response, we have revised the manuscript to better emphasize the novelty and significance of this work, as well as its implications for future developments in atmospheric measurements. The corresponding changes are outlined below.

Specific Comments:

For example, the authors state they “developed an innovative approach of incorporating actual measurements into our estimations” to address systematic uncertainties. However, the text presents a data-informed model and analysis which did not read as new – while the pre-existing dataset itself may be unique or rare. The model was presented, and data (or previously determined correlation factors) was supplemented. Additional detail describing the unique aspects of the “information-content analysis” would strengthen the narrative.

We thank the reviewer for this important comment. We agree that the original wording overstated the novelty of incorporating actual measurements into the instrument performance analysis. While we believe that basing the analysis on measurements rather than purely synthetic data is important for obtaining realistic performance estimates, this is not the main contribution of our study. We have therefore revised the cited sentence accordingly.

Instead, we emphasize that the main contribution of this work is the assessment of the feasibility of monitoring volcanic gases absorbing in the short-wave infrared using scattered sunlight and an FTIR instrument. To strengthen this narrative, we expanded the discussion of the study's significance and included a concrete example illustrating how the proposed optimized instrument could be used to monitor HF and HCl emissions. We also point out that the presented methodology is relevant beyond volcanic applications and may be applicable to observations of large anthropogenic emitters, such as power plants.

Further, the manuscript would benefit from additional discussion around the implications or impact of the findings. It’s clear that the work effort informed the authors’ decision to not proceed with building a new instrument. There is a missed opportunity to emphasize why the reader should be motivated to use the results going forward. For example, a quick comparison of findings with the current measurement approaches – going beyond feasibility/utility and motivating where this technique may best be pursued (particularly for HF and HCl, where responsivity was deemed reasonable) – would help orient and motivate the reader.

We thank the reviewer for this valuable suggestion. We consider this study a contribution towards the monitoring of volcanic gases absorbing in the near-infrared by providing a data-informed assessment of instrument performance. We envision it as a starting point for further investigations of the associated radiative transfer challenges and as a baseline for the development of future measurement concepts.

To better motivate the relevance and impact of this work, we made several changes to the manuscript:

- **Introduction:** We expanded the discussion of the pioneering studies by Love et al. (1998) and Goff et al. (2001), which measured volcanic HF and HCl using sky-looking observations. However, thermal-emission measurements require the presence of sufficiently hot volcanic gases, while observations of sunlight scattered by bright high-altitude clouds are limited to very specific conditions. We believe that discussing these studies provides a clearer motivation for exploring alternative sky-looking observation geometries and places our systematic feasibility analysis into context.
- **Comparison with existing measurement approaches:** In Section 4.4, we directly compare the predicted measurement performance with that of the NOVAC SO_2 network (Galle et al., 2010). NOVAC uses scattered UV sunlight to monitor volcanic SO_2 fluxes worldwide. Our analysis indicates that a NOVAC-like sky scan for HF and HCl is feasible only under favorable observing conditions.
- **Practical measurement concept:** We now discuss a multi-instrument approach combining the proposed FTIR instrument with a co-aligned DOAS instrument and either a NOVAC station or an SO_2 camera. In this configuration, the FTIR provides HF/SO_2 and HCl/SO_2 ratios at minute-scale temporal resolution, while the independently determined SO_2 flux can be used to infer HF and HCl fluxes. This discussion illustrates a concrete application of the proposed instrument design.
- **Future research directions:** We substantially expanded the discussion of the radiative transfer challenges associated with scattered-skylight retrievals. These challenges are to some degree independent of the instrument itself and are particularly important for CO_2, where the atmospheric background is much larger than the volcanic enhancement, requiring accurate light-path modeling. We hope that this discussion will stimulate further research towards enabling routine volcanic observations of HF, HCl, and CO_2.

Finally, we now also emphasize that the methodology presented in this work is not limited to volcanic applications but is also relevant to observations of other extended atmospheric sources, including large anthropogenic emitters.

RC 2

Comment on “Feasibility of Measuring Volcanic Gas Composition Using Sky-Scattered Sunlight and FTIR Spectroscopy” by Tobias Schmidt et al.

General:

This manuscript investigates the feasibility of measuring volcanic gas slant columns, particularly CO_2, using sky-scattered sunlight in the near-infrared spectral region. The concept is scientifically interesting because the use of scattered sunlight would enable measurement geometries that are not accessible with traditional direct-sun observations and could therefore open new possibilities for volcanic gas monitoring.

The authors conclude that the proposed approach is not feasible for CO_2 under the investigated conditions. Although this is a negative result, it is nevertheless an important and scientifically valid outcome. The study addresses a novel measurement concept and provides information that is highly relevant for the atmospheric remote-sensing community. I therefore believe that the manuscript fits within the scope of the journal and should be published after minor to moderate revisions.

In my view, this work represents one of the first systematic investigations of near-infrared retrievals from sky-scattered sunlight for volcanic applications. Even if the approach proves unsuitable for CO_2, the results are valuable because they initiate a broader discussion about the applicability of such techniques to other trace gases and emission sources. While volcanic plumes are relatively weak emitters of CO_2 compared to the atmospheric background, similar methodologies may eventually find applications for large anthropogenic super-emitters such as power plants, industrial facilities, or methane sources. The study is therefore relevant beyond volcanology.

My main concern is that the manuscript does not show in detail measured and simulated spectra demonstrating the retrieval performance. Such examples are essential for evaluating the feasibility of the approach. Discussed microwindows for al 3 Retrievals including measured and simulated spectra, sensitivity studies, and examples of the selected microwindows would greatly strengthen the manuscript and help readers understand the limitations identified by the authors, even if the precision do not allow to find anomalies the simulation of H2O at 5600cm-1 (HCl overtone), CO2 at 6300cm-1 and O2 at (8000 cm-1 HF overtone) should be shown.

A fundamental challenge of using sky-scattered sunlight is that the radiative transfer problem differs substantially from that of direct-solar FTIR observations. In direct-solar measurements, the retrieval can often be interpreted as counting absorption along a relatively well-defined optical path. In contrast, each scattered photon follows a different trajectory through the atmosphere, making the measurement highly sensitive to the vertical distribution of absorbers, atmospheric scattering processes, and particularly aerosols as I imagine. This issue is well known from satellite retrievals, for example in the REMOTEC methane retrieval algorithm used for

TROPOMI, where aerosol-related path-length uncertainties are a major source of error.

In the present application, scattering is simultaneously beneficial and problematic: more scattering increases the signal available for observation, but it also complicates the separation of photons scattered before and after traversing the volcanic plume. This effect will differ substantially between spectral regions, for example for HCl overtone bands near 5600 cm^{-1}, CO_2 absorption near 6300 cm^{-1}, and HF overtone bands near 8000 cm^{-1}.

For this reason, I would like to see representative measured and simulated spectra for the retrieval windows used by REMOTEC. If volcanic absorptions are not detectable in the available datasets, examples showing H_2O near 5600 cm^{-1}, background CO_2 near 6300 cm^{-1}, and O_2 near 8000 cm^{-1} would still be very informative. Such examples would allow the reader to assess the spectral information content and the quality of the radiative transfer modelling.

Overall, I find the study novel, relevant, and worthy of publication. Maybe the manuscript would benefit a little from a more detailed discussion of the underlying radiative transfer problem, additional retrieval diagnostics, and a clearer presentation of measured and simulated spectra and quantity, also I would like to expand the discussion of the instrument design apart from the detector responsivity and noise reduction, to improve of the increase of the “étendue” of the set up.

I hope my following comments make sense and could improve the manuscript, but feel free to ignore them, if they do not make sense to you.

We thank the reviewer for his/her positive evaluation of our manuscript and the excellent summary given. We answer comment-by-comment below.

Major Comments

1. measured and simulated spectra of individual microwindows

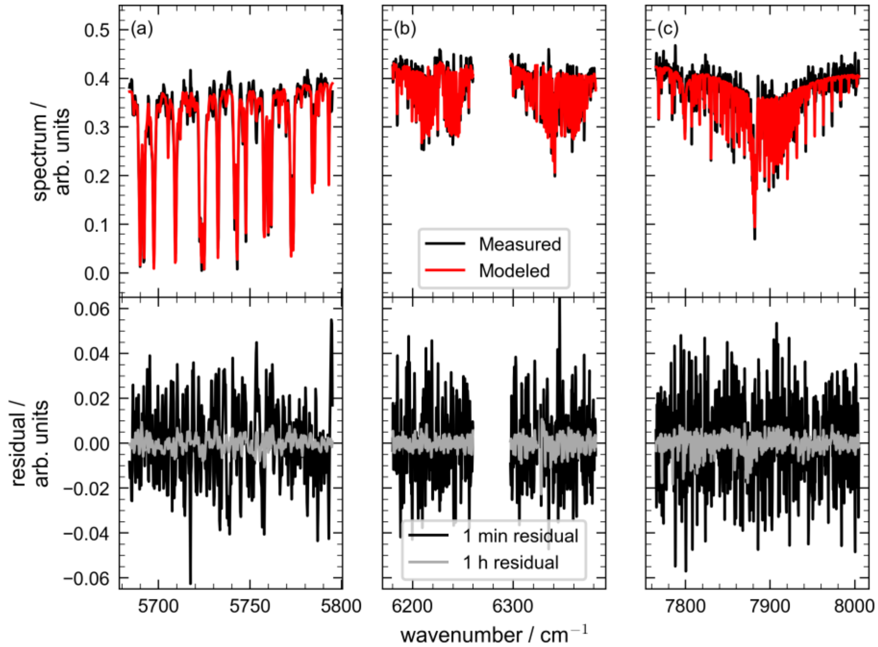
My main concern is that the manuscript does not show any measured or simulated spectra demonstrating the retrieval performance. Since the study concludes that the proposed approach is not feasible for CO_2, it is essential to provide the evidence supporting this conclusion.

I strongly recommend including examples of:

- Measured spectra;
- Simulated spectra;
- Fit residuals;
- Signal-to-noise estimates how they are calculated in detail
- Retrieval diagnostics for the selected microwindows. (slant column Airmassfactore, scattering weights or something similar.)

Even if volcanic absorptions are not visible in the available measurements, examples of retrievals for H_2O (~5600 cm^{-1}), CO_2 (~6300 cm^{-1}), and O_2 (~8000 cm^{-1}) would help the reader understand the information content of the observations and the limitations of the method.

As suggested by the reviewer, we added the following plot of the absorption spectra together with 1-min and 1-h average fitting residuals to illustrate the spectroscopic remote sensing challenge at hand. For the scattering discussion, see comment 2.



For the SNR calculation from the measured spectra, we define the signal as the maximum spectral intensity in the range 6297–6382 cm^{-1}, excluding 6324–6327 cm^{-1}, and the noise as the standard deviation of the spectral intensity in the range 2500–3056 cm^{-1}, which lies beyond the long-wavelength cutoff of the photodiode used in the instrument.

Compared to the preprint manuscript, we corrected a minor error in which the spectral intensity was slightly overestimated due to an unfiltered artifact within the signal window. Excluding this small wavenumber range, as described above, results in lower SNR values for the affected spectra with a given retrieval precision and consequently in the slightly improved performance values shown in Figure 5 (Figure 4 in the preprint). Figure 7 (Figure 6 in the preprint) has been updated accordingly. These changes do not affect the conclusions of the study.

A corresponding description has been added to Section 4.3 of the manuscript.

2. More detailed discussion of radiative transfer

The manuscript would benefit from a more detailed description of the radiative transfer problem solved by REMOTEC and how it differs from the radiative transfer assumptions used in direct-sun FTIR retrieval codes such as PROFFAST, PROFFIT, SFIT, or GFIT.

In direct-solar observations, the optical path is relatively well defined. In contrast, for sky-scattered sunlight, each photon follows a different path through the atmosphere. Consequently, the retrieval becomes highly sensitive to:

- aerosol loading;
- aerosol vertical distribution;
- gas vertical profiles;
- scattering geometry.

A more detailed discussion of these effects would help readers understand why the retrieval problem is fundamentally more challenging than conventional direct-sun observations.

For the fitting, RemoTeC has been used in non-scattering mode (conceptually very similar to PROFFAST, PROFFIT, SFIT, GFIT). The target concentrations were free to be fitted in the lowest atmospheric layer (1001-834 hPa). In a single-scattering view, the lowest atmospheric layer contains the largest fraction of the path component along the instrument viewing direction between the scattering event and the observer while the layers above contain the path component along the solar direction between the sun and the scattering event. This setup delivers the slant column densities (SCDs) i.e. the gas concentrations integrated along the lines-of-sight (see Fig. 1 in the manuscript).

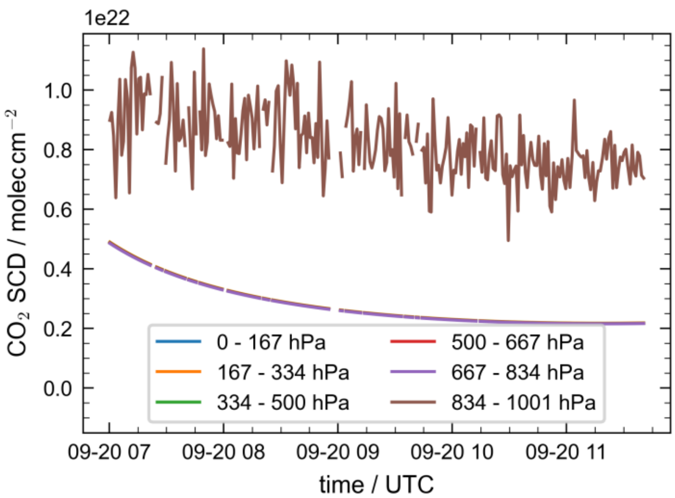
A corresponding paragraph describing this procedure has been added to the manuscript at the end of Section 3.

The figure below shows the retrieved CO_2 SCDs for each of the six retrieval layers. It is visible that the upper five layers remain fixed at their prior values and therefore vary only with solar zenith angle. The lowermost layer exhibits SCDs approximately 2–3 times larger than those of the other layers. This is expected because the shallower viewing zenith angle of 70° results in a longer atmospheric light path, which is further enhanced by multiple scattering.

The reviewer is absolutely right that simulating the radiative transfer in a scattering atmosphere under volcanic plume conditions is a major challenge. Therefore, we believe that it should be addressed by a dedicated study and is beyond the scope of our study here. We added a corresponding discussion of the radiative transfer problem to the discussion section of the manuscript:

> *If an instrument such as the one described here, with potentially better noise performance, were to be deployed for remote sensing at a volcano, the follow-up challenge for obtaining meaningful information about plume gas concentrations would be to develop accurate radiative transfer simulations of the lightpath. The lightpath conceptually divides into a component behind the plume, where sunlight is scattered into the viewing direction, a component inside the plume, where the targeted gas enhancements are accumulated, and*

a component between the plume and the observer (Platt et al. 2018). The solution to this problem largely depends on the scattering properties of the background atmosphere and of the plume in terms of aerosol load, aerosol types, and height distributions, which are all highly variable and poorly constrained by external information for the instantaneous and local measurements envisioned here. The complexity also depends on whether the target gas occurs in large background concentrations such as for CO_2 or whether it only occurs within the plume such as for HF and HCl. For the latter, observations of the plume geometry might deliver sufficient information to obtain a meaningful estimate of the respective plume concentrations, provided the plume is not too opaque, so that the (infrared) sunlight can be assumed to travel through the entire plume cross section. For the former, one would need either accurate radiative transfer simulations fed with accurate aerosol properties or a proxy gas that shares the same lightpath but is not emitted by the volcano. In this case, the radiative transfer problem is similar to the one encountered in UV SO_2 measurements (e.g., Kern et al. 2013). For CO_2, such a proxy is typically methane (CH_4), which has a similar distribution of background concentrations but is not emitted in relevant amounts by the volcano. CH_4 also has absorption features in spectral proximity to CO_2 (around 1650 nm), such that the spectral variation of the optical scattering properties is mild enough to assume very similar lightpaths. Overall, the complexity of radiative transfer is a further major challenge to the type of volcano remote sensing we investigated here.



3. Role of aerosols and scattering-induced dilution

The manuscript should discuss in greater detail how aerosols influence the retrieval.

Scattering simultaneously increases the available signal and introduces uncertainty in the effective light path. The retrieval ultimately depends on distinguishing photons scattered before entering the plume from photons scattered after leaving the plume.

This effect is expected to vary significantly between the HCl, CO_2, and HF retrieval windows because of their different wavelengths and scattering properties.

A sensitivity study investigating aerosol optical depth and aerosol vertical distribution would considerably strengthen the conclusions.

See our reply to comment 2. We added a discussion paragraph that highlights the radiative transfer challenge, including the differences for gases with/without background.

4. Validation of retrieved air-mass factors

The retrievals provide slant columns that are linked to vertical columns through an air-mass factor.

For the Heidelberg region measurements, reasonably accurate estimates of the atmospheric CO_2 and O_2 columns are already available from COCCON and TCCON observations. Therefore, it should be possible to compare the retrieved vertical columns with independent measurements and report the corresponding air-mass factors.

Such a comparison would provide valuable insight into:

- the realism of the retrieval;
- the dependence of air-mass factors on solar zenith angle;
- the relative importance of atmospheric variability, aerosols, and geometry.

This information would also help interpret the variability shown in Figure 4.

See reply to comment 2. We based our SNR analysis on SCDs, not vertical columns. I.e. we did not require running a radiative transfer model, and we did not calculate airmass factors in a scattering atmosphere. It is due to the complexity of the scattering problem, that we believe a dedicated paper should address the radiative transfer problem (in particular, if we decided to actually build and deploy the instrumentation).

In fact, RemoTeC – the algorithm we used – is not able to solve the radiative transfer for an upward looking observer out of the box. The scattering module has been designed for downward-looking observers and due to the sophisticated (and efficient) calculation of the Jacobian matrix (via forward adjoint perturbation theory) the algorithm is not directly applicable to upward-looking geometries. That is, solving the scattering radiative transfer problem would require substantial algorithm developments on our side.

In Section 4.3 of the revised manuscript, we clarified that the Allan deviation shown in Figure 5 (Figure 4 in the preprint) is calculated from retrieved slant column densities. We further explain that the apparent degradation at longer averaging times is not a true loss of performance but rather results from changes in the slant column density caused by the changing solar position, which eventually dominate over the statistical noise.

5. Instrument optimization and alternative observing strategies

The manuscript focuses primarily on detector SNR limitations. However, other approaches to improve the measurement sensitivity should at least be discussed.

For example:

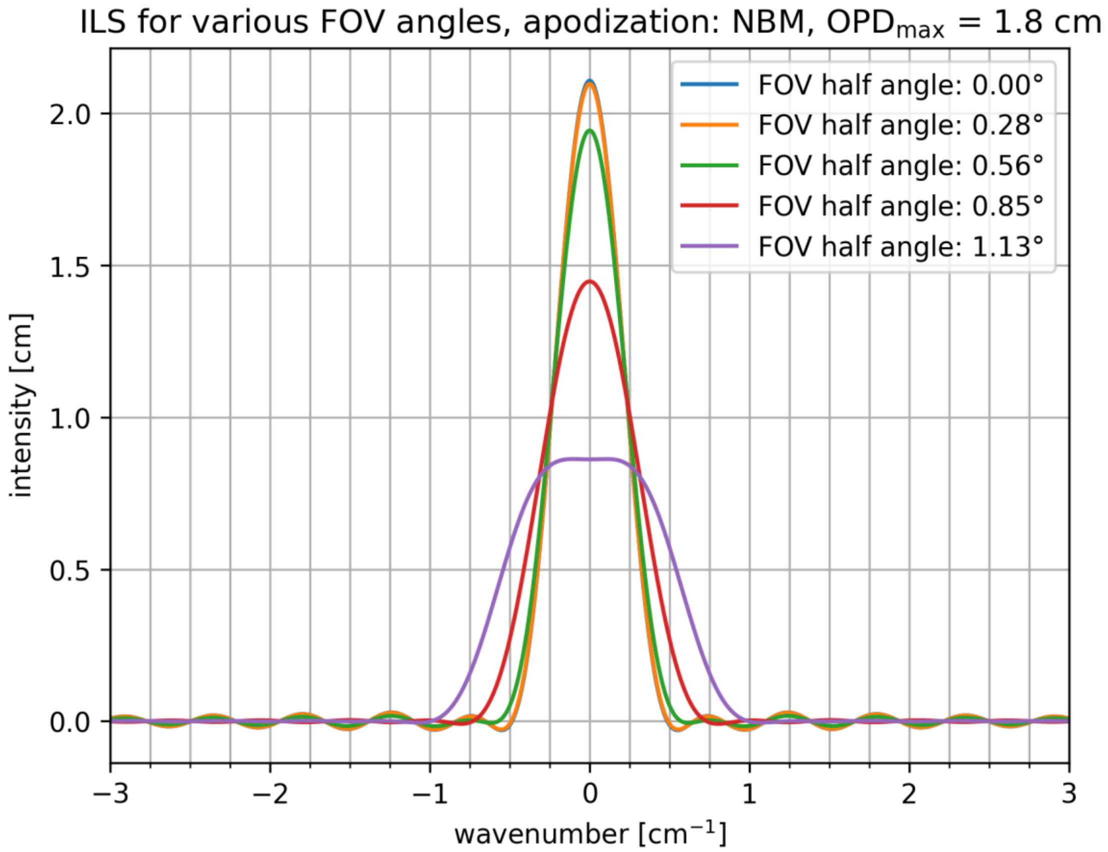
- Increasing the field of view of the detector
- Relaxing requirements on instrumental line shape
- Operating at lower spectral resolution
- Investigating cloud-scattered sunlight as done by Love et al. (1998);
- Direct observations of the sky (plume) without tracker or fiber coupling to a pointing system;
- Alternative optical configurations optimized for signal throughput rather than high-accuracy column retrievals.

Even if such approaches are beyond the scope of the present work, a discussion of their potential benefits and limitations would help justify the negative conclusion.

We thank the reviewer for these valuable suggestions on how to further improve the measurement precision and address them point by point below.

The reviewer is correct that increasing the detector field of view (FOV) is an effective way to increase the instrument's étendue and, consequently, improve the measurement precision. We agree that this possibility was not fully explored in the original manuscript.
Our initial analysis was based on a FOV half-angle of 0.28°, as the resulting instrument line shape (ILS) is nearly identical to the ideal ILS (see figure below). Furthermore, 0.28° is the FOV half-angle of the EM27/SCAv2 instrument used to derive the CO_2 precision shown in Figure 5 (Figure 4 in the preprint). Consequently, the scaling presented there is valid for the corresponding ILS.
However, increasing the FOV half-angle to 0.56° (corresponding to a focal length of 50.8 mm) does not yet lead to a significant degradation of the ILS. We therefore thank the reviewer for pointing out this opportunity and have adopted this larger FOV for the optimized EM27/Volcano instrument in the revised manuscript. As a result, the instrument performance shown in Figure 2 and Figure 7 (Figure 6 in the preprint) approximately doubles. Upon updating the étendue in Table A1, we identified an error in the value reported in the preprint manuscript, which was overestimated by a factor of 4. The value is now corrected and corresponds to a focal length of 50.8 mm. This correction only affects the value displayed in Table A1; all calculations and figures presented in the preprint were already correct.

We believe that a FOV half-angle of 0.56° represents a good compromise for this study. First, the scaling used in Figure 5 assumes a nearly ideal ILS. Using a substantially larger FOV would introduce noticeable ILS degradation and therefore lead us to overestimate the achievable trace gas precision. Second, increasing the FOV further would require focal lengths below 50 mm, which, in our experience, are difficult to realize with 2-inch optics and are more prone to alignment-induced ILS asymmetries. Finally, at even larger FOVs the absorption lines become noticeably shallower, reducing the sensitivity of the trace gas retrieval.



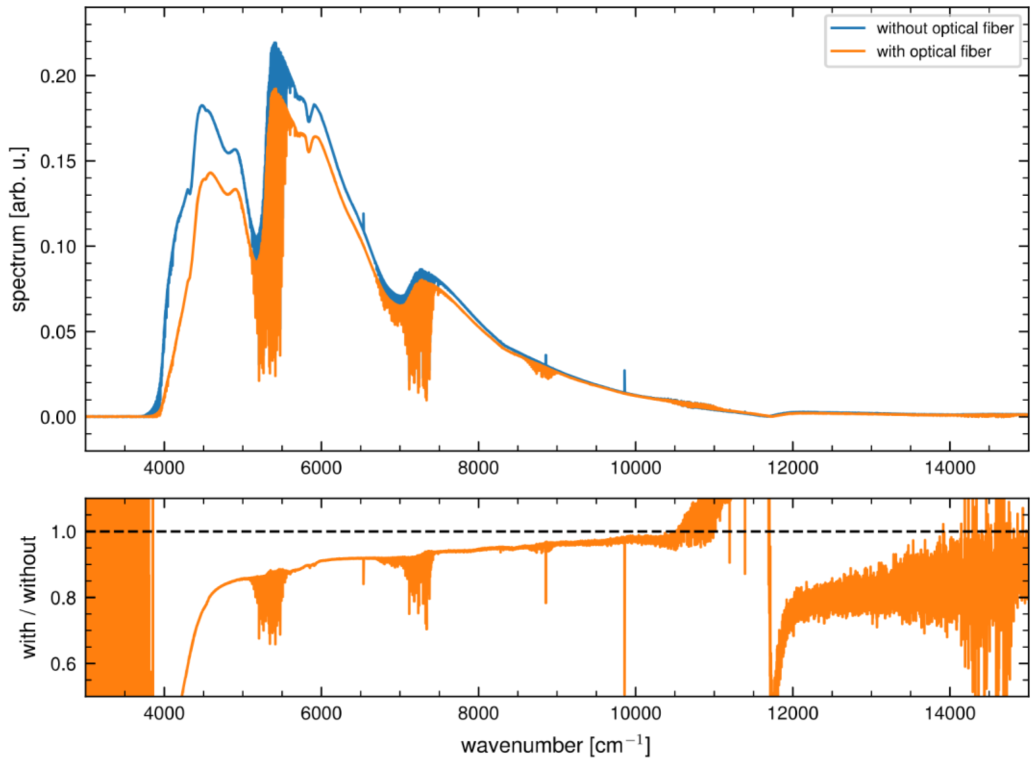
Concerning the spectral resolution, we based our study on the existing Bruker EM27/SUN platform, which has already been successfully deployed for volcanic measurements in direct-sun geometry (Butz et al., 2017). Since this platform does not allow the user to reduce the instrument's spectral resolution, we did not investigate this parameter further. Instead, the aim of this study is to assess the feasibility of volcanic trace gas measurements using an existing FTIR platform, rather than designing a new instrument from scratch.

In principle, lowering the spectral resolution would increase the spectral SNR according to Eq. (10) in the manuscript. At the same time, however, the reduction in spectral sampling decreases the available data points containing information. This scales as the square root of the maximal optical path difference (OPD), resulting in an overall scaling of the information content of the inverse square root of the maximal OPD, so less of an increase as Eq. (10) in itself implies. Furthermore, the broader instrument line shape makes individual absorption lines increasingly difficult to

resolve, reducing the sensitivity of the trace gas retrieval and thereby offsetting part of the gain from the increased SNR.
We therefore expect that optimizing the spectral resolution could yield a modest improvement in measurement precision. However, we do not expect improvements of the orders of magnitude that would be required to make volcanic CO_2 measurements feasible with the proposed instrument concept.

Observations based on cloud-scattered radiance are partially addressed in the manuscript by considering a heavily aerosol-loaded atmosphere, which provides the lower performance bound in Figure 7 (Figure 6 in the preprint). Although cloud-scattered radiance may provide higher signal levels, it is expected to exhibit substantial temporal and spatial variability. This would complicate the observational geometry and limit the available observation periods. Consequently, we believe that such observations are not well suited for routine, continuous measurements, which is the objective of this study.

Concerning the use of a fiber coupled tracker or "direct" observation with the instrument: We measured the transmission of the fiber-coupled solar tracker, including the optical fiber itself, in a laboratory experiment using an integrating sphere equipped with halogen lamps as a spatially homogeneous, broadband light source. The measured transmission is shown in the figure below:



The transmission exceeds 80% across all spectral regions considered in this study (i.e. the spectral windows shown in Figure 4). For the SNR prediction using Eq. (10), we therefore assume an average fiber transmission of 85%-92%. Direct observation, foregoing the fiber, would thus not change the outcome significantly.

And then the minor comments:

Specific comments

Introduction

Page 2, line 45

The sentence

"There were a few attempts to take advantage of IR emission spectroscopy using the thermal emissions of the volcanic gas itself (Love et al., 1998; Goff et al., 2001), but these remained isolated case studies."

could be reformulated in a more balanced way.

For example:

"There were several successful attempts to exploit infrared emission spectroscopy using thermal emission from volcanic gases as well as cloud-scattered sunlight (Love et al., 1998; Goff et al., 2001)."

This wording better acknowledges the pioneering contributions of these studies.

We thank the reviewer for this suggestion and have adopted the proposed wording.

Definition of geometry

Line 67

The terms "on-axis" and "off-axis" are not clearly defined.

Consider replacing them with:

- "on-plume" and "off-plume";

or explicitly defining the axis as the observer–plume viewing direction.

We adopted the proposed wording.

Discussion of Love et al. (1998)

The manuscript briefly cites Love et al. (1998), but the discussion could be expanded.

Although the study relied largely on thermal emission, it also exploited cloud-scattered sunlight. This sounds at least similar as the sky-scattered sunlight approach investigated here.

The comparison may help clarify why the present approach is particularly challenging.

We thank the reviewer for this valuable suggestion. In the revised Introduction, we now address both observation concepts considered in this work: thermal emission and cloud-scattered sunlight.

In the Discussion, we now compare the predicted measurement performance for HF and HCl with the results presented by Love et al. (1998), where a direct comparison is possible. To this end, we had to estimate the atmospheric state to convert their given dry mole fractions into column densities. We settled on a pressure of 500 hPa at 6 km height and a temperature of –25 °C (25 °C temperature at sea level, an adiabatic lapse rate of 10 K/km and the 10 K plume temperature enhancement given in their paper). This yields a number density of 1.5e25 molecules per cubic meter and should be a valid assumption considering, that this is a comparison of scales.

As discussed above, we believe that cloud-scattered observations may provide higher background radiances but are inherently limited in space and time. Consequently, they are not well suited for flexible viewing geometries or continuous monitoring.

Distance to the scattering layer

For volcanic applications, the effective distance and altitude of the scattering layer are important parameters.

This issue has previously been discussed in the context of SO_2 camera measurements (e.g. Campion et al., 2015; Schiavo et al., 2020) and may deserve to be mentioned in the manuscript.

In the revised manuscript, we now cite Kern et al. (2013), which provides a comprehensive discussion of the radiative transfer problem in UV scattered-light observations that is applicable beyond camera measurements. It is cited multiple times within Campion et al. (2015). We believe that many of the concepts discussed there are also relevant for the development of scattered-light retrievals in the NIR, for example for HF and HCl. (see our reply to Major Comment 4).

Retrieval examples from Heidelberg

Since retrievals were successfully performed on the roof in Heidelberg, I encourage the authors to include examples showing:

- measured spectra;
- simulated spectra;
- slant columns;
- vertical columns;
- air-mass factors;
- scattering-weight profiles.

These examples would greatly improve transparency and reproducibility.

Please see our replies to Major Comments 1 and 2. Our response to Major Comment 2 also includes a time series of retrieved CO_2 slant column densities. However, we have not included this figure in the revised manuscript, as we believe it would not add significant information beyond the discussion and figures already presented.

Precisión estimates

The manuscript would benefit from a comparison between:

- analytically predicted retrieval uncertainties;
- empirical precision estimated from consecutive measurements.

Agreement between both estimates would increase confidence in the uncertainty analysis.

At a 1-minute averaging time, the propagated retrieval errors are 7.8×10^{20} molecules / cm^{-2}, while the datapoint at lowest SNR in Figure 5 (Figure 4 in the preprint) which corresponds to the same averaging time yields 1.0×10^{21} molecules / cm^{-2}. These values are in good agreement.

We have added a corresponding statement to the discussion of Figure 5 (Figure 4 in the preprint).

Instrument description

The discussion of the modified EM27/SUN could be expanded.

Compared with the original EM27 (OPAG 22), the EM27/SUN includes several modifications, including changes to detector, beamsplitter, focal length, field stop, and aperture configuration. These changes improve the instrumental line shape, accuracy, and potentially precision, but they all lower the SNR.

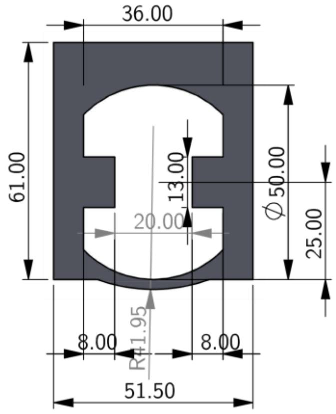
The authors may wish to discuss whether a different aperture configuration could improve signal throughput in the proposed application. I imagin that you will get a different curve in

The instrument used in this study (referred to as the EM27/SCAv2 in the manuscript) is a modified EM27/SUN. The modifications include a different detector, focal length, and aperture.

The figure below shows the custom-built Jacquinot stop inside the instrument. Its shape was designed to maximize the aperture area while preventing the reference laser from reaching the detector, which explains its characteristic peanut shape.

We use the area of this aperture to calculate the instrument étendue and believe that no significant further increase is possible for an instrument based on 2-inch optics.

We have clarified the differences between the EM27/SCAv2 and the EM27/SUN in Section 3 of the revised manuscript, but refrained from giving exact values for the EM27/SUN, since it was never used in the context of this study. But we ensured that the differences between the EM27/SCAv2 and the further optimized EM27/Volcano are clearer.



Etna measurements

If spectra from the Etna campaign are available, I strongly encourage the authors to show them, even if no successful volcanic retrievals were obtained.

Negative examples are scientifically valuable and would help readers understand the practical limitations encountered during the experiment.

We thank the reviewer for this suggestion. Unfortunately, we do not have FTIR spectra acquired in a scattered-skylight geometry at Mt. Etna available for analysis.

Some technical suggestions:

1. Show the measured and fitted spectra of the Microwindows

   We added Figure 4 to the manuscript (see major comment 1).

2. Explain the radiative transfer and the algorithm using equation.

   We added a paragraph to Section 3, describing the retrieval algorithm in greater detail (see major comment 2).

3. Quantify aerosol/scattering effects for the Heidelberg measurements; Validate or at least comment on the air-mass factors some how.

See our reply to major comment 4.

4. Discuss whether the instrument and observing strategy were fully optimized before concluding that the method is not feasible.

   See our reply to major comment 5.

I did not get fully understand the instrumental model and how Table A1 is linked, if it would be possible, It would have liked to have maybe two pairwise instrument configurations orientated on the EM27/sun and the EM27 from Bruker: just add two columns in table A1 and put the numbers and discuss the possibility to improve “étendue”:

We thank the reviewer for the suggestion. After considering it, and following the changes made in response to Major Comment 5, we decided to list the parameters for both the existing EM27/SCAv2, which was used for the validation study, and the proposed EM27/Volcano. We believe that additionally including the original EM27 or the EM27/SUN, neither of which is discussed further in this study, would be more likely to cause confusion than to provide additional benefit.

focal length 10cm-> 3.5 cm-1 (factor 3), aperture (actual maybe 2 cm) -> double (4cm) (factor 4), with pointing optic and without pointing optic, with default detector and with suggested narrow band detector (well described).

See our reply to major comment 5.

And as the authors state the apodization function is somehow a degradation of the spectral resolution, and it is not independent, if you change the field of view of the detector to obtain more Photons you get self apodization, not so important in the mid infrared.

FOV=half of internal field of view=detector_aperture_radius/focal length

Delta v = 0.5*v (FOV/2)^2

Delta v = 0.5*v (1mm/35mm/2)^2= 0.6 cm-1 > resolution EM27/sun

(but not too far away)

Why not using more from the beam diameter /aperture in figure, probably you will also have to use a stronger apodization function.

We use a custom-built aperture that maximizes the optical throughput while preventing the reference laser from reaching the detector. Please see our reply to the reviewer's comment entitled "Instrument description" and to major comment 5.

And why not reduce the spectral resolution, is there a optimal spectral resolution for the geometry?

We thank the reviewer for this valuable comment. We address this point in our response to Major Comment 5.

The settings optimized for direct sun observations might be newly optimized for the new application.

6.What happens if the light goes through a fiber? It would be nice to see it by measurements, if you have an almost homogen atmosphere on the roof of

Heidelberg and nothing special to point to (because you have no direct sun and not a plume), this would be ideal to measure with and without pointing optics.

We have added a plot showing the fiber transmission in our response to Major Comment 5.

I should note that I used ChatGPT to help improve the wording of my comments. Unfortunately, this may have resulted in a tone that sounds somewhat more direct or impolite than I intended. I sincerely apologize if any part of the review came across that way.