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Atmospheric Measurement Techniques
Discussions
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Interactive comment on "Thermal infrared laser heterodyne spectro-radiometry for solar occultation atmospheric CO_2 measurements" *by* Alex Hoffmann et al.

Anonymous Referee #2

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This paper is a thesis–like investigation on a technique to measure atmospheric CO2. The described work includes discussions on the motivation, requirements, theoretical modeling, instrumental development with laboratory and field measurements, and the development and use of retrieval and analysis/interpretation techniques. Results, the authors argue, will enable a compact and efficient method for monitoring the important greenhouse gases in the Earth's atmosphere including CO2, water and other atmospheric constituents. The infrared heterodyne technique in the thermal infrared is a powerful technique for extremely high spectral resolution spectroscopic measurements that measure the true shapes of individual spectral lines. Resolved line shapes contain information on the species abundance and temperature in regions probed, as well as winds which can modify the line positions and shape. The uniquely high spectral resolution enables unambiguous identification and separation of lines of multiple species even in spectrally dense atmospheric wavelength regions. The authors' ultimate goal appears to be to augment and complement ongoing space-based and ground based networks monitoring CO2 and other atmospheric constituents.

Although the authors provide multiple references and discussion on the scientific motivation and current monitoring capabilities and their direct instrumentation heritage, they should address earlier work using infrared heterodyne spectroscopy for Earth and planetary studies. One of the innovations described is the use of quantum cascade semiconductor lasers (QCLs) as local oscillators (LOs). Less capable tunable lead salt semiconductor laser LOs have been used for similar studies in the past (e.g., Frerking and Muehlner, Appl. Optics 16, 526 (1977); Glenar et al. Appl. Optics 21, 253 (1982)). QCL local oscillator use was discussed in Wirtz et al., Spectrochimica Acta Part A 58, 2457 (2002)\ and Sonnabend et al., Journ. of Quantitative Spectroscopy & Radiative Transfer 109, 1016 (2008). Previous measurements and analysis of atmospheric measurements have been discussed in: Menzies and Seals, Science 197, 1275 (1977); Abbas et al. JGR 84, 2681 (1979); Kostiuk, Infrared Physics 35, 243 (1994); and Fast et al. GRL 31, L08109 (2004). Some comparison to these earlier efforts is warranted. Why is their approach better than previous techniques?

On page 2 starting on line 15 the authors describe the importance of retrieving changes in the "lowest 2-3 km" boundary layer, since that is the region of greatest carbon exchange. They seem to imply here and later that the IR heterodyne technique may retrieve a more accurate result. The ultra high spectral resolution is not necessarily a benefit when the line widths are wide as the pressure-broadened line are near the surface. Lines are several GHz wide and the information on the lower atmospheric region is in the wings of the lines and 60 or even 600 MHz resolution are of limited benefit, although the line shapes are not as affected by instrumental functions as in lower resolution spectrometers. Clearly the higher resolution will provide much more accurate information at higher altitude (lower pressure) regions. A direct comparison of expected ultimate accuracy of their mole fraction/column density retrievals vs. current measurement capabilities would be useful, e.g., comparison to TCCON network retrievals. Some of this may be hidden in the text already.

The modeling of a synthetic spectrum from an optimum instrument and fitting it is a good approach in the paper, testing the retrieval program and ultimate sensitivity of the method. The calculated noise level for the resolution bandwidths of 60 and 600 MHz is correct in the modeling. However, the true spectral resolving element is not the 60MHZ or 600 MHz bandwidth as quoted in the subsequent actual measurements. On page 10, paragraph starting with line 9 it is stated that the RF filters defining the bandwidths span 50-80 MHz and 50-350 MHz on each side of the LO. The heterodyne detection is a double sideband process (the photomixer cannot distinguish frequencies below and above the LO frequency) and in each case the noise and signal from each sideband add to give the sum of two 30MHz (300MHz) signals, hence 60MHz and 600MHz. However, the two sidebands are separated by 100 MHz and sample two noncontiguous frequency regions. Therefore, as the QCL LO is tuned over a spectral line each of the two bands samples a different portion of the line, starting at 100 MHz apart and including signal at frequencies increasing in opposite directions over the prescribed bandwidths, e.g., -50 to -80 MHz and 50 to 80 MHz. This introduces a kind of instrumental function that may affect the measured lineshape and change the effective spectral resolution (not 60 MHz). This effect was not addressed in the paper. A similar question arises in the TEC cooled detector measurements (Fig. 8b). Is the single bandwidth 0-50MHz or 50-100 MHz in both sidebands?

I have difficulty determining what the actual degradation from shot noise limited operation of the LHR is. In measurements with a blackbody a factor of ∼2 is quoted. In field measurement (Fig. 7) a factor of 4 is determined (Abstract and page 16), possibly due to “broadband thermal noise”. It would be good to have a list and numbers for the full degradation from ideal performance, including optical losses, detector quantum efficiency, chopping, polarization...etc. It may be just nomenclature, but shot noise limited operation is affected by losses due to all of these. A total factor of 4 seems too good since several of these alone will contribute a factor of 2.

In summary, aside from the comments above, I believe the authors did a good, but difficult job of developing, modeling, and testing the LHR by executing measurements and analyses of the atmospheric CO2 and H2O. They demonstrated that technological improvements enhanced the thermal infrared heterodyne technique for the study of atmospheric constituents. Their goals of a more autonomous, compact design for LHR capable of measurements from multiple locations monitoring changes in our atmospheric composition could be very useful, particularly as probes of lower pressure regions where the very high spectral resolution has a distinct advantage.

Technical comments/corrections:

Page 8, eq. 6: subscript g's are different. Should this be the case?

On page 10 Lines 4 and 5; it is stated that 0.52 mrad represents the coherent FOV and this is 1/8 of the apparent solar disc angle. The solar disc is $\sim$0.5 degrees, $\sim$8.7 mrad. Therefore, the FOV/Solar disc = 0.52/8.7 = 1/16.7. If I understand the text 1/8 should be 1/16, if not some clarification is needed.

Page 17. Line 21: states the TEC-detector has a "narrower" bandwidth. Fig. 8b caption states the bandwidth is 100MHz "double band bandwidth". Is the single bandwidth 0-50MHz or 50-100 MHz in both sidebands? Additional text in the figure would be helpful.

Table 1: A bit confusing. Define the Quantities below the table, e.g., what is - info content H[bit], DFS dx [-] etc. ... Also - error prop. = retrieved error??, Statistical [$\mu \pm \sigma$] = ??. A list/explanation below the table would help and limit searching in the full text.

Figures:

Fig. 2: For the size figure printed some label font inside figures is too small and difficult to read on paper. Some colors do not show well making reading even worse.

Fig. 3: Font size in Fig. (a) is small for the size of the figure as presented. Fig.(a) is an important figure for the instrument discussion. Fig. 3(b) label font is unreadable in paper version. To make it useful either the figure has to be made significantly larger or the size of font increased.

Fif. 5: The word “modulation” here, in other figures and in the text was confusing to me. “Change” would be better. I believe I am right that the direction of the “scan” is from right to left on the page. If so, an arrow indicating that on the figure would be helpful.

Fig. 7: Fig. 7(a) needs more explanation in the caption. Why the SNR bandwidths labeled 30MHz and 300MHz not 60MHz and 600MHz as on the left for dates observed? What does the caption in Fig. (b) mean? A bit more text may help. Fig. 7(c) I assume the black x’s are data points with wide scatter in same day measurements. Some discussion in caption would help.

Fig. 8. See above in text.

Fig. 9: (a) caption needs more description. As in Fig. 2 comment the font size is too small to read on paper. Some colors make it even harder.

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