RC2: 'Comment on amt-2023-101', David Griffith, 17 Jul 2023

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The California Laboratory for Atmospheric Remote Sensing (CLARS) – Fourier Transform Spectrometer is an established facility located at Mt Wilson above the Los Angeles Basin. CLARS maps CO2 and CH4 in the urban atmosphere above LA by measurement of solar radiation scattered from targets around the basin below the observing station. Sequential measurements from a range of targets provides a mapping of CO2 and CH4 concentrations across the basin which can be used through meteorological modelling to elucidate local sources and sinks.

CLARS is a large, fixed spectrometer and thus not mobile or usable at other sites. This paper describes a valuable extension of the measurement principle to using a portable, low resolution FTS (modified Bruker EM27) called the EM27/SCA that can in principle be deployed at other suitable sites with little infrastructure requirement. The paper provides a good technical description of the EM27/SCA, an assessment of SNR and measurement precision, and a month of side-by-side measurements beside CLARS in the LA basin to assess overall performance.

This presentation is of high quality, clear and well presented, and certainly well suited to publication in AMT. However I feel it stops a bit short of its eventual purpose and usefulness and would be a much more valuable paper if the analysis could be extended as described in the general comments below. I have also made specific and minor technical comments directly in the attached pdf version of the manuscript.

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

General comments

1. The neglect of scattering in the RemoTeC retrieval of vertical column densities (VCDs) from spectra leads to significant artificial variability in retrieved VCDs as seen clearly in Fig 14. These are large enough to mask the true variability which is the ultimate aim of these measurements. This is most clearly seen in O2, which should be constant. Why has the analysis stopped at this point, with the neglect of scattering, when it is the main identified cause of the inaccuracy in the measurements? To my knowledge RemoTeC can handle scattering in its forward model, so why not include it? This may not be feasible, in this case the authors should explain why. If it is feasible, Figures such as Fig 14 would be immensely more informative because at present most of the diurnal variability we see is artefact; geophysical features of interest are masked by the artefact.

The retrievals do not include scattering treatment because the radiative transfer model in RemoTeC cannot treat observers situated inside the atmosphere. It can only handle satellite observers. This is due to the fact that the derivatives that populate the Jacobian matrix are calculated via forward adjoint perturbation theory (Hasekamp and Landgraf, 2002, 2005; Hasekamp and Butz, 2008), which is computationally efficient but only little flexible. Updating the code either by implementing observer positions inside the atmosphere or exchanging the entire radiative transfer model is a major conceptual and programming effort. But, we are currently working on an approximate implementation to overcome this limitation which, however, is not operational yet and beyond the scope of our instrument-focused publication here.

Most of the CLARS-FTS studies have used non-scattering retrievals (Fu et al., 2014; Wong et al., 2015, 2016; He et al., 2019; Zeng et al., 2023), and in particular the CH_4/CO_2 ratio. CH_4 and CO_2 undergo approximatly the same lightpath modifications by aerosol scattering. Thus, the scattering effects tend to cancel in the ratio of both. In that context, Fig. 15 shows actual GHG variability and is, methodologically, in line with previous CLARS-FTS studies.

Thus, we believe that our current analyses are valuable and do illustrate the potential of the EM27/SCA to be a portable companion of the CLARS-FTS. But the manuscript clearly highlights the need for developing retrieval techniques that correct for the effects of scattering. The revision includes a more detailed discussion on the need for the treatment of scattering.

2. Although the principal aim of the paper is technical, it would benefit at the end with some simple interpretation of the observed variability (after allowing for the scattering arefacts in 1 above) in terms of local sources and sinks using wind and other meteorological data. This is the ultimate aim of the work, but underrepresented in the present version. A full analysis, say with tomography, is outside the scope of the paper, but some simplified interpretation would improve the paper considerably.

The revision elaborates further on the case study illustrated in Fig. 15 where we find enhanced CH_4/CO_2 that is most likely attributable to upwind CH_4 sources. A quantitative assessment of source/sink patterns would require longer periods of observation and atmospheric modelling to disentangle sources from sinks and both from transport related effects. Our month-long demonstration deployment aims at demonstrating the working principle and the basic performance parameters of the EM27/SCA measurement technique and at identifying further developments needed e.g. in radiative transfer modelling. Next, we will refine the instrument and the retrievals based on the lessons learned during the demonstration deployment and then aim for a permanent deployment of the EM27/SCA.

3. CO precision. The low CO precision is ascribed to low SNR due to the low throughput near the detector cutoff wavelength. What is the impact of solar CO on the retrievals and their accuracy and precision? Much of the solar spectrum is dominated by CO lines at very different conditions (temperature, pressure), and the solar CO lines from different parts of the solar disk are also shifted in wavelength relative to terrestrial CO lines. What is the impact of solar CO on the terrestrial CO retrieval?

We use the full-disk Fraunhofer spectrum assembled by (Toon, 2015) (also recently used by (Coddington et al., 2021) for the TSIS-1 HSRS), that includes the solar CO lines. The assumption is that the solar CO lines are well represented and we have no indications from our analysis here and in previous studies, e.g. using TROPOMI spectra, that solar CO line positions or line shapes are erroneous. Even if there are uncertainties, the reviewer makes the point that the solar CO lines are spectrally shifted with respect to the telluric CO lines and that solar lines are much broader due to the higher temperatures. So, there is no direct spectral correlation between a solar CO line error and the fit to the telluric CO lines. But, of course, there could be indirect correlations via the interfering telluric CH_4 and H_2O lines. All these effects are, however, of systematic nature, while Fig. 12 and 13 clearly illustrate that noise is the limiting factor for CO. Thus, at the current level of precision for CO, we expect no impact of potentially erroneous solar CO line properties on our telluric CO retrieval.

Regarding RC2 specific and minor technical comments (supplement PDF comments)

We adopted your wording corrections, thank you for that. Please find our response to your other inline comments below.

L84: Do you mean adapted from the EM27 camtracker, ie you still use the camtracker mirrors but with manual pointing rather than solar tracking?

Yes, we use a similar alt-azimuthal rotating mirror setup with manual pointing.

L208: Does NCEP really provide O2 profiles? O2 is effectively constant for these purposes, and it is not listed in the NCEP 2000 reference.

We derive our O_2 a priori profile from the pressure information in the NCEP dataset.

L253: Could this simply be the (in)accuracy of the O2 line parameters? Was the O2 continuum included in the fit?

We do not think this is the issue, as retrievals of CLARS-FTS spectra convolved with the EM27/SCA ILS do not show increased residuals in W1. Besides the 5^{th} order polynomial accounting for the spectral continuum, we fit a pseudo-absorber with corresponding cross sections to account for O_2 collision induced absorption (c.f. Table 1). We added a sentence in the revised manuscript to clarify this.

Fig. 12: Could you add a horizontal zero line to each plot? This would make it immediately easier for the reader to assess the differences CLARS-EM27.

This is indeed helpful, we added the horizontal zero line to each plot. Thank you for the input.

We want to emphasize though, that this figure does not contain data from CLARS-FTS. As described in the caption and L265f. (Sect. 5.2), the differences shown are between EM27/SCA data and its own 30min rolling average to assess the scatter.

L286: Do you see similar differences to the CLARS data retrieved with the CLARS retrieval code? In general it would be informative to add the CLARS retrievals to this comparison. In Fig 13, I assume the CLARS spectra are retrieved with RemoTeC, in which case these are essentially a replot of the data of Fig 12.

Indeed Fig. 13 uses CLARS spectra retrieved with RemoTeC. We looked additionally at the correlation between EM27/SCA RemoTeC retrievals and CLARS-FTS retrievals using their standard retrieval algorithm (modified GFIT, see (Fu et al., 2014)). Fig. AC5 shows the correlation plots for CO_2 and CH_4. For O_2 however the target measurements show a substantial bias. It is unclear why this bias occurs. We speculate that this difference could be a result of our choice of regularization. With total column scaling, errors due to the neglect of scattering would also be attributed to regions with lower pressure where absorption lines saturate faster, leading to an underestimation of the total SCD. However, we currently have no proof for this hypothesis. When switching to the lower-partial-column retrieval as described in the manuscript, we found a decrease in the total SCD for target measurements. This effect was strongest for O_2.

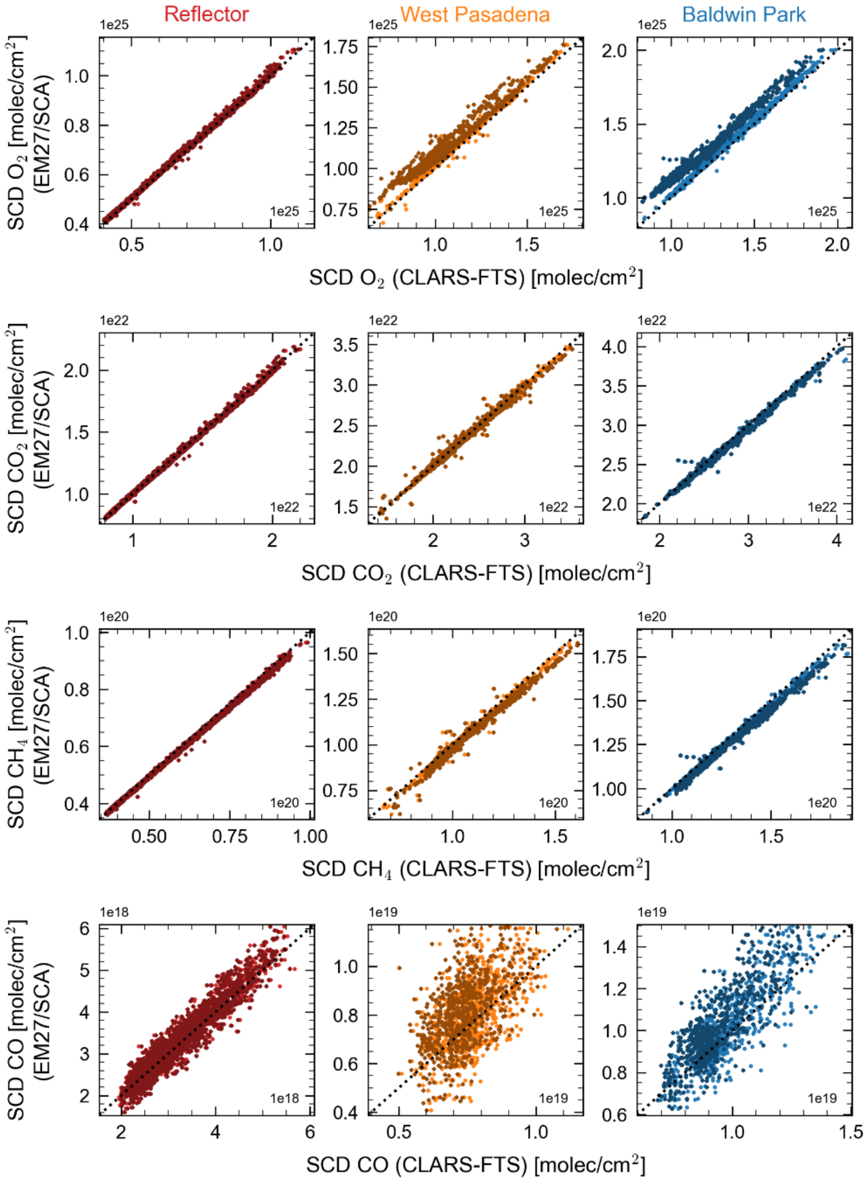

Figure AC5: Correlation of O_2, CO_2, CH_4 and CO SCDs (top to bottom) between simultaneous EM27/SCA and CLARS-FTS measurements. EM27/SCA SCDs are retrieved with RemoTeC, while CLARS-FTS SCDs are retrieved with both RemoTeC (bright dots) and the CLARS-FTS standard retrieval software (modified GFIT (Fu et al., 2014), dark dots). This figure was produced in the same way as Fig. 13 in the manuscript.

L296: Why is CLARS precision higher? Lower resolution generally leads to higher precision (lower noise bandwidth), so what else contributes? Throughput?

This is correct. Due to the higher throughput of CLARS-FTS and longer co-adding the spectral SNR of both instruments is comparable. CLARS-FTS has a larger beam diameter at similar FOV. Additionally CLARS-FTS measurements integrate over 3 min, while we report 1 min measurements. The higher spectral resolution leads to deeper absorption features. This in turn results in more precise retrievals at similar SNR.

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