

Atmospheric CO₂ measurement using dispersive infrared spectroscopy with a scanning Fabry-Pérot interferometer sensor



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Introduction

Carbon dioxide (CO₂) is the most important greenhouse gas in the atmosphere with serious impacts on radiative warming of planet earth. Currently, infrared spectroscopic based technique is the most common method for CO₂ measurement. However, this measurement technique is strongly depends on the influence of other infrared absorbing gases. Moreover, non-linear absorption is often an issue for infrared spectroscopic measurement.

We present a novel approach for infrared absorbing gases measurement by using a newly developed scanning Fabry-Pérot interferometer (FPI) sensor with auto-correction retrieval algorithm for both non-linear effect of absorption and interference from other co-existing infrared absorbers. This measurement technique is first applied for atmospheric CO₂ measurement. Modeled data and comparison measurements were used to examine the performance of the retrieval.

Experimental Setup

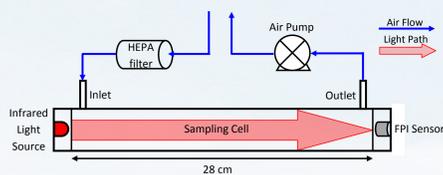


Figure 1: Schematic diagram of the experimental setup of the dispersive infrared spectroscopy measurement.

Retrieval algorithm

The non-linearity absorption effect is mainly due to the low spectral resolution of instrument spectrograph that is not able to fully reproduce the absorption signal. Light intensity measured by an instrument after passing through a volume of absorbing matter can be described by the Lambert-Beer law (Equation 1).

$$I(\lambda) = \left[I_0(\lambda) \cdot \exp \left(-L \sum_i \sigma_i(\lambda) \cdot c_i \right) \right] * F(\lambda') \quad (1)$$

- I : measured light intensity
- I_0 : initial light intensity
- L : optical path length
- c_i : concentrations of gas i
- σ_i : absorption cross-sections of gas i
- F : instrument function

The problem can be linearized by assuming the optical densities of the gases are low. Equation 1 can be rewrite as Equation 2.

$$I(\lambda) = \left[I_0(\lambda) \cdot \exp \left(-L \sum_i \sigma_i(\lambda) * F(\lambda') \cdot c_i \right) \right] \quad (2)$$

However, these assumption is not valid for high optical densities and low spectral resolution measurements. Standard non-dispersive infrared measurement techniques employ a look-up calibration table to correct for the non-linear effect. However, there are multiple gases absorbing in the wavelength band making these correction difficult. Alternatively, in the current approach with multiple wavelength absorption information from the FPI sensor, the non-linear effect can be corrected by using an iterative retrieval algorithm with major influencing gases included in the fit. The absorption cross sections can be modified according to the

gases mixing ratio by equation (3). The modified absorption cross sections $\sigma_{i,n}$ are used in the fit. The fit result (slant column densities/ mixing ratios) is then used to further modify the absorption cross sections.

$$\sigma_{i,n+1}(\lambda) = \frac{-\ln(\exp(-L \cdot \sigma_{i,n}(\lambda) \cdot c_{i,n}) * F(\lambda'))}{L \cdot c_{i,n}} \quad (3)$$

In our procedure, the loop proceeds until the change in one iteration is less than half of the estimated error of the fit or number of iteration reaches 100. A reasonable initial estimate of the gas mixing ratios as input of the retrieval can avoid unnecessary calculations. More iteration steps are expected if the difference between the initial estimate and the final retrieved result is large.

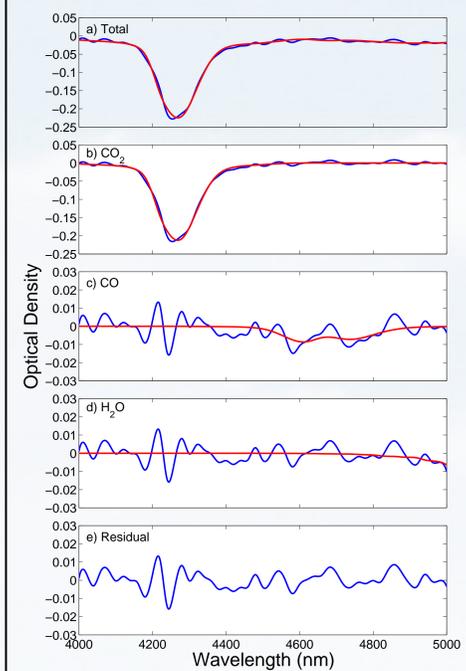


Figure 2: Example of a CO₂ retrieval using the iterative algorithm. The spectrum is taken on 26th April 2013 at 13:44 (local time) with CO₂ mixing ratio of (423.2 ± 6.3) ppmv. Fitting of the absorption cross sections (red curves) and the measured optical densities (blue curves) are shown for a) all absorptions, b) CO₂, c) CO and d) H₂O. Panel e) shows the residual of the fit.

Validation

This approach is validated by both model simulation and experimental measurements. Monte Carlo simulations were employed to estimate the influence of different error sources and parameterization of the retrieval on the measurement result. The effect of instrument noise on the retrieval result is estimated numerically through Monte Carlo simulations. The retrieval was applied to simulated absorption spectra with different CO₂ mixing ratios and noise levels to estimate how instrument noise could affect the solutions of the retrieval algorithm.

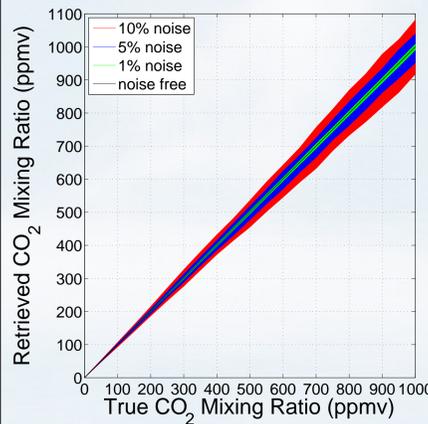


Figure 3: Averaged CO₂ mixing ratios retrieved from simulated spectra with different CO₂ mixing ratios and noise levels. 500 Monte Carlo simulations are done for each CO₂ mixing ratio and noise level. Normally distributed random noise with standard deviation of 10%, 5% and 1% of the absorption signal are added to the simulated absorption spectra. Result from error free spectra is also shown for reference. The shaded areas show the 1 σ standard deviation variation of the CO₂ retrieval results. A Gaussian shaped instrument resolution function with FWHM of 78.8 nm is assumed in the simulation.

The measurement results from the dispersive infrared instrument was validated by using a series of known concentration standard CO₂ gases (Linde Group). Ten concentration points were used spanning from 100 to 1000 ppmv supplied by a gas distribution system and compared to the retrieved CO₂ concentration using our retrieval algorithm.

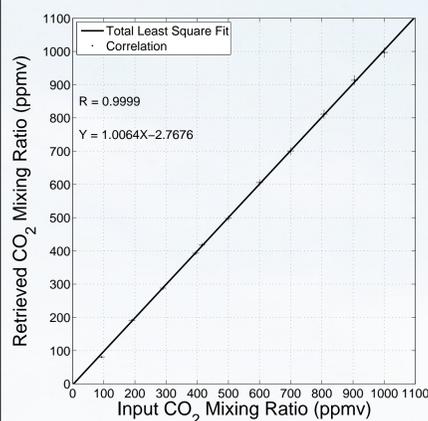


Figure 4: Validation of the dispersive infrared measurement of CO₂. X-axis shows the input CO₂ mixing ratio from the gas distribution system. Y-axis shows the dispersive infrared retrieved CO₂ mixing ratio. The error bars of the X-axis indicate the uncertainty of the gas distribution system which is about 10 ppmv for CO₂, while the error bars of the Y-axis represent the measurement error of CO₂.

Field Measurement and Comparison with Reference Analyzer

In addition to the laboratory validation, the performance of the dispersive infrared spectroscopy instrument was evaluated in the field for ambient measurement by comparing side by side with a commercial non-dispersive infrared CO₂ analyzer. Figure 5a shows the time series of the atmospheric CO₂ measurements obtained from the dispersive infrared instrument and the NDIR analyzer. An overall very good agreement was achieved with a Pearson correlation coefficient (R) of 0.9921 and a slope of 0.9912. The offset between the two measurements is 1.34 ppmv. This small offset might result from the uncertainty of the calibration for the reference analyzer.

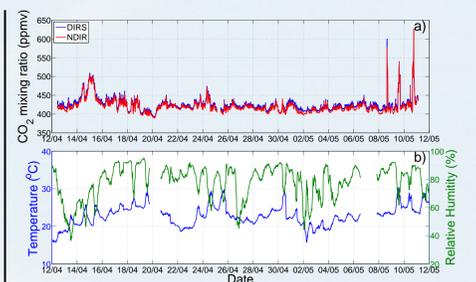


Figure 5: Time series of ambient CO₂ measured by the dispersive infrared instrument (blue curve) and the commercial NDIR instrument (red curve) are shown in a). b) shows the temperature (blue curve) and relative humidity (green curve) data recorded by the weather station at City University of Hong Kong. 10 minutes averaged data are shown.

Figure 6 shows the mean diurnal cycle of atmospheric CO₂ measured by the dispersive infrared instrument during the measurement period of one month. The bimodal CO₂ profile in the diurnal cycle is mainly related to increase in traffic load during the morning and evening rush hours. Relatively lower traffic emissions during the night probably result in lower observed CO₂ levels.

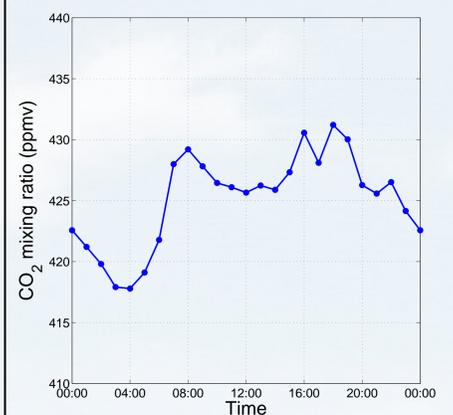


Figure 6: Mean diurnal cycle of atmospheric CO₂ measured by dispersive infrared instrument from 12th April 2013 to 11th May 2013.

Conclusion

- A new method for infrared absorbing gases (greenhouse gases) measurement is developed by using a newly developed scanning Fabry-Pérot interferometer (FPI) sensor.
- Retrieval algorithm with auto-correction for both non-linear effect of absorption and interference from other co-existing infrared absorbers is developed.
- This measurement technique is validated by both model simulation and laboratory measurement.
- The instrument is deployed for atmospheric CO₂ measurement. Result shows good agreement with reference sensor.
- The bimodal diurnal cycle of CO₂ indicated the CO₂ levels in Hong Kong is mainly related to traffic emission.

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