

The manuscript by Schmitt et al. demonstrates the development and deployment of a near-infrared open-path dual-comb spectroscopy system for CO<sub>2</sub> (and CH<sub>4</sub>) observation over the city of Heidelberg. The manuscript is well-written and describes the dual-comb system well. While the authors demonstrate the good precision of the system for the concentration of CO<sub>2</sub>, the ambiguous position of CH<sub>4</sub> in the manuscript needs to be addressed before this manuscript can be recommended for publication.

### Specific comments:

1. My primary concern in this manuscript is that CH<sub>4</sub> is in some parts of the text presented with a more central position in this work than in other parts. For instance, while the title and abstract do not mention CH<sub>4</sub>, the second sentence of the conclusion mentions that the system “*allows to retrieve column-averaged dry-air mole fractions of CO<sub>2</sub> and CH<sub>4</sub>*”. The manuscript, however, does not contain any column-averaged dry-air mole fraction of CH<sub>4</sub>, though section 3 does discuss the problems in fitting CH<sub>4</sub> in detail. I would suggest either removing the content on CH<sub>4</sub> from the manuscript altogether, if the authors wish to focus solely on CO<sub>2</sub>, or (and this would be my personal suggestion) giving CH<sub>4</sub> a more central role in the manuscript, mentioning it in the abstract (and potentially the title).

In the latter case, I would suggest elaborating further on the fits of CH<sub>4</sub>: could the fit be improved using different reference spectra, either using observational CH<sub>4</sub> spectra or the older HITRAN08 line list (as mentioned in the HITRAN24 paper p.17, many “remote sensing” groups prefer this version as it gives the most reliable methane fits).

On p.8 of the current manuscript, a spectrum of CH<sub>4</sub> is shown, and the text describes the problems with the fit. However, what are the actual, retrieved concentrations of CH<sub>4</sub> using the current best fits? It would be interesting to learn what the achieved precision is and whether the line list problems only cause a large bias (since the line list is static and its error should therefore also be systematic). Figure 4(a) shows absorption lines with a strength around ~0.1, with residuals of those peaks generally within 0.0025, so one would quickly estimate a bias to be around or lower than 2.5% of the reported value. Is this correct?

2. The comparison to the co-deployed FTIR yields a positive bias of the DCS instrument of 0.16 ppm CO<sub>2</sub>. Since this is a major outcome in this work, further elaboration on the nature of this bias would be appropriate. For example, do the residuals of both instruments for the longest possible averaging time (thus minimal noise) show certain differences in how well absorption lines are fitted? Do both instruments fit the same absorption lines of CO<sub>2</sub> or could the difference be explained by line list discrepancies between the different absorption lines?

And is the bias stable over the fluctuations in pressure and temperature, or is the bias a temperature effect? Particularly, an analysis of the bias over varying ambient temperature could be interesting.

3. In section 2, the authors describe the data acquisition of the interferograms, which consists of a rudimentary averaging step, centering the maxima of the interferograms. While I understand that the active stabilization in principle enables this approach, I would invite the authors to comment on the level of phase noise their system has and whether this does (or does not) obstruct their averaging without any phase-noise correction.
  4. In lines 140-143, p.6, the authors describe that fitting pressure from their spectra reduced the CO<sub>2</sub> concentration precision due to the uncertainty in the retrieved pressure. Could the authors provide additional information about this finding, such as a spectral residual plot comparison of both methods or an example of the difference in retrieved concentrations for both methods over time?
  5. How big is the difference between the locally measured temperature and the path-averaged temperature from the spectrum (Line 168, p.8)? It would be interesting if the retrieved temperatures are also shown in the manuscript, similarly to the retrieved concentrations. This would help to interpret the precision of temperature retrieval as well as provide insights into the possible application of retrieving a path-averaged temperature.
  6. The authors provide a comprehensive overview of laser-based open-path spectroscopy, but have a very strong focus on the work developed by NIST. While this group has certainly been instrumental in the development of this technology, I suggest removing the statement “All of these advances originated from NIST, Boulder, Colorado, and collaborating institutions.” (line 62, p.3), as readers can interpret this from the references themselves, and “*all of these advances*” could easily be interpreted as “*all advances*” which would not do justice to other institutes working on this technique. For example, regarding open-path DCS, the authors could consider the relevant work on QCL-based open-path DCS [1]. Further, while “*Open-path Fourier transform infrared (FTIR) [...] [is] fundamentally constrained by the limited spectral radiance of thermal sources*” (line 37-40, p.2), this has been demonstrated to be overcome when combining a (laser-based) supercontinuum source with FTIR [2]. And besides the work mentioned on TDLAS, QCLs have also been employed for such laser-based open-path spectroscopy [3].
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1. Westberg, J. *et al.* Urban open-air chemical sensing using a mobile quantum cascade laser dual-comb spectrometer. *APL Photonics* **8**, 120803 (2023).

2. Krebbers, R. et al. Ultra-broadband coherent open-path spectroscopy for multi-gas monitoring in wastewater treatment. *Environmental Science and Ecotechnology* **25**, 100554 (2025).
3. Rodrigo, P. J. et al. Fast horizontal radial plume mapping of N<sub>2</sub>O using open-path absorption spectroscopy with a quantum-cascade laser. *Atmospheric Environment* 120510 (2024).
  
7. The statement in the abstract that “The results demonstrate that high-quality, long-term open-path DCS operation is achievable with readily available hardware” (line 13) is not very strong, given that long-term open-path DCS has been demonstrated in many papers by other authors. It feels like a deeper analysis of the results obtained in the work might give a stronger comparison to the existing co-deployed FTIR and other available techniques, resulting in a stronger, more substantial conclusion on the applications enabled by this DCS system.

**Technical corrections:**

1. Please check the caption of Figure 8 for language issues. For example, datapoints instead of “data-points”, timeseries instead of “time-series”, and Gaussian noise instead of “gaussian noise”.
2. I would suggest making the processed data (retrieved concentrations, temperatures, etc.) available as supplementary data.