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**Framework for
comparing remotely
sensed and in-situ
CO₂ VMRs**

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A framework for comparing remotely sensed and in-situ CO₂ concentrations

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Abstract

A framework that allows validating CO₂ column averaged volume mixing ratios (VMRs) retrieved from ground-based solar absorption measurements using Fourier transform infrared spectrometry (FTS) against measurements made in-situ (such as from aircrafts and tall towers) has been developed. Since in-situ measurements are done frequently and at high accuracy on the global calibration scale, linking this scale with FTS total column retrievals ultimately provides a calibration scale for remote sensing. FTS, tower and aircraft data were analyzed from measurements during the CarboEurope Regional Experiment Strategy (CERES) from May to June 2005 in Biscarrosse, France. Carbon dioxide VMRs from the MetAir Dimona aircraft, the TM3 global transport model and Observations of the Middle Stratosphere (OMS) balloon based experiments were combined and integrated to compare with FTS measurements. The comparison agrees fairly well with differences resulting from the spatial variability of CO₂ around the FTS as measured by the aircraft. Additionally, the Stochastic Time Inverted Lagrangian Transport (STILT) model served as a “transfer standard” between the in-situ data measured at a co-located tower and the remotely sensed data from the FTS. The variability of carbon dioxide VMRs was modeled well by STILT with differences coming partly from uncertainties in the spatial variation of carbon dioxide.

1 Introduction

There has been much evidence that increasing global temperatures for the past 50 years can be attributed to human activity and that anthropogenic influence would continue to change the composition of the atmosphere in the next years. Due to man’s insatiable need for energy and industrialization, carbon dioxide (CO₂), a by-product of fossil fuel combustion and biomass burning (brought about by land use change) has become the most significant greenhouse gas (IPCC, 2005). Due to this, much attention is being given on the absorption characteristics of CO₂ as well as its contribution

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to possible climate changes due to its increased concentration in the atmosphere (McCartney, 1983).

Currently, global transport models utilize in-situ measurements of carbon dioxide from a global network of surface sites for analyzing, estimating and predicting its concentrations as well as determining regional scale exchanges of CO₂ (Rödenbeck et al., 2006; Peylin et al., 2005; Peters et al., 2007). These in-situ surface measurements have the advantage that they are highly accurate. However, they have a limited spatial coverage and an increasing number of measurements are performed within the proximity of local sources and sinks with networks of tall tower observatories over the continents. This makes model estimates susceptible to transport errors, such as errors in vertical transport processes (moist convection and turbulent mixing in the boundary layer), especially for continental regions (Washenfelder et al. 2006; Gerbig et al. 2007). This, in turn, provides uncertainties in the geographic (spatial) and temporal distributions of CO₂ sources and sinks (Dufour et al., 2004; Gerbig et al., 2007). The uncertainties imply that difficulties would come about in predicting the response of carbon dioxide due to climate and land-use changes (Yang et al., 2002), as well as in projecting the future rate of increase of atmospheric CO₂ (Dufour et al., 2004).

Space-borne or satellite measurements, such as the Orbiting Carbon Observatory (OCO) (whose planned launch is in 2009) (Crisp et al., 2004), the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) (Burrows et al., 1990) and the Greenhouse Gases Observing Satellite (GOSAT), may offer the solution to the problem of sparse spatial and temporal distributions of carbon dioxide sources and sinks by providing global column measurements of CO₂ (Yang et al., 2002). To supplement and validate the satellite data, ground-based solar absorption spectroscopy in the infrared or Fourier transform infrared (FTIR) spectrometry is employed (Warneke et al., 2005). It measures the same quantity (column concentrations) as the satellite and exhibits less spatial and temporal variability as compared to in-situ data while retaining information about the surface fluxes and the diurnal behavior of carbon dioxide. It also complements existing in-situ networks and provides information

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about CO₂ exchange on regional scales (Washenfelder et al., 2006). The Total Carbon Column Observing Network (TCCON), which is a system of high-resolution ground-based FTIR spectrometers, provides this capability (<http://www.tcon.caltech.edu/>).

In this paper, CO₂ column abundances from solar absorption FTIR measurements during the CarboEurope Regional Experiment Strategy (CERES) in Biscarrose, France are presented as well as a method to calibrate these measurements against aircraft data. To provide for a “transfer standard” between incomparable measurement techniques, such as in-situ tower data and column concentrations from FTIR measurements, the Stochastic Time Inverted Lagrangian Transport (STILT) model (Lin et al., 2003) was utilized. The study is not about showing the full capability of solar absorption FTIR measurements for column retrievals of CO₂, since the instrument used in CERES is inferior to the ones targeted and in operation for TCCON. The main aim is to provide a framework that allows validating the FTIR retrievals against measurements made in-situ from aircraft as well as from tall towers. Such in-situ measurements are made regularly with high accuracy on the global calibration scale, and linking this scale with FTIR retrievals ultimately provides a calibration scale for remote sensing.

2 Determining CO₂ concentrations

Fourier transform infrared (FTIR) spectroscopy measurements were performed during the CarboEurope Regional Experiment Strategy (CERES) from May to June 2005. CERES aims to come up with a comprehensive database of atmospheric CO₂ concentrations, fluxes, as well as meteorological parameters at the regional scale. An overview of the experiment is given in Dolman et al. (2006). The experiment area is a 250 km×150 km region located Southwest of France bounded to the west by the Atlantic ocean with a shoreline almost rectilinear along a north-northeast orientation. The Les Landes forest dominates the western half of the domain with 80% incorporated in the regional experiment area. It is mainly composed of maritime pines containing clearings of different sizes and are composed of agricultural land, mainly crop, and

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also grassland and pasture. Historically, a plantation forest was originally planted in the area to drain the marshlands. Now, the region is managed as a commercial forest with regular harvests and crop rotations (Dolman et al., 2006).

During the measurement campaign, carbon dioxide was analyzed in the near infrared region of the electromagnetic spectrum due to its proximity to the solar Planck function maxima, which then maximizes the signal-to-noise ratio. Atmospheric oxygen was also retrieved to provide a means to determine the dry air mixing ratio, avoiding uncertainties from the surface pressure and the water vapor column. The Fourier transform spectrometer (FTS) was stationed in Biscarrosse, France at 44°22'40'N latitude, 1°13'52'W longitude and 67.6 m (above sea level) altitude. A total of 4908 spectra were analyzed during the CarboEurope regional experiment encompassing measurements from 8 May 2005 to 26 June 2005. The Bruker 120 M (Mobile) Fourier transform spectrometer was utilized during the campaign. A maximum optical path difference of 30 cm was employed and a resolution of 0.03 cm⁻¹ was used. The 120 M has a focal length of 220 mm and an aperture size of 0.5 mm was used during the dates mentioned. This produces a field of view of 2.3 mrad. Forward and backward scans were taken totaling an average acquisition time of 24.0 s for each spectrum.

Beside the FTS station is a tower instrumented by the Laboratoire des Sciences du Climat et de l'Environnement (LSCE). It houses a continuous in-situ monitoring station called CARIBOU, which includes a LICOR analyzer that measures CO₂ concentrations with a ±0.5 ppm precision. The tower is located at a latitude of 44°22'40.6'N, a longitude of 1°13'52.5'W with the inlet at 114.71 m (above sea level). It also houses a pressure sensor located at 106.81 m (above sea level) (Galdemard et al., 2006). Several aircraft measurements were also performed during the regional experiment. Among them is the METAIR Dimona (Dimona), a touring motor glider (TMG), in which CO₂ is measured onboard using a combination of a fast, open path LICOR 7500, a slower, more precise closed path LICOR 6262 (Neininger et al., 2001), and flask samples that are analyzed for CO₂ in the laboratory at the Max Planck Institute for Biogeochemistry (MPI-BGC) in Jena, Germany with an accuracy of 0.1 ppm. The overall precision of the

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combined CO₂ dataset at 1 Hz is 0.5 ppm.

To aid in the interpretation of the data and to serve as a “transfer standard” between different incomparable kinds of measurements such as the FTS and the in-situ tower data, the Stochastic Time Inverted Lagrangian Transport (STILT) model was utilized (Lin et al., 2003). It is based on the HYSPLIT model (Draxler and Hess, 1997; Draxler and Hess, 1998), using a similar mean advection scheme but employing a different turbulence module. It has been further modified to use winds, surface sensible heat and momentum fluxes, and computed convective mass fluxes from ECMWF assimilated meteorological fields (Gerbig et al., 2007). Being originally designed for comparisons with in-situ measurements (single receptors or single measurement locations), STILT was modified for comparisons with column measurements (multiple receptors). The multiple receptor scheme is depicted in Fig. 1. Receptor points were placed at equal intervals along the vertical for each altitude range. The altitude ranges were set at 1–500 m, 500–3000 m, 3–6 km, 6–11 km and 11–18 km. For each receptor location, x_r , representative particles were released at a time t_r giving rise to particle densities, $\rho(x_r, t_r | x, t)$ at x and time t . From the particle densities, the surface influence or footprint, $S(x, t)$, which relate surface fluxes (sources or sinks) to the concentration, $C(x_r, t)$, at the measurement location (the receptor), can be determined. The initial boundary tracer conditions are taken from the TM3 global transport model (Heimann and Körner, 2003). For more details on the STILT model refer to the papers from Lin et al., 2003 and to Gerbig et al., 2003. The model was run at a 0.125°latitude×0.083°longitude resolution and 3 days backward in time. The CO₂ concentration output from the model (in ppm) is determined by

$$\text{CO}_2 = \text{CO}_{2,\text{background}} + \text{CO}_{2,\text{fossil;fuel}} + \text{CO}_{2,\text{photosynthetic;uptake}} + \text{CO}_{2,\text{respiration}} \quad (1)$$

where $\text{CO}_{2,\text{background}}$ is the background carbon dioxide obtained from the TM3 global transport model boundary fields, $\text{CO}_{2,\text{fossil;fuel}}$ comes from fossil fuel emissions due to combustion estimated using the recent greenhouse gas emissions inventory from the Institute of Economics and the Rational Use of Energy (IER), University of Stuttgart (<http://carboeurope.ier.uni-stuttgart.de/>), $\text{CO}_{2,\text{photosynthetic;uptake}}$ is the carbon dioxide

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concentration taken up by the vegetation and $\text{CO}_{2,\text{respiration}}$ is the amount of CO_2 released by plants. The biospheric exchange is based on the diagnostic model GSB (greatly simplified biosphere) using light and temperature response and 3 vegetation classes namely forests, shrubs and crops (Gerbig et al., 2006).

3 Results

The next sections discuss results from the FTIR retrievals, comparisons with the MetAir Dimona aircraft, results from the STILT model and the effect of clouds on the retrieved O_2 and CO_2 columns.

3.1 Retrieval

CO_2 and O_2 vertical columns were retrieved using the GFIT nonlinear least squares spectral fitting algorithm (version 2.40.2) developed by NASA/JPL (Toon et al., 1992). O_2 was analyzed in the $7765\text{--}8005\text{ cm}^{-1}$ band centered at 7885 cm^{-1} with H_2O as an interfering gas. CO_2 was retrieved in the $6180\text{--}6260\text{ cm}^{-1}$ band centered at 6220 cm^{-1} . Interfering gases in the 6220 cm^{-1} CO_2 band are H_2O , HDO and CH_4 .

The retrieved O_2 column was compared to 20.95% of the total dry pressure column, $P_{\text{dry,column}}$. The dry pressure column was determined using

$$P_{\text{dry,column}} = \frac{P_{\text{obs}}}{m_{\text{dry}}g} - \text{H}_2\text{O}_{\text{column}} \left(\frac{m_{\text{H}_2\text{O}}}{m_{\text{dry}}} \right) \quad (2)$$

where P_{obs} is the observed surface pressure, m_{dry} is the mean molecular mass of dry air, $m_{\text{H}_2\text{O}}$ is the mean molecular mass of water vapor, g is the density weighted acceleration due to gravity and the $\text{H}_2\text{O}_{\text{column}}$ is the water vapor column retrieved in the O_2 window (Washenfelder et al., 2006). From this, a linear fit with zero intercept was done from which the slope (1.0432) was used to scale down the O_2 column to make it correspond with the known atmospheric O_2 concentration (0.2095). This is depicted in

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Fig. 2. The correlation coefficient between the O₂ column and 20.95% of the total dry pressure column is 0.82. The residuals range from ±1%. The O₂ volume mixing ratio (VMR), O_{2,vmr}, as shown in Fig. 3, was then determined by dividing the O₂ column with the total dry pressure column. The O₂ VMR varies from 0.2061 to 0.2132.

The upper limit of the precision of the O₂ VMR was determined from its diurnal variation as shown in Fig. 4. The O₂ diurnal variation is given by

$$O_{2,\text{diurnal}} = 100 \times \left(\frac{O_{2,\text{vmr}}}{\langle O_{2,\text{vmr}} \rangle} - 1 \right) \quad (3)$$

where $\langle O_{2,\text{vmr}} \rangle$ is the daily mean of the volume mixing ratio of oxygen. One way of estimating the upper limit of the precision of CO₂ is to use the diurnal variation of the CO₂ column average VMRs (Yang et al., 2002) shown in Fig. 5. However, since there is a natural variability in the CO₂ column average volume mixing ratio over the course of the day due to diurnally varying surface sources and sinks (mostly biospheric), this method only gives an upper limit of the precision. The CO₂ diurnal variation is given as

$$CO_{2,\text{diurnal}} = 100 \times \left(\frac{CO_{2,\text{vmr}}}{\langle CO_{2,\text{vmr}} \rangle} - 1 \right) \quad (4)$$

where CO_{2,vmr} is defined as 20.95% of the CO₂/O₂ column ratio for individual measurements and $\langle CO_{2,\text{vmr}} \rangle$ is the mean of the day. The CO₂/O₂ column ratio minimizes systematic errors such as errors present in the pressure and in the instrumental line shape (Warneke et al., 2005) and at the same time retaining the diurnal source/sink signals.

Quantiles were used to quantitatively assess the diurnal variations, specifically quartiles and the central 90% ile. Using quartiles, for O₂, the first quartile is at -0.2110%, the median is at 0.0007%, the third quartile is at 0.2072%, the interquartile range is 0.4182% and the quartile deviation is 0.2091%. The minimum O₂ diurnal variation is -1.6128% and the maximum O₂ diurnal variation is 1.7793%. For CO₂, the first quartile is at -0.2046%, the median is at 0.0038%, the third quartile is at 0.2098%, the

interquartile range is 0.4144% and the quartile deviation is 0.2072%. The minimum CO₂ diurnal variation is -1.2337% and the maximum CO₂ diurnal variation is 1.2826%. This means that approximately 50% of the measured data have diurnal variations between ±0.21% for both O₂ and CO₂. Using the central 90% ile, for O₂, the 5% ile is at -0.5596%, the 95% ile is at 0.5558%, the 95% ile-5% ile range is 1.1154% and the central 90% ile deviation is 0.5577%. For CO₂, the 5% ile is at -0.5349%, the 95% ile is at 0.5181%, the 95% ile-5% ile range is 1.0530% and the central 90% ile deviation is 0.5265%. Approximately 90% of the measured data have diurnal variations between ±0.56% for both O₂ and CO₂. This is depicted in Fig. 6. The outliers in the diurnal variations result from influences of clouds (Warneke et al., 2006).

3.2 Aircraft comparison

The accuracy of the CO₂ retrievals was determined by comparing the FTS CO₂ VMRs with integrated aircraft carbon dioxide volume mixing ratios. Of the mentioned measurement dates, simultaneous Dimona and FTS measurements were available during five days, 25 May, 26 May, 27 May, 6 June and 14 June. During these days, only those data from the aircraft that fell within a 50 km radius from the FTS station were selected. From this, seven instances were identified wherein the Dimona and the FTS measured simultaneously with the same airmass. These instances are summarized in Table 1. The spatial coverage of the Dimona flight paths for the instances defined is shown in Fig. 7.

The Dimona reached a maximum altitude of approximately 3 km during the CarboEurope experiment. It was thus necessary to append CO₂ profiles above the aircraft ceiling. For the free troposphere portion of the profile, data were taken from the TM3 global transport model, which was coupled to surface fluxes from fossil fuel emissions as well as to the BIOME-BGC model to include biospheric exchange (Heimann and Körner, 2003). For the stratospheric part of the profile, in-situ balloon data from the Observations of the Middle Stratosphere (OMS) experiment performed in Fort Sumner, New Mexico (35° N, 104° W) on 17 September 2004 were utilized. The balloon profile was

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corrected for age using the annual increase rate of CO₂. It was then coordinate transformed to potential temperature using simultaneous measurements of pressure and temperature during the balloon flight. The potential temperature was then converted to altitude using the equation formulated by Knox (1998)

$$z = \frac{\ln\left(\frac{\theta}{350}\right)}{0.045} + 13 \quad (5)$$

where θ is the potential temperature in Kelvin and z is the altitude in km. It was then converted back to pressure using NCEP altitude-pressure-temperature profiles for the specific aircraft overpass dates and the CO₂ concentration values were then interpolated. A ± 0.75 ppm uncertainty was assigned based on the precision of the balloon data and on the uncertainties in the mean age of air in the stratosphere from CO₂ balloon measurements. The CO₂ concentrations for the aircraft have an uncertainty of ± 0.5 ppm. For the model, a pressure dependent uncertainty in the CO₂ profile was assigned ranging from ± 0.5 ppm at the aircraft ceiling increasing to a maximum of ± 0.75 ppm at the tropopause.

To compare the aircraft carbon dioxide measurements with the FTS data, it is necessary to consider the different characteristics of the observing systems. Derived quantities, such as total columns, may then be compared properly among different measurement platforms. In this case, the aircraft data is said to be “simulated” by the FTS retrievals using the FTS a priori and averaging kernels (Rodgers et al., 2003). The column averaging kernels, corresponding to each instance, as well as the a priori CO₂ used in the retrievals were applied to the aircraft data using the expression

$$\text{CO}_{2,\text{simulated}} = \text{CO}_{2,a,\text{priori}} + A(\text{CO}_{2,\text{aircraft+MODEL+balloon}} - \text{CO}_{2,a,\text{priori}}) \quad (6)$$

where $\text{CO}_{2,a,\text{priori}}$ is the a priori CO₂ profile used in the retrieval, A is the column averaging kernel (shown in Fig. 8 for instance 7) and $\text{CO}_{2,\text{aircraft+MODEL+balloon}}$ is the aircraft data appended with the model and balloon data. The simulated CO₂ profile, as shown in Fig. 8, was then integrated with respect to pressure using a trapezoidal numerical

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integration to determine the column CO₂. The column averaged volume mixing ratio is then determined by dividing the column CO₂ by the dry pressure column.

Figure 9 shows the comparison of the averaged (retrieval error weighted) FTS CO₂ VMR (20.95% of the CO₂/O₂ column ratio) for the aforementioned instances to the integrated aircraft CO₂ VMRs. CO₂ columns were reduced by 1.0331 wherein the scaling factor was determined from the slope of a zero-intercept linear fit. The correlation coefficient is 0.69 and the residuals vary between ±1 ppm. Two instances, 4 and 6, deviated more than expected from the one-to-one line due to differences in the surface influences between the FTS and the Dimona (see Discussion).

3.3 STILT

The STILT model was used for comparison to carbon dioxide concentration time series from the Biscarrosse tower data using a single receptor placed at the same latitude and longitude as the tower with an above ground level height of 47 m. Figure 10 shows the time series comparison between STILT and the tower data. The root-mean-square (RMS) difference between the model and the measured data is 7.06 ppm. The standard deviation of the differences is 6.78 ppm and the mean difference is -1.95 ppm.

The STILT model was then extended for comparison to vertical column concentrations of CO₂ using multiple receptors along the column (see Fig. 1). Similar to what was done with the aircraft profiles, OMS in-situ balloon data, corrected for age and transformed in coordinates, were appended above the STILT model. The modeled carbon dioxide profile is shown in Fig. 11 for instance 7 compared to the Dimona-TM3-OMS CO₂ profile. The FTS retrieval a priori CO₂ and its averaging kernel were also applied (Eq. 6) to the modeled CO₂ profile before integrating the column. The column averaged VMRs of carbon dioxide from the STILT model and the FTIR data were then compared. This is shown in Fig. 12. For the STILT-FTIR comparison, the standard deviation of the differences is 0.97 ppm, the mean difference is 2.20 ppm and the correlation coefficient is 0.51. Aside from this, the column results from STILT and the FTIR data were also compared with the Weather Research and Forecasting-Vegetation Pho-

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tosynthesis and Respiration Model (WRF-VPRM) modeling system. WRF-VPRM is a coupled modeling system designed to simulate high-resolution atmospheric CO₂ concentration fields. Here, WRF is the state of the art mesoscale meteorological model and it is coupled to the diagnostic biospheric model VPRM. VPRM produces biospheric CO₂ fluxes and passes to WRF, which performs atmospheric CO₂ tracer transport simulation. The modeling system also takes into account anthropogenic CO₂ fluxes. The comprehensive description of the modeling system and setup can be found in Ahmadov et al. For the WRF-VPRM-FTIR comparison, the standard deviation of the differences is 1.02 ppm, the mean difference is 3.08 ppm and the correlation coefficient is 0.24 taking note that the WRF-VPRM was not simulated by the averaging kernel and the a priori of the FTS. For the comparison between the two models, therefore, between WRF-VPRM and STILT the standard deviation of the differences is 0.652 ppm and the mean difference is -0.78 ppm.

3.4 Effect of clouds on O₂ and CO₂ precision

To quantitatively assess the effect of clouds on the precision of the retrieved O₂ and CO₂ VMRs, measurements from a clear day and a partly cloudy day during the campaign were compared. As shown in Fig. 13, 2.75-min averaged data were compared from measurements during a clear day (18 June 2005) and during a partly cloudy day (14 May 2005) characterized by thin high altitude cirrus clouds. The standard deviation of the O₂ VMR during the clear day is $\pm 3.86 \times 10^{-4}$ while for the partly cloudy is $\pm 7.44 \times 10^{-4}$. For the CO₂ VMRs, comparisons were made between the O₂ normalized carbon dioxide concentrations and the pressure normalized CO₂ vmr (CO₂ column divided by 20.95% of the dry pressure column). The standard deviation increased from ± 0.62 ppm for the clear day measurement to ± 1.09 ppm for partly cloudy day spectra for the O₂ normalized CO₂ volume mixing ratio while a larger increase in the standard deviation is observed for the pressure normalized CO₂ VMR from ± 0.48 ppm (clear day) to ± 1.31 ppm (partly cloudy day). This also shows the improved precision by normalizing with O₂ (which minimizes systematic errors) particularly for partly cloudy day

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measurements (Washenfelder et al., 2006). These are outlined in Table 2.

4 Discussion

Surface influence functions, or footprints, which quantify the contribution of surface fluxes to the concentration of the aircraft measurement as well as of the FTIR column, can be used to assess potential reasons for disagreement between the two types of measurements. The time integrated footprints shown in Fig. 14 have been determined using STILT. They show that the surface influences for instances 4 and 6 have a significant difference for the FTS and for the Dimona aircraft. For instance 4, where the CO₂ column averaged VMR of the FTS is lower compared to Dimona (see Fig. 9, instance 4), the FTS footprint has a discontinuity in the area of northern Spain. Surface fluxes in this region would therefore not affect the FTS measurements as it does for Dimona producing the mentioned difference. This discontinuity can be attributed to particles rising above the surface hence producing no surface influence at that region. Aside from this, the aircraft is also more confined in a smaller region for this instance compared with the other instances (see Fig. 7, instance 4). This gives it a rather limited sampling area, in which other processes can influence the aircraft data as compared to the FTIR. For instance 6, the FTS column averaged VMR is higher than Dimona (see Fig. 9, instance 6). The footprints of the Dimona show more influences on land than the FTS (see Fig. 14, instance 6), consistent with the flight track covering more vegetated areas (see Fig. 7, instance 6). Given that the land region at that time of the year is a much stronger sink for CO₂ as compared to the ocean due to the active land biosphere, explains the lower CO₂ observed by the aircraft.

In Fig. 15, decomposition of the CO₂ concentration for the different altitude ranges is shown. The lower altitude ranges (1–500 m and 500–3000 m), show significant influence from the biosphere in the CO₂ variability. These altitude ranges, which are well within the planetary boundary layer where significant turbulence is experienced (hence more vertical mixing), get more contributions from vegetation photosynthetic

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uptake and respiration. Higher up, from 3 km to 18 km, the carbon dioxide is dominated mostly by the background values with little effect from the vegetation in its variability. This also proves the point in the aircraft comparisons with the FTS that when the air masses come from the ocean, larger CO₂ is observed by the FTS compared than the aircraft which sample data along vegetation regions (instance 6) since the FTS measures carbon dioxide from areas with less biospheric influence. The altitude range and source/sink decomposition is shown in Fig. 16 for instance 6 and instances 2 and 3.

Referring to Fig. 7, one can see that there are instances (instances 4 and 6) where the Dimona was taking samples in locations where the FTS was not pointing. One might say that this could be a potential source of disagreement between the FTIR spectrometer and the aircraft. However, looking at the FTS slant and vertical column averaged VMRs in Fig. 17, one can see that taking slant or vertical column averaged VMRs does not matter. This was also verified with the WRF-VPRM also shown in Fig. 17.

The comparison between the carbon dioxide column averaged VMRs measured with the FTIR spectrometer and the MetAir Dimona aircraft can be considered to be in agreement with each other since the error bars fall within the one-to-one line (see Fig. 9). The most significant source of error for the FTS CO₂ column averaged volume mixing ratio is the precision of the instrument (120M) used in the CarboEurope experiment. As stated earlier, the 120M is inferior to the ones targeted and in operation for TCCON. TCCON uses more high-resolution instruments. For the integrated aircraft carbon dioxide VMR, the most significant source of uncertainty is in the spatial variability of CO₂ particularly in the planetary boundary layer (see Fig. 8). The spatial variability is a result of taking aircraft data within a 50 km radius around the FTS station. FTS and aircraft validations can therefore be improved by using higher resolutions in measuring solar absorption spectra and by flying within a closer vicinity of the FTS station.

After validating the FTS carbon dioxide column averaged VMRs with the aircraft, a meaningful next step would be to compare FTS measurements with in-situ tower

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data. The problem of directly comparing in-situ and remotely sensed data is that the quantities are different in nature to start with. One needs a tool to mediate between the two measuring techniques and whose role is to assess whether both the in-situ and the FTS data are consistent with each other. This is provided by the STILT model.

5 The comparison between the tower and STILT performed fairly well with differences partly coming from using the greatly simplified biosphere (GSB) vegetation grid in the STILT model. For the STILT-FTIR comparison, the differences is obviously not so much due to the GSB used in STILT, since STILT and WRF-VPRM agree considerably well and since column measurements are not much affected by local sources and sinks,
10 but more due to the spatial variability in the aircraft data partly used to scale the CO₂ columns. This spatial variability is not that evident in the modeled profile (see Fig. 11). Additional information on this spatial variability will be available from the simulation of CO₂ along the flight track, however, this is beyond the scope of this paper and will be presented in a future publication focusing on the airborne data. On the other hand, the
15 STILT model was able to capture the variability in the column averaged volume mixing ratio measured by the FTS as evidence from a relatively low standard deviation in the differences between STILT and FTIR. Both STILT and WRF-VPRM also captured the decreasing trend of CO₂ as seen from the FTS data (see Fig. 12). The decrease is part of the seasonal variation of CO₂. From Fig. 10 and Fig. 12, and from the statistics of
20 the comparisons, STILT, which used identical model parameters for the in-situ (single receptor) and the column (multiple receptors) calculations, can therefore function as a “transfer standard” between in-situ and remotely sensed measurements.

5 Conclusions

25 Ground-based solar absorption measurements using Fourier transform infrared spectrometry (FTS) were performed during the CarboEurope Regional Experiment Strategy (CERES) from May to June 2005 in Biscarrosse, France. Near-infrared spectra from a Bruker 120 M FTIR spectrometer were then analyzed to retrieve carbon dioxide con-

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centrations using a non-linear least squares fitting algorithm developed by NASA JPL (GFIT). The Stochastic Time Inverted Lagrangian Transport (STILT) model was used to facilitate the comparison of the FTIR CO₂ retrievals to simultaneous in-situ measurements made at a surface monitoring station and aboard an aircraft.

5 As an internal check on the quality of the measurements, O₂ was retrieved in the 7765–8005 cm⁻¹ band. It was then compared to 20.95% of the dry pressure column resulting in a reduction factor of 1.0432 for oxygen and a correlation coefficient of 0.82. Using the diurnal variation of oxygen, the upper limit of the retrieval precision was determined. Its diurnal variation had an interquartile range of 0.4128% and its quartile deviation is 0.2091%. Its 95% ile–5% ile range is 1.1154% and its central 90% ile deviation is 0.5577%. The central 90% ile deviation shows that 90% of the retrieved O₂ columns fall within ±0.56% of the diurnal variations.

10 Carbon dioxide, on the other hand, was analyzed in the 6180–6260 cm⁻¹ band. The diurnal variation of CO₂ had an interquartile range of 0.4144% and its quartile deviation is 0.2072%. Its 95%–5% ile range is 1.0530% and its central 90% ile deviation is 0.5265%. The retrieved carbon dioxide columns were then calibrated using data from the METAIR Dimona aircraft. Seven instances in which both the FTS and the Dimona simultaneously measured were identified. To compare the FTS and the Dimona, pressure and CO₂ model data from the TM3 global transport model and in-situ balloon data from OMS measurements were appended above the aircraft ceiling to produce CO₂ profiles during the instances. The profiles were then “simulated” using the a priori and the averaging kernels used in the FTS retrievals and were then integrated to come up with the column concentrations. The CO₂ columns were then reduced by 1.0331 after comparisons with the aircraft. The FTS and the Dimona carbon dioxide data had a 0.69 correlation coefficient. Two instances (4 and 6) deviated larger than expected from the one-to-one line and these instances were identified to have FTS and Dimona footprints that differ relatively more in terms of influence regions than the other instances. Additionally, the STILT column model outputs showed that altitudes from 0–3 km receive significant biospheric influence while altitudes above 3 km are mostly

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affected by globally advected carbon dioxide. The difference between the slant and vertical column averaged VMRs of carbon dioxide were also analyzed and verified with the Weather Research and Forecasting - Vegetation Photosynthesis and Respiration Model (WRF-VPRM). The difference in the slant and vertical column averaged volume mixing ratio turned out to be negligible. For future FTS validation experiments using aircrafts, this means that vertical profiles should be flown in close proximity given the horizontal variability of carbon dioxide, but there is no need to adopt a slanting aircraft profile.

Time series concentrations of carbon dioxide from the STILT model were compared to in-situ tower data and to FTS column concentrations. The difference between the STILT model and the tower data partly comes from using the greatly simplified biosphere (GSB) diagnostic model that only uses 3 classes of vegetation. However, given the agreement between STILT and WRF-VPRM, another reason to consider is the scaling factor used to reduce the CO₂ columns. The scaling factor was determined from aircraft measurements that sampled at a 50 km radius from the FTS and this introduces spatial variability around the FTS. However, the variability of the column averaged VMRs measured by the FTS was modeled fairly well by STILT as seen from the calculated standard deviation between STILT and the FTIR data differences (0.97 ppm).

Since identical model parameters were used for land-atmosphere fluxes when STILT was compared with in-situ tower data (single receptor) and with column measurements from the FTS (multiple receptors), STILT can be used as a “transfer standard”. Using STILT for comparing remotely sensed CO₂ data with tower measurements of carbon dioxide provided a framework that allowed validating the FTIR retrievals versus measurements made in-situ. Since these in-situ measurements are done frequently and at high accuracy on the global calibration scale, linking this scale with FTIR retrievals ultimately provides a calibration scale for remote sensing.

Acknowledgements. The authors would like to thank O. Schrems and the A. Wegener Institute for the FTS used in the campaign, P. Galdemar for the tall tower information and data, Rebecca Washenfelder for assisting in the aircraft comparisons, G. Aleks, G. Toon and P. Wennberg for

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assistance and suggestions on the FTIR retrievals and B. Daube for providing OMS balloon data.

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Table 1. Instances. Dates and times where both the MetAir Dimona aircraft and the FTS had measurements.

Instance	Date	Time (UTC)	Max. Altitude of Dimona (km)
1	25 May	10:00–13:00	2.7249
2	26 May	10:00–11:00	3.0655
3	26 May	13:00–15:00	2.5573
4	27 May	07:00–09:00	3.2324
5	27 May	12:00–14:00	2.6851
6	6 June	09:00–13:00	2.5615
7	14 June	10:00–15:00	2.6441

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Table 2. Clear and Partly Cloudy Days (2.75-min averages). Normalizing CO₂ by O₂ increases the precision particularly for partly cloudy days.

	18 June (Clear Day)	14 May (Partly Cloudy Day)
O ₂ VMR	$0.2095 \pm 3.86 \times 10^{-4}$	$0.2096 \pm 7.44 \times 10^{-4}$
CO ₂ VMR (O ₂ Normalized)	377.52 ± 0.62 ppm	379.37 ± 1.09 ppm
CO ₂ VMR (Pressure Normalized)	377.55 ± 0.48 ppm	379.61 ± 1.31 ppm

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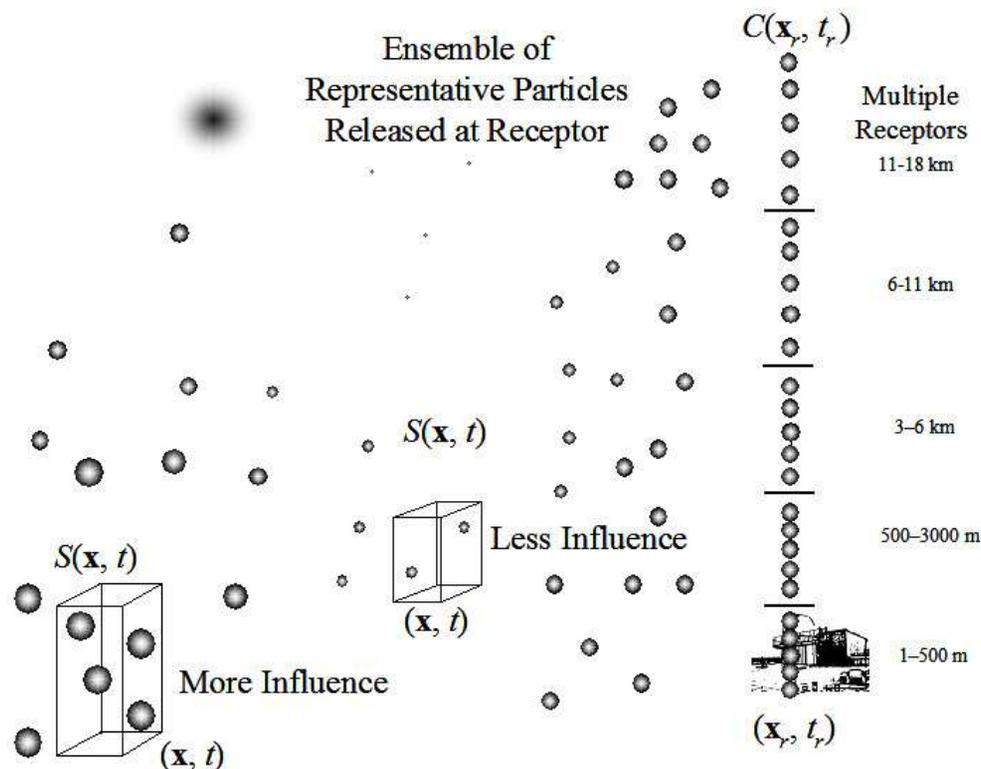


Fig. 1. STILT Applied to Column Measurements. Receptor points were placed at equal intervals along the vertical column for each altitude range. Altitude ranges are from 1–500 m, 500–3000 m, 3–6 km, 6–11 km and 11–18 km. The released particles give rise to particle densities at certain locations wherein influences can be calculated.

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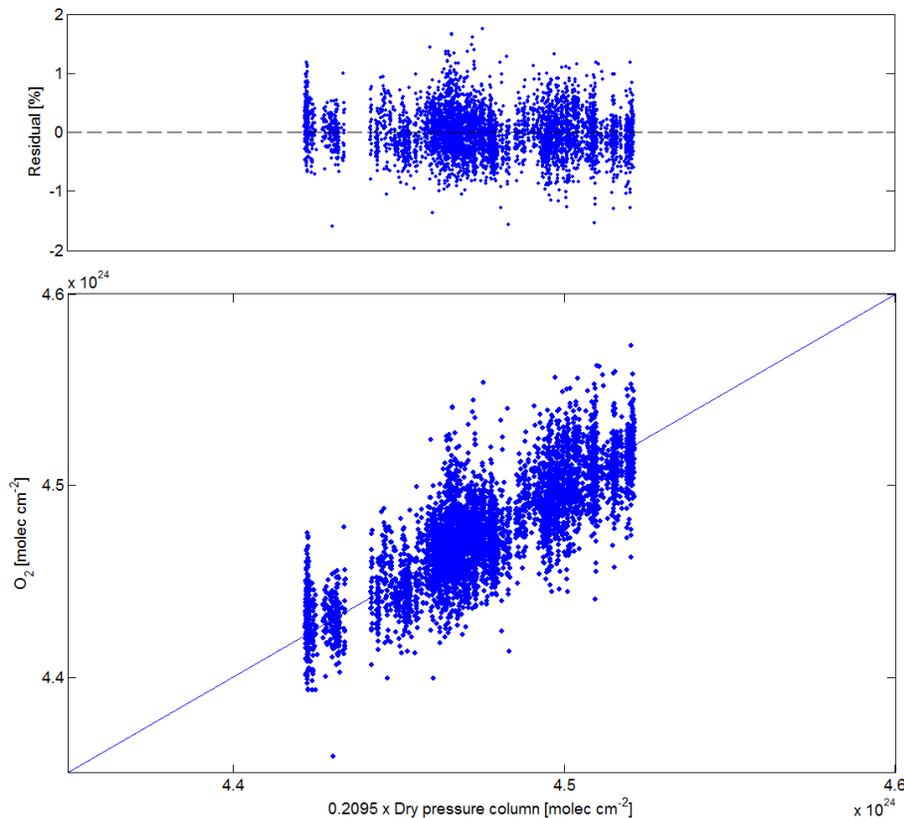


Fig. 2. Comparison of the O₂ column with 20.95% of the total dry pressure column. The correlation coefficient is 0.82. A line of slope 1.0432 with zero intercept was fitted to the data and this slope was used to reduce the O₂ column to make it correspond with the known O₂ concentration of 0.2095.

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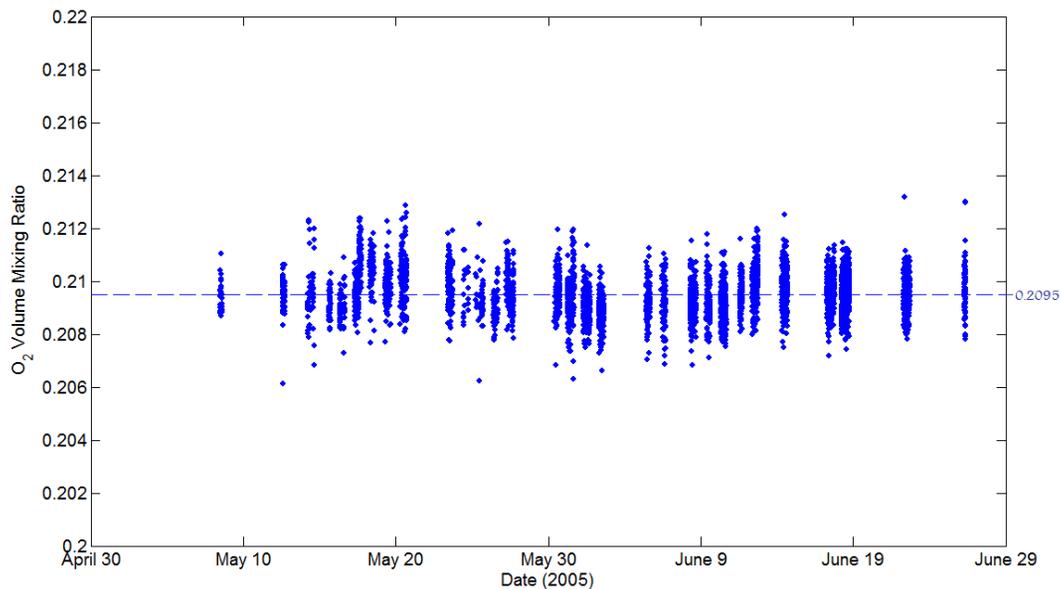
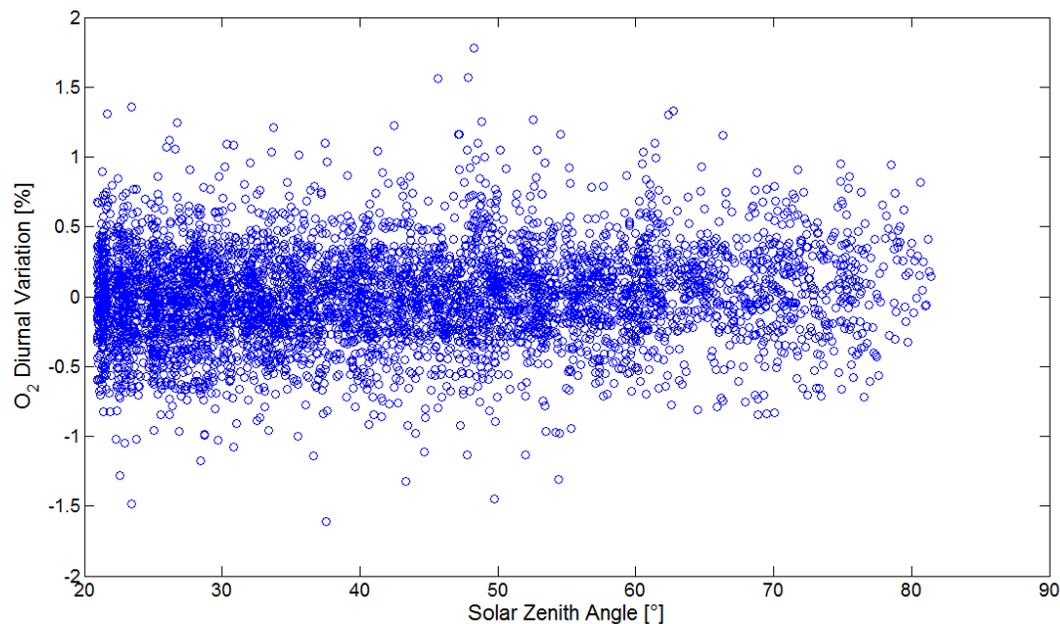


Fig. 3. O₂ Volumn Mixing Ratio. The O₂ vmr varies from 0.2061 to 0.2132.

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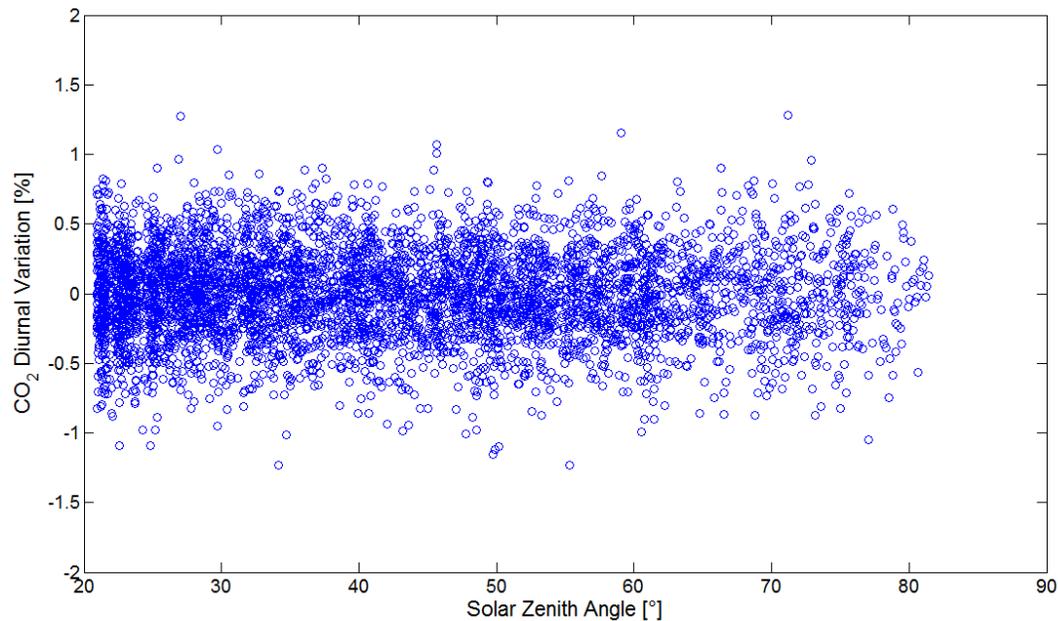
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**Fig. 4.** O₂ Diurnal Variation.[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

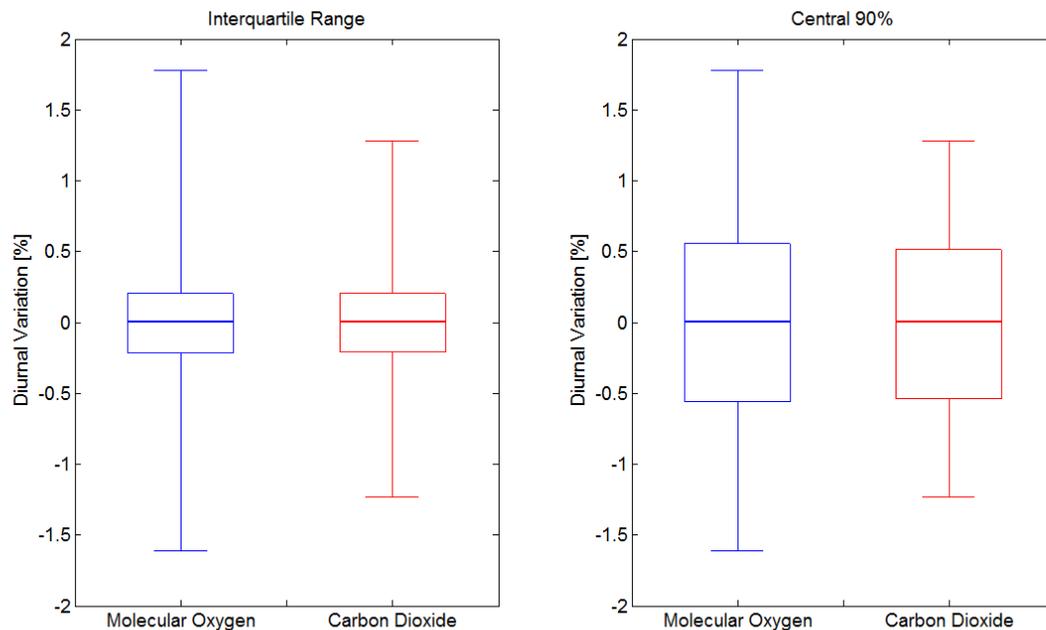
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**Fig. 6.** Quantiles of the Diurnal Variations of O₂ and CO₂.[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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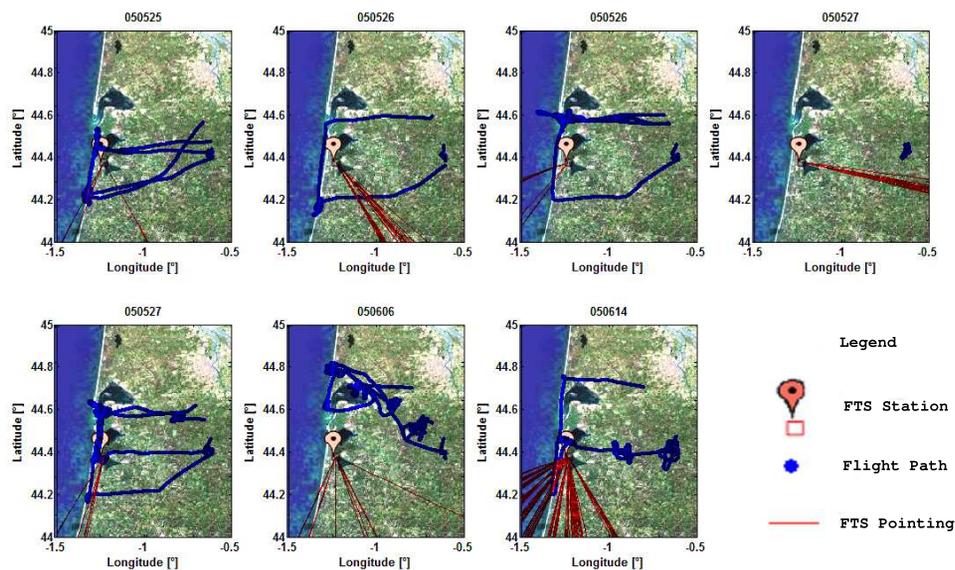


Fig. 7. Spatial Coverage. Shown is the location of the FTS station, the flight paths of the MetAir Dimona and the direction where the FTS was pointing for instances 1–7.

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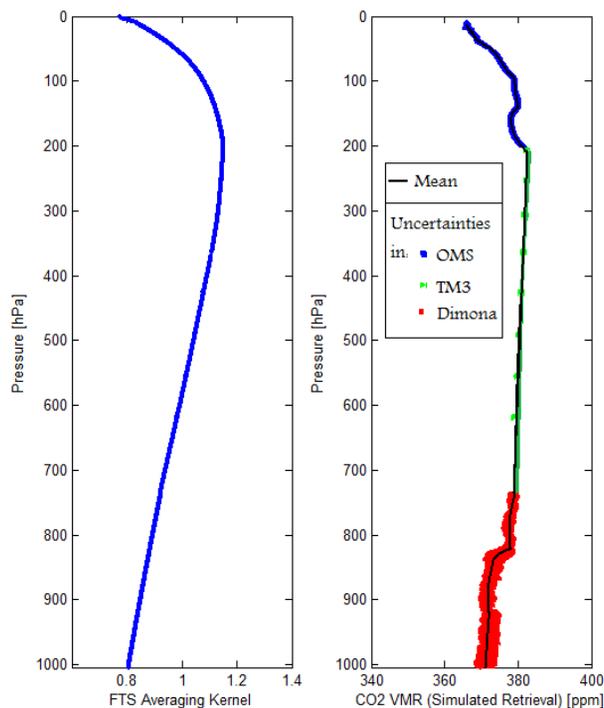


Fig. 8. CO₂ Profile for Instance 7 (14 June 2005; 10:00–15:00 UTC). (Left) The averaging kernel used in the FTS retrieval was applied to the aircraft data to make a comparison with the Fourier transform spectrometer CO₂ concentration data using the expression $CO_{2,\text{simulated}} = CO_{2,a,\text{priori}} + A(CO_{2,\text{aircraft+MODEL+balloon}} - CO_{2,a,\text{priori}})$. (Right) Simulated CO₂ profile after pressure and CO₂ concentrations from the TM3 global transport model and from OMS in-situ balloon data were appended above the aircraft ceiling. CO₂ concentration data for the aircraft have an uncertainty of ± 0.5 ppm. Above the aircraft ceiling, the modeled CO₂ data was assigned to have a pressure dependent uncertainty varying from ± 0.5 ppm to ± 0.75 ppm. The uncertainty in the balloon data was estimated to have a ± 0.75 ppm based upon the variability of the measured CO₂ data and the mean age of air in the stratosphere.

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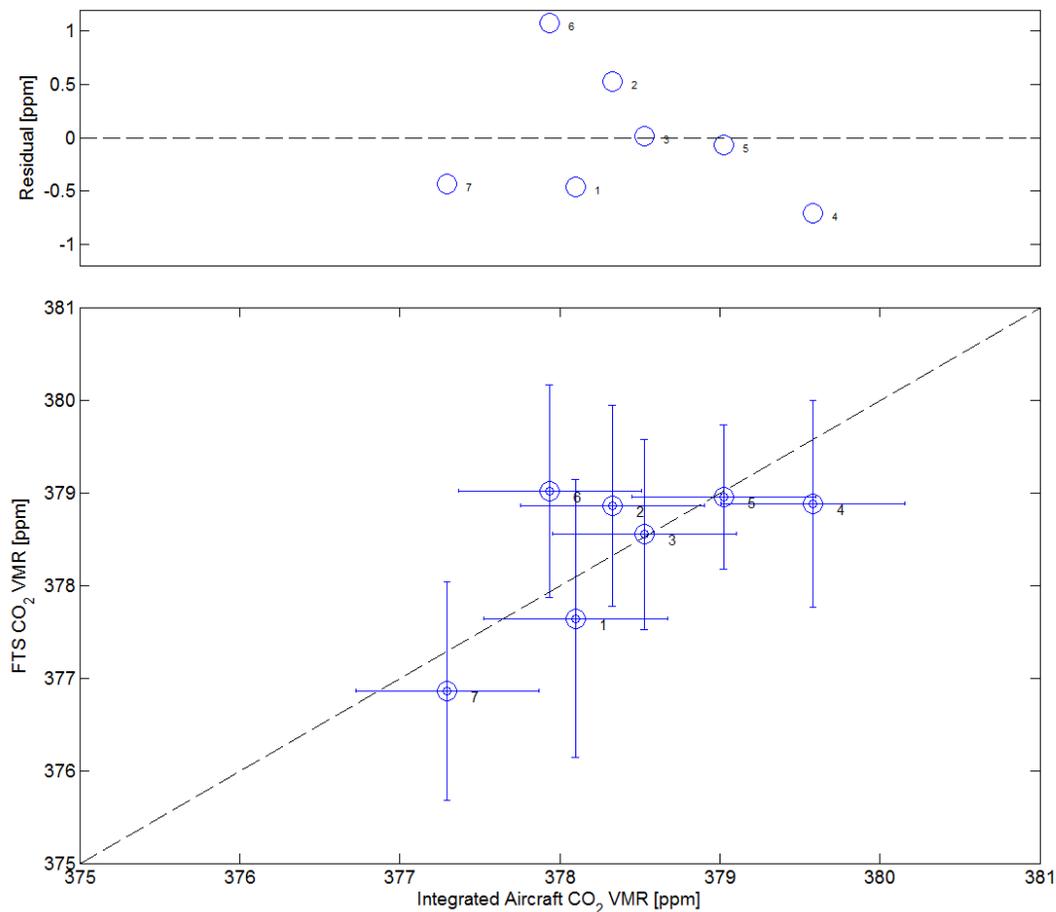


Fig. 9. FTS and Integrated Aircraft CO₂ Comparison. CO₂ columns were reduced by 1.0331 determined from the slope of a zero intercept linear fit. The correlation coefficient is 0.69 and the residuals vary between ± 1 ppm.

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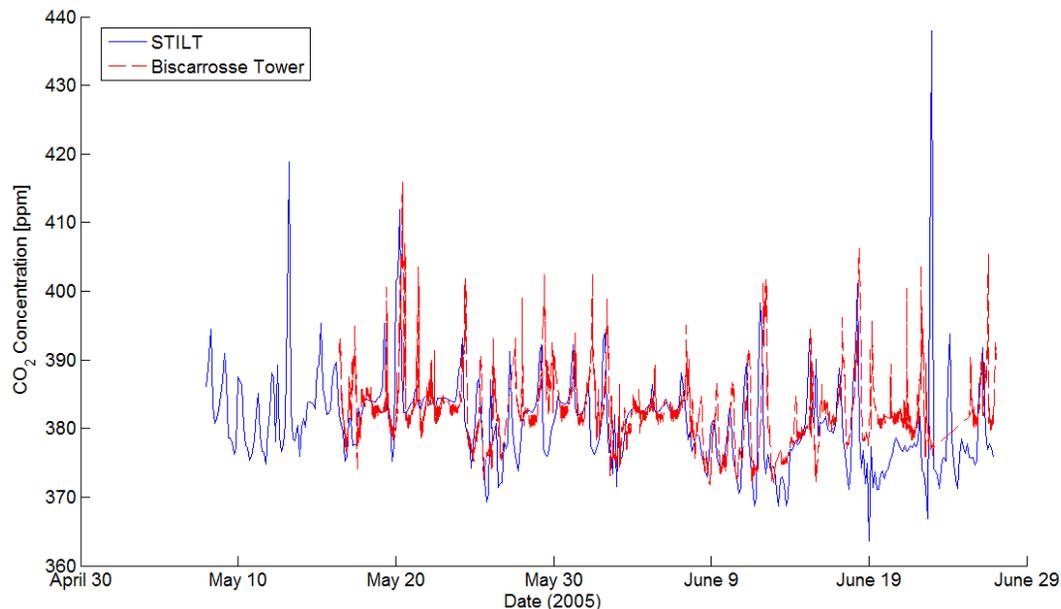
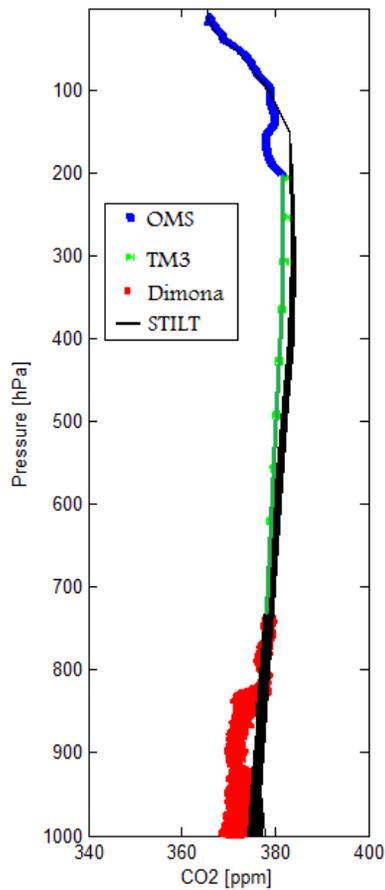


Fig. 10. STILT-Biscarrosse Tower Comparison. The RMS of the differences between the two data sets is 7.06 ppm, the standard deviation of the differences is 6.78 ppm and the mean difference is -1.95 ppm.

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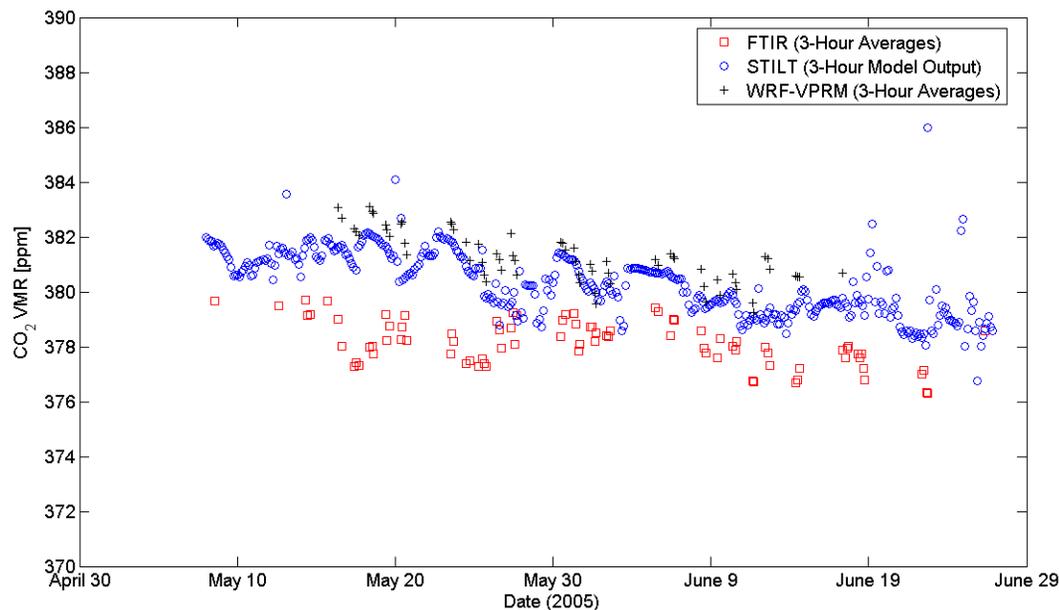
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**Fig. 11.** Dimona-TM3-OMS and STILT Carbon Dioxide Profiles for Instance 7.[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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**Fig. 12.** STILT, WRF-VPRM and FTIR Comparisons.[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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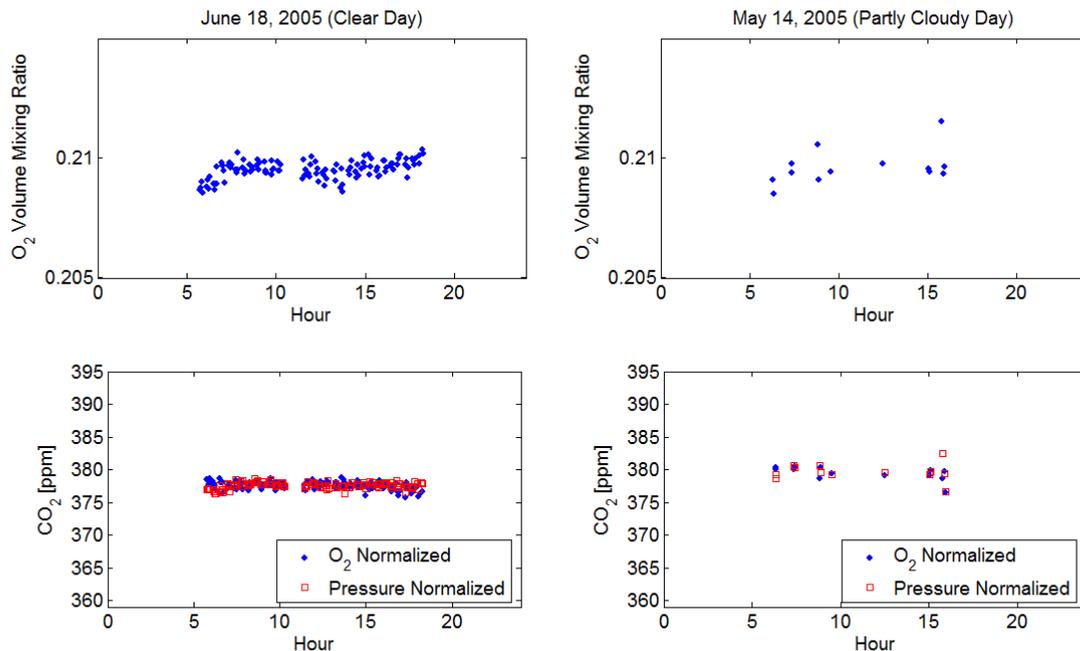


Fig. 13. Clear and Partly Cloudy Days (2.75-min averages). Thin cirrus clouds affect the precision of both O₂ and CO₂ VMRs.

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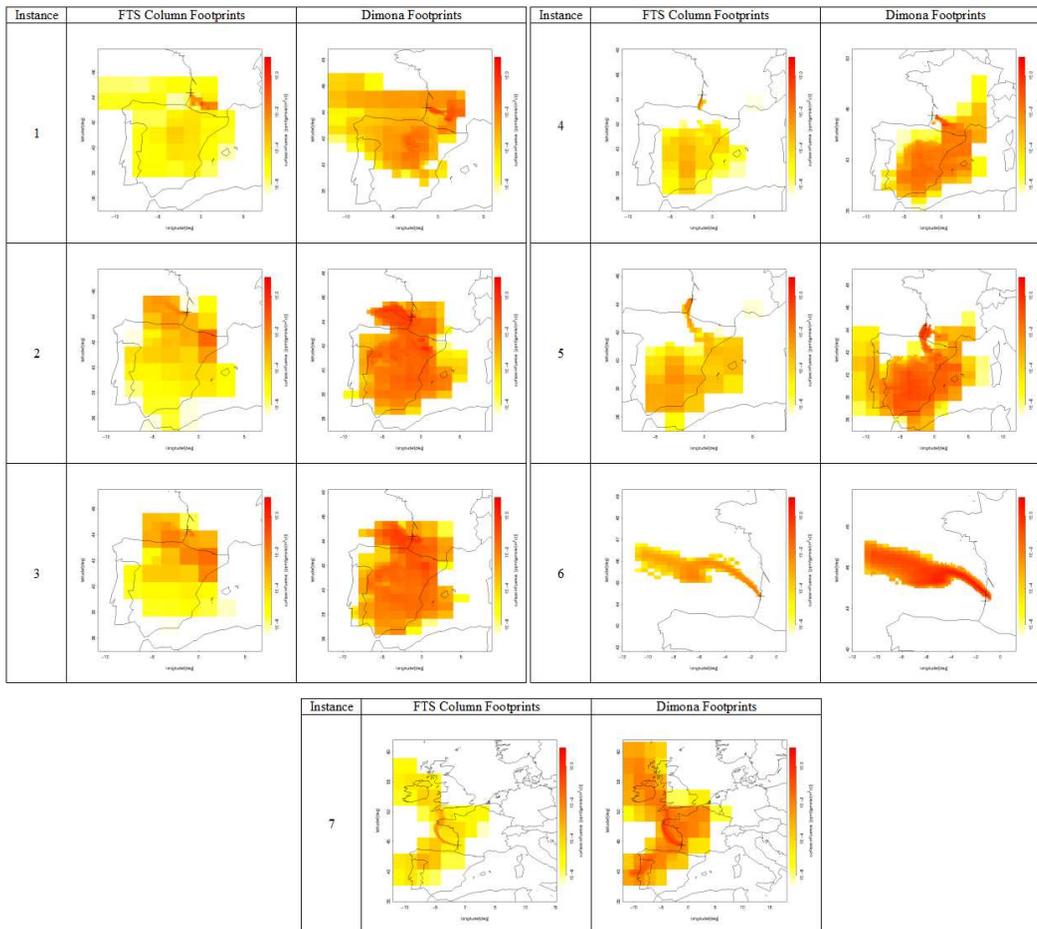


Fig. 14. Footprints for Instances 1 to 7. Instances 4 and 6 footprints have a significant difference between the FTS and the Dimona explaining their larger than expected deviation from the one-to-one line.

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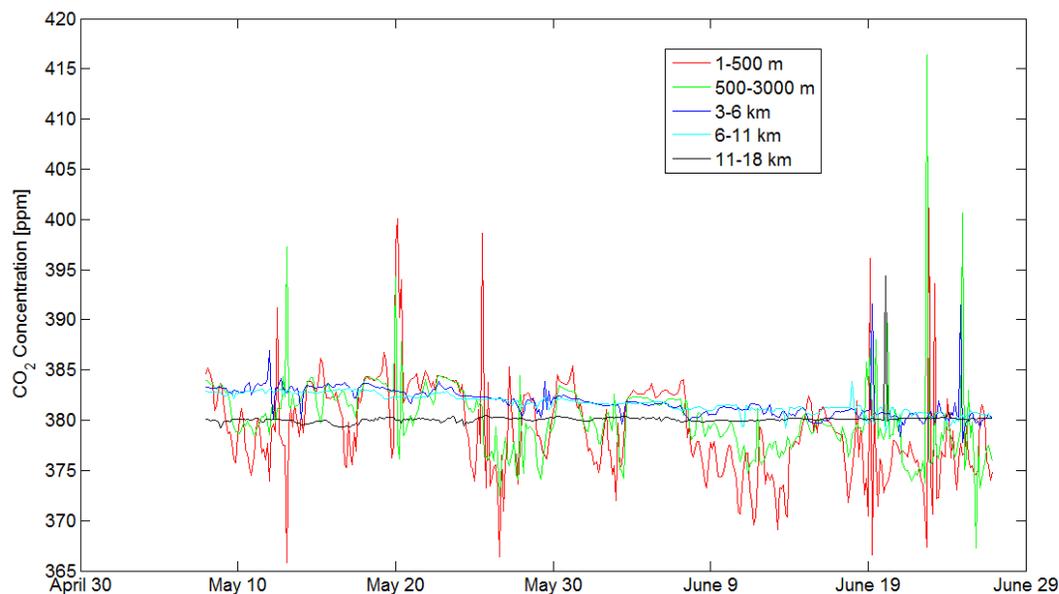


Fig. 15. STILT Decomposition of CO₂ by Altitude Range. The CO₂ multiple receptor signal is decomposed into the different altitude ranges of 1–500 m, 500–3000 m, 3–6 km, 6–11 km and 11–18 km.

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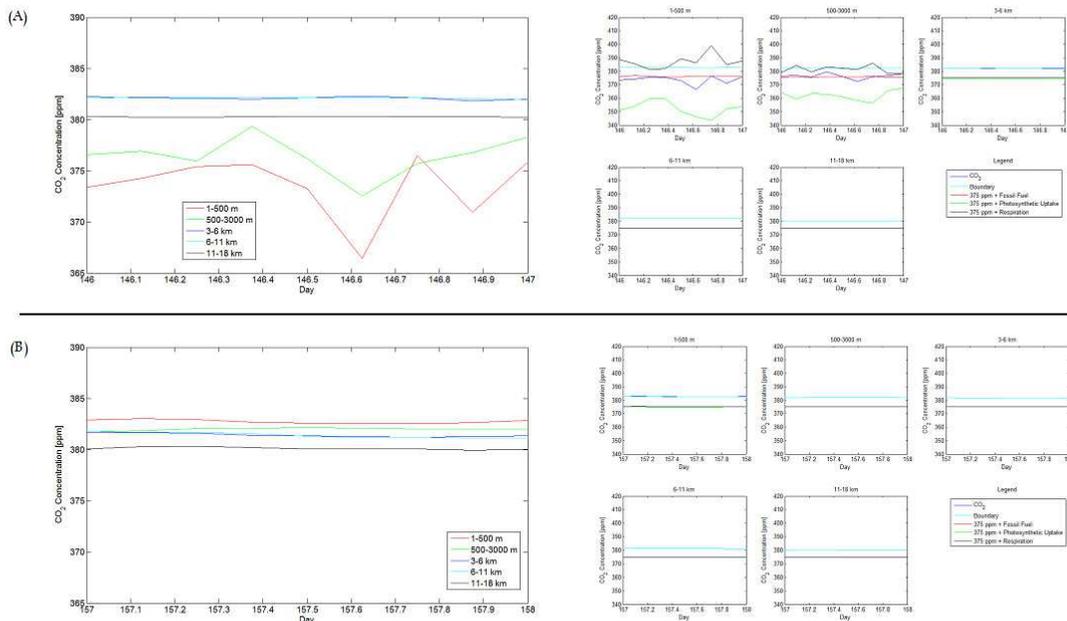


Fig. 16. STILT Decomposition of CO₂ by Altitude Range and Sources/Sinks for **(A)** Instances 2, 3 and **(B)** Instance 6. Instance 6 receives less influence from the biosphere since the footprints originate mostly from the ocean. This produces a higher CO₂ value detected by the FTS than the aircraft (sampling over vegetation).

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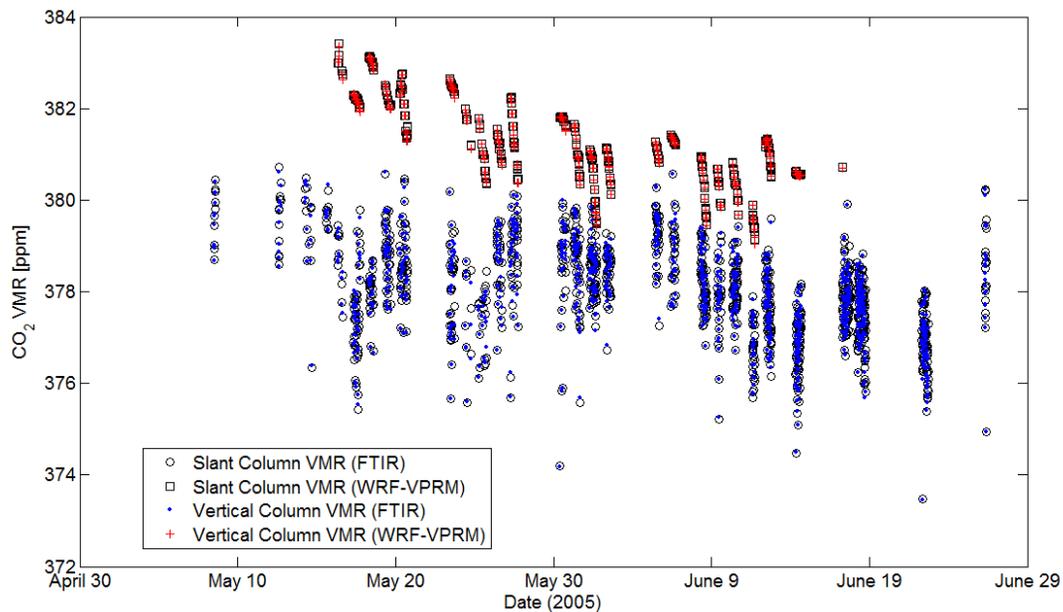


Fig. 17. 2.75-min averaged Slant and Vertical Columns from the FTS and the WRF-VPRM Model.

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