Authors response to the reviewer

We would like to thank the reviewer for their careful reading and thoughtful evaluation of our manuscript. We are encouraged by the recognition of the significance of our work, especially the potential of using a widely tunable diode laser in laser heterodyne radiometry (LHR) for atmospheric remote sensing. We also appreciate the positive comments regarding the information content analysis, and channel selection strategy. In response to the reviewer's suggestions, we have revised the manuscript to improve clarity, particularly in terms of the retrieval setup, the definition of state and measurement vectors and the role of background channels in the information content analysis. We have also expanded acronym definitions and addressed the technical questions and semantic points raised in both the general and line-by-line comments. Our aim throughout the revision has been to improve the accessibility of the manuscript for both atmospheric remote sensing specialists and those coming from a laboratory spectroscopy background.

Reviewer Comment: 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.

Response: We have revised the manuscript to ensure that all acronyms are defined at first mention. We also provide, where relevant, references to foundational publications describing these networks/missions.

Reviewer Comment: You write a lot about your Model and Retrieval, but in the end it is a bit vague to me 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.

Response:

To address this point, we have added a paragraph early in the retrieval section that clearly defines:

- The state vector, which includes the vertical profile of CO_2 volume mixing ratios (VMR) on a fixed grid. Depending on the scenario, we may also retrieve scaling factors for temperature profiles, and in some cases, interfering species such as H_2O.
- The measurement vector consists of calibrated radiance spectra derived from observations. These are calculated by multiplying the solar spectrum measured during atmospheric absorption with the SOLAR-ISS spectrum (see response to comment on Line 94).

These clarifications are now explicitly included in Section 3.2 of the manuscript for better illustration of the retrieval components.

Reviewer Comment:

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. **As I understand it, 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 in my opinion that you can measure full rot-vib bands and get constraints on the temperature - which is degenerated with the gas amount for a single line or a few close ones. Could you see any improvements here?**
3. **Are you proposing to simply limit the used channels in a retrieval or also to limit the measured spectral bandwidth?**

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.

Line 9f: "I find the first sentence a bit vague and not adding anything of value – the same could be said about many methods."

Response: We have revised the opening sentence to make it more specific to our study and its context within LHR-based remote sensing. The new phrasing emphasizes the unique combination of broadband tunability and heterodyne resolution enabled by our setup, rather than making a general statement about LHR.

Line 10f: "Semantically wrong in my opinion. Heterodyne detection is a method, not an instrument and thus can not be 'transportable' or similar."

Response: We have revised the phrasing accordingly. The text now refers to the instrumental setup employing heterodyne detection as being portable, rather than attributing this characteristic to the detection method itself.

Line 23: "I would say the 'radiative impacts on the atmosphere' of higher GHG concentrations is well sorted since a few decades and 'studying the effects of climate change' with an instrument as presented sounds to me at best like the analysis of changes in biological sources and sinks of GHGs."

Response: We have rephrased this sentence to clarify that the system is intended to contribute to monitoring and understanding the spatiotemporal variations in greenhouse gas concentrations, which in turn supports the study of emission sources, sinks, and atmospheric transport.

Line 24ff: "This is quite a generic reference with an even broader reference. If you cite the latest IPCC report for something like this, I would ask you to be more precise where in the thousands of pages you get that from."

Response: We have now replaced the generic IPCC citation with a more targeted reference and section number from the most relevant IPCC report chapter (e.g., AR6 WG1 Ch. 1 & Ch. 10), which specifically discusses the role of trace gas measurements in climate monitoring.

Line 31: "I wouldn't call the logistical requirements of COCCON 'low'. While the EM27/SUN is portable (unlike the instruments of the TCCON network), the logistics behind operating a network of them, ensuring the comparability of the instruments, etc. is quite substantial and an achievement."

Response: Thank you for pointing this out. We have revised the description to acknowledge that while the instruments themselves are portable, the operation and maintenance of an extensive instrument network, like COCCON, is indeed non-trivial and requires significant coordination and effort. We now frame this differently rather than implying minimal effort.

Line 35: "If you argue via cost effectiveness, could you give a rough number/price?"

Response: We now specify that the cost of our prototype LHR system is approximately 20% of that of a typical EM27/SUN spectrometer. This estimate reflects our current setup and highlights the potential cost advantage of heterodyne detection, although we note that the final price is highly dependent on the choice of laser source and detector.

Line 35 (continued): "What sensitivity limits are you talking about?"

Response: The sensitivity limits referred to here, is the vertical sensitivity.

Line 55: "Since lock-in amplifiers are not necessarily known to everybody in the target group of this journal, I would ask you to add half a sentence explaining why you modulate the light source with a chopper."

Response: We updated the text to briefly explain the rationale: … A mechanical chopper that modulates light to enable phase-sensitive detection by the lock-in amplifier, isolating the heterodyne signal from low-frequency noise. This modulated sunlight…

Line 68: "Can you also give details on the product concerning the 'square law detector'? I think this could help avoid misunderstandings, since many readers will think 'photodiode' when reading this in the context of this journal and the topic."

Response: Indeed, this is not a photodiode, but a Schottky diode that's is used to extract the absorption signature, which is the envelope of the amplitude of the radio frequency signal. The output of such a detector is proportional to the square of the amplitude of the input beat signal.

Line 77: "I think this equation is not clear at all, even after reading the cited reference. At a minimum, the chosen definition for SNR should be reiterated and the relevant assumptions stated (relative strengths of signals, shot noise limits, etc.)."

Response: In this response, we provide a more detailed explanation of the SNR definition as reported in the cited reference, and we outline the main noise sources in laser heterodyne systems.

While we agree that a more complete treatment would improve clarity, a full discussion of the noise model goes beyond the scope of the present manuscript and will be addressed in a forthcoming technical paper. For this reason, we have opted not to include these details in the current version but now briefly mention the primary noise contributions for transparency.

Sunlight signal:

$$E_S(\mathrm{t}) = A_S \cos(\omega_s \mathrm{t} + \varphi_s) \quad (1)$$

Local oscillator laser:

$$E_{LO}(\mathrm{t}) = A_{LO} \cos(\omega_{LO} \mathrm{t} + \varphi_{LO}) \quad (2)$$

Total field intensity on the photodetector:

$$P = (E_S(\mathrm{t}) + E_{LO}(\mathrm{t}))^2 = (A_S \cos(\omega_s \mathrm{t} + \varphi_s) + A_{LO} \cos(\omega_{LO} \mathrm{t} + \varphi_{LO}))^2 = \frac{{A_S}^2 + {A_{LO}}^2}{2} + \frac{{A_S}^2}{2} cos(2\omega_s \mathrm{t} + 2\varphi_s) + \frac{{A_{LO}}^2}{2} cos(2\omega_{LO} \mathrm{t} + 2\varphi_{LO}) + A_S A_{LO} \{cos[(\omega_s + \omega_{LO})t + \varphi_s + \varphi_{LO}] + cos[(\omega_s - \omega_{LO})t + \varphi_s - \varphi_{LO}]\} \quad (3)$$

Filtering out high-frequency and DC components, the intermediate frequency (IF) signal is:

$$P_{IF} = A_S A_{LO} cos[(\omega_s - \omega_{LO})t + \varphi_s - \varphi_{LO}] = 2\sqrt{P_S \times P_{LO}} cos[(\omega_s - \omega_{LO})t + \varphi_s - \varphi_{LO}] \ (4)$$

The photocurrent after photodetector is:

$$i_{IF} = 2\frac{\eta e}{h\nu}\sqrt{P_S \times P_{LO}} \cos[(\omega_s - \omega_{LO})t + \varphi_s - \varphi_{LO}] \quad (5)$$

The photocurrent amplitude is:

$$i_{IF} = 2\frac{\eta e}{h\nu}\sqrt{P_S \times P_{LO}} \quad (6)$$

Main Noise Sources in Laser Heterodyne Radiometer

1. Johnson noise:
   a) The thermal noise of the photodetector:

   $$< i_j^2 >= \frac{4kT_m\Delta f}{R_m} \quad (7)$$

   b) The thermal noise of the amplifier:

   $$< i_A^2 >= \frac{4kT_A\Delta f}{R_A} \quad (8)$$

2. Coherently detected thermal noise:

$$< i_{CDT}^2 >= \frac{4\eta^2 e^2}{hv} \delta_s P_{LO} \Delta f \qquad (9)$$

$$\delta_s = \left[\exp\left(\frac{hv}{kT_s}\right) - 1\right]^{-1} \qquad (10)$$

3. Laser-induced noise

a) The relative intensity noise:

$$< i_{RIN}^2 >= R_{IN} \Delta f \left(\frac{\eta e}{hv} P_{LO}\right)^2 \qquad (11)$$

b) The shot noise:

$$< i_s^2 >= 2e i_{DC} \Delta f = 2e \frac{\eta e}{hv} P_{LO} \Delta f \qquad (12)$$

Signal-to-noise Ratio

Based on the above analysis, the signal-to-noise ratio (SNR) of the LHR system can be expressed as:

$$SNR = \frac{<i_{IF}^2>}{<i_j^2>+<i_A^2>+<i_{CDT}^2>+<i_{RIN}^2>+<i_s^2>} \qquad (13)$$

The LHR system was designed to operate in the shot noise–limited regime by optimizing the local oscillator laser power, such that the total system noise is dominated by LO-induced shot noise.

$$< i_s^2 > >> < i_j^2 > ; < i_s^2 > >> < i_A^2 > \qquad (14)$$

Therefore, the thermal noise can be ignored. Meanwhile, a balanced detector in LHR was used for heterodyne signal detection, eliminating the relative intensity noise of the local oscillator laser. Thus, the relative intensity noise is also ignored.

Therefore,

$$SNR = \frac{<i_{IF}^2>}{<i_{CDT}^2>+<i_s^2>} \qquad (15)$$

By substituting formulas (6), (9) and (12) into formula (15), SNR can be expressed as

$$SNR = \frac{1}{\Delta f} \frac{\eta P_S}{hv(1+2\eta\delta_s)} \qquad (16)$$

The signal light power received by the heterodyne system can be expressed as

$$P_s = 2T_0 hv \Delta f \delta_s \qquad (17)$$

Hence, the SNR can be expressed as

$$SNR = \frac{2T_0\eta}{2\eta + \exp\left(\frac{h\nu}{kT_S}\right) - 1} \qquad (18)$$

The final SNR at the output of the RF filter with the bandwidth Δf and an integration time τ is given by:

$$SNR = \frac{2\eta T_0\sqrt{\Delta f \tau}}{2\eta + \exp\left(\frac{h\nu}{kT_S}\right) - 1} \qquad (19)$$

Line 82: "You state the theoretical SNR and then introduce your measurements, but I do not find how well the actual measured SNR compares to the theoretical one."

Response: The actual measured SNR is approximately 200, based on a single scan, in contrast to the FTIR measurements where multiple scans are averaged. The reduced SNR can be attributed to several factors, primarily the absence of spectral averaging. Additional contributors include suboptimal detector performance such as lower-than-expected quantum efficiency, elevated dark current, and electronic noise sources including amplifier and digitizer interference. We have now added a quantitative comparison between the theoretical and measured SNR values in the main text, along with a brief discussion to interpret the observed discrepancy. While current measurements yield a lower SNR (~200), an SNR of 700 is achievable through additional scan averaging or improved detector performance. We therefore use SNR = 700 to assess the theoretical information content under optimal conditions, which will be targeted in future measurement campaigns.

Line 84: "'An information content study'?"

Response: We have corrected this phrase.

Line 90: "Why a Gaussian line profile? This would be rather unusual (and wrong). You should at least use Voigt, but even this is not necessarily up to current standards."

Response: This is an important point. We agree that the original phrasing was misleading. In our retrievals, the absorption lines are modeled using Voigt profiles, consistent with standard spectroscopic practice. The use of a Gaussian line shape refers specifically to the Instrument Line Shape (ILS), which is convolved with the Voigt-profiled spectrum. This choice is based on the characteristics of the LHR instrument, whose optical and detection system yields a response that is better approximated by a Gaussian. We have revised the text to clearly distinguish between the line profile and the ILS to avoid further confusion.

Line 94: "What is the LATMOS function?"

Response: We have now defined the "LATMOS function" explicitly in the text. It refers to a custom radiative transfer routine developed at LATMOS, which we used for the forward simulation of absorption spectra. This routine relies on the SOLAR-ISS spectrum, a high-resolution solar reference spectrum constructed by combining existing solar spectra with SOLAR/SOLSPEC measurements using known slit functions. SOLAR-ISS provides an accurate representation of the solar irradiance during the 2008 solar minimum, especially in the ultraviolet, visible, and infrared regions.

Line 103f: "To make any statement about the consistency a plot of the residuals between measurement and forward model is required."

Figure 2: "Please be more clear in the layout and caption of the figure what is measured data and what is simulation."

Response: We added a new panel for illustrating the residuals between measured and simulated spectra. The figure caption and layout have been revised to distinctly label measured data and simulated spectra as well as a clear legend for better readability.

Line 144: "I 'I' a unity matrix?"

Response: Yes, here 'I' denotes the identity matrix. We have added this information to the text.

Line 146f: "You say that 'Sm is calculated […]' but then proceed to give an equation for Smeas."

Response: Indeed, we have corrected the notation to consistently use S_{meas} in the text.

Line 161: "I don't get the division by 100 in the equation. Looks to me like a conversion from percent to a straight number, but perror is sometimes given as absolute value including units (i.e. for the temperature) in your table."

Response: In our implementation (based on ARAHMIS), the uncertainties are initially expressed in relative terms (i.e., as percentages). However, since the subsequent calculations are performed using absolute values, a conversion from percent to fractional form (i.e., division by 100) is necessary.

Line 190: “To my understanding 10° measurements are rather unrealistic for high quality direct sun observations.”

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 (Kattar et al., 2020).

Line 200f: “In a sense, this is obvious - higher up, lower pressure, less molecules in a fixed height layer. This is one of the reasons why the atmospheric retrieval codes I am familiar with use equidistant levels in pressure, which result in (roughly) equal amounts of molecules per layer. So here, it is unclear to me, how much if not all of the described effect is due to the lower number of molecules.”

Response: We agree that reduced pressure and molecular density at higher altitudes are major contributors to the reduced sensitivity of LHR measurements. However, this factor alone does not fully explain the effect.

At high altitudes, absorption lines become narrower due to reduced pressure broadening. These narrow lines are more difficult for instruments with finite spectral resolution to resolve, which diminishes the strength of the observed signal even when the total number of molecules is accounted for. In addition, the line strength itself can decrease due to altitude-dependent temperature effects.

Instrumental limitations such as spectral resolution, signal-to-noise ratio, and observational geometry (e.g., reduced path length through the absorbing layer) further compound the reduction in sensitivity. This behavior is consistent with satellite observations as well: for instance, in limb-viewing geometries, satellites traverse long atmospheric paths but still exhibit low sensitivity at high altitudes for similar reasons, not just low density, but also narrow line widths and weaker absorption features.

To further clarify this point, and following the editor’s suggestion, we computed an information content analysis using a full covariance matrix. The results show that even in high-altitude regions with low molecular abundance, the averaging kernels can be comparable to those at lower altitudes. This suggests that reduced sensitivity cannot be attributed to molecule number alone, but results from a combination of spectral, instrumental, and geometric factors.

Line 208: “Maybe change the order of the tables? Table 4 is needed before Table 2.”

Response: We added the DOFs values mentioned in Table 4 to Table 2 to improve the logical flow in the text, as Table 4 is needed in a later section.

Figure 3: "Please add a better explanation (maybe linked to your formalism for A) what the different lines are."

Response: The explanation of the different lines of A are added both in the main text and in the caption of Figure 3.

Line 235: "Optical path difference? You are talking about a longer path in the atmosphere I assume?"

Response: Yes, the term "optical path difference" refers to a longer path through the atmosphere. This expression has been clarified to avoid confusion with the instrument's optical path difference.

Table 3: "What definition of SNR is utilized here? Is it comparable between the different measurements/works? Also: What CHRIS is remains unclear to somebody not familiar with the corresponding paper."

Response: We have added the integration times alongside the SNR value in Table 3. We confirm that, with this information, the SNR is comparable to the other measurements. Additionally, the acronym CHRIS is now defined in the table caption, with a reference to the corresponding publication for readers who may be unfamiliar with it.

Figure 5: "Axis ticks very hard to read."

Response: The figure has been reformatted to improve the readability of the axis ticks. As recommended by the editor, it now includes the baseline in the Jacobian, and the spectrum is shown instead of the Jacobian for better visualization.