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Measuring atmospheric CO₂ from space using Full Spectral Initiation (FSI) WFM-DOAS

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Abstract

Satellite measurements of atmospheric CO₂ concentrations are a rapidly evolving area of scientific research which can help reduce the uncertainties in the global carbon cycle fluxes and provide insight into surface sources and sinks. One of the emerging CO₂ measurement techniques is a relatively new retrieval algorithm called Weighting Function Modified Differential Optical Absorption Spectroscopy (WFM-DOAS) that has been developed by Buchwitz et al. (2000). This algorithm is designed to measure the total columns of CO₂ (and other greenhouse gases) through the application to spectral measurements in the near infrared (NIR), made by the SCIAMACHY instrument on-board ENVISAT. The algorithm itself is based on fitting the logarithm of a model reference spectrum and its derivatives to the logarithm of the ratio of a measured nadir radiance and solar irradiance spectrum. In this work, a detailed error assessment of this technique has been conducted and it has been found necessary to include suitable a priori information within the retrieval in order to minimize the errors on the retrieved CO₂ columns. Hence, a new CO₂ retrieval algorithm called Full Spectral Initiation (FSI) WFM-DOAS has been developed which generates a reference spectrum for each individual SCIAMACHY observation using the known properties of the atmosphere and surface at the time of the measurement. Initial retrievals over Siberia during the summer of 2003 show that the measured CO₂ columns are not biased from the input a priori data and that whilst the monthly averaged CO₂ distributions contain a high degree of variability, they also contain significant spatial features.

1 Introduction

The carbon cycle is one of the most important physical systems on the Earth and its response to increased levels of atmospheric CO₂ and to global warming is one of the factors that will shape the future climate. In the natural cycle, carbon is continually transported between the atmosphere, ocean and terrestrial biosphere through fluxes

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that are of order of tens of billion tons per year ([Intergovernmental Panel on Climate Change, 2001](#)). Over the last 200 years humans have perturbed this natural cycle by adding comparatively small, but nevertheless significant amounts of carbon into the atmosphere through the burning of fossil fuels, deforestation and by the industrial production of cement, lime and ammonia. Consequently during this period the concentration of atmospheric CO₂ has risen by about 30%, soaring from 270 to 370 ppmv (parts per million by volume). Only approximately half of the total anthropogenic carbon emitted into the atmosphere actually remains there, the rest being absorbed by the oceans and terrestrial biosphere. Carbon that eventually reaches the deep oceans is considered to be removed from the climate system for several hundred years whereas that sequestered by the biosphere may subsequently be released back to the atmosphere over much shorter timescales. The variability and efficiency of the oceanic and terrestrial fluxes will therefore be critical in determining future atmospheric CO₂ levels.

Currently these fluxes are estimated using inverse methods that employ transport models coupled with surface measurements of the atmospheric concentration (e.g. [Rödenbeck et al., 2003](#)). However, such models are hampered firstly by the accuracy of the meteorological fields that drive them and secondly by the sparseness and inhomogeneous geographical distribution of the approximately one hundred CO₂ measurements sites that provide the observational constraints ([GLOBALVIEW-CO₂, 2005](#)). Although this approach firmly suggests a Northern Hemisphere terrestrial sink ([Tans et al., 1990](#); [Ciais et al., 1995](#)), it is somewhat limited as it cannot fully resolve the North American and Eurasian components nor can it determine the extent of the fluxes in the Southern Oceans ([Gurney et al., 2002](#)). Knowledge of such regional scale fluxes and the processes that control them can be vastly improved through sampling the atmosphere more comprehensively. The optimal way to achieve this is to make observations from space.

However, this is a challenging observational problem as CO₂ is considered to be well mixed within the atmosphere owing to its long lifetime. As it is chemically inert, its distribution is influenced by transport processes and through the spatial and temporal

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variability of both natural and anthropogenic surface fluxes. Seasonal and geographical trends and gradients are evident, not only at the surface but also within the free troposphere and lower stratosphere, typically with an observed variability of about 1–20 ppmv (Nakazawa et al., 1991, 1992; Conway et al., 1994; Anderson et al., 1996; Nakazawa et al., 1997; Vay et al., 1999; Kuck et al., 2000; Levin et al., 2002; Matsueda et al., 2002; Sidorov et al., 2002). That aside, total columns of CO₂ exhibit about only half the variability at the surface with the diurnal fluctuations rarely exceeding 1 ppmv (Olsen and Randerson, 2004). Such small fluctuations in the CO₂ concentration will only therefore produce correspondingly small changes in radiance at the top of the atmosphere (Mao and Kawa, 2004). Since carbon dioxide is a strong absorber, any space-borne sensor will need to detect such minor responses against what is a dominant background signal. The detection of small radiance changes against this strong background requires measurements to be made at high precision, challenging the instrumental limits of the current suite of available sensors. If satellite observations are to improve over the existing ground network, monthly averaged column data at a precision of 1% (2.5 ppmv) or better, for an 8° × 10° footprint are needed (Rayner and O'Brien, 2001).

Over the last several years various sensitivity analyses have been undertaken to assess if this 1% precision is attainable by satellite instruments. These have mostly focussed on the retrieval of total columns from either the near infrared (NIR) using differential absorption optical spectroscopy (DOAS) (O'Brien and Rayner, 2002; Dufour and Bréon, 2003; Kuang et al., 2002) or from the close examination of the CO₂ emission bands in the thermal infrared (Chédin et al., 2003). Light in the thermal IR originates from the mid-troposphere in contrast to the reflected sunlight of the near infrared. Thus the near surface sensitivity of the NIR makes it the ideal spectral region for observing surface fluxes (even though the absorption bands at 4 μm and 15 μm are much stronger). In addition the NIR is less sensitive to temperature and water vapour.

Since the launch of the SCIAMACHY instrument on-board the European ENVISAT satellite, there is now an ability to measure total columns of CO₂ and other greenhouse

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gases from spectral measurements in the NIR. Observations of CO₂ (Houweling et al., 2005), carbon monoxide (CO) (Frankenberg et al., 2005b; Buchwitz et al., 2004) and methane (CH₄) (Frankenberg et al., 2005a) have been performed. From these studies, a global CO₂ data set for 2003 has been prepared by Buchwitz et al. (2005), using a relatively new retrieval algorithm called “Weighting Function Modified Differential Optical Absorption Spectroscopy” (WFM-DOAS). In this paper the SCIAMACHY/WFM-DOAS approach is used but developed further with the aim of increasing the precision and accuracy of space-borne CO₂ retrieval. An initial assessment of this algorithm’s sensitivity is given, highlighting the necessity for the inclusion of suitable a priori information within the retrieval in order to constrain the errors on the retrieved CO₂ columns. Furthermore, a new CO₂ retrieval algorithm called “Full Spectral Initiation” (FSI) WFM-DOAS is introduced that is biased towards the known properties of the atmosphere and surface at the time of the SCIAMACHY observation.

The structure of this paper is as follows. In Sect. 2 the SCIAMACHY instrument is briefly described before the WFM-DOAS retrieval technique and an error assessment of its performance is discussed in Sect. 3. In Sect. 4 the FSI WFM-DOAS retrieval algorithm is introduced with some initial results shown in Sect. 5. The paper finishes with a summary in Sect. 6.

2 The SCIAMACHY instrument

The SCanning Imaging Absorption spectrometer for Atmospheric CHartography (SCIAMACHY) is a passive hyper-spectral UV-VIS-NIR grating spectrometer (Bovensmann et al., 1999). It was launched onboard the ENVISAT satellite in March 2002 into a polar sun-synchronous orbit, crossing the Equator on its descending node (i.e. southwards) at 10.00 a.m. local time. The instrument covers the spectral range 240–2380 nm, non-continuously, in eight separate channels, with a moderate resolution of 0.2–1.4 nm. From its orbit, SCIAMACHY can observe the Earth from three distinct viewing geometries nadir, limb and lunar/solar occultation, with the majority of the or-

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bit consisting of measurements in an alternating limb and nadir sequence. The total columns of CO₂ are derived from nadir observations in the NIR, focussing on a small wavelength window within channel six, centered on the CO₂ band at 1.57 μm. The radiation in this wavelength interval consists of solar energy either reflected from the surface or scattered from the atmosphere. In nadir mode, the atmosphere directly below the sensor is viewed producing retrievals with good spatial but poor vertical resolution. A characteristic set of observations consists of the nadir mirror scanning across track for 4 s followed by a fast 1 s back-scan. This is repeated for either 65 or 80 s according to the orbital region. The ground swath viewed has a fixed dimensions of 960×30 km², (across × along track). For channel 6, the nominal size of each pixel within the swath is 60×30 km², corresponding to an integration time of 0.25 s. Global coverage is achieved at the Equator within 6 days.

3 Assessment of the WFM-DOAS retrieval algorithm

3.1 WFM-DOAS

The WFM-DOAS algorithm (Buchwitz et al., 2000; Buchwitz and Burrows, 2004) defined in Eq. (1), is based on a linear least squares fit of the logarithm of a model reference spectrum I_i^{ref} and its derivatives, plus a quadratic polynomial P_i , to the logarithm of the measured sun normalized intensity I_i^{meas} .

$$\left\| \ln I_i^{\text{meas}}(\mathbf{v}^t) - \left[\ln I_i^{\text{ref}}(\bar{\mathbf{v}}) + \frac{\partial \ln I_i^{\text{ref}}}{\partial \bar{V}_{\text{CO}_2}} \cdot (\hat{V}_{\text{CO}_2} - \bar{V}_{\text{CO}_2}) + \frac{\partial \ln I_i^{\text{ref}}}{\partial \bar{V}_{\text{H}_2\text{O}}} \cdot (\hat{V}_{\text{H}_2\text{O}} - \bar{V}_{\text{H}_2\text{O}}) + \frac{\partial \ln I_i^{\text{ref}}}{\partial \bar{V}_{\text{Temp}}} \cdot (\hat{V}_{\text{Temp}} - \bar{V}_{\text{Temp}}) \right] \right\|$$

$$+ P_i(a_m) \Big] \Big] \Big] \equiv \|\text{RES}_i\|^2 \rightarrow \min \text{ w.r.t } \hat{V}_j \text{ \& } a_m \quad (1)$$

The subscript i refers to each detector pixel of wavelength λ_i and the true, model and retrieved vertical columns are represented by $\mathbf{V}^t = (V_{\text{CO}_2}^t, V_{\text{H}_2\text{O}}^t, V_{\text{Temp}}^t)$, $\bar{\mathbf{V}} = (\bar{V}_{\text{CO}_2}, \bar{V}_{\text{H}_2\text{O}}, \bar{V}_{\text{Temp}})$ and \hat{V}_j respectively (where subscript j refers to the variables CO₂, H₂O and temperature). Here V_{Temp} is not a vertical column as such, but rather a scaling factor applied to the vertical temperature profile. Each derivative (or column weighting function) represents the change in radiance as a function of a relative scaling of the corresponding trace gas or temperature profile. Effectively the column weighting functions are replacing the absorption cross sections found in normal DOAS methods. In order to retrieve carbon dioxide derivatives for CO₂, H₂O and (optionally) temperature are required. In the case of the temperature derivative, the term $(\hat{V}_{\text{Temp}} - \bar{V}_{\text{Temp}})$ represents a uniform shift in the entire temperature profile. The fit parameters are the trace gas columns \hat{V}_{CO_2} and $\hat{V}_{\text{H}_2\text{O}}$, the temperature scaling factor \hat{V}_{Temp} and the polynomial coefficients a_m .

Although an initial error analysis was performed by [Buchwitz and Burrows \(2004\)](#) a more thorough study has been conducted, and summarized here, to assess possible biases and retrieval errors. It is not the aim of this paper to repeat their efforts but rather to highlight the most important sources of inaccuracy. To achieve this synthetic measurements for a given atmospheric state and instrument set-up were generated using the radiative transfer model SCIATRAN ([Rozanov et al., 2002](#)). Simulated retrievals were then performed using a standard reference spectrum, so that in each case the retrieved CO₂ column could be compared to its true value. This reference spectrum was constructed using the US Standard atmosphere ([McClatchey et al., 1972](#)) together with a default aerosol scenario (unless indicated otherwise). The CO₂ profile was scaled to coincide with its current concentration of 370 ppmv. Aerosols were parameterized by the LOWTRAN scheme ([Kneizys et al., 1986, 1996](#)) the default setting being a spring/summer season with maritime aerosols in the boundary layer (visibility

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23 km and humidity 80%). Background tropospheric and stratospheric aerosol conditions were assumed in conjunction with a normal mesospheric loading. Both the solar zenith and the relative azimuth angles (both 45°) and the albedo at 0.2 were kept constant throughout the studies (unless indicated). Each spectrum was created on a high resolution wavelength grid and then convolved with a Gaussian slit function of 1.4 nm (FWHM) before being interpolated onto the SCIAMACHY wavelength grid. The fitting window used throughout this sensitivity analysis was confined to the wavelength region 1561.03–1585.39 nm (see Sect. 4) which, besides the CO₂ band, contains some weak water vapour absorption lines. Weighting functions for CO₂ and H₂O, together with the quadratic polynomial were always included in the WFM-DOAS fit. However for each scenario the simulated retrieval was performed both with and without the temperature weighting function, thus allowing its contribution to be assessed. In this sensitivity study, the relative column error E_{rel} , that is the percentage difference between the retrieved CO₂ column \bar{V}^{ret} and the actual column \bar{V}^{actual} , is used to gauge the accuracy of each simulated retrieval (Eq. 2).

$$E_{\text{rel}} = \frac{\bar{V}^{\text{ret}} - \bar{V}^{\text{actual}}}{\bar{V}^{\text{actual}}} \times 100\% \quad (2)$$

3.2 Sensitivity to CO₂ vertical profile

The CO₂ profile associated with the US Standard atmosphere has a uniform volume mixing ratio of 370 ppmv, up to an altitude of 80 km. For the tropical regions, where vertical mixing is rapid, this is a fair representation. For higher latitudes however, the shape of the profile is more strongly influenced by dynamics and biospheric activity thus varying seasonally (Anderson et al., 1996; Nakazawa et al., 1997). To ascertain the sensitivity of the algorithm to the vertical CO₂ distribution, simulated retrievals were performed using spectra generated from profiles more representative of the global latitudinal and seasonal CO₂ distribution (illustrated in Fig. 1). These were taken from a new climatology prepared by J. Remedios (University of Leicester, private communica-

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tion, 2005). In each simulation the CO₂ profile was interpolated onto the US Standard pressure scale and used in conjunction with the US Standard water vapour profile.

The results from these simulations show that the shape of the CO₂ profile used to create the model reference spectra is important (Fig. 2). Using a uniform CO₂ profile tends to lead to an over-estimation of the true CO₂ concentration irrespective if the temperature weighting function is fitted. The relative error in the retrieved column varies throughout the year and is most pronounced in the Northern Hemisphere where it follows the seasonal cycle, peaking in the month of April at ~0.5%. In the Southern Hemisphere, where the CO₂ distribution is dominated by inter-hemispheric mixing, this seasonal variation is not observed and the error is smaller, typically less than 0.2%. Failure to use realistic a priori CO₂ vertical profiles within the WFM-DOAS algorithm will therefore introduce time-dependent biases.

3.3 Sensitivity to temperature and water vapour profiles

The U.S. standard atmosphere, used to generate the baseline scenario, represents an average climatology whereas the real environment itself is obviously more varied especially between the distinct latitude bands of the tropics, mid latitudes and sub-polar regions. Water vapour and temperature profiles representative of these areas (McClatchey et al., 1972) were consequently used to produce a series of simulated measurements in a similar manner to Buchwitz and Burrows (2004). However, to create a much wider range of atmospheric states both the CO₂ and H₂O trace gas profiles were also multiplied by constant factors (where the CO₂ profile here, refers to that of the US Standard atmosphere). In an addition to this, a further set of simulated measurements was created from 16 ECMWF (un-scaled) temperature and water vapour profiles selected from the months of January and July 2003. These were chosen on the basis of being strongly deviant from the U.S. standard atmosphere (see Fig. 3).

These simulations reveal that the accuracy of the retrieved CO₂ column is highly dependent on the inclusion of the temperature weighting function within the WFM-DOAS fit. If the temperature derivative is included, then in the case of the regional profiles

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(Table 1) the errors are less than 1% even when there is coincidental uniform scaling of the CO₂ profile. When more realistic ECMWF profiles are used to generate the synthetic measurements, these errors are larger typically <1.5%, without any additional scaling of the CO₂ concentration (Table 2). Using a climatological mean profile to produce the baseline spectrum is hence inadequate, suggesting that a priori knowledge of the temperature and water vapour profiles is needed when applying WFM-DOAS to real SCIAMACHY spectra. Even if the reference spectra are created from assimilated atmospheric profiles, the temperature derivative must be always included within the retrieval to limit this source of error. This is also clearly demonstrated by examining the retrieval around the temperature linearization point. To accomplish this, a third set of synthetic measurements was created by applying small temperature shifts of 1 K to the US Standard atmosphere (up to a maximum of ±10 K). The subsequent retrievals reveal that not fitting the temperature derivative can produce a relative error in the CO₂ column of as much as ±1.3% for a uniform ±5 K difference between the true and reference temperature profiles (Fig. 4). If the temperature weighting function is fitted, this error is kept to less than 0.1%.

3.4 Sensitivity to CO₂ profile and atmospheric profile

Sections 3.2 and 3.3 have shown that the sufficient knowledge CO₂, temperature and water vapour profiles is necessary to produce accurate retrievals. It is possible to investigate the combined effect of using alternative CO₂ and atmospheric profiles by interpolating the CO₂ profiles, from the new climatology (Sect. 3.2), onto the appropriate tropical, mid-latitude and sub-arctic regional profiles. The consequent simulations show that relative errors in the retrieved column in the order 0.5% can be produced (Fig. 5). Unlike the analysis in Sect. 3.2, the temperature derivative has a much bigger effect. Whilst in the tropics these errors are almost constant throughout the year, at mid and high latitudes they vary seasonally. During the winter months the mid-latitude columns are systematically overestimated by about 0.75% as compared to the summer where they are underestimated by an average of 0.13%. These biases will increase if

the CO₂ profiles are interpolated onto the more lifelike ECMWF profiles. Retrievals performed without using realistic CO₂ profiles *and* realistic temperature and water-vapour profiles and will therefore result in the terrestrial biosphere fluxes in the Northern mid-latitudes being overestimated. Since the natural variability of atmospheric CO₂ total columns is only about 3% these errors are not negligible.

3.5 Sensitivity to the surface elevation and pressure

The surface pressure must be carefully considered in any CO₂ retrieval algorithm. Firstly, because gradients in the atmospheric CO₂ concentration created by variations in the surface topography and low altitude weather systems can often mask those produced by surface fluxes (Kawa et al., 2004) and secondly because the measurement is sensitive to pressure, owing to the broadening of absorption lines (Frankenberg et al., 2005c). The error in the retrieved column as a function of the surface pressure was investigated by varying the surface elevation within the SCIATRAN model. Inspection of the results (Fig. 6) shows that the retrieved column is systematically under-estimated when the surface elevation is increased relative to that used in the reference scenario. The magnitude of this error is approximately -0.31% per 50 m increase in surface elevation, when the temperature weighting function is not fitted, as opposed to approximately -0.34% per 50 m when it is included.

3.6 Sensitivity to the surface albedo

The Earth's surface reflectance within the near infra-red can vary from as little as 0.001 over the oceans to as much 0.4 over the desert (Dufour and Bréon, 2003). This affects not only the overall amount of solar radiation reflected back to space (and subsequently the signal to noise ratio of SCIAMACHY measurements) but also the relative depth of CO₂ and H₂O absorption lines by influencing the path-length traveled by the photons (Buchwitz and Burrows, 2004). To gauge this effect, the column error as a function of the surface reflectance was examined using a reference spectrum produced with

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an albedo of 0.1 (so as to be consistent with current implementations of WFM-DOAS (Buchwitz et al., 2005). Variations in the surface albedo alone, from 0.05 to 0.4, can cause an error in the retrieved column of up to 2% (Fig. 7). The errors increase further still at very low albedos. Employing a fixed albedo within the algorithm, will thus introduce what are essentially unnecessary errors.

3.7 Sensitivity to aerosols

Retrievals of CO₂ in the near infra-red are affected by the presence of aerosols (Dufour and Bréon, 2003). For example, systematic errors caused by Saharan dust, have already been clearly identified in CO₂ columns retrieved from SCIAMACHY (Houweling et al., 2005). Aerosols can both scatter and absorb light depending on their composition. Aerosol scattering mainly shortens the photon path-length leading to an underestimation of the CO₂ column. To determine their influence on WFM-DOAS, a set of scenarios were created based on the aerosol types defined by Hess et al. (1998) using a parameterization developed by Hoogen (1995). Aerosol extinction profiles were taken (and adapted) from the LOWTRAN library (Kneizys et al., 1996). These aerosols were confined to the boundary layer with background tropospheric and stratospheric conditions assumed. Retrievals simulated using the default scenario show that the errors are lower than 2%, except in extreme cases, and that they increase if the temperature derivative is included within the fit (Table 3). The background marine conditions associated with the reference spectrum however produces biases when applied to contrasting conditions, for example with urban, biomass burning and desert scenarios. This may have repercussions for trying to estimate CO₂ emissions over large urban areas or regions affected by wild fires. However, the inclusion of some background aerosols within the reference spectrum is clearly better than retrievals performed on the basis of an aerosol free atmosphere.

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3.8 Fitting of height resolved weighting functions

The non-linearity of DOAS fitting within the near infra-red spectral region, owing to strong absorbers and the effects of pressure broadening, was clearly identified by Frankenberg et al. (2005c). To account for this difficulty, scaling factors for different height layers were used within the IMAP retrieval algorithms state vector. To determine whether the sensitivity of WFM-DOAS could be improved by using this approach, the trace gas columns and associated weighting functions of the reference spectrum, were grouped into three individual height layers: 0–3 km, 3–12 km and 12 km to top of the atmosphere (each column derivative being simply the sum of a set of height resolved weighting functions). The simulated retrievals were then repeated and the resulting errors compared to those obtained using the original column derivatives. In contrast to the findings of Frankenberg et al. (2005c) there was little evidence of increased sensitivity from using this method.

4 Full Spectral Initiation (FSI) WFM-DOAS

The WFM-DOAS algorithm is able to detect changes in the atmospheric CO₂ concentration. However if the CO₂ column is retrieved using a reference spectrum created from a scenario that differs from the true atmospheric state, significant errors could be introduced. As demonstrated, factors which affect the calculated model spectrum are the surface albedo, surface pressure, aerosols and the vertical profiles of CO₂, water vapour and temperature. When varied alone each of these parameters creates an error in the column of typically less than about 2%. However, in the real atmosphere none are known exactly, thus the errors propagate accordingly and are especially significant when considering the high precision measurements that are needed to observe the small gradients associated with atmospheric CO₂. To reduce these errors a reference spectrum that is very close to the real measurement must be used, i.e. as much “a priori” information must be included in the retrieval as possible. It is on this basis that

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5 a new retrieval algorithm called Full Spectral Initiation or (FSI)-WFM-DOAS has been developed. In FSI, a reference spectrum is generated for every single SCIAMACHY measurement to obtain the best linearization point for the retrieval. Subsequent iterations are not performed, in order to limit the computational time. Were this not an issue, then the retrieved parameters would be fed back into the algorithm iteratively. To calculate the best possible reference spectrum all the known or rather estimated properties of the atmosphere and surface at the time of the observation, serve as input for the radiative transfer model. Once the sun-normalised radiance is obtained, a WFM-DOAS fit is then performed and the vertical column retrieved.

10 The FSI algorithm is applied to calibrated radiances using the fitting window 1561.03–1585.39 nm. This region was chosen in order to minimize interference from water vapour and not to extend into channel 6+, where the Indium Gallium Arsenide (InGaAs) detectors were doped with higher amounts of Indium leading to different behavioural characteristics (Lichtenberg et al., 2005). The number of fitting points within this micro-window is usually thirty-two, with each detector pixel spanning a wavelength interval of 0.7 nm. At this resolution, SCIAMACHY is close to under-sampling the CO₂ band. This spectral interval is also advantageous in that, unlike channels 7 and 8, it is unaffected by orbital variations of the dark current and the build up of ice-layers on the detectors (as discussed by Gloudemans et al., 2005). It is also stable with regard to so called “dead and bad pixels” created by thermal and radiation degradation (Kleipool, 2004). The SCIAMACHY pixel mask, however is checked and if necessary, updated for each orbit using the standard deviations of the dark current, as proposed by Frankenberg et al. (2005b). Detector pixels are also discarded if erroneous spikes occur in the measured radiance. All measurements have been corrected for non-linear effects (Kleipool, 2003b) and the dark current (Kleipool, 2003a). The FSI algorithm also uses a solar reference spectrum with improved calibration, provided to the SCIAMACHY community by ESA, courtesy of Johannes Frerick (ESA, ESTEC), in preference to that in the official L1C product. An optimised systematic shift, based on the inspection of the fit residuals, of 0.15 nm is applied to the observed spectra to align it to the synthetic

radiances calculated by SCIATRAN. To improve the quality of the FSI spectral fits, the latest version of the HITRAN molecular spectroscopic database has been implemented in the radiative transfer model (Rothman et al., 2005).

Each SCIAMACHY observation has an associated reference spectrum created from several sources of atmospheric and surface data. The CO₂ profile is selected from the new climatology (Sect. 3.2) and is chosen according to the time of the observation and the latitude band in which the ground pixel falls. To obtain the closest atmospheric state to the observation temperature, pressure and water vapour profiles, derived from operational 6 hourly ECMWF data (1.125° × 1.125° grid), are interpolated onto the local overpass time and centre of the of the SCIAMACHY pixel. To determine the linearization point for the surface albedo, a look-up table of mean radiances, for the defined fitting window, has been generated as a function of both the surface reflectance and solar zenith angle. From using the mean radiance of the SCIAMACHY observation and the solar zenith angle at the corresponding time, it is possible to infer an approximate value for the albedo. To account for aerosols, three scenarios are incorporated into the retrieval algorithm. Maritime and rural scenarios are implemented over the oceans and land respectively. In both cases, the boundary layer visibility is 23 km. If a major city occurs within the SCIAMACHY footprint then an urban scenario is selected with a reduced visibility of 5 km. For all three cases the LOWTRAN aerosol model is employed, with the relative humidity set to 80% in summer and 70% in winter. Background conditions are assumed for the free troposphere, stratosphere and mesosphere.

The viewing geometry of SCIAMACHY allows it to scan to ±30° around the nadir line of sight. Unfortunately every model spectrum must be calculated for the exact nadir position, as the computational time for off axis spectra is very high. To account for the increased path-length in off-axis viewing geometry, a correction has been applied to the final retrieved column (Buchwitz et al., 2000). This is not an ideal solution, as it can cause an over correction of up to 2–3% for pixels at the extremities (Buchwitz and Burrows, 2004). All SCIAMACHY observations are cloud screened prior to retrieval processing, with cloud contaminated pixels flagged and disregarded using the current

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version of the Heidelberg Iterative Cloud Retrieval Utilities (HICRU) database (Grzegorski et al., 2005). Where gaps occur in this database or if the HICRU algorithm fails then an alternative method is employed which uses the polarization measurement devices, specifically PMD1, flagging a pixel if a pre-defined threshold (set at 70 000 BU) is exceeded (de Beek et al., 2004). Despite this, it is evident that cloudy pixels are still processed. Such columns are easily noticeable by their extremely low mixing ratios and low correlation with the surface elevation. These are subsequently filtered out after processing. Back-scans along with observations that have solar zenith angles greater than 75° are also excluded. All columns are normalized to the ECMWF surface pressure.

5 Preliminary results

The calculation of a reference spectrum for each SCIAMACHY observation, whilst felt necessary, is still time-consuming at approximately 5 min per ground pixel. This means that, at this stage, FSI-WFM-DOAS can only be applied to selected “target regions” that are of particular scientific interest. In this first demonstration of the FSI algorithm, Siberia has been chosen as an example test region, specifically the area bounded by the latitudes 48° N–80° N and the longitudes of 45° E–180° E, with just the summer months processed (excluding August during which SCIAMACHY underwent a decontamination phase to remove ice on the NIR detectors). To ensure only valid retrievals are examined, only columns that have retrieval errors less than 5% and lie within a quite broad range of 340–400 ppmv are accepted. These results are summarized in Table 4. In this initial analysis, comparisons to chemical transport models, for example the TM3 model (Heimann and Körner, 2003) or ground station data are not made, as this will be the goal of future research. Instead the focus is on the quality of the spectral fits and possible errors and biases associated with the FSI WFM-DOAS retrieval. Hence, in this study neither the retrieved columns (in molecules per cm²) or the normalized columns (in ppmv), have had scaling factors applied as in other studies (e.g.

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Yang et al., 2002 or Buchwitz et al., 2005), thus only the CO₂ spatial distribution can really be discussed.

The spectral fits produced by the FSI algorithm are promising, with a typical fit shown in Fig. 8. The CO₂ fit indicates that there is good sensitivity to the CO₂ vertical column unlike temperature, where the fit is much worse. If the retrievals are repeated but this time only fitting the CO₂ and H₂O weighting functions, then the that magnitude of the vertical columns increases but their relative spatial distribution remains the same. Fitting the temperature derivative therefore only creates a negative offset, suggesting that the current algorithm is more sensitive to other factors, possibly calibration issues. Nevertheless, it is always included, following the sensitivity study presented in Sect. 3. By averaging over all retrievals, the mean fit residual should ideally reflect only measurement noise. However, this is not the case, illustrated by Fig. 9, as it contains quite, stable systematic structures, the origin of which has yet to be clarified. It is most likely that such features will be reduced (or removed) by improving the calibration of the SCIAMACHY spectra and through continually including the latest spectroscopic data in the radiative transfer model. The residuals are also strongly influenced by the slit function used in the convolution of the reference spectra and weighting functions. From close examination of the fit residuals, a Gaussian slit function of width 1.4 nm has been implemented as this yields the lowest retrieval errors and best fits. The root-mean-square (RMS) error, over the time-period considered, is extremely consistent at about 0.2% showing that the FSI algorithm is very stable (Table 4).

Although the algorithm uses several sources of a priori information it is important to ensure that the retrieved columns are not biased from the input data. This is especially relevant with regards to the a priori CO₂ vertical columns, the surface albedo and surface pressure. The a priori and retrieved CO₂ vertical columns for June (Fig. 10) demonstrate that whilst the spatial distribution of input columns is moderately smooth (essentially following the surface topography) the measured columns exhibit much more variation. The correlation between the two sets of columns, though relatively high at 0.82, is acceptable considering the low variability in the total column (Fig. 11).

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For the surface albedo no such correlation is found (Fig. 12), although the retrievals errors increase when the surface reflectance becomes progressively lower (i.e. as SCIAMACHY's signal to noise ratio decreases). Concerning the input ECMWF surface pressure, the correlation with the retrieved vertical column densities is again, quite high at 0.82 but is to be somewhat expected with CO₂ being well mixed in the atmosphere. If comparisons are made to the surface elevation (Fig. 13) however, which is on a finer resolution grid, then a slight bias is noticeable in that a significant fraction of the columns lie below the 3% (and indeed 5%) variability associated with the US Standard atmosphere. This in itself, is not indicative that retrieval algorithm produces this bias as it is possible that a large number of these low column densities may still be partially cloud contaminated. Alternatively, these low values maybe a calibration related effect but whatever the origin this systematic underestimation requires further examination. Checks for trends and biases in the retrieved CO₂ columns related to the longitudinal and latitudinal distribution of the ground pixels; the line of sight and solar zenith angles and the a priori and retrieved water vapour columns were also performed. In all circumstances no correlation was found. This analysis was also repeated for the retrieval error associated with CO₂ columns with again, no biases detected.

Having considered the quality of the FSI retrieval algorithm, its results are shown in Fig. 14 with all normalized columns averaged on to a 1°×1° grid. The most noticeable feature of these monthly fields is the fairly high variability of the CO₂ concentration, with the 1σ deviation about each monthly mean being ~10 ppmv. Whilst some of this variability may be realistic it is probable that some of the deviations maybe removed by normalizing the retrieved CO₂ columns with oxygen (O₂) columns, instead of the surface pressure. By taking the ratio with oxygen, which can also be retrieved from SCIAMACHY, the effects of aerosols and clouds on the light path may cancel out. This so-called dry mixing ratio denoted XCO₂ (=CO₂/O₂×0.295) is used to reduce systematic errors in satellite retrievals and is already implemented by Buchwitz et al. (2005) and proposed for the future Orbiting Carbon Observatory (OCO) mission (Crisp et al., 2004). This notwithstanding, distinct evolving patterns within the CO₂ distribution are

visible, for example there is an enhanced region over the Yablonovyy mountain range (approximately 115° E 49° N) in May and what seems to be a persistent dip in the CO₂ concentration over the West Siberian Plain in the vicinity of 75° E 60° N. Similarly the seasonal uptake of CO₂ by the biosphere appears to be detectable with a decline in the concentration from May to July before increasing again by September. Whether these features are created by CO₂ surface fluxes is yet unproven and warrants further investigation. Nevertheless it is encouraging that some structure within the CO₂ fields is visible and may possibly provide information on important carbon processes occurring within the Siberian region. As more SCIAMACHY data is processed the identification of CO₂ surface fluxes will be the goal of future research efforts.

6 Conclusions

The ability of the WFM-DOAS retrieval algorithm to measure total columns of atmospheric CO₂ has been closely examined. A detailed sensitivity analysis has been performed that has identified several non-negligible error sources and time dependent biases which affect the accuracy of the retrieved CO₂ columns. These inaccuracies originate from using a reference spectrum which is created from a differing atmospheric state to that of the measurement. To minimize these errors a new CO₂ retrieval algorithm called Full Spectral Initiation (FSI) WFM-DOAS has been developed, the foundation of which is the use of a priori data to obtain the best possible linearization point for the retrieval. The FSI algorithm has then been applied to SCIAMACHY observations with both the quality of the spectral fits and possible biases assessed. Although some of the CO₂ columns appear to be under-estimated there isn't any evidence of a priori influence in the retrieved data. Preliminary results for the summer months over Siberia have shown that the retrieved monthly fields exhibit a significant amount of variability but that some structure in the CO₂ concentration is visible.

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Table 1. Relative errors produced in the retrieved CO₂ column created from using alternative temperature and water vapour profiles. Scaling factors of 0.95, 0.97, 1.0, 1.03 and 1.05 applied to the CO₂ column yield mixing ratios of 351.5 ppmv, 358.9 ppmv, 370 ppmv, 381.1 ppmv and 388.5 ppmv respectively. No TWF: temperature weighting function not fitted. TWF: temperature weighting function fitted.

Climatology	Water vapour Scaling	Relative column error (%) CO ₂ Column scaling									
		0.95		0.97		1.00		1.03		1.05	
		No TWF	TWF	No TWF	TWF	No TWF	TWF	No TWF	TWF	No TWF	TWF
Tropical	0.5	-2.72	-0.29	-2.68	-0.26	-2.66	-0.25	-2.68	-0.28	-2.71	-0.32
	1.0	-2.78	-0.32	-2.73	-0.29	-2.70	-0.28	-2.71	-0.30	-2.74	-0.34
	1.5	-3.03	-0.46	-2.98	-0.42	-2.94	-0.40	-2.94	-0.42	-2.96	-0.45
Mid Lat Summer	0.5	-3.17	-0.82	-3.00	-0.67	-2.79	-0.49	-2.64	-0.37	-2.56	-0.32
	1.0	-3.20	-0.84	-3.02	-0.69	-2.81	-0.51	-2.65	-0.39	-2.57	-0.33
	1.5	-3.35	-0.94	-3.17	-0.78	-2.94	-0.60	-2.78	-0.47	-2.70	-0.40
Mid Lat Winter	0.5	1.69	-0.04	1.86	0.12	2.06	0.31	2.20	0.45	2.26	0.52
	1.0	1.68	-0.05	1.85	0.11	2.05	0.31	2.19	0.45	2.26	0.51
	1.5	1.66	-0.06	1.83	0.11	2.03	0.30	2.17	0.44	2.24	0.51
Sub Arc Summer	0.5	-1.57	-0.54	-1.40	-0.39	-1.18	-0.20	-1.03	-0.07	-0.95	-0.01
	1.0	-0.02	-0.57	-1.42	-0.41	-1.20	-0.22	-1.04	-0.08	-0.96	-0.02
	1.5	-1.70	-0.63	-1.51	-0.47	-1.29	-0.28	-1.12	-0.13	-1.04	-0.07
Sub Arc Winter	0.5	3.74	-0.31	3.91	-0.13	4.11	0.08	4.26	0.23	4.33	0.32
	1.0	3.73	-0.31	3.90	-0.13	4.11	0.08	4.25	0.24	4.32	0.32
	1.5	3.72	-0.31	3.89	-0.13	4.10	0.08	4.26	0.24	4.32	0.32

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Table 2. The error produced in the retrieved CO₂ column brought about from using ECMWF temperature and water vapour profiles instead of the U.S. Standard atmosphere. Each ECMWF profile number corresponds to the indexing in Fig. 3.

ECMWF profile	Relative column error %	
	No TW	TW
1	1.04	-0.70
2	1.01	-1.50
3	3.31	0.90
4	3.86	1.17
5	4.87	-0.89
6	5.26	-0.48
7	-3.99	-1.24
8	2.35	0.94
9	-4.04	-1.29
10	-4.00	-1.31
11	-3.96	-1.24
12	-2.57	-0.39
13	-2.43	-0.32
14	-2.66	-0.41
15	-2.72	-0.24
16	-2.74	-0.25

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Table 3. Aerosol error analysis. Inc. Aerosols = Simulated retrievals performed using a reference spectrum with the default aerosol scenario. Exc. Aerosols = Simulated retrievals performed using a reference spectrum without any aerosols present. The component contributions are defined as: waso = watersoluble, inso = insoluble, ssam = sea salt accumulation mode, sscm = sea salt coarse mode, mnuc = mineral nucleation mode, macc = mineral accumulation mode. The biomass burning scenarios were taken from [Holzer-Popp et al. \(2002\)](#). The extinction coefficients within the planetary boundary layer (0–2 km) for the urban smog and biomass burning I & II scenarios were 19.49 km^{-1} , 1.50 km^{-1} and 4.00 km^{-1} .

Aerosol Scenario	Relative Column Error %				Comments
	Inc. aerosols		Exc. aerosols		
	No TWF	TWF	No TWF	TWF	
Continental Clean Summer	1.78	1.88	3.51	3.69	Remote Continental Areas (waso: 99.998%; inso: 0.002%)
Continental Clean Winter	0.98	1.04	2.70	2.85	
Continental Average Summer	1.04	1.11	2.76	2.92	Anthropogenically influenced continental areas (waso: 45.79%; inso: 0.01%; soot: 54.20%)
Continental Average Winter	0.22	0.26	1.94	2.06	
Urban Polluted Summer	−0.44	−0.45	1.27	1.35	Polluted urban areas (waso: 31.39%; inso: 0.01%; soot: 68.6%)
Urban Polluted Winter	−4.07	−4.17	−2.39	−2.41	
Urban Smog Summer	−32.42	−32.84	−31.13	−31.04	Highly polluted urban areas (waso: 31.39%; inso: 0.01%; soot: 68.6%)
Urban Smog Winter	−32.49	−32.80	−31.03	−31.34	
Desert Summer	1.63	1.72	3.36	3.36	Desert regions (e.g. Sahara, Gobi) (mnuc: 93.19%; macc: 6.81%)
Desert Winter	1.27	1.35	3.00	3.15	
Clean Maritime Summer	1.47	1.54	3.20	3.35	Remote marine conditions (waso: 98.68%; ssam: 1.32%)
Clean Maritime Winter	1.05	1.12	2.78	2.92	
Polluted Maritime Summer	1.38	1.45	3.10	3.26	Maritime region under anthropogenic influence (waso: 39.4%; ssac: 0.02%; sscm: 0.03%; soot: 60.4%)
Polluted Maritime Winter	0.65	0.70	2.38	2.51	
Biomass Burning Summer I	−4.06	−4.15	−2.38	3.35	Regions affected by wild fires (waso: 82.00%; inso: 0.01%; soot: 16.60%)
Biomass Burning Winter I	−4.05	−4.14	−2.37	−2.38	
Biomass Burning Summer II	−15.82	−16.30	−14.23	−14.23	Regions affected by heavy biomass burning (waso: 54.00%; inso: 2.00%; soot: 44.00%)
Biomass Burning Winter II	−16.09	−16.56	−14.49	−14.92	

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Table 4. Summary of FSI WFM-DOAS retrieval results for the Siberian test region (see Fig. 14) during the summer of 2003.

Month	Number of pixels	Mean CO ₂ VMR [ppmv]	1 σ VMR [ppmv]	Mean retrieval Error [%]	Mean RMS Error [%]
May	13 612	371.24	11.34	2.52	0.22
June	15 888	365.12	10.10	2.46	0.23
July	21 399	362.15	9.72	2.46	0.24
Sep	24 022	365.15	10.56	2.41	0.27

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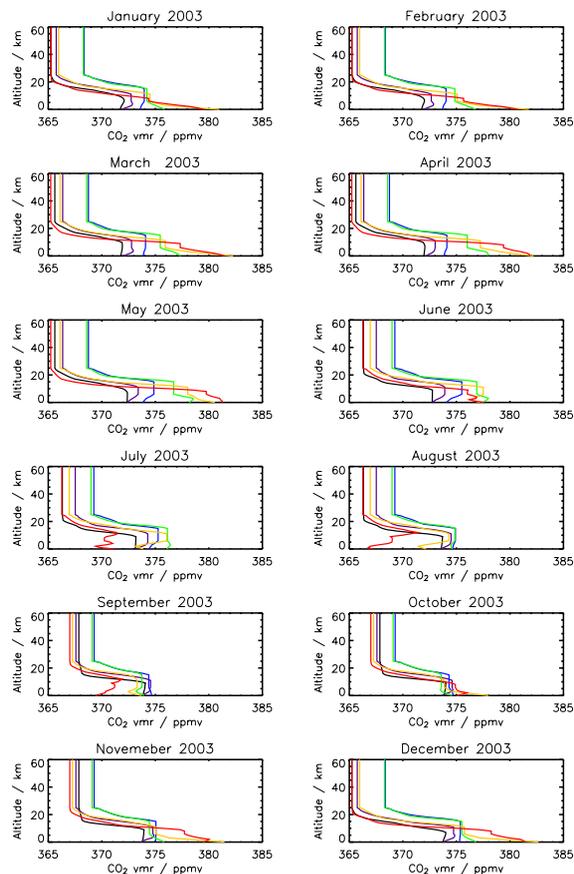


Fig. 1. Vertical CO₂ profiles used in the sensitivity analysis. This data set, for 2003, contains a total of 72 CO₂ profiles, constructed from the flask measurements made by the GLOBALVIEW network. Each month has six profiles centered on the latitudes 75° N (red), 45° N (yellow), 10° N (green), 10° S (blue), 45° S (purple) and 75° S (black), each representative of a 30° latitude band.

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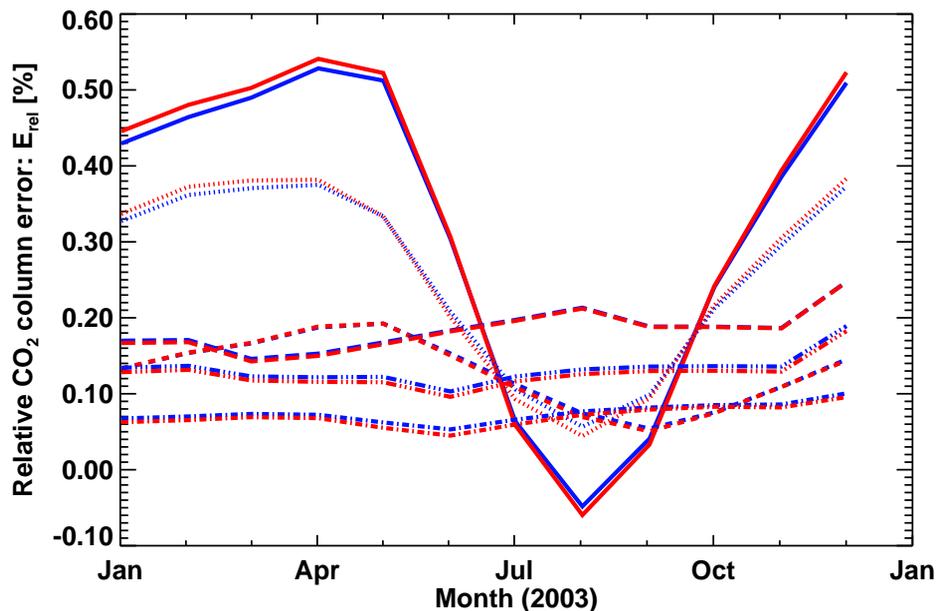


Fig. 2. The relative errors in the retrieved CO₂ column for 75° N (solid), 45° N (dotted), 10° N (dashed), 10° S (dash dot), 45° S (dash dot dot) and 75° S (long dashes) produced by using CO₂ profiles other than the US Standard atmosphere (associated with the reference spectrum), with the simulated retrievals performed both with (red) and without (blue) the temperature weighting function.

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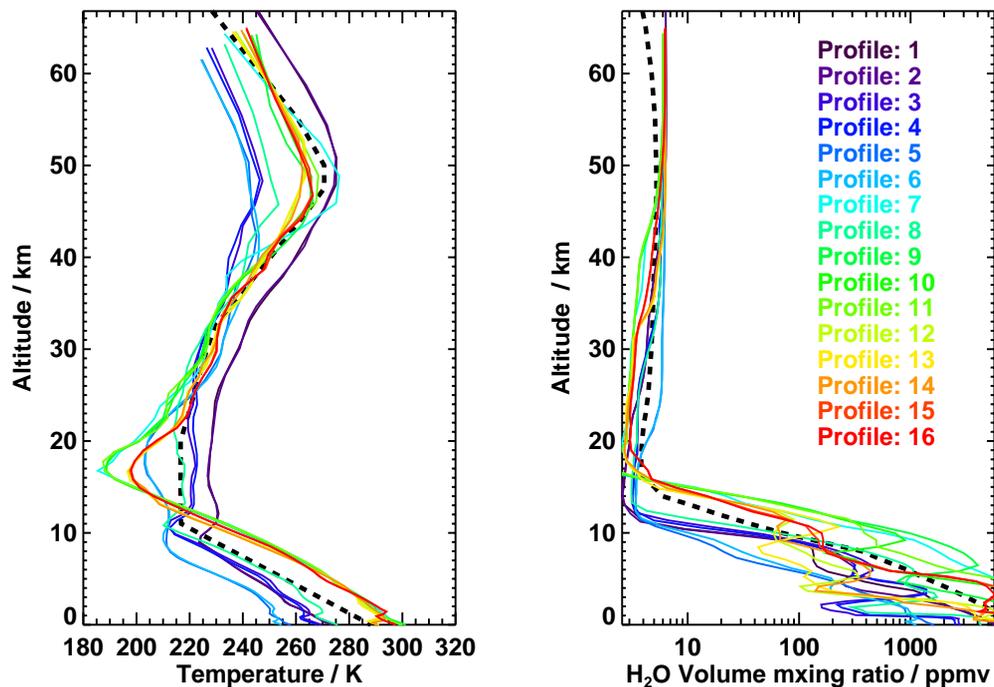


Fig. 3. The ECMWF temperature profiles (left) and corresponding water vapour profiles (right) used in the sensitivity analysis, together with the US standard atmosphere (black dashed line).

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FSI WFM-DOAS

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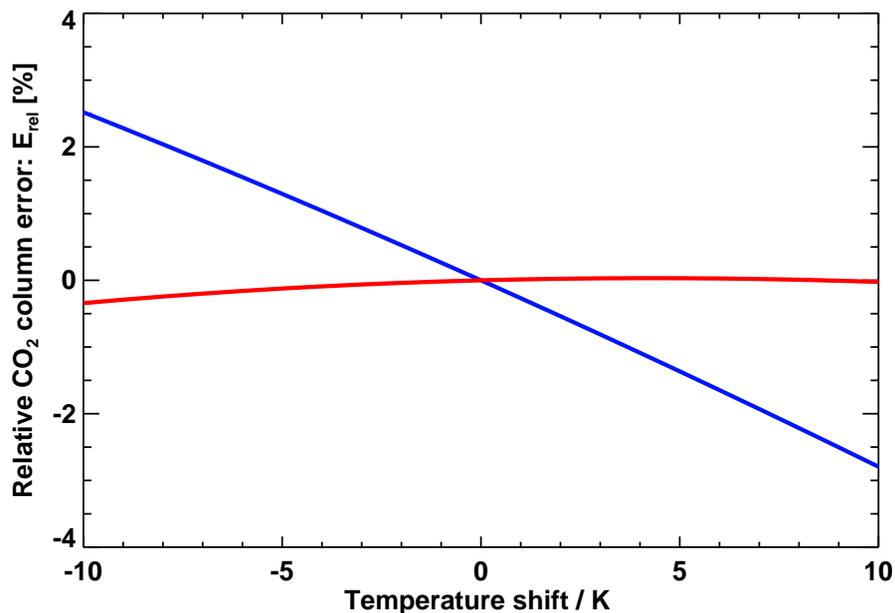


Fig. 4. The sensitivity of the WFM-DOAS algorithm to the temperature linearization point, shown here as a temperature shift of the US Standard Atmosphere, for simulations performed both with/without (red/blue) the temperature weighting function.

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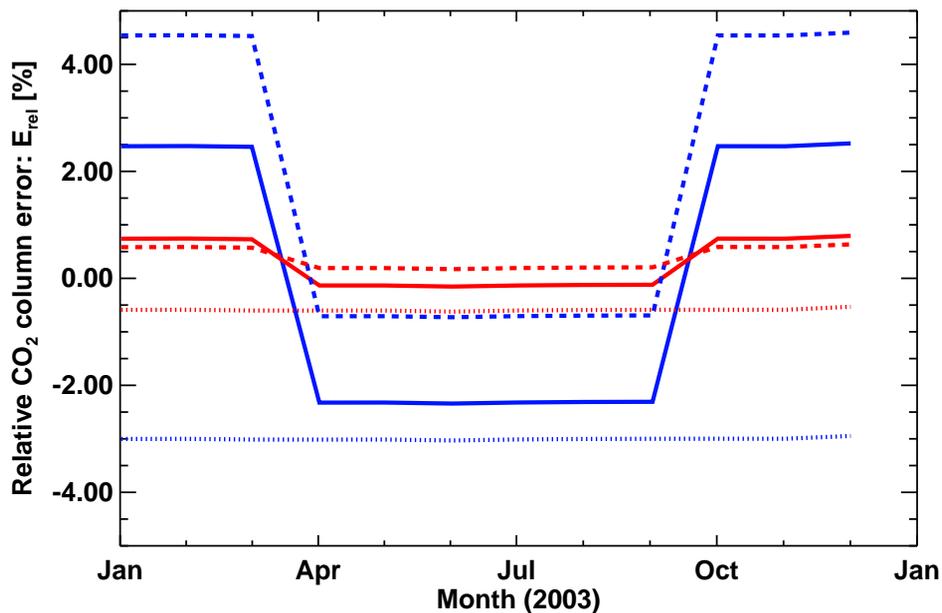


Fig. 5. The relative column error produced by using realistic monthly CO₂ profiles and temperature and water vapour profiles representative of the Northern Hemisphere tropics (dotted), mid-latitudes (solid) and sub-arctic (dashed), for simulated retrievals performed both with/without (red/blue) the temperature weighting function.

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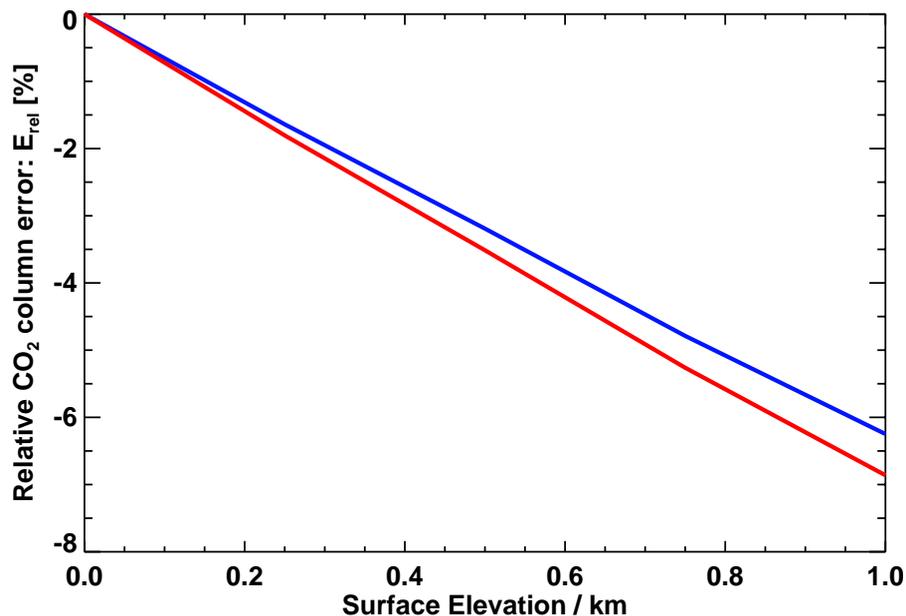


Fig. 6. The relative column error produced as a function of the surface elevation, for simulations performed both with/without (red/blue) the temperature weighting function.. Here the retrieved CO₂ column is systematically under-estimated.

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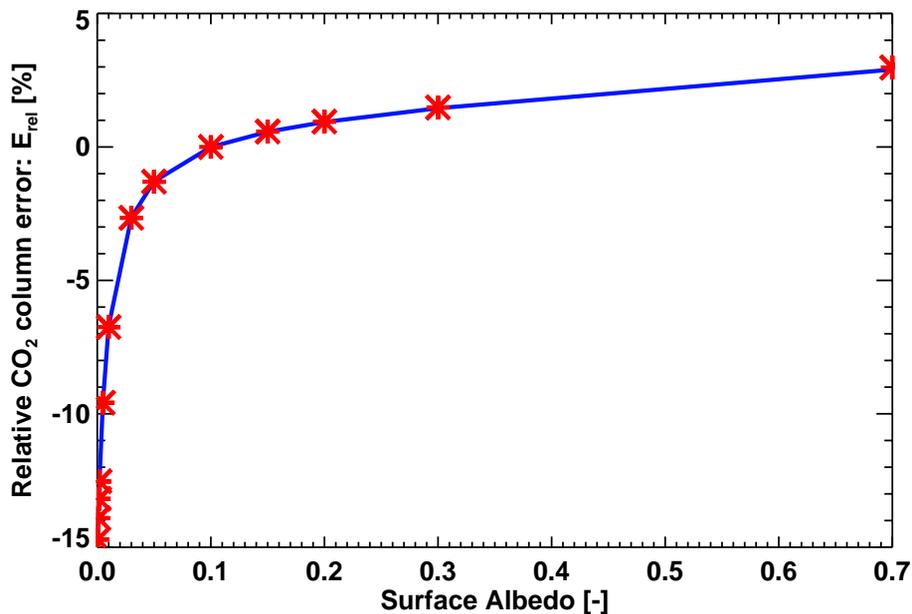


Fig. 7. The relative column error produced as a function of the surface albedo. In this instance the errors produced by fitting (red) and not fitting (blue) the temperature weighting function are barely discernible.

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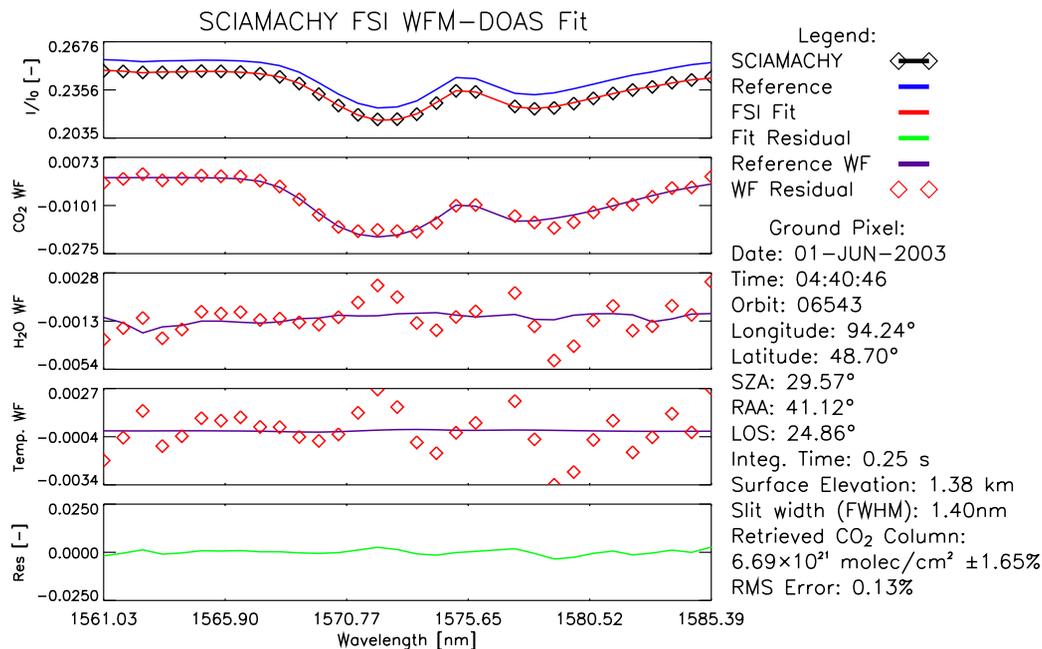


Fig. 8. A typical example of a FSI WFM-DOAS fit. Top panel: The reference spectrum (blue) generated by the retrieval (from the a priori data), the sun-normalized radiance measured by SCIAMACHY (black diamonds) and the FSI WFM-DOAS fit (red) to this measurement. Following three panels: **(i)** The CO₂ fit. This shows CO₂ total column weighting function (WF) scaled by $(\bar{V}_{\text{CO}_2} - \bar{V}_{\text{CO}_2})$ (purple) and the CO₂ fit residuum (red diamonds), which is the scaled CO₂ WF plus the difference between the measurement and fit (i.e. the fit residual) **(ii–iii)** Similar but for H₂O and temperature. Bottom panel: The fit residual with a root-mean-square (RMS) difference of 0.13%.

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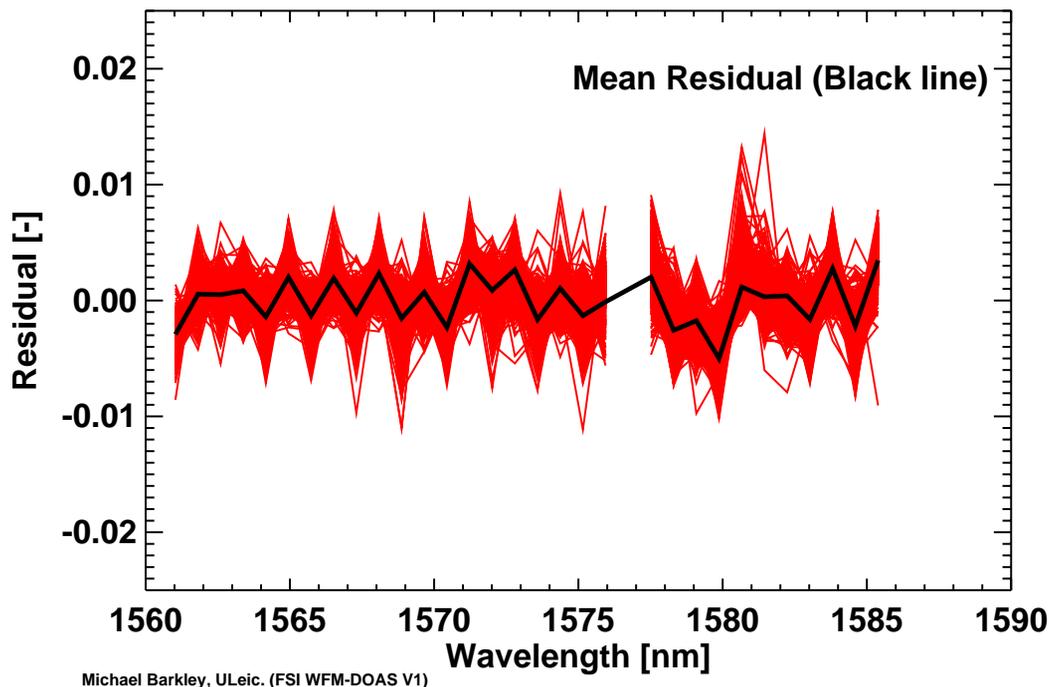


Fig. 9. FSI WFM-DOAS fit residuals for the Siberian test region during May 2003, over-plotted with the mean fit residual (black). The mean RMS error is 0.22%. The detector pixel, of wavelength 1576.72 nm, is omitted in the retrieval as this always worsens the quality of the fit and increases the error on the retrieved column.

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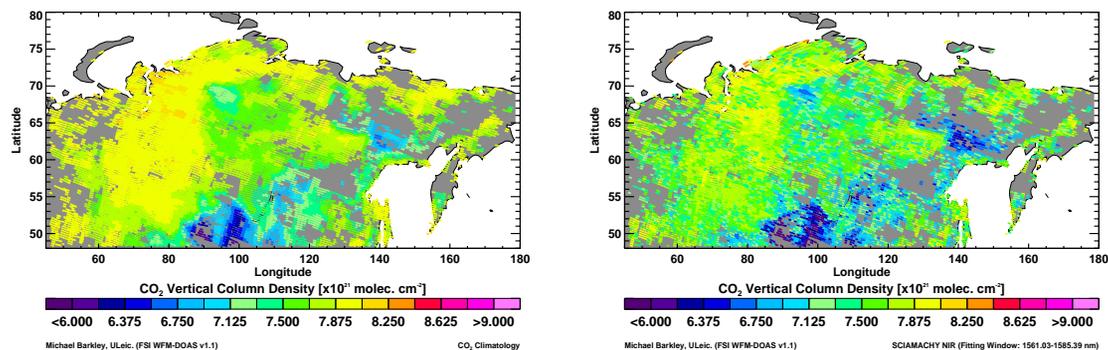


Fig. 10. Left panel: a priori CO₂ columns used in the retrieval for June, 2003, constructed from the CO₂ climatology. Right panel: The FSI WFM-DOAS retrieved CO₂ columns for the same month.

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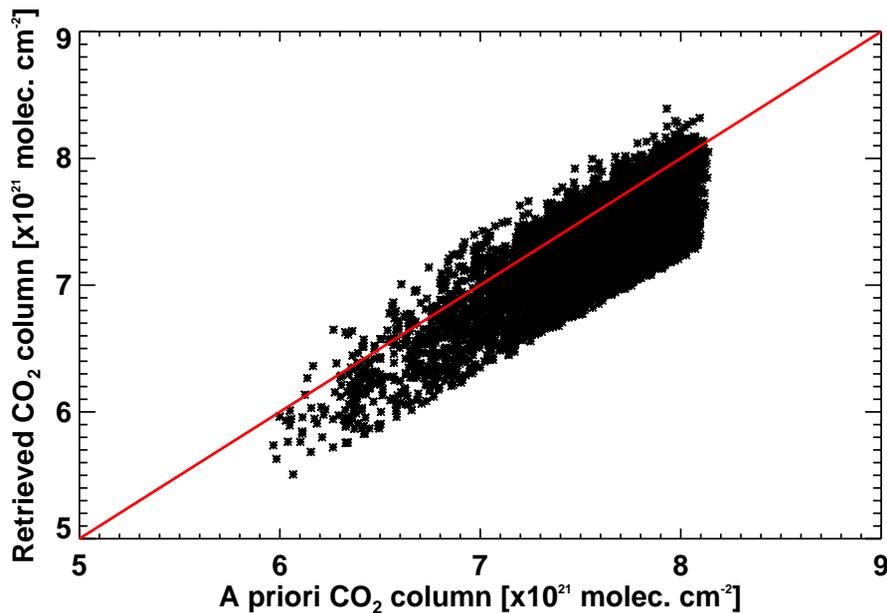


Fig. 11. The retrieved CO₂ columns as a function of the a priori CO₂ columns (shown in Fig. 10) for June 2003. The red line indicates a one-to-one relationship. The linear correlation coefficient between the data is 0.82.

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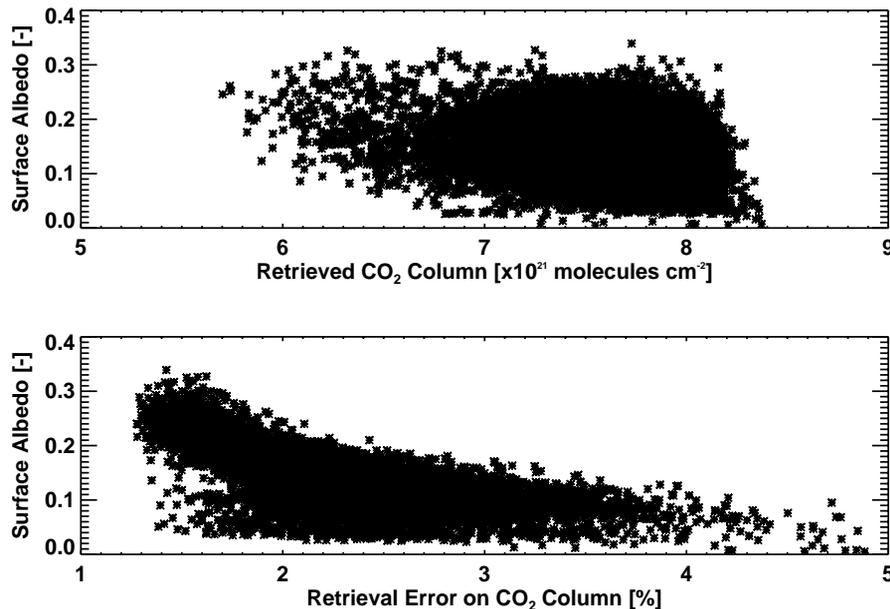


Fig. 12. Top panel: The retrieved CO₂ columns, shown here for the month of May, haven't any dependency on the a priori surface albedo. Bottom panel: The retrieval errors clearly increase when the a priori surface reflectance is small.

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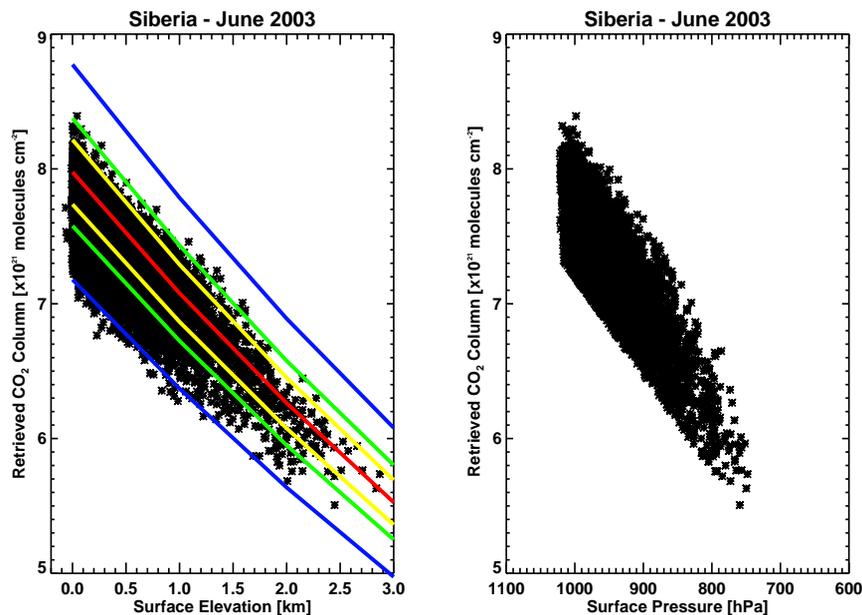


Fig. 13. Left: Comparison between the surface elevation and the retrieved CO₂ columns. The surface elevation is taken from the GLOBE Digital Elevation Model (DEM) (<http://www.ngdc.noaa.gov/mgg/topo/globe.html>) and averaged to a 0.25° × 0.25° grid. The linear correlation coefficient is -0.815 . Over-plotted are the variations, of the vertical column density with surface elevation, for the US Standard atmosphere (red), the US Standard atmosphere $\pm 3\%$ variability in the total column (yellow), the US Standard atmosphere $\pm 5\%$ variability (green) and the US Standard atmosphere $\pm 10\%$ variability (blue). Right: The variation of the retrieved CO₂ column with the a priori ECMWF surface pressure, which has a correlation coefficient of 0.82.

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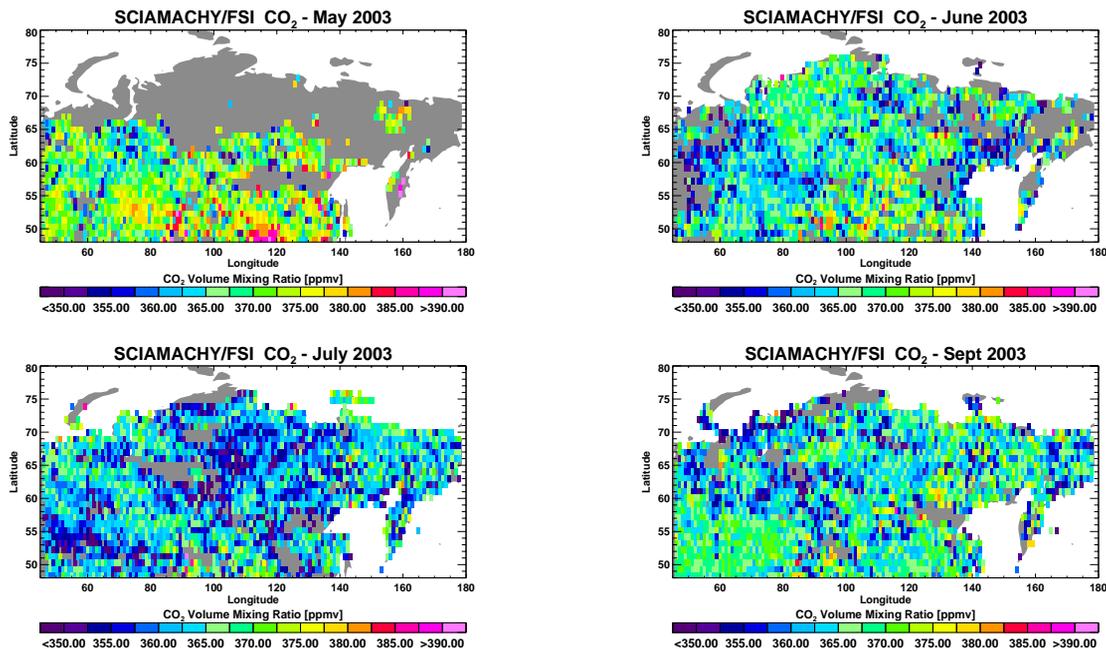


Fig. 14. Normalized CO₂ columns, retrieved over the Siberian test region for the summer of 2003, averaged on to a 1° × 1° grid.

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