Author’s response

This document provides detailed responses to the editor’s comments and the main reviewer comments that require substantive revisions. For the reviewer responses, we focus on addressing the major and general comments. Minor corrections and line-by-line editorial suggestions from the reviewers are not included here, as they have already been addressed in our individual responses to the reviewer reports.

Editor

Comment (Editor): Section 3.1, line 90

I can hardly believe a Gaussian line shape is assumed for modelling the absorption lines; I guess you refer to the Voigt profile?

Author’s response: You are correct, and as the reviewer also pointed out, the Voigt profile is used to model our spectral lines, while the Instrument Line Shape (ILS) is Gaussian. The simulated spectrum is therefore the result of a convolution between the Voigt profile and this Gaussian ILS.

Change in manuscript:

Page 4, lines 109–111: The absorption spectrum of gases is derived using the updated HITRAN 2020 database (Gordon et al., 2022), with spectral lines represented by Voigt profiles. The resulting spectrum is convolved with a Gaussian Instrument Line Shape (ILS), which reflects the optical and detection characteristics of the LHR system.

Comment (Editor): Section 3.1, line 102

Please rephrase: “... while TCCON a-priori information is used for CO_2 and H_2O atmospheric profiles.”

Author’s response: We have rephrased this sentence as suggested.

Change in manuscript:

Page 5, line 127-129: A priori profiles of CO2 and H2O used to construct the state vector and prior covariance matrix are derived respectively from the AirCore launches from the MAGIC campaigns (see Section 4.1) and the Orléans TCCON station, which is the closest operational site to Dunkirk.

Comment (Editor): Figure 2

The calculated H_2O signatures seem undetectable in the measured spectrum — is the slant column used for the calculation not matched properly? In the figure description, please specify SZA of the observation and total integration time.

Author's response: We now specify the SZA and total integration time in the caption. We also investigated the mismatch between the modeled and measured H_2O features. The slant column has been adjusted accordingly to improve agreement.

Change in manuscript:

Page 6, Figure 2 caption: Comparison of measured and simulated LHR transmittance spectra under clear-sky conditions in Dunkirk, for an SZA of 55° and a total integration time of 15 minutes.

Comment (Editor): Section 3.2

In the opening section of this treatment, information needs to be given concerning which quantities are fitted in the retrieval (please add a table listing all components of the state vector)…

Author's response: We have added a table in Section 3.2 listing all retrieval parameters in the state vector, including the volume mixing ratios of CO_2 and H_2O. Spectral alignment parameters such as shift and scale are handled during preprocessing, not in the state vector. A stable H_2O absorption line is used for spectral calibration.

Change in manuscript:

Page 9, Table 1: Table 1 is updated

Page 6, line 148-150: A scaling factor α is derived from the observed and theoretical line positions to correct the solar spectral abscissa. This correction is performed during preprocessing and is not part of the state vector.

Comment (Editor): Section 3.3 – Prior covariance and information content

The construction of the a-priori (see Table 1) is so oversimplified…

Author's response:

- We constructed realistic S_a matrices for CO_2 and temperature using publicly available AirCore datasets, notably from the MAGIC campaign, covering the period from 2016 to 2023. For H_2O, we used data from the ERA5 reanalysis over the same period.
- In all cases, full covariance matrices were retained, including off-diagonal elements, to preserve the vertical correlation lengths. This approach ensures more realistic estimates of the smoothing error and enhances the accuracy of the vertical information content assessment. The diagonal covariance matrices are also kept as a comparison tool.

A short explanation of the method has been added to Section 4.1 of the manuscript.

Change in manuscript:

Page 8-13: We now use realistic prior covariance matrices for CO_2 and T from the MAGIC AirCore dataset (2016–2023) and for H_2O from ERA5 reanalysis. These sections are revised and changed.

Comment (Editor): XGAS error propagation: T and SZA

… the propagation of T errors into an O_2 retrieval from the 1.26 µm band needs to be included…

Author's response: We appreciate this important point regarding the propagation of temperature and SZA errors in a column-ratio retrieval framework. However, we respectfully note that in our current LHR configuration, we are not yet able to retrieve O_2 columns, as we lack a laser source covering the 1.26 µm O_2 absorption band. Procuring such a laser is a planned future upgrade to enable direct XCO_2 retrieval via the CO_2/O_2 column ratio, consistent with the approach used in TCCON and COCCON. In the absence of an O_2 measurement, we do not currently compute XCO_2, and the uncertainty budget is expressed in terms of vertically integrated CO_2 profile uncertainty, rather than in terms of XCO_2. To clarify this and avoid confusion with standard total column quantities, we have revised our terminology throughout the manuscript. Specifically, we now refer to the 'total column uncertainty' as the 'integrated profile uncertainty' to clearly distinguish it from column-averaged quantities like XCO_2. We plan to quantify the error cancellation benefits once O_2 retrievals become available with the upgraded setup.

Regarding solar zenith angle errors, we agree that these can partially be cancelled when using a gas ratio approach. However, since our current implementation does not include such a ratio, we retain the SZA uncertainty contribution in the LHR error budget for completeness.

Change in manuscript:

Page 12-13, Section 5.2: To clarify that XCO_2 is not retrieved in our current system, we now refer to the reported uncertainty as 'integrated profile uncertainty'. This section is corrected accordingly.

Comment (Editor): Ground pressure as a model parameter

Ideally, it should be included in the error analysis…

Author's response: We agree that ground pressure is an important parameter, particularly given the high spectral resolution of the LHR and the absence of O_2-based column normalization as used in TCCON and COCCON. In our current setup, we use ground pressure measurements from a Vaisala PTU radiosonde with an accuracy of ±0.3 hPa. These values are used to overwrite the default ground pressure in the retrieval algorithm prior to inversion. Given the high accuracy of this input, and to keep the focus on dominant sources of uncertainty, we have not included ground pressure in the error analysis in this study. However, we acknowledge its potential impact and will consider its contribution explicitly in future studies.

Change in manuscript:

Page 5, lines 124-126: The calculations depend on the concentration of the target atmospheric profile, along with associated data profile such as temperature, pressure, and relative humidity, which are obtained from a nearby PTU300 Vaisala radiosonde, with manufacturer-specified uncertainties of ±0.2°C for temperature, ±0.3 hPa for pressure, and ±1% for relative humidity.

Comment (Editor): Table 3 – Add integration times

Author's response: Integration times have been added for all cases to allow for meaningful SNR comparison.

Change in manuscript:

Page 14, Table 3: A new column labeled "Integration Time" has been added to all rows.

Comment (Editor): Table 4 – Decompose column errors

Author's response:

- Measurement error,
- Model parameter uncertainty (T, SZA),
- Smoothing error.

Change in manuscript:

Page 13, Table 2: We decompose the errors for the LHR in this manner and refer to the previous study in Table 4 for comparison.

Comment (Editor): Section 4 – On channel selection

Realistic retrievals need background channels in addition to absorption peaks…

Author's response: We have revised this section to emphasize that realistic retrievals require not only strong CO_2 absorption channels, but also the identification of a set of informative channels that includes spectral regions free of absorption (i.e., baseline). The new analysis is calculated with a Jacobian that includes this baseline. Furthermore, we now present the top 100 channels ranked by information content, and importantly, we find that nearly 30% of these selected channels are located in baseline regions with little or no CO_2 absorption.

Change in manuscript:

Page 16, lines 361–364: Additionally, in **Figure 5**, we present the first 100 selected channels ranked by their information content with respect to our Jacobian. The first 30 channels are shown in red, channels 31 to 60 in blue, and channels 61 to 100 in green. Notably, the information is primarily concentrated around three absorption lines in the range 6362-6365 cm^{-1}. Interestingly, nearly 30% of the top 100 channels lie in baseline regions with little to no CO_2 absorption.

Reviewer 1

Comment (Reviewer 1): Acronyms

Many acronyms are not explained (TCCON, COCCON, MAGIC, FORUM). With some this might be fine (i.e., citation to the network main paper), but with others not.

Author's response: We have carefully revised the manuscript to ensure that all acronyms are defined at their first occurrence. Where appropriate, we also cite the primary publications describing each network or mission to guide the reader toward more detailed information.

Change in manuscript:

Page 2, lines 32 (example): The COllaborative Carbon Column Observing Network (COCCON)...

Comment (Reviewer 1): State and measurement vectors

You write a lot about your model and retrieval, but in the end it is a bit vague what exactly you use in your state vector (for the gases: mixing ratios, concentrations, column densities) and in your measurement vector (radiances, transmittance, ...). I would consider this the most relevant information on a higher level of how your retrieval is designed.

Author's response:

To clarify the retrieval design, we have added a dedicated paragraph at the beginning of Section 3.2 that describes the structure of both the state and measurement vectors:

- The **state vector** contains the vertical profile of CO_2 volume mixing ratios (VMRs) on a fixed grid. Depending on the setup, it may also include temperature scaling parameters and interfering species (e.g., H_2O).
- The **measurement vector** consists of calibrated radiance spectra derived from solar absorption observations. These are processed using the high-resolution solar reference spectrum from SOLAR-ISS (see also our response to the comment on Line 94).

Change in manuscript:

Page 6, lines 144–150: Depending on the retrieval scenario, the state vector may also include additional parameters, such as a scaling factor for atmospheric temperature. The measurement vector y comprises calibrated radiance spectra derived from observed solar absorption, computed by multiplying the solar spectrum (transmittance) with the SOLAR-ISS spectrum (see Section 3.1). Prior to retrieval, all measured spectra are corrected for spectral shift and solar abscissa scale by calibrating against a stable, unsaturated H_2O absorption line. A scaling factor α is derived from the observed and theoretical line positions to correct the solar spectral abscissa. This correction is performed during preprocessing and is not part of the state vector.

Comment (Reviewer 1): Information Content Analysis – Missing Considerations

Regarding your results of the information content analysis of the spectrum (Figure 4 and 5), I am not completely convinced, since I miss a few points in the discussion:

1. *You do all of this analysis in some type of absorbance space—but to get there from measured radiances, a "background channel" is definitely needed, which I do not see represented in your results.*
2. *You are only considering the CO_2 information at the moment, but the large advantage of using a widely tunable laser is that you can measure full rot-vib bands and get constraints on temperature—degenerate with gas amount for a single line or few lines.*
3. *Are you proposing to simply limit the used channels in a retrieval or also to limit the measured spectral bandwidth?*

Author's response:

1. If the "background channel" here refers to a reference radiance spectrum without CO_2 absorption (a clean solar background spectrum), unfortunately, such a measurement is not possible in our case because atmospheric absorption is always present along the sunlight's path. Instead, we adopted a commonly used method in solar absorption spectroscopy: we applied a baseline fitting procedure over a broad spectral window to approximate the background continuum. At the same time, we corrected for variations in sunlight and local oscillator laser intensity. This gives us a transmittance spectrum without needing a separate background measurement. Also, while a lab-based LHR system can measure its own heterodyne background, this isn't a valid replacement for the solar background.

2. We agree that one of the major advantages of using a widely tunable diode laser is its ability to span entire rotational-vibrational bands, providing sensitivity to temperature through the shape and relative intensities of spectral lines. However, the primary aim of this paper is to introduce a new, time-efficient LHR system that can be deployed in field campaigns and achieves accurate retrievals comparable to those from FTIR systems. Unlike FTIR, the larger the spectral range we cover, the longer the acquisition time. Therefore, it's important to find a good balance between spectral coverage and acquisition duration. The integration time should not exceed 5 minutes; otherwise, the air column may become too mixed and the optical path length may vary.

3. Our intention is to identify and prioritize informative spectral channels within the measured range to improve retrieval stability and reduce computational cost. Depending on the scenario, and specifically in campaign measurements, we propose to limit the measurement bandwidth, especially given the high integration time.

Change in manuscript:

Page 16, lines 364–365 (example for 3): This suggests that, in future acquisitions, the combined range can be used to enable faster measurements while preserving a small scan step.

<u>Reviewer 2</u>

Comment (Reviewer 2): Manuscript organization and clarity

While the scientific content is of interest, the organization of the manuscript would benefit from improvement… I suggest restructuring Section 3 to reflect a clearer progression: Theory → Application → Results → Comparison. Also, the current use of "a priori" in Sections 3.3.2 and 3.3.3 is confusing.

Author's response: We have reorganized Section 3, as the reviewer asked, as follows:

- Section 3: Theory now includes the forward model and information content analysis (3.1 and 3.2).
- Section 4: Application to the LHR Instrument, now includes the specifics of the a priori state, measurement errors, and non-retrieved parameter treatment (revised from 3.3).
- Section 5: Results → Information Content and Uncertainty, contains the analysis based on LHR simulations (previously 3.4).
- Section 6: Comparison with Existing Networks, presents the comparison with TCCON and COCCON systems (previously 3.5).

We have also clarified the use of the term "a priori" in Section 4.1. In our revised manuscript, we now define this term more precisely to include parameters such as temperature and humidity profiles that are not retrieved but are incorporated as input into the forward model with associated uncertainties. These inputs contribute to the total error budget and are treated using an ensemble of perturbations, as clarified in Section 4.3.

Change in manuscript:

- Page 4–14: Sections reorganized to match the logical structure: Theory → Application → Results → Comparison.
- Page 6, lines 140–150: Clarified distinction between retrieved state vector and non-retrieved model inputs.

Comment (Reviewer 2): Clarification of EM27/SUN and spectral resolution

In the introduction (lines 30–36), I recommend expanding the description of the EM27/SUN spectrometer to improve clarity. For example, line 31 should clearly refer to it as the Bruker EM27/SUN, and you can also include the spectral resolution for comparison against the earlier stated IFS125HR spectral resolution...

Author's response:

- In the Introduction (lines 30–36), we now explicitly refer to the Bruker EM27/SUN, and include its nominal spectral resolution of 0.5 cm^{-1}, in contrast to the IFS125HR's 0.02 cm^{-1}.

- We have revised the sentence about portability and spectral resolution to clarify that reduced resolution arises from design trade-offs in optical path length due to compactness, not portability per se.
- We now cite Herkommer et al. (2024) and Mostafavi Pak et al. (2023) to highlight that the EM27/SUN still performs remarkably well in CO_2 retrievals. Please refer to answer 6 to reflect on whether the increased resolution of LHR leads to meaningful improvements in retrieval accuracy.

Change in manuscript:

- Page 2, lines 33–40: These spectrometers are relatively easy to operate and enable measurements in locations inaccessible to larger systems, with a spectral resolution of 0.5 cm^{-1} (Table 3), a trade-off from their compact design which limits the maximum optical path difference. While their portability allows for flexible deployment, maintaining network-wide consistency and coordination remains a significant logistical and technical achievement (Frey et al., 2019). Moreover, several studies have directly compared the performance of the high-resolution IFS125HR with the portable EM27/SUN spectrometers, including Pak et al. (2023) and Herkommer et al. (2024), showing that CO_2 retrievals from the EM27/SUN differ by only about 0.1%, a remarkable result considering its lower spectral resolution.

Comment (Reviewer 2): Radiosonde and ancillary data

In Section 3.1, where you describe the use of PTU Vaisala radiosondes and ancillary data from the TCCON database, I suggest adding more specific information to improve transparency and reproducibility…

Author's response: We have expanded the description in Section 3.1 (now Section 3) as follows:

- For the PTU Vaisala radiosonde (PTU300), we now provide typical manufacturer-specified uncertainties: ±0.2°C (temperature), ±0.3 hPa (pressure), and ±1% RH. These values are referenced and used to estimate perturbations in temperature and humidity profiles for the uncertainty analysis in Section 4.3.
- We clarify that ancillary data refers to a priori profiles of CO_2 and H_2O used to construct the state vector and prior covariance matrix. In our case, these are derived respectively from the AirCore launches from the MAGIC campaigns and the Orléans TCCON station, which is the closest operational site to Dunkirk from 2016 to 2023.

Change in manuscript:

- Page 5, lines 124–129: The calculations depend on the concentration of the target atmospheric profile, along with associated data profile such as temperature, pressure, and relative humidity, which are obtained from a nearby PTU300 Vaisala radiosonde, with manufacturer-specified uncertainties of ±0.2°C for temperature, ±0.3 hPa for pressure, and ±1% for relative humidity. A priori profiles of CO_2 and H_2O used to construct the state vector and prior covariance matrix are derived respectively from the AirCore launches from the MAGIC campaigns (see Section 4.1) and the Orléans TCCON station, which is the closest operational site to Dunkirk.

Comment (Reviewer 2): Averaging kernel comparison

In Section 3.5, include a plot comparing averaging kernels from LHR and FTS instruments.

Author's response: We agree that a direct visual comparison would enhance the interpretation of our results. However, overlaying the averaging kernels significantly reduces the clarity of the figure, as more than 160 lines become indistinguishable. Therefore, we refer the reader to our previous study for a detailed comparison of these averaging kernels.

Change in manuscript:

Page 14, lines 310-313: A comparison of averaging kernels (cf. El Kattar, Auriol and Herbin, 2020) with FTS instruments reveals sharper peaks and a more homogeneous vertical distribution than CHRIS, EM27/SUN and IFS125HR, suggesting higher sensitivity at higher altitudes though the a posteriori error S_x is significantly reduced in the lower atmosphere.

Comment (Reviewer 2): "Channel selection" terminology

The term "channel" may be misleading; consider using "line selection" or define your usage clearly.

Author's response: To avoid confusion with terminology used in the TCCON and EM27/SUN communities, we have now explicitly defined the term "channel" at the beginning of Section 7 (previously Section 4). In this study, "channel" refers to an individual spectral point (i.e., a specific wavenumber bin) in the measured radiance spectrum. We have also updated the caption of Figure 5 to reflect this definition and added the term "micro-window selection" where appropriate to clarify that this selection is based on information content per spectral point.

Change in manuscript:

- Page 15, line 326-327: To optimize acquisition, we preselect the most informative spectral points, hereafter referred to as channels, prior to measurement. Each channel corresponds to an individual wavenumber bin in the radiance spectrum.
- Page 18, Figure 5 caption updated to reflect this definition.

Comment (Reviewer 2): 2.74% uncertainty vs. mature networks

The reported 2.74% CO_2 uncertainty at 10° SZA appears high. TCCON reports 0.16% for XCO_2. How does the LHR improve over existing systems?

Author's response: We fully agree that the current level of uncertainty appears high compared to the operational performance of mature networks such as TCCON and COCCON. However, we would like to clarify that the reported 2.74% corresponds to the vertically integrated profile retrieval uncertainty, not to a total column XCO_2 uncertainty derived from a ratio of CO_2 and O_2 columns as in TCCON/COCCON. Since our current setup does not yet include an O_2 channel (due to the lack of a suitable laser source in the 1.26 µm region), a true XCO_2 product cannot yet be derived. For this reason, and to avoid confusion, we have renamed the reported quantity "integrated profile uncertainty" in the revised manuscript.
We agree that the high spectral resolution of the LHR holds great potential to reduce smoothing errors and improve retrieval quality. A full profile retrieval for CO_2 is currently under development and will be presented in a future study. We expect that this, combined with the future addition of an O_2 channel, will enable a direct and fair comparison with TCCON/COCCON XCO_2 error budgets, including potential advantages in vertical sensitivity. In this study, we focus on the initial demonstration of information content and error propagation for a profile retrieval from a compact LHR instrument, while acknowledging that further development is needed before it can match or surpass operational standards for satellite validation.

Change in manuscript:

Page 13, lines 282–287: It's important to note that TCCON's method removes systematic errors common to both the target gas and O_2 columns, which is not possible here. In our current LHR configuration, we are not yet able to retrieve O_2 columns, as we lack a laser source covering the 1.26 µm O_2 absorption band. Procuring such a laser is a planned future upgrade to enable direct XCO_2 retrieval via the CO_2/O_2 column ratio, consistent with the approach used in TCCON and COCCON. In the absence of an O_2 measurement, we do not currently compute XCO_2, and the uncertainty budget is expressed in terms of vertically integrated CO_2 profile uncertainty, rather than in terms of XCO_2.