Authors response to the reviewer

The manuscript presents an innovative approach for measuring atmospheric CO_2 using a portable Laser Heterodyne Radiometer (LHR) operating in the near-infrared (NIR) region. This work is highly relevant to the scope of Atmospheric Measurement Techniques (AMT), and the manuscript provides a thorough description of the experimental setup, along with a theoretical framework for information content analysis. The authors apply this framework to quantify the Degrees of Freedom (DOF) of the LHR instrument and compare its performance against established systems such as the Total Carbon Column Observing Network (TCCON) and the Collaborative Carbon Column Observing Network (COCCON).

While the scientific content is of interest, the organization of the manuscript would benefit from improvement to enhance the logical flow and clarity for the reader. Several key concepts require more detailed explanation to ensure they are accessible to a broader audience. Below are some general comments that I believe would help strengthen the manuscript:

1. I suggest restructuring Section 3 to better reflect the logical progression of the work, which currently mixes theoretical background, instrument-specific inputs, and results into a single extended section. Reorganizing the content under a clearer functional structure: — **Theory → Application → Results → Comparison** —would significantly improve clarity and readability. Specifically:

   Theory: Sections 3.1 and 3.2 present the forward model and the framework for information content analysis. These form the core theoretical basis of the study and could be grouped together under a dedicated section on theory.

   Application: Section 3.3 introduces the a priori information, measurement error covariance, and uncertainties in non-retrieved parameters as they pertain to the LHR. This section represents the application of the theoretical framework to the specific case of the LHR instrument and should be distinguished from the more abstract theory above.

   Results: Section 3.4, which applies the framework to retrieve CO_2 information content and uncertainty from LHR simulations, could be promoted to its own section—e.g., “Information Content and Uncertainty Estimation for the LHR”. This would clearly signal the shift to presenting results derived from the defined retrieval setup.

   Comparison: Section 3.5, which compares the LHR with existing systems like TCCON and COCCON, should also be elevated to a standalone section, such as “Comparison with Existing Networks”, to help readers easily locate this critical performance evaluation.

   The current placement of Sections 3.3.2 and 3.3.3 under the heading “A priori information” may be misleading. While Section 3.3.1 appropriately discusses the CO_2 profile and its covariance matrix as part of the a priori state vector, Section 3.3.2 refers to measurement error, and Section 3.3.3 introduces parameters such as temperature, humidity, and SZA as non-retrieved. However, in many retrieval frameworks, temperature and humidity profiles are typically treated as a priori inputs. Please clarify your definition of ”a priori“ to avoid confusion regarding the role of these parameters in the forward model versus the retrieval.

2. 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.
   Additionally, the statement "the drawback of being portable is that the FTS reduces the spectral resolution" is somewhat misleading. The reduced resolution is not inherently due to portability but rather results from design trade-offs in optical path difference: smaller instruments have shorter maximum optical path lengths, which limits achievable resolution.
   You could also expand on the consequences of lower spectral resolution. Specifically, lower resolution can limit the ability to resolve narrow absorption lines, potentially leading to increased interference from neighboring lines, reduced retrieval precision, and sensitivity to pressure broadening effects.
   Moreover, there are published studies that directly compare the performance of the IFS125HR and the EM27/SUN, such as Herkommer et al. (2024) and Mostafavi Pak et al. (2023), which show that CO_2 retrievals from the EM27/SUN differ by only approximately 0.1%, which is quite impressive given its lower spectral resolution. This raises an important question for the present study: does the LHR system, with its much higher spectral resolution, offer a meaningful improvement over this offset?

3. 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.
   For the PTU Vaisala radiosonde, please include the typical accuracy specifications for temperature, pressure, and relative humidity. These values are likely used to define the uncertainties in your forward model or retrievals later in the analysis (e.g., Section 3.3.3), so it would be helpful to establish them clearly at this stage.
   Regarding the TCCON database, it would be beneficial to specify which TCCON station the ancillary CO_2 and H_2O data are derived from, especially considering the measurements are conducted in Dunkirk (51.035°N, 2.369°E). Are you using a nearby TCCON site (e.g.,Orléans)? Additionally, please clarify what do you mean by ancillary data? Do you mean the a priori profiles?

4. In Section 3.5, you describe differences in averaging kernels between the LHR and existing FTS instruments (e.g., EM27/SUN and IFS125HR). To support this comparison more effectively, I recommend including a plot with the averaging kernels from those FTS instruments overlaid on top of the LHR kernel.

5. In Section 4, the term "channel selection" is used to describe the identification of individual absorption lines with the highest information content. However, in the TCCON and EM27/SUN communities, "channel" typically refers to detector channels (e.g., InGaAs vs. Si), rather than specific spectral lines or intervals within an absorption band. This difference in terminology may lead to confusion for readers familiar with those systems. To improve clarity, consider using more precise terms such as "line

selection" or "micro-window selection", or alternatively, explicitly define your use of "channel" at the beginning of the section.

6. In the conclusion, you report a 2.74% error in total column CO_2 at 10° SZA. This level of uncertainty appears quite high, especially when compared to existing ground-based systems:
   TCCON reports an error budget of 0.16% for XCO_2, and the COCCON network shows an average offset of 0.1% relative to TCCON (e.g., Herkommer et al., 2024; Mostafavi Pak et al., 2023).
   Given that one of the key motivations stated in the introduction is that LHR's higher spectral resolution should improve retrieval quality, the reported uncertainty seems to contradict this expectation. It would be important to clarify how this instrument would compete with EM27/SUN in operational or satellite-validation contexts.

Response to Reviewer Comments

We thank the reviewer for their thorough and insightful review. We appreciate the recognition of the novelty and relevance of our approach, as well as the constructive suggestions to improve the manuscript. Below, we respond point-by-point to the comments and outline the corresponding revisions made.

1. Reorganization of Section 3

We have reorganized Section 3, as the reviewer asked, as follows:

- Section 3: Theoretical Framework, 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.

2. Clarification of EM27/SUN description and spectral resolution

We appreciate the reviewer's suggestions regarding the description of the EM27/SUN spectrometer:

- 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.

3. Radiosonde accuracy and ancillary data clarification

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.

4. Averaging Kernel comparison plot

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.

5. Terminology clarification on "Channel Selection"

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.

6. Reported XCO_2 uncertainty and comparability to TCCON/COCCON

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.

Minor corrections and comments:

• Line 9: Please be more specific, what type of sensitivity you are referring to. What kind of resolution is meant, spectral, temporal, vertical?

Response: We have revised this sentence to clarify that we are referring specifically to *vertical sensitivity* enabled by the high *spectral resolution* of the LHR instrument.

• Line 16: ... an extensive analysis...

Response: Corrected as suggested.

• Line 36: "heterodyne spectro-radiometer" is not a method, maybe you mean measurement technique?

Response: We agree and have changed "heterodyne spectro-radiometer" from being described as a method to "measurement technique" for accuracy.

• Line 52-71: You seem to be switching from the present tense (Solar radiation is captured ...) to the past tense (The modulated radiation was split by ...). I recommend using the present tense throughout, since this is a description of the standard setup.

Response: We have revised this section to consistently use the present tense.

• Line 121-122: The variables A and Sx are introduced before they are defined in equations 3 and 5. Please consider restructuring the paragraphs accordingly.

Response: We have restructured the text to ensure that variables A (averaging kernel matrix) and S_x (posterior error covariance matrix) are first introduced conceptually before being formally defined in Equations 3 and 5, respectively.

• Line 158: ”The a priori error covariance matrix Sa can be evaluated using in-situ data or climatology, but diagonal matrices are often used for space-based retrievals.” The use of “but” in the sentence implies a contrast that does not really exist.

Response: The sentence has been revised especially since we add a part where we use an a priori covariance matrix.

• Line 163: Define perr.

Response: The variable p_{err} is now defined.

• Line 198: please be more specific what do you mean by Kernels. do you mean posterior(total), measured, etc?

Response: We now clarify that “Kernels” refers specifically to *averaging kernels* associated with the posterior solution, calculated via Equation 4. This clarification has been added in the revised text.

• Line 232: The sentence “the total column uncertainty is calculated by adding up the concentration of each layer, adjusted by the dry air column (Figure 3)” is unclear and may be misleading. Summing layer concentrations gives the total column amount, but uncertainty in the total column requires proper error propagation.

Response: The sentence has been revised (see answer 6).

• Line 235: the term OPD appears to be misused. If you are referring to the increased atmospheric path length at high solar zenith angles, “slant path” would be the correct terminology.

Response: “OPD” was misused here. We have replaced “OPD” with atmospheric path length at higher SZA.

• Line 251: By green line and violet line, it seems like you are referring to Figure 3, please mention it.

Response: We have revised the sentence to explicitly reference Figure 3.

• Table 1: I recommend adding a more comprehensive caption that explains what each state vector element refers to (e.g., whether CO_2 refers to a profile or total column scaling). Be more specific about what you mean by TCCON database.

Response: We have expanded the caption to clarify:

- Whether each state vector element refers to a profile or scaling factor (e.g., CO_2 is a profile, SZA is a scalar).
- That "TCCON database" refers to publicly available Level 2 products from the Orléans station, which were used to define a priori CO_2 and H_2O profiles.

• Figure 2: please clearly indicate which curve corresponds to the measured LHR spectrum and which one to the ARAHMIS simulation. In addition, could you clarify why CH_4 was not included in the forward model simulation shown? I would also recommend adding a residual plot (i.e., measured – modeled) below the main panel.

Response:

- We now label which curve corresponds to the measured LHR spectrum and which corresponds to the ARAHMIS simulation**.**
- CH_4 was excluded from the forward model in this case for clarity and because its absorption lines do not overlap with the selected CO_2 micro-window.
- As suggested, a residual plot (observed – calculated) has been added below the main panel to help visualize the fit quality.

• Table 2: why is SZA = 10° used as the minimum value? At your measurement site in Dunkirk, the lowest achievable SZA is around 30° in summer. Using a range like 30°–80° would be more realistic and representative of actual observing conditions.

Response: We agree that a 10° solar zenith angle is rather unrealistic for high-quality direct sun observations. In our study, the 10° (and subsequently 80°) cases are used primarily as theoretical scenario to demonstrate the two extremes of the instrument's operating range, rather than to represent typical observation conditions. Our aim was to explore the range of sensitivity under idealized geometries and to facilitate comparison with previous work.

• Table 3: you present the full spectral ranges of the EM27/SUN and IFS125HR instruments. However, it would be more informative to also include the specific CO_2 micro-windows typically used for retrievals with these instruments. This would allow for a more direct and meaningful comparison with the spectral region covered by the LHR.

Response: We have now added a new row to the table listing the typical CO_2 micro-windows used in retrievals for the EM27/SUN (6173 to 6390 cm^{-1}) and the IFS125HR (6300 cm^{-1} band). This allows for a clearer comparison with the spectral region targeted by the LHR.

• Figure 3: The left panel displays numerous colored lines representing averaging kernels, but the caption and legend do not explain what these colors signify. Additionally, the right

panel legend includes five color-coded components, but do they apply to the left panel? Please consider separating or clarifying the legends to avoid ambiguity. In addition, please explicitly state in the caption that the figure corresponds to a solar zenith angle (SZA) of 10°.

Response:

- We revised the legend and caption to clearly explain the color coding of the left panel, which shows the averaging kernels for each retrieval level.
- We clarified that the right panel's legend applies only to the uncertainty decomposition.
- The caption now explicitly states that the figure corresponds to a solar zenith angle (SZA) of 10°.