**A DOAS-like method
for X_{CO_2}**

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A DOAS-like method for total column of CO_2 from ground-based FTS measurements of the direct solar beam

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Abstract

A new algorithm, similar to that of DOAS method, is developed to retrieve the total column CO_2 from the ground-based hyper-spectral measurements of direct solar beam. Other than spectrum fitting method, which is generally used in the optimal estimation algorithm, the radiances ratio at two wavelength named channel pair, where one is of weak and the other is of relatively strong absorption, is used to retrieve the total column CO_2 in the Short Wave InfraRed(SWIR) band. Sensitivity studies show that this DOAS-like method is less dependent on the model parameters such as aerosols, water vapor, surface pressure, temperature, wavelength shift and signal noise, and the pairs of channels are carefully selected based on the sensitivity studies. To validate the algorithm, the FTS measurements located at Xi'Chong astronomical observatory are used to derive the total column CO_2 amount, 272 pairs ratios are used in the retrieval and the results agree very well with that of GOSAT, which shows that the DOAS-like method could give reasonable value of X_{CO_2} .

1 Introduction

Carbon dioxide (CO_2) has been considered to be the main cause of the current climate change (Solomon et al., 2007), though there exists many other scientific explanations. The disagreements about this issue mostly due to the lacks of long term records of CO_2 measurements, especially for large area measurements and its sources and sinks (Canadell et al., 2010; Stephens et al., 2007), which is the advantages of satellite observation. At present, only satellite datasets from the SCIAMACHY (SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY) on board the ENVISAT (ENVironmental SATellite) and the TANSO-FTS (Thermal And Near infrared Sensor for carbon Observation Fourier-Transform Spectrometer) on board the GOSAT (Greenhouse gases Observing SATellite), can be used to evaluate CO_2 sources and sinks (Bovensmann et al., 1999; Kuze et al., 2009). Both instruments use the reflected

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solar radiation in the SWIR (Short Wave Infra-Red) spectral region, which are sensitive to near-surface CO_2 concentration variations. Unfortunately, the low resolution of the SCIAMACHY limits its inversion accuracy (Buchwitz et al., 2005; Reuter et al., 2011), while the CO_2 accuracy of CO_2 from GOSAT has reached 0.3–1 % (1–4 ppm) (Guo et al., 2012).

Compared with satellite observations, ground-based observations have higher spectral resolution, higher signal to noise ratio (SNR), and therefore ground-based observations can achieve higher accuracy and precision in determining CO_2 total column concentrations. At present, the TCCON (Total Carbon Column Observing Network) is the only existing network to validate the satellite measurements of total column concentration of CO_2 from ground-based Fourier Transform Spectrometers (FTS), and the uncertainty of X_{CO_2} is 0.8 ppm after correcting for an air mass-dependent artifact (Wunch et al., 2011a; Wunch et al., 2011b).

As planning, a Chinese satellite will be launched into space in 2015, and several ground stations with the abilities to measure the hyper-spectrum of solar beam in the SWIR bands has been setup. To derive the total column amount of CO_2 from the spectral measurements, the optimal estimation method or say, a best fitting algorithm between the simulated and measured spectrum are often used for part of the absorption band. In this paper, a new DOAS-like algorithm was developed. In this method, the ratios of absorbed solar beam in two wavelengths are used, we call it channel pair. one of the pair for relative strong absorption and the other for weak absorption, and the pair are carefully selected depends on their sensitivities to the surface pressure and air temperature. Compared with the optimal estimation algorithm, this method is less dependent on the model parameter error.

2 Retrieval algorithm

Our retrieval algorithm is based on the fact that the ratio of channel pair is proportional to X_{CO_2} if the surface pressure and temperature profile are known. Other than the

whole absorption band are used to derive the total column amount of CO₂ by minimum of least square fitting method, only tens of pairs are used to reduce or eliminate the effect surface pressure and temperature.

2.1 Physical basis

- 5 Based on the Lambert–Beer's law, a ground-based measurement of the direct solar beam for a fixed wavelength can be expressed as:

$$I_{\lambda} = I_{\lambda}^{\text{sca}} + I_{0,\lambda} \cdot e^{-\tau/\mu_0} \quad (1)$$

- 10 where I_{λ} is the radiance measured at the bottom of atmosphere for wavelength λ , I_{λ}^{sca} is the forward scattering contribution in the incident direction, and I_0 is the incoming solar radiance at the top of the atmosphere. μ_0 is the cosine of the SZA (Solar Zenith Angle). τ is the total optical depth in the optical path, which can be written as:

$$\tau = \tau_{\text{CO}_2} + \tau_{\text{H}_2\text{O}} + \tau_{\text{aer}} + \tau_{\text{Ray}}, \quad (2)$$

- 15 where the terms subscribed as CO₂, H₂O, aer, and Ray are due to the absorption by CO₂, water vapor, scattering of aerosol and molecule, respectively. The scattering term I_{λ}^{sca} in Eq. (1) is negligible for small aerosol optical depth (AOD), therefore, the radiance can be simplified as:

$$20 \quad I_{\lambda} = I_0 \cdot e^{-(\tau_{\text{CO}_2} + \tau_{\text{H}_2\text{O}} + \tau_{\text{aer}} + \tau_{\text{Ray}})/\mu_0}, \quad (3)$$

in a very limited spectral range, the variations of τ_{aer} , τ_{Ray} along the wavelength are very small or can be ignored. Therefore we have:

$$\frac{I_{\lambda_1}}{I_{\lambda_2}} = \frac{I_{0,\lambda_1}}{I_{0,\lambda_2}} \cdot e^{-((\tau_{\text{CO}_2,\lambda_1} - \tau_{\text{CO}_2,\lambda_2}) - (\tau_{\text{H}_2\text{O},\lambda_1} - \tau_{\text{H}_2\text{O},\lambda_2}))/\mu_0}. \quad (4)$$

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The optical depth τ_{CO_2} due to CO_2 absorption is proportional to the number of total CO_2 molecular in the atmosphere:

$$\tau_{\text{CO}_2} = N_{\text{CO}_2} \cdot \bar{\sigma}_{\text{CO}_2}, \quad (5)$$

- 5 where N_{CO_2} is the number of CO_2 in the atmosphere, $\bar{\sigma}_{\text{CO}_2}$ is the absorption coefficient. Substitute τ_{CO_2} with Eq. (5) in Eq. (1), and let $r = I_{\lambda_1}/I_{\lambda_2}$, $r_0 = I_{0,\lambda_1}/I_{0,\lambda_2}$, Eq. (4) can be rewrite as:

$$r = r_0 \cdot e^{-[N_{\text{CO}_2} \cdot (\bar{\sigma}_{\text{CO}_2,\lambda_1} - \bar{\sigma}_{\text{CO}_2,\lambda_2}) - (\tau_{\text{H}_2\text{O},\lambda_1} - \tau_{\text{H}_2\text{O},\lambda_2})] / \mu_0}. \quad (6)$$

- 10 So we have:

$$N_{\text{CO}_2} = -\mu_0 \frac{\ln r - \ln r_0}{\bar{\sigma}_{\text{CO}_2,\lambda_1} - \bar{\sigma}_{\text{CO}_2,\lambda_2}} - \frac{\tau_{\text{H}_2\text{O},\lambda_1} - \tau_{\text{H}_2\text{O},\lambda_2}}{\bar{\sigma}_{\text{CO}_2,\lambda_1} - \bar{\sigma}_{\text{CO}_2,\lambda_2}}. \quad (7)$$

- For the weak water absorption, $\tau_{\text{H}_2\text{O}}$ is proportional to water vapor vertical column amount (VCA), $\bar{\sigma}_{\text{CO}_2}$ is a function of pressure and temperature, and N_{CO_2} can be simplified as:

$$N_{\text{CO}_2} = \mu_0 \cdot f(P_s, T) \cdot (\ln r - \ln r_0) + g(P_s, T) \cdot \text{VCA}_{\text{H}_2\text{O}}, \quad (8)$$

- where, $\text{VCA}_{\text{H}_2\text{O}}$ is the VCA of H_2O . P_s and T are the surface pressure and temperature, respectively. Both f and g are functions of P_s and T . As X_{CO_2} is proportional to N_{CO_2} , when P_s and T are known, X_{CO_2} can be expressed by

$$X_{\text{CO}_2} = \mu_0 \cdot a \cdot (\ln r - \ln r_0) + b \cdot \text{VCA}_{\text{H}_2\text{O}}, \quad (9)$$

where a and b are coefficients corresponding to f and g , respectively.

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2.2 Channel selection

The DOAS-like method could greatly eliminate the scattering effects by atmospheric molecular and aerosols, but more inversion errors may be introduced due to the temperature profile and wavelength shift. So the channel pairs must be carefully selected to reduce the effects due to the model parameter errors such as temperatures, water vapor and wavelength shift in the algorithm.

In the SWIR Band located at 1.6 μm, the main factors that affect the accuracy of CO₂ retrieval are temperature, pressure, water vapor, signal-noise, and wavelength shift. Therefore, the pairs where the parameter *a* (Eq. 9) is insensitive to pressure, temperature and small *b* are selected to reduce the inversion errors due to the inaccuracy of pressure, temperature and water vapor.

The noise is randomly produced during photoelectric conversion, and related to the instrument SNR. The retrieval errors due to the signal noise named random error. The higher SNR, the smaller random errors of the retrieval. Figure 1 illustrates the retrieval error due to SNR, its standard deviation is almost linearly related to the number of pairs used in the inverse method, and the errors are acceptable when more than 242 pairs are used.

If the spectrum is not correctly registered, a wavelength shift happens. Generally the shift is consistent in a limited spectral range. If the pairs of channels on the same side of absorption line are selected, the errors due to the inaccuracy of the wavelength registration, as shown in Fig. 2, increase as the increasing line shift. While, if the pairs almost evenly distributed on both side of the absorption line, the error could be reduced significantly due to the opposite behavior of the shift effect on the two side of the line. Moreover, the errors could be even suppressed by increasing the number of pairs used in the algorithm (Fig. 2).

Furthermore, a pair of wavelength must be close enough to reduce the scattering effects of atmospheric molecular and aerosols. The channels on the baseline and on

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the absorption peak should not be selected. It is also necessary to avoid the solar Fraunhofer line. The channels used in algorithm are demonstrated in Fig. 3.

3 Sensitivity analysis

Sensitivity analysis of the model parameters such as temperature profile and pressure, is given under assumption of a Lambertian land surface, and the US standard atmosphere is used. The whole atmosphere is divided into 30 layers: 1000 hPa, 975 hPa, 950 hPa, 925 hPa, 900 hPa, 850 hPa, 800 hPa, 700 hPa, 600 hPa, 500 hPa, 400 hPa, 300 hPa, 250 hPa, 200 hPa, 150 hPa, 100 hPa, 70 hPa, 50 hPa, 30 hPa, 20 hPa, 10 hPa, 7 hPa, 5 hPa, 3 hPa, 2 hPa, 1 hPa, 0.7 hPa, 0.5 hPa, 0.3 hPa, 0.2 hPa, and 0.1 hPa. The radiative transfer models of DISORT and LBLRTM with assumption of Voigt line shape are used to simulate the observation, the resolution of 0.0075 cm^{-1} and sample rate of 0.00377 cm^{-1} , which is related to our FTIR measurement. The aerosol optical properties are calculated thorough Mie theory. The schematic of the whole simulation is shown in Fig. 4.

To estimate the effect of temperature on the retrieval of CO_2 , two kind of errors are consider for the temperature profile, one is the shift of temperature profile with unchanged shape of its vertical distribution, another one is that of a random difference added to the different level of the profile. Figure 5 shows the retrieval errors if +3 K (upper panels) and +5 K (lower panels) shift of temperature profile, Fig. 6 show the errors introduced by +3 K and +5 K random errors of the temperature profiles The results derived by the optimal estimation method are also shown in the right two panels in Figs. 5 and 6. Compared with that of the optimal estimation methods, the DOAS-like method is less dependent on the uncertainty of the temperature profile, especially for larger difference of the temperature profiles.

Figure 7 shows the errors due to the uncertainties of surface pressure. Results of +1 hPa, +2 hPa and +3 hPa difference in surface pressure are respectively illustrated in the upper, middle and lower panels in Fig. 7. The smaller of the surface pressure

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uncertainties, the smaller of the retrieval errors of the CO₂. Similar with that of temperature analysis, this method results in smaller errors than that of the optimal estimation for the same difference of the surface pressure.

In short, the DOAS-like method which using the ratio of radiance of two channels is less dependent on the model parameters such as temperature, surface pressure, wavelength shift and more accurate retrieval of CO₂ could be given.

4 Case studies and comparisons

To validate the algorithm, the hyper-spectral measurements from a ground-based FTS instrument (IFS 125M), located on a mountain in Xi'chong astronomical observatory (22.4825° N, 114.5559° E) in Shenzhen, south of China, are used in the algorithm. FTS is the typical instrument in validation of satellite data of CO₂ in TCCON (Total Carbon Column Observing Network). The surface pressure data is from the automatic meteorological station. The temperature profile and water vapor VCA are from ECWMF dataset. The results of X_{CO₂} in clear sky between 1 September 2011 and 29 February 2012 are shown in Fig. 8, the black dot are results of this method, and results of GOSAT are dotted with red color. An average of 391.1 ppm with standard deviation of 3.7 ppm (40 points) are given by the DOAS-like method, while that of GOSAT are near 392.2 ppm with standard deviation of 2.7 ppm (6 points). Considering the systematic bias caused by noises in Fig. 1, a bias of 0.9 ppm will be added to the retrievals when SNR of FTS is about 300, therefore the average of our results is 392.0 ppm, which are consistent with that of GOSAT.

5 Conclusions

To derive the total column CO₂ from the ground hyper-spectral measurements of the direct solar beam, we develop a new algorithm which uses ratios of radiance at two wavelengths, where one is weak and the other is relative strong absorption for CO₂.

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This algorithm is similar to that of DOAS method and we name it DOAS-like algorithm. The pairs of the channels should be carefully selected and they must be located on both side of the absorption line, otherwise more error will be introduced. Sensitivity studies shows that DOAS-like method is less dependent on the surface pressure, temperature profiles, aerosols, water vapor, spectral shift and signal noise. The retrievals with the FTS measurements at the Xichong Astronomical observatory between September 2011 and 29 February 2012 show an average of 391.1 ppm with standard deviation of 3.7 ppm, while the results of GOSAT is of 392.0 ppm in average. As there is no direct measurement of the total column of CO_2 , the comparison with that of GOSAT shows that this algorithm could results in reasonable value of X_{CO_2} , but further validation is necessary and will be done in the future by comparing with the data of balloon soundings measurement.

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Fig. 1. Variation of X_{CO_2} mean inversion error and its standard deviation caused by noises with the number of channel pairs for different SNR.

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Fig. 2. Two channel pairs on the different sides of absorption line and their errors with line shift (upper panel), errors with line shift according to different channel pairs (lower panel).

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Fig. 3. Simulated normalized spectrum produced by FTS.

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Fig. 4. Flowchart of forward model.

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Fig. 5. Errors introduced by 3 K (upper panels) and 5 K (lower panels) of the temperature shift. The left two panels are results of this method, and the right two panels are given by the method of optimal estimation.

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Fig. 6. The same as Fig. 5 but for random errors of the temperature profile.

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Fig. 7. Errors introduced by +1 hPa (upper panels), +2 hPa (middle panels) and +3 hPa (lower panels) difference in surface pressure. The left three panels are results of this method, and the right three panels are given by the method of optimal estimation.

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Fig. 8. Comparison of inversion results from FTS and that of GOSAT.

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