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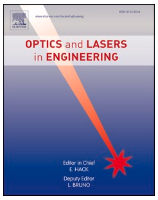

Design and evaluation of a portable frequency comb-referenced laser heterodyne radiometer

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ABSTRACT

In this paper, we present the design of a laser heterodyne radiometry instrument that combines, for the first time, frequency comb calibration and a remarkably high level of portability. A design that can, therefore, be more than capable of addressing the current need for accurate ground-based greenhouse gases monitoring in urban areas and other emission hot spots. Indeed, the compact, battery-powered system allows the acquisition of atmospheric spectral characterizations, at any location, without restrictions. As its most prominent feature, the system is equipped with an electro-optic frequency comb reference that provides a set of calibration ticks from which an accurate characterization of the absorption line shape can be obtained. Besides this, the spectrometer has been designed to promptly switch between traditional operation and wavelength modulation, so the performance of future inversion models may benefit greatly by this complementary data. The system has been tested in different locations in the Madrid region (Spain), where measurements have been carried out under a wide variety of conditions. Here, a set of highly representative results is presented clearly illustrating the capabilities of the developed system.

1. Introduction

Over the last two centuries, human activity has released massive amounts of greenhouse gases (GHG) into the atmosphere, which has led to a significant increase in the Earth's surface temperature. The consequences of this problem are many and obvious, from rising sea levels to increased severity of extreme weather events, which has made global warming a major global concern today. If the problem is to be tackled in the most efficient way possible, it is essential to identify and characterize GHG sources and sinks on a global scale, but also to accurately monitor atmospheric concentrations of GHGs. To this end, a number of satellite-based and ground-based initiatives are currently in operation.

Among the current satellite-based missions, the JAXA Greenhouse gases Observing SATellite (GOSAT), the GOSAT-2, the NASA Orbiting Carbon Observatory-2 and 3 (OCO-2 and 3), or the ESA TROPOspheric Monitoring Instrument (TROPOMI) missions stand out [1–3]. Additionally, a number of ground-based networks currently collect and archive high-quality and relevant GHG-related atmospheric measurements: point concentrations, isotopes and turbulent fluxes (e.g., GAW, ESRL, and ICOS); total column amounts (TCCON, COCCON, and NDACC) and balloon-based vertical profiles (AirCore) [4]. Nonetheless, whereas on the first (satellite based-infrastructures) interrogation volumes are very wide and the overpass frequencies scarce, the later have a sparse and irregular geographical coverage, being originally set up to focus on GHG observations of background environments. All this has led to many hot spots being critically under sampled, with the data provided not allowing the study of local but important emission centers, such as urban areas with large populations or major industrial facilities. To name but one, this is corroborated by the European Commission green report in the framework of the Operational Anthropogenic CO_2 Emissions Monitoring and Verification Support (MVS) capacity [5], that states that these existing ground-based networks currently do not meet all operational requirements for the Copernicus CO2 MVS capacity due to the lack of in situ measurement data from urban areas and other emission hot spots. Unfortunately, this deficiency of the GHG monitoring networks is very difficult to tackle with traditional spectrometer technologies, which are normally bulky, expensive and highly technically demanding.

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In this scenario, the development of lightweight, portable Laser Heterodyne Radiometry (LHR) [6–10] systems have become one of the best alternatives, as evidenced by the notable increase in the number of articles published using this method for atmospheric exploration in recent years [11–16]. This technique, which combines an ultra-high spectral resolution and a sensitivity close to/equal to the quantum limit with rugged and compact system implementations, is ideal for the characterization of the vertical distribution of the target gas concentration or the total column of many atmospheric constituents. Nonetheless, for its future routine use as part of current monitoring networks, several challenges still need to be addressed. A specially challenging aspect of the practical use of LHR for atmospheric studies is the accurate characterization of the frequency axis. It should be remembered that the operating frequency of these spectrometers is determined by the frequency at which the local oscillator laser is tuned into; this parameter needs, therefore, to be carefully controlled. However, the dependence between the current injected into the laser and the tuning frequency is non-linear, and this dependence is also strongly affected by temperature. All these factors contribute to generate a large uncertainty in the determination of the important frequency axis of the spectral analysis. Unfortunately, such uncertainty directly impacts the ability of inversion models to determine the profile of concentration of the atmospheric compounds of interest. Thankfully, the emergence of new high-performance optical sources has made it possible to improve many aspects of traditional spectroscopy, which has also been the case for the LHR method. Very recently the use of an optical frequency comb for calibrating the frequency axis of the spectral measurement was demonstrated [17], reaching unprecedented levels of performance. However, this technology was only validated within the laboratory, which severely restricts its use and, consequently, its potential impact on future atmospheric characterization.

This paper presents the design and characterization of the first fieldable, frequency-comb calibrated, LHR for the study of atmospheric CO_2. Nonetheless, the architecture can be straightforwardly adapted for the detection of any other analyte of interest. The battery-powered system, which is thoroughly described, can operate in both traditional and wavelength modulation modes, and has been characterized in detail and also tested in a large city, one of the areas of greatest interest for atmospheric survey systems today.

2. Material and methods

2.1. Laser heterodyne radiometry and practical frequency-comb calibration

In general terms, the LHR technique has very similar fundamentals to those of a heterodyne radio-frequency spectrum analyzer, except that the local oscillator is a laser and that the mixing with the signal whose spectrum is to be analyzed is done directly in a photodetector. When the laser emission frequency is swept, a radio frequency power meter connected to the photodetector (after a chain of amplification and filtering) allows the spectrum of the incident optical signal to be recovered. The development of the LHR method in the 1970s for spectroscopic applications must be credited to the Jet Propulsion Laboratory of NASA [18] and, for many decades, their use has been restricted to some of the leading metrology laboratories. Today, and without significant changes to its architecture, important technological developments in terms of the performance of the components used (availability of robust, compact laser sources and small, high-efficiency, low-noise detectors) have made its use in the field feasible. Indeed, significant efforts have been made in recent years to ensure the reliability and maximize the promising performance of the LHR technique for atmospheric remote sensing applications [10,16].

A simplified block diagram of a LHR is shown in Fig. 1 (comprehensive analysis of the basics of LHR have been presented in [8,19–21]); the incoming signal is combined with the light from the local oscillator and the resulting beam is focused into the photodetector. The output of the photodetector, now providing a downshifted radio-frequency (RF) copy of the spectrum of the input signal, is, normally, amplified and then filtered, in order to configure the optical resolution of the radiometer. As final step, the RF power is detected by a RF power meter. Thus, when the laser is tuned across the spectral feature of interest, the readings of the power meter allow recovering the spectrum of the incident signal with a super optical resolution. In virtually all experimental implementations of the method the incoming signal is intensity modulated (using normally a chopper) with the dual objective of eliminating different offsets in the final signal and maximizing the signal-to-noise ratio. Thus, a lock-in amplifier is used after the power meter to retrieve the spectrum of the input signal. To this regard, it should be added that in the last years, several research groups have started to explore some of the advantages of using a wavelength modulated local oscillator laser for laser heterodyne radiometry (WM-LHR) [11,22–25], for which minimum changes in the traditional scheme are required. Simply, a wavelength modulated local oscillator laser replaces the intensity modulation of the input signal in WM-LHR. The system presented in this paper is capable of

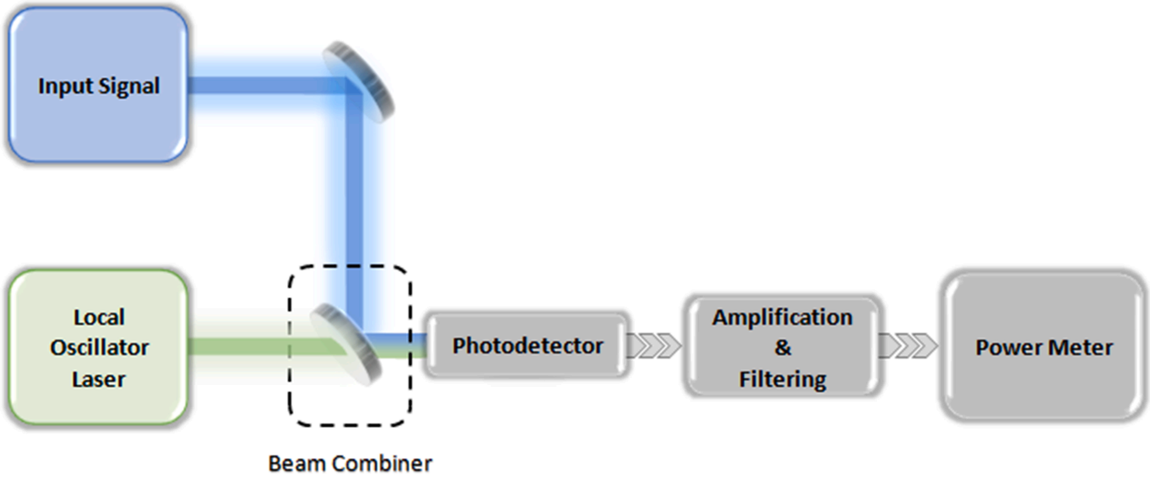

Fig. 1. Basic block diagram of a laser heterodyne radiometer.

operating in both traditional and WM-LHR modes.

As presented in the introduction, one of the most important technological challenges so far of LHR systems was the calibration of the frequency axis, a real hurdle that has been only very recently overcome through the use of frequency comb technology in the laboratory. However, if the advantages of this method are to be exploited in field measurements, the high complexity and cost of most comb generators must be properly addressed. In the system described in this work, this necessary reduction in size, complexity and cost of the system has been guaranteed after considering that in the vast majority of current atmospheric inversion models the center frequency of the absorption line is completely irrelevant, being one of the parameters to be fitted during data processing. This (as long as any changes in the offset frequency are kept below the optical resolution of the instrument for the time that it takes to sweep the optical span, being this rather short) allows the use of a simple electro-optic comb, which is not self-referenced or referenced to a hydrogen maser, but still provides the required set of frequency calibration marks that ensures accuracy in determining the absorption line shape; being this the really important aspect for the inversion model. In electro-optic comb generators, the accuracy in the separation of the teeth (that become markers for frequency calibration), is dictated by the accuracy of the RF generator employed [26], that can easily be of less than a single ppm for narrow combs, which results in an absorption line shape being characterized with levels of accuracy that can be orders of magnitude higher than that of non-frequency comb calibrated systems.

Two additional considerations should be added: the optical resolution employed in atmospheric studies is normally 600 MHz and the common optical span of a LHR system is limited, by the tuning range of the laser, to less than 100 GHz (for regular laser diodes). Therefore, the low repetition rate and wide optical span characteristic of regular mode locked lasers-based frequency combs make them unsuitable for the calibration of the vast majority of LHR systems. These features can be precisely controlled on an electro-optic comb generator, making it the, by far, most appropriate technology to calibrate LHR instruments.

2.2. *Instrument design and implementation*

As introduced above, in this paper we present the design of a LHR that brings together two of the previously highlighted developments, WM-LHR operation and frequency-comb calibration, with the portability that guarantees the ability to operate in urban areas and other emission hot spots. Indeed, the compact, battery-powered, system allows atmospheric characterization, even at remote locations, without restrictions. The complete block diagram of the implemented LHR is shown in Fig. 2(a). The system has been designed to automatically switch (electronically) from the traditional operation mode (absorption line sweeping) to WM-LHR, so the performance of the instrument can be reinforced by means of the combination of the benefits of both approaches. In this way, either the intensity of the incoming signal, via electro-optic modulator, or the laser current (and, therefore, its wavelength) is modulated to enable a given operation mode. This functionality is achieved by the use of a data acquisition system driving both the electro-optic modulator and the current source of the laser diode. The use of an electro-optic modulator provides an important benefit when the LHR operation mode is selected, as it allows the use of light-gathering optics of a much larger size than would be possible if an optical chopper had to be used. However, this feature of the system will also make the performance of the WM-LHR mode degraded compared to LHR, due to the insertion losses of the modulator. Following the signal path, after passing through the electro-optic modulator, the incoming signal is combined in a 2 by 2 fiber coupler with the local oscillator laser, the intensity of which is adjusted using an optical attenuator to avoid detector saturation. An amplified balanced detector is finally employed for the realization of the optical signal mixing. To maximize measurement consistency and signal stability, only polarization maintaining fiber components were employed in the set-up. The resulting heterodyne signal is then band pass filtered from 20 MHz to a configurable frequency that ranges from 100 MHz to 300 MHz, which allows providing dual-sideband optical resolutions that go from 0.0066 cm^{-1} to 0.02 cm^{-1}. After filtering, the RF signal is amplified, using an ultra-low noise RF amplifier, and detected with an USB power sensor that directly provides a digitized reading of the signal power averaged during an integration time that is configurable in steps of 20 ms. Even though this approach requires a software-implemented lock-in amplifier as the first stage of the signal processing; it provides the obvious advantage of a tremendous simplification in system hardware, very beneficial for meeting the stated objective of portability. It should be noted that a second optical input has been integrated into the system to allow the frequency comb to be injected into the instrument for frequency calibration. In pursuit of maximum simplification of the instrument, a single-stage electro-optic frequency comb generator was employed for calibration. Hence, a laser, driven by low noise electronics, was modulated by a single electro-optic modulator to generate a frequency comb (with repetition rates that can, in our system, be freely adjusted between 10 MHz and 15 GHz). As anticipated above, this approach allowed us to implement the frequency comb generator in a, again, battery-operated, ultra-compact format. Finally, it should be added that in-house developed solar tracker, implemented using two motorized rotation stations guided by the readings of a sensor (consisting of a quadrant detector with a pinhole placed 40 mm in front of it), was employed in the tests. A C80APC-C (Thorlabs) large-beam fiber optics collimator was employed capturing the sunlight. In its current version, the system provides an autonomy of around 15 hours of uninterrupted use without charging or replacing batteries, which, on the other hand, can be done in a matter of seconds. A photograph showing the complete system, in which the radiometer, at the bottom, can be distinguished from the frequency comb generator, middle, and the solar tracker, operating on top, is shown in Fig. 2(b).

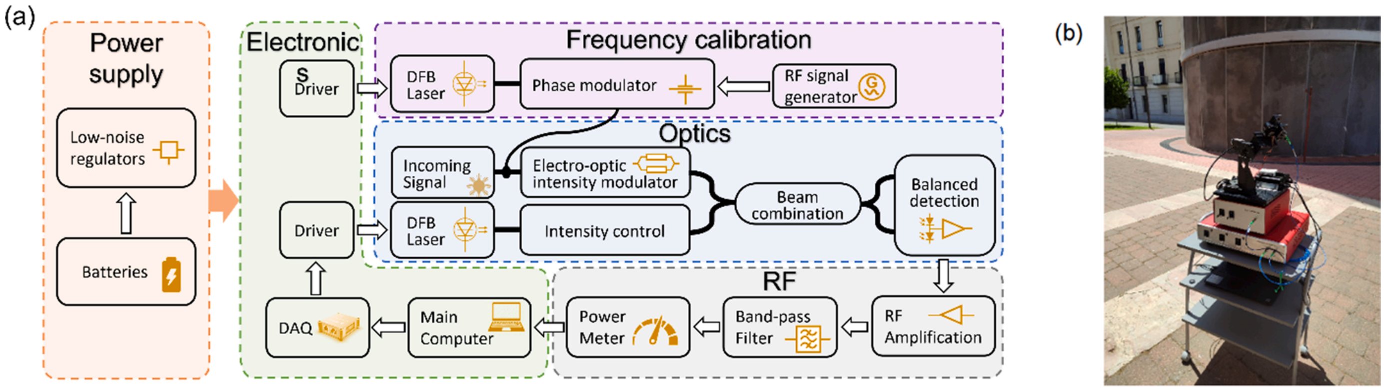

Fig. 2. (a) Detailed LHR system architecture. (b) Photograph of the implemented system during testing.

3. Results and discussion

The developed system has been evaluated in a city; this set of tests has been carried out at various locations, at different times of the day and under a variety of irradiation conditions. A representation of the results obtained is presented below.

The system was tested in different sites located mainly within several urban centers on the outskirts of Madrid, Spain. Spectral measurements were made during different seasons at times ranging from sunrise to sunset with various ambient conditions. In this test campaign the three CO_2 absorption lines located at wavelengths of around 1570.28 nm, 1570.54 nm and 1570.82 nm were targeted. An optical resolution of 600 MHz (0.02 cm^{-1}), standard for ground-based remote sensing GHGs monitoring networks [2], was employed throughout. Spectral measurements were taken at many different point spacings (induced by controlling the injection current of the laser), from 600 MHz, for future processing by inversion models, to very dense characterizations. The integration time per point was configured at 20 ms, obtaining the final spectrum by averaging a variable number of measurements. This allows for a high measurement speed that provides the ability to operate in changing atmospheric conditions.

Being this one of the main novelties of the system presented, this LHR spectrometer allows for frequency calibration to be performed during the measurement process, whereby the frequency comb spectrum is acquired and stored for a subsequent post-process that yields an accurate frequency determination. This involves the identification of all the teeth of the frequency comb within the span of the instrument, extracting their positions within the measurement vector. The process allows the complete rearrangement of the frequency axis so that the calibration marks (teeth) spacing matches the characteristics of the reference frequency comb. A second order fitting is used to maximize the performance of the frequency calibration process, which ensures a high level of accuracy in the post analysis of the absorption line shape that is of vital importance for atmospheric composition retrieval. Depending on the actual measurement requirements, the density of teeth can be adjusted to match different optical resolutions and wavelength spans. As an example, Fig. 3 shows the detected intensity, after frequency calibration, when a narrow optical frequency comb with a repetition rate of 4 GHz in is injected into the instrument. It should be highlighted that the spectral shape of the calibration comb, in terms of teeth intensity, is a direct consequence of the use of a single-stage phase modulation comb generation scheme and has no influence on the performance of the frequency calibration algorithm.

As highlighted at the beginning of the section, the system has been tested under many different atmospheric conditions. These detailed tests, in conjunction with the frequency calibration capabilities, have allowed us to identify one of the main drawbacks of LHR systems for use in the field: the enormous influence of temperature on the wavelength sweep of the local oscillator laser. To these authors, it is noteworthy that this point has remained unexplored in virtually all previous publications. In order to properly characterize this strongly influential factor, the different sub-systems of the LHR spectrometer have been placed into a climatic chamber (Dycometal CCK 180) in which temperature sweeps of between 10°C and 30°C have been carried out. On the one side, the readings of the LHR interrogator have been analyzed while the frequency comb calibration module was kept outside of the climatic chamber and at a constant temperature (a wavelength meter together with a RF frequency measurement corroborated an adequate stability of the calibration comb).

The reference comb processing presented in the paragraph above was used to determine both the shift in the central frequency of the LHR and also the consistency in the estimation of the linewidth of the spectral feature. The later can be illustrated by quantifying the separation between optical teeth. With regard to the central frequency shift, Fig. 4(a) shows a huge deviation that reaches a value close to 3 GHz (for a temperature drift of 20 °C). Nonetheless, as presented before, this parameter is normally fitted by the inversion model and it is, therefore, of no great significance. Much more worrying is the variation in the wavelength tuning profile which causes errors in FWHM estimation. Tests have determined that the linewidth measured by a typical LHR in which frequency calibration system is not employed can lead to a more than notable error. In our experiments, the frequency difference between teeth, shown in Fig. 4(b), changes by more than 100 MHz for two teeth spaced 10 GHz, which results in an accuracy error greater than 10,000 ppm in the estimation of the linewidth of the spectral feature. This will inevitably lead to significant inaccuracies in the determination of the vertical gas profile. To illustrate the ability of the frequency comb calibration to overcome this problem, our frequency comb calibrator was introduced into the climatic chamber and differences in the separation between frequency teeth of less than 15 Hz were observed (for the same frequency and temperature range utilized before). This shift is evidently associated to minute frequency drifts on the simple frequency generator due to temperature changes, which translates into potential accuracies in the ppb level in clear contrast to the performance of the uncalibrated instrument (frequencies were measured using a Rb referenced RF spectrum analyzer (Agilent Technologies N9001A). However,

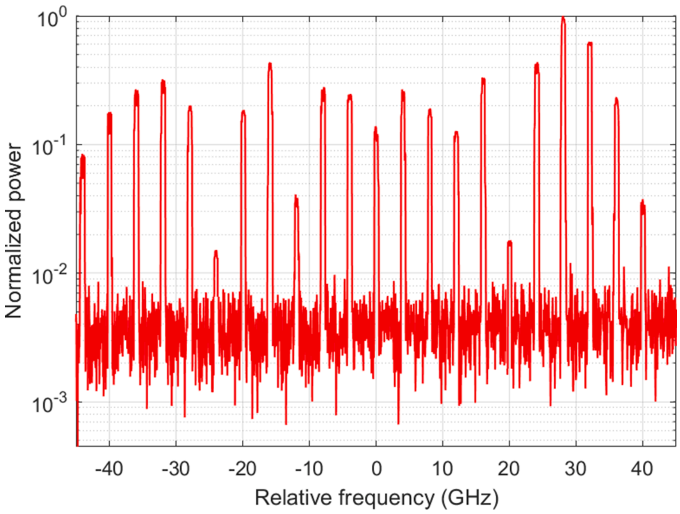

Fig. 3. Intensity detected by the system when an optical frequency comb with a repetition rate of 4 GHz is fed to the system.

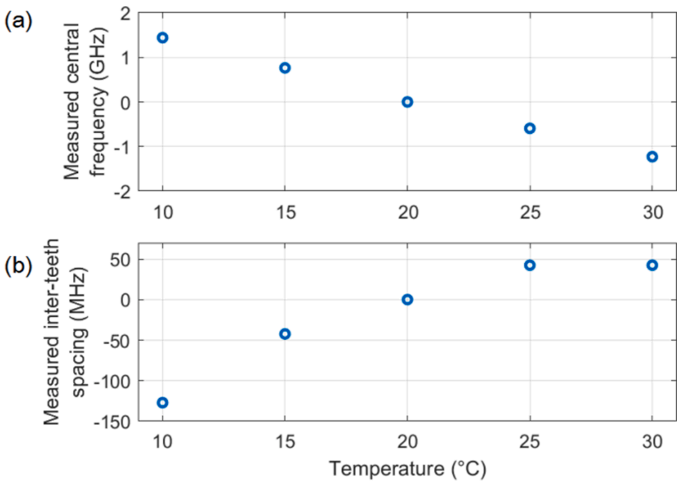

Fig. 4. Performance representation of the uncalibrated LHR system. (a) Shift in the central operation frequency of the system with temperature, which can be compensated by the inversion model. (b) Temperature drift of the inter-teeth spacing relative to 20 °C, which can be corrected by calibration with the optical frequency comb reference.

the effect of changing ambient temperature distorting the frequency axis determination must also be considered. If a normal change of 0.5 mK/s is assumed, during a 10 s scan, a shift of 1 MHz in the laser frequency can be expected, which results in a shift in the 10 GHz mode spacing of 100 kHz (considering an optical span of 100 GHz). This translates directly into an error of 10 ppm, four orders of magnitude lower than that achieved with the uncalibrated system. It can, of course, be added that such distortion can be drastically reduced if measurement times are reduced and, for example, spectrum averaging is used to maintain the desired signal to noise ratio. In summary, the results obtained in these tests lead to the conclusion that the potential of the proposed system to overcome the significant impact of temperature on the measurement in current LHR instruments is more than outstanding.

With regard to the actual atmospheric measurements, and beyond system accuracy characterization, Fig. 5 shows different results, with various optical spans and operation modes. As an example of a typical in-city result, the measurements shown are not taken on a perfectly sunny day, in order also to demonstrate the operational capabilities on days when conditions are less than perfect (irradiance levels of around 800 W/m^2 and a solar elevation of approximately 30°). In order to better appreciate the performance of the developed system, raw measurements, without any further data processing other than frequency calibration (and therefore have the slope associated with the laser source sweep), are shown. Regarding the frequency calibration procedure, even though different approaches are possible, the comb reference was superimposed to the incoming signal at periodic intervals in order to acquire a frequency calibration measurement. This same procedure it is also employed in the wavelength modulation operation mode.

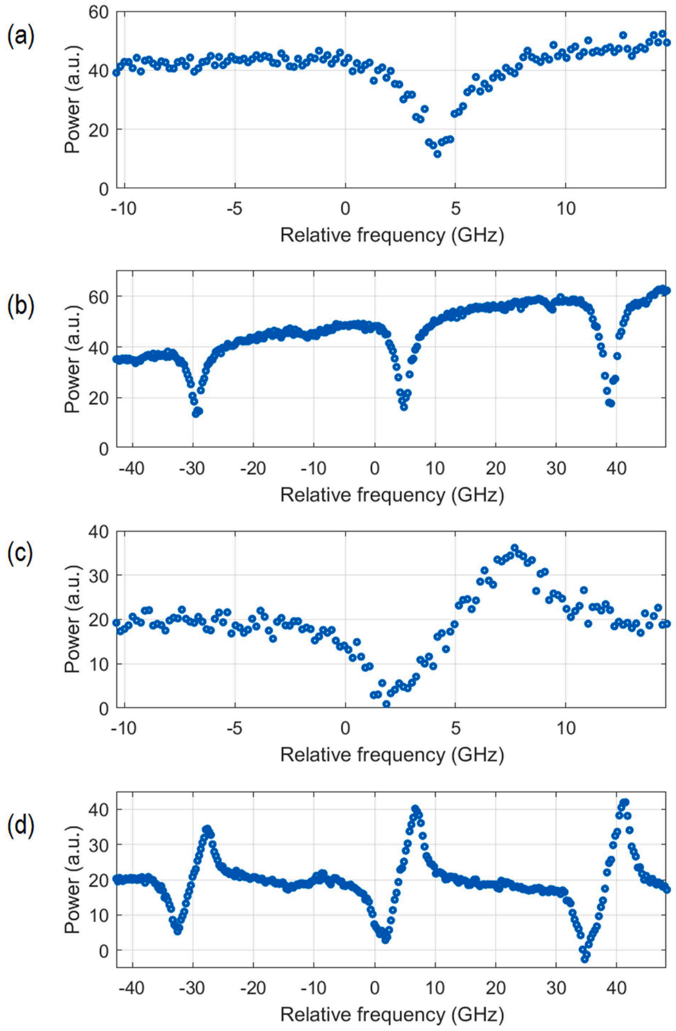

Fig. 5. Results for (a) single feature, 1570.54 nm, and traditional operation mode, (b) three absorption lines, at 1570.28 nm, 1570.54 nm and 1570.82 nm, with traditional operation mode and (c) a single and (d) three spectral features with WM-LHR mode (see text for further details). These particular measurements were taken on October 17, 2022.

Fig. 5(a) and (b) present a typical result of the characterization, using the traditional LHR operation mode, of a single (1570.54 nm) and the three (1570.28 nm, 1570.54 nm and 1570.82 nm) CO_2 absorption lines that the instrument can cover (without changes in the laser source temperature). It should be added that weaker absorptions at approximately -40 GHz, -10 GHz, 30 GHz and 45 GHz are also clearly visible. The frequency axis has been obtained after comb calibration, setting the zero reference arbitrarily (as this will be fitted by the atmospheric model). The single absorption feature measurement shows the results of a single spectral sweep, without any averaging, with an integration time per point of 20 ms and a total of 130 points. The total measurement time is slightly greater than 2.8 s due to processing requirements. The data shown for the three absorption lines is the result of averaging 5 subsequent spectra acquired for an integration time of, as before, 20 ms per point. This results on an equivalent integration time of 100 ms per point, considering that the number of points has now been expanded to 300, the total measuring time is of around 33 s.

Fig. 5(c) and (d) show the signals obtained with the instrument operating in WM-LHR mode for the same configuration as for traditional LHR (no averaging for the single line measurement and 5 spectra averaging for the three-line characterization). As before, measurements have exclusively been processed to calibrate the frequency axis. In the case of the measurements shown in this paper, the modulation depth was set approximately at 6.5 GHz in order to maximize the signal amplitude [23]. It is worth noting the particular shape of the WM-LHR data, corresponding roughly to the first derivative of the absorption line shape.

As introduced previously, the system has the ability to take both, traditional and WM-LHR, spectral characterizations for subsequent periodic measurements. In the authors' view, the distinctive WM-LHR line shape provides important complementary information to that of the traditional LHR for the inversion model, which should have a positive impact on its performance. A detailed study of this matter is currently being carried out.

With the configuration mentioned in the previous sections, the measurements shown exhibited spectral SNRs of roughly 70 for the traditional LHR operation scheme and 85 for WM-LHR for the three-line measurement (100 ms total averaging time per point). In the case of the traditional operating mode, the SNR is calculated as the ratio between the depth of the absorption feature and the standard deviation found in the wings of the absorption lines (in the spectral window around -20 GHz, which has no significant molecular transitions). For WM-LHR, the SNR is calculated by dividing the peak-to-peak value of the signal obtained by, again, the standard deviation found in the wings of the molecular transition.

4. Conclusion

In this paper, we have presented a fieldable, frequency comb calibrated LHR capable of potentially addressing the need for ground based GHG monitoring systems in urban areas and other emission hot spots in the very near future. Even though the particular experimental validation of this architecture has been carried out detecting atmospheric CO_2, many other atmospheric components can be targeted by simply replacing or combining various local oscillator lasers. As its most prominent aspect, the system features a simple and cost-effective optical frequency comb reference providing a set of calibration ticks from which an accurate characterization of the absorption line shape, of vital importance for retrieving the vertical profile, can be obtained. By taking advantage of a specific feature of most atmospheric inversion models, the irrelevance of the absolute frequency, and that the optical resolution and span of LHR systems, an electro-optic comb is used as the ideal frequency calibrator. Additionally, the system features a dual operation mode by which traditional spectra or WM-LHR characterizations can be performed interchangeably. This most certainly will improve the performance of inversion models by bringing together, in a complementary way, the advantages of the two approaches. The system has been tested in different locations in various urban centers on the outskirts of Madrid (Spain), in which atmospheric measurements were performed in different conditions. Some representative results obtained in this test campaign, including that of the use of the frequency calibration module, are shown in the paper.

CRediT authorship contribution statement

Aldo Moreno-Oyervides: Methodology, Software, Validation, Investigation, Writing – original draft, Visualization, Writing – review & editing. **Oscar Elías Bonilla-Manrique:** Software, Investigation, Writing – review & editing. **Omaira García:** Conceptualization, Writing – review & editing, Supervision, Funding acquisition. **Pedro Martín-Mateos:** Conceptualization, Methodology, Investigation, Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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