RC1: 'Comment on amt-2023-101', Anonymous Referee #1, 04 Jul 2023

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GENERAL COMMENTS

The large portions of the GHG gases emit from global mega cities and point sources. Remote sensing form upper sky is a powerful tool to capture potential emission plumes, however, the amount of data with satellites and airplanes is limited. Local time of the existing satellites are around noon. The emission estimates from satellite data have large uncertainties due to local weather conditions such as wind speed and direction. Diurnal variation data from the fixed point will improve emission monitoring. The modification of the light source from a direct solar light (COCCON) to scattered light instrument and its characterization tests are well described in the manuscript. Technical portions are good. However, scientific discussions are needed. I have the following general comments. The discussions on additional characterization and applications will improve the scientific significance. Major revisions are needed.

We thank the reviewer for the appreciation of our work and the helpful comments. Find our point-by-point reply below.

(1) Retrieval

The present retrieval seems to be modification of the direct solar measurement such as COCCON to slant column densities. Aerosols over LA Basin causes large fluctuations with the large off nadir geometry. Are the authors planning to retrieve several parameters such as aerosol optical thickness, surface albedo, surface pressure from EM27 spectra?

Yes, we plan to investigate the impact of aerosols on our measurement. For future studies, we will develop a simultaneous retrieval of GHGs and aerosol properties plus surface albedo from the EM27/SCA spectra. However, this is a major complication. Our radiative transfer and retrieval software (RemoTeC) can, in principle, treat aerosol scattering but, for technical reasons (see reply to comment RC2), it cannot treat observers that are positioned inside the atmosphere (as opposed to at top-of-the-atmosphere such as a satellite). The related developments are not within the scope of our instrument performance assessment, here, but they deserve an extra study (and extra time). We added a corresponding paragraph to the discussion section of the manuscript.

Does the surface reflectance over the LA basin has strong dependency on solar zenith angles? Are there critical angles in viewing and solar zenith?

We retrieve a spectral background polynomial (c.f. L203). This removes the broadband effects originating from spectral surface reflectance for each measurement individually. If there were critical angles in terms of BDRF angle dependencies, they would show up, for example, in SNR dependencies on SZA or VZA, as the signal level directly relates to surface reflectance. We do not find such cases (c.f. Fig. 9). Additionally, since we neglect aerosol scattering within our retrieval, the directionality of surface reflectance is not incorporated in our forward model. Therefore, the retrieved background polynomial represents the surface brightness for the current viewing geometry. This would be different if the forward model included multiple scattering e.g. between an aerosol layer on the ground.

A remaining possibility for the directionality of surface reflectance to impact our retrieval would be that specular reflection spots apear within the FOV, adding to scene inhomogeneity within the FOV.

As mentioned in the manuscript, we selected the ground scattering targets to exhibit as homogeneous reflectance as possible (c.f. L134). We cannot identify critical angles due to SZA or VZA dependent scene inhomogeneity in the SZA dependence on fit RMS (Fig. AC1). For individual measurements, we occasionally observe point-like specular reflections in the imaging camera images, but their contribution to the overall signal is negligible.

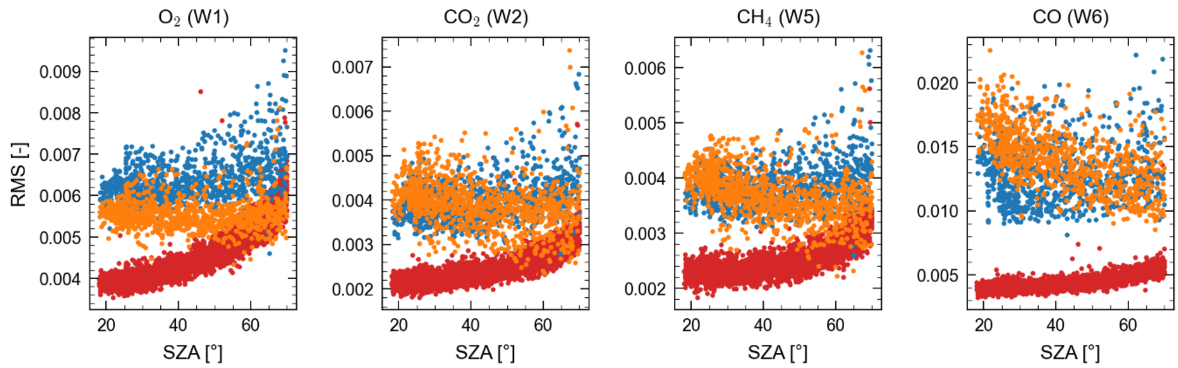

Figure AC1: RMS over SZA for the different retrieval windows. We do not observe distinct angles critical for the fit quality.

Discussions on which parameters are retrieved and assumed will suggest the importance of the measurements.

As specified in L177f. we retrieve absorber column densities with one degree of freedom simultaneously with a broadband background polynomial (L203). For reflector measurement this is equivalent to a column scaling approach employed by GFIT and PROFFIT (COCCON). For the LA basin target observations however, only the column below instrument level is scaled. This is, because we expect the main variability to occur in this portion of the light path: (1) the emission signal is located there, (2) the pathlength in this region is longest and (3) light path modifications arising from aerosol scattering also arise mainly there. Given this implementation, errors in the a priori above instrument level are mistaken as changes in the lowest layer, which is not a problem since we use only the total SCD for further analysis (c.f. Eq (3)). The remainig error is caused by attributing the molecules in the total column to regions with different pressure and temperature. The alternative implementation of scaling the entire profile would attribute the variability coming largely from below instrument level to the entire total column which would lead to substantially larger errors.

We added this discussion as paragraph after L195 and an overview table summarizing retrieved and assumed parameters to the manuscript.

(2) Instrument Resources

The EM25 spectrometer is still heavy and expensive, if we install several systems from different location. Is it possible to reduce size and weight by relaxing spectral resolution?

We consider the EM27 spectrometer quite portable and versatile, given that one instrument alone can remotely sample various locations for mapping entire regions.

Resolution is not easily tradable for size and weight, as the maximum optical path difference (OPD), on which the FTS resolution mainly depends, has only a minor contribution to the overall instrument size and weight as long as OPDs are on the order of a few cm (as typical for our case). Reducing the maximum OPD further from 1.8 cm will have a neglible influence on instrument size and weight. There is a smaller instrument available from Bruker (IRCube) with the same OPD as the EM27 but with a higher level of integration of the components and a smaller throughput (beam diameter). We have chosen for the more spacious EM27, since we needed to replace optics and electronics parts without major mechanical integration issues.

By significantly reducing the spectral resolution, we would lose the ability to resolve individual absorption lines. Doing so, we would no longer leverage the contrast between absorption line and continuum and run the risk to confound spectral structures of surface albedo with atmospheric signal. Wilzewski et al. 2020, showed that spectrally degrading spectra of the GOSAT satellite leads to worse XCO_2 consistency with ground-truth measured by TCCON, but also to biases correlating with surface albedo and particle scattering parameters.

Thus, we would not recommend relaxing spectral resolution further for mapping out rather small GHG gradients as showcased here. Worse resolution is acceptable, if the goal is to quantify emissions of localized hotspots with large GHG enhancements, as illustrated by our study using a hyperspectral camera (Knapp et al., 2023).

(3) Polarization sensitivity?

EM27/SUN for the direct sun does not care the input light polarization. However, surface reflected light and aerosol scattered light are polarized. Aerosol scattering is a major contamination source for slant viewing measurement over megacities. Do the authors characterize the instrument polarization? Do they try to measure the polarization of the input light by installing and rotating the polarizer in the front optics?

FTS are in principle sensitive to the polarization of incoming light, however the effect is less severe compared to grating spectrometers. The main effect for FTS is the polarization dependent reflectivity of the beamsplitter (e.g. (Griffiths and de Haseth, 2007) chapter 5.7). Following the Fresnel equations, the reflectivity of the beamsplitter differs for light polarized parallel and perpendicular to the plane of reflection. In our case this reduces the throughput for light polarized in the plane of the interferometer (horizontally). In laboratory measurements we found that the EM27/SCA is roughly 10% less sensitive to horizontally polarized light compared to vertical polarization (see Fig. AC2), in the spectral range 5600-8000 cm^{-1}, relevant for the CO_2, CH_4 and O_2 retrieval windows (W1-W5). The sensitivity difference increases towards lower wavenumbers. Considering that aerosol and surface reflection only polarize the light partially, we consider this a minor issue.

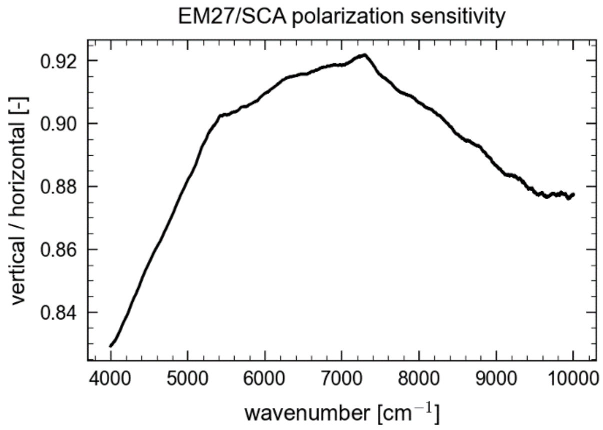

Figure AC2: Polarization sensitivity of the EM27/SCA. We show the ratio of averaged spectra recorded with horizontally and vertically polarized light.

Since, at this point, our retrieval is a transmittance calculation neglecting any scattering effects, we do not expect that polarization of the scattered light has an impact on our retrievals. Or in other words: the entire neglect of scattering causes much larger errors than the polarization effects through scattering. In first order, variable polarization of the incoming lightbeam would only lead to broadband transmittance changes, which are absorbed in the background polynomial.

We include the polarization characterization in section 2 and note its possible relevance when retrieving aerosol properties in the future (discussion section of the revised manuscript).

SPECIFIC COMMENTS

(1) P11, Line 189 “geometric assumptions, … not uniform”

It is not clear. More detailed description is needed.

Thank you for the feedback. We updated L189f. of the manuscript with a more explicit explanation.

(2) P14, Lines 283-284, “SNR …. is most compact”

How the authors mean by “compact”? Does it mean calculated SNR has low variation?

Yes, compact means that SNR has low variation around the expected SZA dependence. Expected variation for reflector measurements is a sqrt(cos(SZA)) relationship, because the signal depends on the angle under which the Lambertian reflector plate is illuminated.

(3) Page 19 Figure 14 and Page 21, Figure 15,

Discussion on wind speed and direction, possible CO2 and CH4 emission sources and ground measured surface pressure in the LA basin will improve the readers understanding. Does the TCCON data at Caltech, Pasadena show the similar trend of the diurnal variation of that day with the West Pasadena data?

Ad Figure 14: As discussed in lines 302 ff., we cannot interpret the diurnal variation shown in Figure 14, as the main effect introducing variablility is light scattering by aerosols.

Ad Figure 15: For the CH_4/CO_2 ratio presented in Figure 15, this is different: The TCCON station at Caltech is approximately 5 km away from the WP target location. The afternoon enhancement of CH_4/CO_2 we see in the WP measurements is also visible in the TCCON data (see Fig. AC3). However, a quantitative comparison between the partial VCD measured in reflected-sun geometry and the total column measurement of TCCON is not straightforward. The amplitude of the CH_4/CO_2 signal differs substantially between the measurement geometries. So, despite the high precision of the TCCON measurements, the signal is barely larger than the data scatter. While, for the less precise EM27/SCA data, the signal is clearly observable. This illustrates the value of the reflected sunlight measurement geometry.

Regarding information on wind speed and direction, these provide useful information, indeed, and the comment pointed us to an inconsistent interpretation in the manuscript. Looking at the NOAA High-Resolution Rapid Refresh (HRRR) dataset, we find mostly wind directions coming from the south-west / west direction, where major CH_4 sources are located (see Fig. AC4 below).

We adjusted our discussion of Figure 15 accordingly and added Fig. AC3 and AC4 to the appendix of the revised manuscript. Thank you for the input.

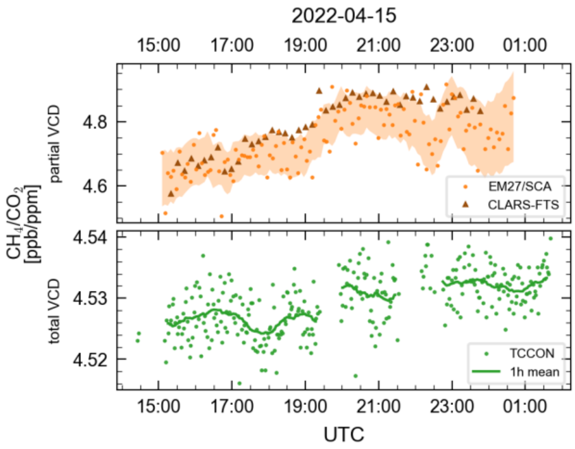

Figure AC3: Qualitative comparison of CH_4/CO_2 ratio between partial VCD below instrument level (upper panel) measured with EM27/SCA (orange dots) and CLARS-FTS (orange triangles) and total VCD measured by TCCON at CalTech, Pasadena (lower panel, (Wennberg et al., 2022)).

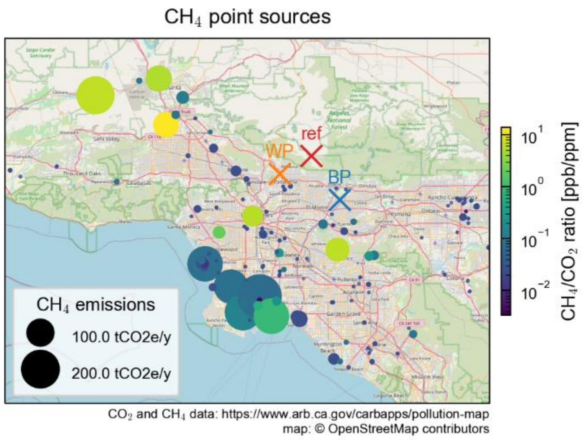

Figure AC4: CH_4 point sources as registered in the California Air Resources Board (CARB) inventory (CARB Pollution Mapping Tool v2.6, 2023). Point size corresponds to CH_4 emission strength, color corresponds to the CH_4/CO_2 emission ratio for the respective CH_4 source.

TECHNICAL CORRECTIONS

(1) Page 16, Figure 11 “RMS”

Dese it mean “RMS of SCD.

RMS refers to the root mean square error of the spectral residuals, as introduced in line 248. We will specifically write “RMS of the spectral residuals for measurements between […]” in the caption of Fig. 11.

(2) Page 24, References. Journal title abbreviation

Examples for reference types are available at https://www.atmospheric-measurement-techniques.net/submission.html.

You are correct, we corrected this in the revised manuscript, thank you.

References

CARB Pollution Mapping Tool v2.6: https://www.arb.ca.gov/carbapps/pollution-map, last access: 3 August 2023.

Griffiths, P. R. and de Haseth, J. A.: Fourier Transform Infrared Spectrometry, 2. ed., Wiley-Interscience, 704 pp., https://doi.org/10.1002/047010631X, 2007.

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