

Ocean-glint as a diagnostic tool

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This discussion paper is/has been under review for the journal Atmospheric Measurement Techniques (AMT). Please refer to the corresponding final paper in AMT if available.

Using ocean-glint scattered sunlight as a diagnostic tool for satellite remote sensing of greenhouse gases

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Received: 25 March 2013 – Accepted: 7 May 2013 – Published: 16 May 2013

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

Spectroscopic measurements of sunlight backscattered by the Earth's surface is a technique widely used for remote sensing of atmospheric constituent concentrations from space. Thereby, remote sensing of greenhouse gases poses particularly challenging accuracy requirements for instrumentation and retrieval algorithms which, in general, suffer from various error sources. Here, we investigate a method that helps disentangle sources of error for observations of sunlight backscattered from the glint spot on the ocean surface. The method exploits the backscattering characteristics of the ocean surface which is bright for glint geometry but dark for off-glint angles. This property allows for identifying a set of clean scenes where light scattering due to particles in the atmosphere is negligible such that uncertain knowledge of the lightpath can be excluded as a source of error. We apply the method to more than 3 yr of ocean-glint measurements by the Thermal And Near infrared Sensor for carbon Observation (TANSO) – Fourier Transform Spectrometer (FTS) onboard the Greenhouse Gases Observing Satellite (GOSAT) which aims at measuring carbon dioxide (CO₂) and methane (CH₄) concentrations. The proposed method is able to clearly monitor recent improvements in the instrument calibration of the oxygen (O₂) A-band channel and suggests some residual uncertainty in our knowledge about the instrument. We further assess the consistency of CO₂ retrievals from several absorption bands between 6400 cm⁻¹ (1565 nm) and 4800 cm⁻¹ (2100 nm) and find that the absorption bands commonly used for monitoring of CO₂ dry air mole fractions from GOSAT allow for consistency better than 1.5 ppm. Usage of other bands reveals significant inconsistency among retrieved CO₂ concentrations pointing at inconsistency of spectroscopic parameters.

1 Introduction

When observing sunlight backscattered from the Earth's surface, the glint-spot on the ocean surface typically offers a bright target. Directional, specular reflection dominates

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over diffuse backscattering as long as the ocean surface is not too rough. For off-glint observing geometry, in contrast, the ocean surface is dark in the entire solar spectral domain. Therefore, satellite sensors such as the Greenhouse Gases Observing Satellite (GOSAT) and the Orbiting Carbon Observatory (OCO) have been designed to regularly point at the glint-spot when they overfly ocean surfaces (Crisp et al., 2004; Kuze et al., 2009).

GOSAT and OCO attempt highly accurate measurements of atmospheric greenhouse gas total column concentrations by exploiting absorption signatures of the target species (carbon dioxide – CO_2 – for OCO, CO_2 and methane – CH_4 – for GOSAT) in shortwave infrared (SWIR) spectra of sunlight backscattered by the Earth's surface and atmosphere. These observations are challenged by the ambitious precision and accuracy requirements which, typically, amount to fractions of a percent of the retrieved gas concentrations (e.g. Miller et al., 2007). Sources of error for such solar backscatter measurements in the SWIR are manifold including amongst others instrument noise, uncertain knowledge and degradation of instrument parameters, spectroscopic uncertainties as well as approximations inherent to the retrieval forward model such as the approximate treatment of light scattering by aerosols, cirrus and thin clouds. In practice, it is difficult to disentangle types of error and to conclude on effective counter measures.

Butz et al. (2011) proposed a method how to discriminate between errors related to unaccounted light scattering effects and other sources of error using ocean-glint observations. The method has been demonstrated using measurements of the Thermal And Near infrared Sensor for carbon Observation (TANSO) – Fourier Transform Spectrometer (FTS) onboard GOSAT. Figure 1 motivates the general concept by comparing the total column concentration of molecular oxygen ($[\text{O}_2]$) retrieved from TANSO-FTS O_2A -band ($\sim 13\,100\text{ cm}^{-1}$) spectra to $[\text{O}_2]$ calculated with high accuracy from meteorological support data. The retrievals neglect any (particulate as well as molecular) light scattering effects and thus, deviations between meteorological and retrieved $[\text{O}_2]$ are to be expected. Figure 1, however, shows that these deviations exhibit a quite

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different pattern for land-nadir and ocean-glint observations. For land-nadir, the distribution scatters rather symmetrically about the occurrence peak with a long tail to strong overestimation as well as underestimation. For ocean-glint, in contrast, the distribution exhibits a sharp “upper edge” and wide lower tail. The occurrence peak for ocean-glint is offset to lower retrieved $[O_2]$ with respect to the land-nadir retrievals.

The rationale to explain the observed pattern assumes that, for ocean-glint, virtually all scattering effects result in a shortening of the lightpath while, for land-nadir, scattering effects can have a lightpath enhancing as well as a lightpath shortening effect. Lightpath enhancement typically requires a scattering event at the Earth’s surface which, however, is dark for the off-glint ocean. Retrievals of $[O_2]$ from ocean-glint observations therefore virtually always suffer from unaccounted net lightpath shortening and thus, yield an underestimation of the true $[O_2]$ if the retrieval algorithm neglects light scattering effects. For land-nadir, the trade-off between lightpath shortening and lightpath enhancement is largely controlled by surface albedo which typically varies substantially for land surfaces. This rationale fits the pattern observed in Fig. 1 explaining the sharp “upper edge” and wide lower tail for ocean-glint as well as the rather symmetric distribution for land-nadir.

If this argument is true, simple “non-scattering” $[O_2]$ retrievals from the O_2A -band can be used to find an ensemble of clean ocean-glint scenes where lightpath modification due to scattering effects is negligible. Following our rationale above, the “upper edge” of ocean-glint retrievals, i.e the retrievals which show the least underestimation of the expected meteorological O_2 concentration, are the scenes with the least contamination by scattering effects. For the “upper edge” ensemble, light scattering as a source of error is negligible making it a good candidate ensemble to examine other sources of error such as spectroscopic uncertainties or instrumental deficiencies.

Here, we systematically assess the usefulness of the “upper edge” method through retrieval simulations and exemplary application to more than three years of TANSO-FTS ocean-glint measurements. Sections 2 and 3 introduce the employed methods and discuss the concept with the help of typical retrieval simulations. Section 4 shows how

to select the “upper edge” ensemble and how to use it for diagnosing calibration errors of the TANSO-FTS O₂A-band channel. Section 5 uses the “upper edge” ensemble to assess the spectroscopic consistency among several CO₂ absorption bands in the SWIR.

2 Methods

The main tools used here are variants of the retrieval algorithm RemoTeC jointly developed at the Netherlands Institute for Space Research (SRON) and the Karlsruhe Institute of Technology (KIT). RemoTeC has been designed to retrieve greenhouse gas concentrations from solar backscatter measurements in the SWIR spectral range. It has been used for performance simulations as well as for retrievals of CO₂ and CH₄ total column concentrations from TANSO-FTS in a variety of studies (e.g. Butz et al., 2011, 2012; Schepers et al., 2012). The algorithm is able to simulate spectra of sunlight backscattered by the Earth’s surface and atmosphere in the presence of aerosols, cirrus particles, and thin clouds (Hasekamp and Landgraf, 2002, 2005; Hasekamp and Butz, 2008).

2.1 Retrieval method

RemoTeC’s capability to simulate spectra in particle loaded atmospheres is used in Sect. 3 to generate synthetic TANSO-FTS-like measurements. For the most part of this study, however, the retrieval method employed here is a simplified variant of RemoTeC which neglects light scattering by particles. Henceforth, we refer to this simple RemoTeC variant as the “non-scattering” retrieval. In contrast to the [O₂] retrievals shown in Fig. 1, the “non-scattering” method employed here is slightly optimized for our purposes by actually taking into account molecular Rayleigh scattering and by using a narrower window within the O₂A-band for the [O₂] retrievals. The latter choice has been made in order to enhance sensitivity to scattering effects.

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As listed in Table 1, spectral windows used further on cover the R-branch of the O₂A-band, several weakly absorbing CO₂ bands around 6200 cm⁻¹, and strongly absorbing CO₂ bands around 4900 cm⁻¹. The RemoTeC forward model simulates radiances of backscattered sunlight based on molecular absorption properties provided by the line-mixing model of Tran and Hartmann (2008) for O₂ and by a standard Voigt line-shape model for CH₄ and H₂O driven by HITRAN 2008 (Rothman et al., 2009). The spectroscopic model of the O₂A-band further includes continuum absorption due to collision-induced-absorption. For CO₂, we construct a line-mixing correction from the line-mixing code by Lamouroux et al. (2010). The line-mixing correction is then used to correct the standard Voigt line-shape driven by HITRAN 2008.

First guess and a priori gas concentration profiles for CO₂ and CH₄, respectively, come from CarbonTracker (code version 2011) model output for the years 2009 and 2010 (Peters et al., 2007) and from TM4 model output for the year 2007 (Meirink et al., 2008). For O₂ and H₂O, initial guess and a priori are calculated from our meteorological support data. The latter are in particular pressure, temperature, humidity, and surface windspeed provided by the 6 hourly ERA-Interim analysis of the European Centre for Medium-Range Weather Forecast (ECMWF) with a horizontal resolution of 0.75° × 0.75° latitude × longitude. The algorithm models reflection at the glint-spot on the ocean surface through a wind-speed driven Cox-and-Munk reflection model (Cox and Munk, 1954) and a Lambertian albedo term that varies with wavelength (up to cubic order). Polarization effects are neglected.

Retrieval parameters are the 4-layer partial column profiles of the target absorbers O₂ and CO₂, the total column concentrations of other interfering absorbers, spectral shift parameters (per window), and 3 parameters (per window) driving the Lambertian part of surface reflection. In the O₂A-band, we further fit a radiance offset parameter to account for non-linearity of the detector electronics (Kuze et al., 2012). Wind speed is not retrieved but interpolated from the meteorological fields. The absorber concentrations to be retrieved are defined per window (Table 1) implying that the retrieval is free

to find different concentrations for the same chemical species retrieved from different windows.

Given forward modelled radiances, an inverse method estimates the retrieval parameters from the measurements. The inverse method used here is based on a Phillips–Tikhonov scheme which minimizes a cost function composed of two terms. The least-squares term contains the (squared) error-weighted mismatch between measured and modelled radiances. The side-constraint term is the first-order finite difference operator for the partial column profiles of the target absorbers and vanishes for all other retrieval parameters. The regularization parameter weighting the side-constraint against the least-squares term is chosen such that each absorber vertical profile gets one degree of freedom. Thus, the inverse method yields absorber profiles with the same shape as the a priori profiles while the total column absorber concentration is only constraint by the least-squares term.

2.2 Simulation method

For generating simulated measurements, we use the standard variant of RemoTeC which takes into account particle scattering in the atmosphere. In order to limit the number of simulations to typical cases, we select one aerosol and one cirrus particle type. The aerosol particles are weakly absorbing with sizes following a bimodal log-normal distribution with a dominating coarse mode. Assuming sphericity of the aerosols, optical properties are calculated from Mie theory. The cirrus particles are a mixture of randomly oriented planar and columnar crystals with sizes between 0.003 and 1.3 mm (for the longest side). Their optical properties are modeled by the raytracing code of Hess (1998). The simulations assume generic height distributions of the scattering particles in the atmosphere characterized through the center height z_s and the width w_z (full-width-at-half-maximum) of a Gaussian or a box-like distribution. As for the retrieval method, the simulations treat reflection from the ocean-glint spot by a wind-speed driven Cox-and-Munk reflection model. In simulated ocean-glint geometry, viewing zenith angle equals exactly solar zenith angle and the relative solar and

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viewing azimuth is zero. Instrument properties such as spectral sampling and instrument line shape are taken from the analysis of actual TANSO-FTS spectra. Simulations are performed noiseless.

2.3 Measurements

Radiances submitted to the retrieval algorithm are either simulated spectra as described above or spectra recorded by the TANSO-FTS on board GOSAT. The TANSO-FTS observations exploited here are spectra of sunlight backscattered from the ocean surface in ocean-glint geometry. We use the ocean-glint criterion defined by Wunch et al. (2011b) to identify suitable scenes. Ocean-glint observations are a regularly scheduled operation pattern when GOSAT flies over the ocean. TANSO-FTS records radiances in three SWIR spectral bands and, for each band, in two orthogonal polarization directions from which we calculate the total backscattered radiance according to Yoshida et al. (2011). Instrument line shape and radiometric calibration are provided through GOSAT's instrument support with recent updates e.g. by Kuze et al. (2012) and Yoshida et al. (2012). The standard measurement set used here are TANSO-FTS spectra generated by the level 1 processor version 141.141 and later. A sensitivity study in Sect. 4 uses level 1 versions older than 141.141 (version 130.130 and older) in order to illustrate the usefulness of ocean-glint observations for diagnosing errors in the level 1 processor.

3 O₂A band retrieval simulations

The simulations aim at showing that, following the rationale outlined in Sect. 1, the “upper edge” of [O₂] retrievals are indeed scenes where lightpath modification due to light scattering by particles is negligible. To this end, we perform “non-scattering” [O₂] retrievals from simulated ocean-glint observations of the O₂A-band (W1 in table 1). Aben et al. (2007) performed similar simulations for retrievals of the CO₂ total

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column concentration ($[\text{CO}_2]$) from synthetic ocean-glint and land-nadir measurements of the Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY) in the weakly absorbing CO_2 band around 6350 cm^{-1} (similar to W2 in table 1). They found that the neglect of aerosol and cirrus scattering effects yields underestimation of the true $[\text{CO}_2]$ for all considered ocean-glint scenarios. For Lambertian land surfaces, however, overestimation of the true CO_2 concentration due to unaccounted lightpath enhancement is the usual case for moderate to high surface albedo.

Figure 2 illustrates the effect of the “non-scattering” assumption on retrieved $[\text{O}_2]$ if scattering particles are present in the atmosphere. The test scenarios cover aerosols in the lower troposphere (Gaussian layer, $z_s = 1 \text{ km}$, $w_z = 2 \text{ km}$), and cirrus particles in the upper troposphere (Gaussian layer, $z_s = 10 \text{ km}$, $w_z = 2 \text{ km}$). Simulations were performed for a range of particle optical thickness (τ_s , given at the O_2A -band), solar zenith angle (SZA), and surface wind speed.

The simulations confirm that for the considered aerosol and cirrus scenarios retrieved $[\text{O}_2]$ increasingly underestimates the true concentration with increasing particle optical thickness. Due to the elevated altitude, the cirrus layer has a more severe screening effect than the low-altitude aerosol layer. Higher wind speed roughens the ocean surface and reduces the amount of directly reflected sunlight. Thus, the contribution of scattered light and the penalty for neglecting scattering in the retrieval generally becomes larger for higher wind speeds. The effect of solar zenith angle in particular depends on the details of the scattering phase function which is driven by particle type and size. In accordance with our introductory argument, overestimation of the true O_2 concentration does not occur for these simple generic simulations.

Overestimation of true $[\text{O}_2]$ could occur in a scenario where an optically thick particle layer at very low altitude is overlaid by an elevated optically thin scattering layer. The lower layer acts as a bright reflector and due to its low altitude only shields a small fraction of the O_2 column. Multiple scattering between the upper and the lower particle layer could result in a net light-path enhancement. In that case, retrieved $[\text{O}_2]$ could overestimate the expected concentration. Figure 3 illustrates such a scenario. It assumes

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a double layer of low-altitude particles (box layer) overlaid by an high-altitude cirrus layer (Gaussian layer, $z_s = 10$ km, $w_z = 2$ km) with small optical thickness $\tau_{\text{cir}} = 0.2$. If optical thickness of the lower layer is small, the shielding effect of the particle layers dominates causing severe underestimation of the true $[\text{O}_2]$. With increasing optical thickness of the lower layer, however, multiple scattering between the layers and, for large optical thickness, multiple scattering within the lower layer itself tend to offset the screening effect. This leads to $[\text{O}_2]/[\text{O}_{2,\text{true}}]$ exceeding 1 for large particle optical thickness. The geometric extent of the lower particle layer controls the magnitude of the screening effect and thus, the amount of multiple scattering events necessary to offset the overall screening.

Thus, there are cases conceivable where a combination of low clouds and elevated scattering layers could result in seemingly accurate or even high-biased $[\text{O}_2]$ ocean-glint retrievals. This contradicts the general applicability of the proposed method. In order to avoid scenes with multiple scattering layers contaminating our findings from GOSAT, we attempt screening such scenes using two criteria. The first criterion is based on the retrieved Lambertian albedo correction (0th order) to the wind-speed driven Cox-and-Munk glint reflection model. Figure 3 shows that for large optical thickness of the lower scattering layer the correction term becomes large as well. Putting an upper limit of 0.05 on the retrieved Lambertian term screens all the simulated scenes which could contaminate the “upper edge” ensemble. The second screening criterion discards observations contaminated by thin cirrus clouds similar to the test proposed by Yoshida et al. (2011). The latter screening method exploits the optically thick water vapor (H_2O) absorption band around 5160 cm^{-1} .

4 TANSO-FTS O_2 A-band

The retrieval simulations in Sect. 3 confirm our mechanistic understanding of radiative transfer in ocean-glint geometry. In the next step, we process TANSO-FTS spectra recorded in ocean-glint geometry between June 2009 and September 2012 by the

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“non-scattering” method. As for the simulations, the target quantity is the $[O_2]/[O_{2,meteo}]$ ratio. The “non-scattering” retrievals are subject to some basic quality filters which check for anomalies of the TANSO-FTS instrument, non-convergence of the algorithm, bad quality of the spectral fit ($\chi^2 > 4$) and cirrus contamination.

Figure 4 shows a timeseries of the $[O_2]/[O_{2,meteo}]$ ratio. The retrievals are binned in ~ 10 – 12 day clusters, which are separated by ~ 1 – 3 days for which GOSAT did not conduct ocean-glint measurements. These temporal bins are selected manually since the operation pattern is not entirely regular. For each of the temporal bins, we generate an occurrence count similar to Fig. 1 by counting the occurrence of $[O_2]/[O_{2,meteo}]$ values in 0.002 wide slots and normalize it such that a value of 1 indicates the slot with most counts.

As for Fig. 1, we find that overall there is a cloud of low-biased $[O_2]$ retrievals bound by a relatively sharp “upper edge”. In contrast to Fig. 1, the retrievals shown here consider Rayleigh scattering by molecules and use a narrower O_2A -band retrieval window (Table 1). In order to identify the “upper edge”, we select all retrievals between the 95 % and 99 % percentile of $[O_2]$ retrievals, separately for each 10–12 day temporal bin.

Butz et al. (2011) used a different “upper edge” criterion based on the retrieval noise error which worked well for the considered 3-months period. Here, we examine an extended time series and aim at determining the “upper edge” per 10–12 day bin in order to assess temporal variability. The noise criterion failed to determine a reasonable “upper edge” for several of the bins. Therefore, we decided to use a conceptually very simple criterion based on ad-hoc chosen percentiles. The ambiguity related to the definition of the “upper edge” is to be considered as part of the intrinsic uncertainty of the method. Most conclusions elaborated on below are not sensitive to the exact choice of the “upper edge”.

Generally, the “upper edge” does not align with the expected O_2 concentration at a ratio $[O_2]/[O_{2,meteo}] = 1$ but a factor 1.03 to 1.04 to higher values. The latter finding has been detected previously by Butz et al. (2011) from the first three months of the time series, though Butz et al. (2011) find the “upper edge” at slightly lower values

around 1.030. There, we suggested to introduce a scaling factor for the line strengths of O_2 absorption lines. Differences between the study here and Butz et al. (2011) are due to a narrower O_2 retrieval window here and due to a different definition of the “upper edge” ensemble.

Figure 5 shows the selected “upper edge” ensemble of standard retrievals together with the same ensemble reprocessed using spectra generated by older versions of the level 1 processor (level 1 version 130.130 and older). The latter are only available up to December 2012. Beside the overall scaling factor, Fig. 5 shows substantial temporal variability of the “upper edge” of both, the standard retrievals as well as the retrievals from old level 1 versions. The abrupt increase of the latter in April 2011 coincides with the update of the TANSO-FTS level 1 processor to version 130.130 which includes an erroneous correction algorithm for residual non-linearity of the O_2A -band detector module (Kuze et al., 2012). The attempted correction in particular results in an undesired change of the instrument line shape (ILS). Since this was unforeseen, $[O_2]$ retrievals from level 1 version 130.130 suffer from an erroneous ILS model. The new level 1 versions (141.141 and later) clearly remedy this effect.

We find further temporal variability which, so far, cannot be attributed unambiguously to instrumental effects. In particular, the first year of the timeseries reveals a bowl-shape pattern that rather abruptly ends in August 2010. On 10 August 2010, the GOSAT team switched on the image motion compensation (IMC) mechanism for ocean-glint operations. IMC makes the TANSO-FTS stare at the same ground spot during the ~ 4 s acquisition time of the interferogram. Before 10 August 2010, ocean-glint operations were conducted with a fixed pointing mirror orientation making the ground spot move along with the satellite track. Given the apparent effect of the IMC on our “upper edge” O_2 retrievals, one might speculate about slightly erroneous pointing information or its erroneous treatment by the retrieval algorithm before 10 August 2010. So far, however, we are not able to identify the root cause for this behaviour.

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5 TANSO-FTS CO₂ bands

Section 4 describes how we select the “upper edge” and use it for testing our forward model of the TANSO-FTS O₂A-band observations. Here, we investigate whether our forward model allows for consistent retrievals of the CO₂ total column concentration ([CO₂]) from different CO₂ absorption bands covered by the TANSO-FTS. To this end, we reprocess the “upper edge” ensemble by the “non-scattering” method and retrieve [CO₂]_{*i*} as a target absorber separately from window *W_i* with *i* = 2, 3, 4, 5, 6. As emphasized in Sect. 2, the method is setup such that the windows are uncoupled and [CO₂]_{*i*} retrieved from one window is free to vary independently from the other windows. If the forward model of the considered windows is consistent, we expect retrieved [CO₂]_{*i*} not to vary among the windows, since the “upper edge” ensemble should be free of (wavelength dependent) particle scattering effects.

Figure 6 shows the relative difference of [CO₂] retrievals from windows W3, W4, W5, and W6 with respect to the retrievals from window W2, defined as

$$\Delta[\text{CO}_2]_i = \frac{[\text{CO}_2]_i}{[\text{CO}_2]_2} - 1, \quad (1)$$

with *i* = 3, 4, 5, 6. We find that on average W3 and W6 retrievals are different from W2 by 0.33 % (1.3 ppm out of 390 ppm) and 0.17 % (0.7 ppm out of 390 ppm), respectively. W4 and W5 retrievals show larger average differences of 1.57 % (6.2 ppm out of 390 ppm) and 2.22 % (8.7 ppm out of 390 ppm) with respect to W2. No systematic temporal pattern is detectable. Variability is dominated by data scatter which, for W3 and W4, can be explained by the retrieval noise error calculated via error propagation from the out-of-band noise of the TANSO-FTS spectra. For W5 and W6, retrieval noise amounts to roughly half of the observed standard deviation which could point to some residual scattering effects contaminating our selection of the “upper edge”.

The observed differences in [CO₂]_{*i*} could merely reflect sensitivity differences between the individual retrievals. Vertical sensitivity of the total column retrieval from

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window W_i is typically described by the column averaging kernel a_i (Rodgers, 2000; Connor et al., 2008). Typical column averaging kernels for the various retrieval windows are shown in Fig. 7. The column averaging kernels have a very similar shape for W_2 and W_3 , and for W_5 and W_6 since the respective bands contain very similar absorption structures of CO_2 . Thus, we expect the sensitivity effect to be smallest when comparing W_2 with W_3 and W_5 with W_6 . In fact, Fig. 6 indicates that there is differences between W_5 and W_6 that are larger than differences between W_5 and W_2 . Consequently, the observed difference between W_5 and W_6 is most likely not attributable to sensitivity effects alone.

In order to assess the effect of different a_i on our comparison of $[\text{CO}_2]_i$ retrieved from the various windows, we estimate the smoothing effects by calculating

$$\Delta_s[\text{CO}_2]_i = \frac{a_i x_a}{a_2 x_a} - 1, \quad (2)$$

where $i = 3, 4, 5, 6$ and x_a is the a priori CO_2 vertical profile derived from CarbonTracker as noted in Sect. 2. Similar to Fig. 6, $\Delta_s[\text{CO}_2]_i$ is then plotted for the “upper edge” ensemble and each window (not shown for conciseness). As expected, we find the smallest sensitivity effect when comparing W_2 and W_3 amounting to average $[\text{CO}_2]$ differences of 0.002 %. The greatest $[\text{CO}_2]$ sensitivity difference is found between W_2 and W_5 , W_6 retrievals with an average difference of 0.17 %. Average sensitivity differences between W_2 and W_4 amount to 0.10 %. The averaging kernels a_i exhibit some temporal variability due to observation geometry, signal-to-noise, and concentration distributions changing over time. Thus, $\Delta_s[\text{CO}_2]_i$ also exhibits a time dependence the amplitude of which is typically on the order of the observed average differences.

Considering the magnitude of the effect of retrieval sensitivity on $[\text{CO}_2]_i$, we conclude that the agreement found for our ocean-glint CO_2 retrievals from W_2 , W_3 , W_6 is within the range which could be affected by sensitivity issues. Strictly, the detected difference between W_2 and W_3 retrievals of 0.33 % is significantly greater than the effect expected from sensitivity differences (less than 0.01 %). So, we are tempted to attribute this small difference to an actual shortcoming of the input data to our algorithm. CO_2 retrievals

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from W4 and W5, however, show large differences when compared to retrievals from W2, W3, W6. These differences greatly surpass the effect expected from different averaging kernels. Our forward model for W4 and W5 must suffer from an inconsistency that causes this pattern.

The obvious candidate for such an inconsistency is the calibration of the absorption line strengths among the various CO₂ retrieval windows. The use of erroneous instrument line shape functions would be another candidate that could cause these inconsistent CO₂ retrievals. The later seems unlikely since all CO₂ windows are extracted from band 2 and band 3 of the TANSO-FTS SWIR observations which exhibit accurate knowledge of the instrument line shape. For W4, we note a strong dependence of retrieved [CO₂]₄ on the polynomial order of the albedo correction. Since we are not able to unambiguously determine the root cause for this algorithm deficiency, we recommend not using W4, W5 together with W2, W3, W6 for CO₂ retrievals or to uncouple W4, W5 from W2, W3, W6 when setting up the retrieval method. Alternatively, we will consider a similar strategy as for our O₂A-band forward model where we proposed to scale the O₂ absorption line strengths by a constant factor to make the O₂A-band forward model consistent with meteorological surface pressure. For CO₂ retrievals from W2 through W6, one could introduce similar scaling factors to make the forward model consistent among the various windows.

6 Conclusions and discussion

Observations of sunlight backscattered from the glint spot on the ocean surface offer a tool to select an ensemble of observations that do not suffer from errors due to uncertain knowledge of the lightpath. Since lightpath modification by atmospheric particles in general is a dominating source of uncertainty for the retrieval of atmospheric constituent concentrations from solar backscatter measurements, our method allows for assessing other sources of error often masked by lightpath related errors.

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We suggest to identify the ensemble by performing $[O_2]$ retrievals from ocean-glint spectra of the O_2A -band. Retrievals are performed under the assumption of negligible particle scattering in the atmosphere and thus, are computationally cheap. Simulations confirm, that light scattering by particles is negligible for the “upper edge” of these retrievals i.e. the retrievals which show the least underestimation with respect to $[O_{2,meteo}]$, the O_2 concentration expected from meteorological support data. A caveat applies for scenes where an optically thick particle layer such as a cloud at low altitude acts as a bright reflector and is overlaid by an optically thin elevated scattering layer such as cirrus. Independent cloud screening can help avoid contamination of the “upper edge” ensemble by such scenes.

For real measurements such as from the TANSO-FTS onboard GOSAT, selection of the “upper edge” to some extent depends on the identification criterion. Here, we use a conceptually very simple criterion attributing soundings to the “upper edge” which yield 95 to 99% of the largest $[O_2]/[O_{2,meteo}]$ ratios. Further uncertainty is introduced by the meteorological and geolocation data which are used to calculate $[O_{2,meteo}]$ for each sounding. Case studies have shown that ECMWF surface pressure used here for meteorological input is accurate to a few hPa (King, 2003; Lammert et al., 2008).

We derive the “upper edge” ensemble for 10–12 day periods over more than 3 yr of TANSO-FTS ocean-glint observations between June 2009 and September 2012. The corresponding $[O_2]/[O_{2,meteo}]$ ratios do not align at the expected ratio of unity, but at a ratio 1.03 to 1.04 which largely confirms findings by our precursor study (Butz et al., 2011). There, we speculated on a potential scaling factor of 1.030 to be applied to the O_2 line strengths in the O_2A -band. Crisp et al. (2012) derive a similar scaling factor of 1.025 based on their surface pressure retrievals from TANSO-FTS nadir-land observations. Given the uncertainty in determining the “upper edge” by our method, the findings by Crisp et al. (2012) and Butz et al. (2011) appear consistent. Yoshida et al. (2013) find a smaller scaling factor of 1.010 using the “upper edge” method and Cogan et al. (2012) conclude on a surface pressure bias of only 4 hPa when not scaling the O_2 line strengths. Clearly, findings on a potential spectroscopic scaling factor depend on

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the spectroscopic parameters used to drive the retrieval forward models which might not be identical among different retrieval methods. Nevertheless, an inconsistency between the former and latter studies remains.

The “upper edge” $[O_2]/[O_{2,meteo}]$ ensemble obtained by processing the most recent TANSO-FTS level 1 spectra (version 141.141 and later) reveals a substantial time dependence in particular in the first year of the mission. An abrupt change of the $[O_2]/[O_{2,meteo}]$ ratios coincides with activation of the image motion compensation for ocean-glint observations in August 2010. After August 2010, time dependencies are small. For an older level 1 dataset (version 130.130), the $[O_2]/[O_{2,meteo}]$ ratios clearly monitor the detrimental impact of an erroneous non-linearity correction scheme on constituent retrievals.

We further use the “upper edge” ensemble to conduct $[CO_2]$ retrievals from several absorption bands covered by the TANSO-FTS and to be covered by future satellite missions. The CO_2 concentrations found are consistent to better than 1.5 ppm among the bands (W2, W3, W6 around 6350, 6200, 4850 cm^{-1}) which are heavily used for CO_2 retrievals from TANSO-FTS, the upcoming Orbiting Carbon Observatory (OCO-2), and the ground-based Total Carbon Column Observing Network (TCCON) (Wunch et al., 2011a). Considerable effort has been put into determining the related spectroscopic parameters with high accuracy (e.g. Lamouroux et al., 2010; Devi et al., 2010). Substantially larger retrieval differences are observed for two other CO_2 absorption bands (W4, W5 around 6000, 4970 cm^{-1}) pointing to errors in the spectroscopic parameters. The detected inconsistencies are unacceptably large in the view of the stringent accuracy requirements for remote sensing of CO_2 concentrations and thus, demand further investigation if these bands are to be used for monitoring of CO_2 or for simultaneous use with the other bands.

Acknowledgements. AB is supported by Deutsche Forschungsgemeinschaft (DFG) through the Emmy-Noether programme, grant BU2599/1-1 (RemoteC). Access to GOSAT data was granted through the 2nd GOSAT research announcement jointly issued by JAXA, NIES, and MOE. ECMWF ERA Interim analyses are provided through

http://data-portal.ecmwf.int/data/d/interim_daily/. CarbonTracker data are provided by NOAA ESRL, Boulder, Colorado, USA from the Web site at http://carbontracker.noaa.gov.

The service charges for this open access publication
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Helmholtz Association.

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Table 1. Definition of spectral windows used in this study. Boldface indicates target absorbers for the retrieval algorithm.

Window name	Wavenumber range/cm ⁻¹	Wavelength range/nm	Main absorber	Interfering absorber
W1	13 123.0–13 195.0	757.9–762.0	O₂	–
W2	6301.2–6380.8	1567.2–1587.0	CO₂	H ₂ O
W3	6170.0–6278.5	1592.7–1620.7	CO₂	H ₂ O, CH ₄
W4	5945.9–6138.0	1629.2–1681.8	CH ₄	H ₂ O, CO₂
W5	4946.0–5006.0	1997.6–2021.8	CO₂	H ₂ O
W6	4806.0–4896.0	2041.5–2080.7	CO₂	H ₂ O

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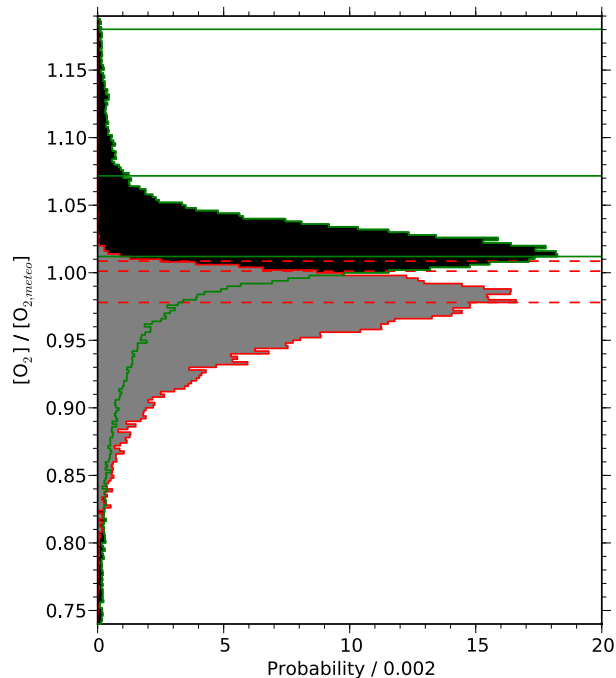


Fig. 1. Histogram of the ratio between the total column O_2 concentration retrieved from TANSO-FTS O_2A -band spectra ($[O_2]$) and the expected meteorological O_2 total column concentration ($[O_{2,meteo}]$). Histograms are shown separately for retrievals from nadir soundings over land (green lines, black fill) and from glint-spot soundings over the ocean (red lines, grey fill). The horizontal lines depict the 99, 95% percentiles and the occurrence peak (from top to bottom). Retrievals neglect molecular as well as particulate light scattering effects and cover more than 3 yr of GOSAT operations from June 2009 to September 2012 (~ 1 million nadir soundings, ~ 0.3 million glint soundings). Basic quality screening is based on convergence of the iterative algorithm, instrument anomaly flagging, goodness of the fit, and cirrus contamination detectable at the 5160 cm^{-1} water vapor absorption band.

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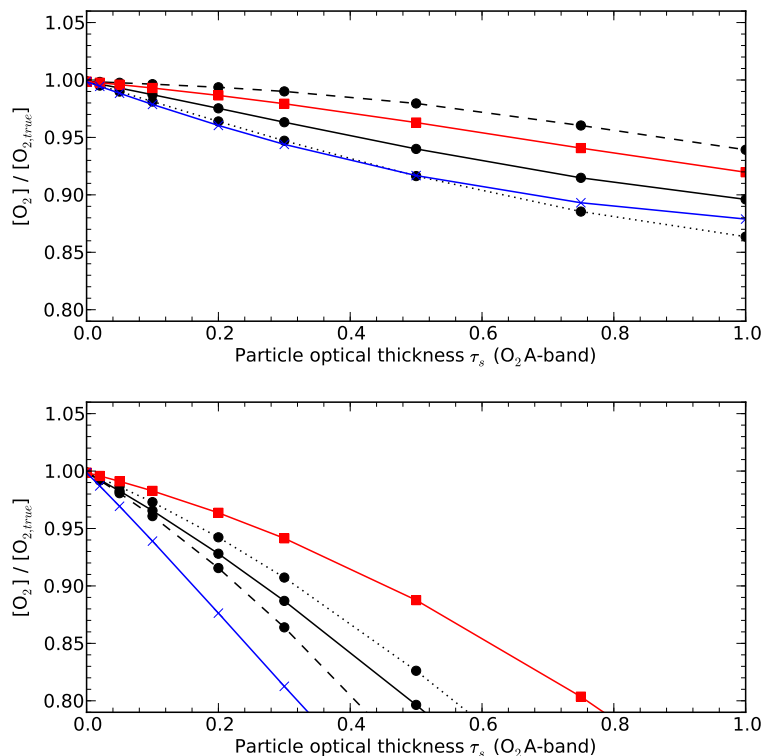


Fig. 2. Simulated ratio of retrieved ($[O_2]$) and true O_2 ($[O_{2,true}]$) total column concentration for various particle optical thickness τ_s (at the O_2A -band). Scenarios cover a coarse particle aerosol layer in the lower atmosphere (upper panel, Gaussian, $z_s = 1$ km, $w_z = 2$ km) and an elevated layer of cirrus particles (lower panel, Gaussian, $z_s = 10$ km, $w_z = 2$ km). Simulations are performed for wind speeds 2, 5, 10 m s^{-1} (red squares, black dots, blue crosses). In the case of 5 m s^{-1} wind speed, retrievals are shown for solar zenith angle 5, 15, 45° (dotted, solid, dashed lines), which covers the SZA range of 5 to 30° typical for GOSAT ocean-glint observations.

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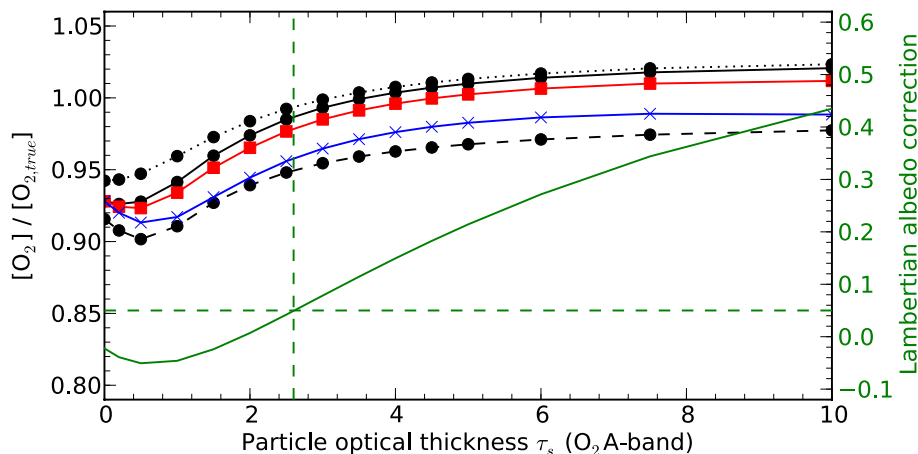


Fig. 3. Similar to Fig. 2 but for a double layer of low-altitude aerosols with variable optical thickness ($\tau_s=0$ to 10) overlaid by a cirrus layer (Gaussian, $z_s = 10$ km, $w_z = 2$ km) of small optical thickness ($\tau_{cir} = 0.2$). The geometric extent of the low-layer aerosol is assumed box-like extending from the ground to 200, 500, and 800 m (black dots, red squares, blue crosses) altitude. Simulated retrievals are performed for wind speed 5 m s^{-1} . For the case of 200 m geometric extent of the lower layer, simulations cover solar zenith angle 5, 15, 45° (dotted, solid, dashed lines). For the later case with solar zenith angle of 15° , the retrieved Lambertian albedo correction (green solid line, 0th order term) is shown on the right (green) ordinate scale. Horizontal and vertical, dashed, green lines illustrate the threshold used for screening contaminated TANSO-FTS scenes.

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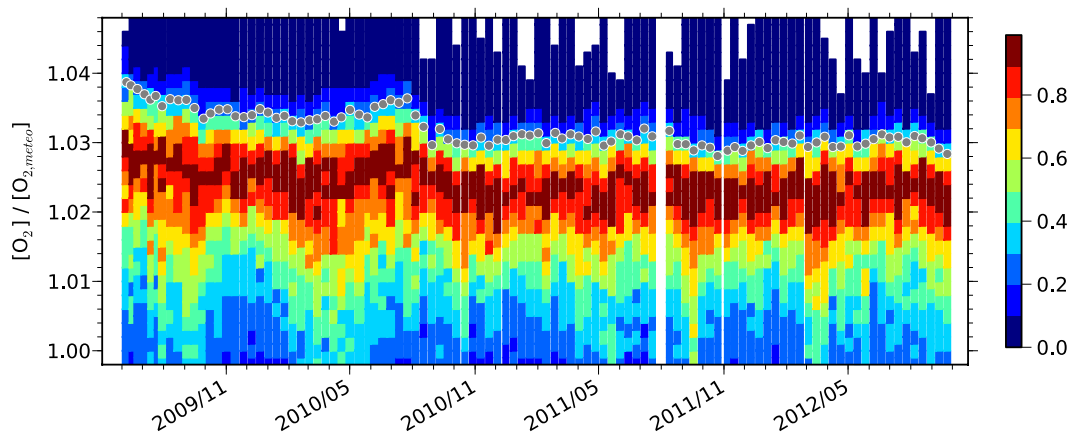


Fig. 4. Timeseries of the $[O_2]/[O_{2,meteo}]$ ratio as found by “non-scattering” retrievals from TANSO-FTS measurements between June 2009 and September 2012 (level 1 version 141.141 and higher). The color code illustrates the normalized occurrence of retrievals per 10–12 day abscissa (temporal) bin and 0.002 ordinate ($[O_2]/[O_{2,meteo}]$ ratio) bin. For each temporal bin, the occurrence is normalized such that the ratio bin with most counts has a normalized occurrence of 1. We call the “upper edge” all soundings between the 95 and 99 % percentile for each temporal bin. The median value within the “upper edge” is shown as grey circles. White color indicates no data.

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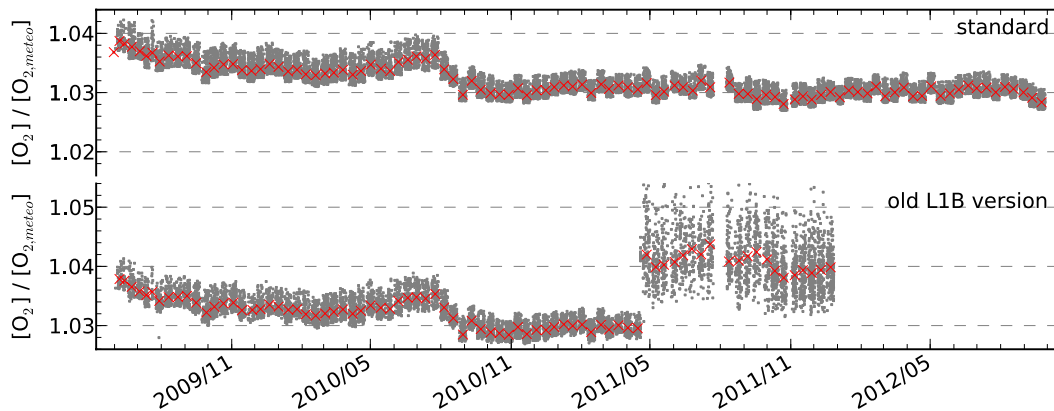


Fig. 5. Timeseries of the “upper edge” $[O_2]/[O_{2,\text{meteo}}]$ ratio. Standard “non-scattering” retrievals (upper panel, level 1 version 141.141 and later) are compared to retrievals from old level 1 versions (lower panel, version 130.130 and older). Individual soundings are shown as grey squares. Red crosses indicate the median per 10–12 day temporal bin.

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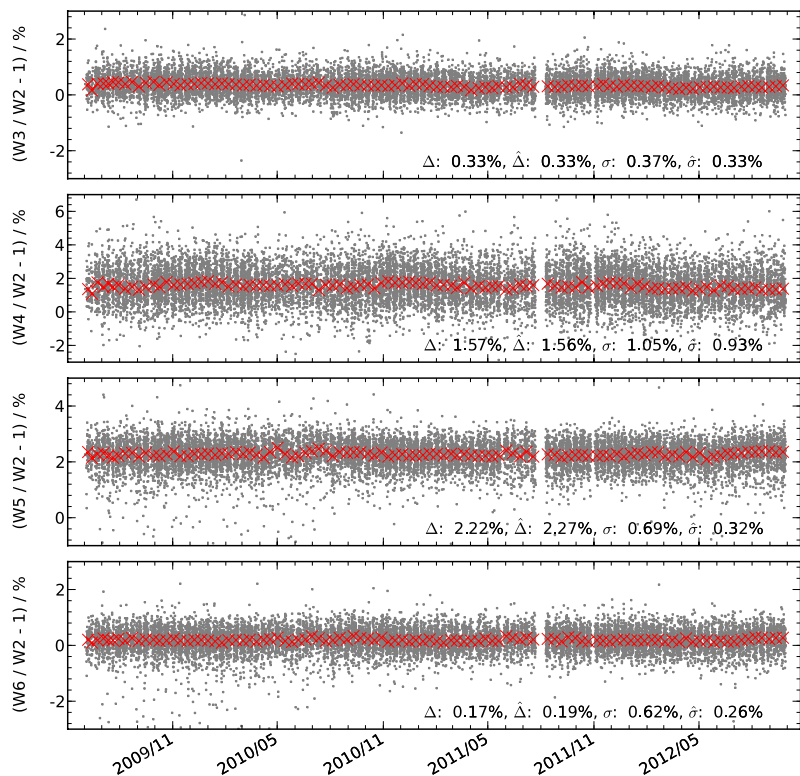


Fig. 6. Relative difference between $[\text{CO}_2]$ retrieved from various windows. Retrievals from window 3 (uppermost panel, W3), window 4 (upper middle panel, W4), window 5 (lower middle panel, W5), and window 6 (lowermost panel, W6) are referenced to retrievals from window 2 (W2). Individual soundings are shown as grey squares. Red crosses indicate the median per 10–12 day temporal bin. For each panel, the mean difference (Δ), median difference ($\hat{\Delta}$), standard deviation of differences (σ), and the average retrieval noise error ($\hat{\sigma}$) are given in percent.

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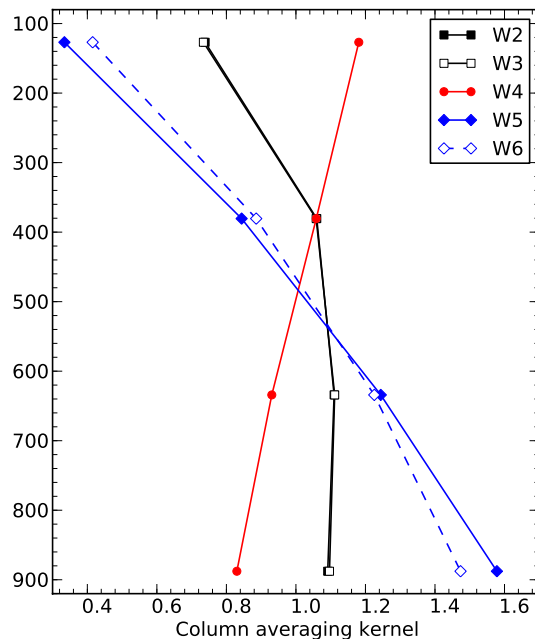


Fig. 7. Column averaging kernels for $[\text{CO}_2]$ retrieved from windows W2 (black, solid squares), W3 (black, open squares), W4 (red, solid circles), W5 (blue, solid diamonds), and W6 (blue, open diamonds). Averaging kernels for W2 and W3 are almost indistinguishable. For each window, the mean column averaging kernel among the “upper edge” ensemble is shown.

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