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Co-located column and in situ measurements of CO_2 in the tropics compared with model simulations

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Abstract. The first ground-based remote sensing measurements of the column averaged volume mixing ratio of CO_2 (XCO_2) in tropical South America have been obtained at Paramaribo, Suriname (5.8° N, 55.2° W). The remote sensing observations are complemented by surface air-samples collected at the site, analyzed for CO_2 and $^{13}CO_2$. The surface in-situ measurements are strongly influenced by local sources. From the isotopic composition of the air samples the local source component is suggested to be dominated by the terrestrial biosphere. Using $\delta^{13}C$ from the NOAA/ESRL stations Ascension Is. (ASC), 7.9° S, 14.4° W, and Ragged Point (RPB), 13.2° N, 59.4° W, the data has been corrected for the local source component. Due to the migration of the Intertropical Convergence Zone (ITCZ) over the measurement site the sampled air masses belong to the Northern or Southern Hemisphere depending on the time of the year. Comparison to analyzed CO_2 fields based on TM3 model simulations using optimized fluxes indicate agreement for the seasonality in XCO_2 as well as for the corrected CO_2 mixing ratios at the surface for the long dry season, when Paramaribo belongs to the Southern Hemisphere. A slightly worse agreement during the short dry season is attributed to a larger representation error during this time of the year. Overall the comparison indicates that the TM3 model is capable to simulate the seasonal variation of surface concentrations as well as column densities of CO_2 correctly at Paramaribo. It has been also shown that the column measurements currently performed at Paramaribo have a limited precision and lack the link to the in situ measurements. Solutions for future improvements have been suggested.

1 Introduction

Carbon dioxide (CO_2) is the most important anthropogenic greenhouse gas. Human activities, primarily fossil fuel combustion and deforestation, are responsible for a continuing increase of its atmospheric concentration. The oceans and terrestrial ecosystems currently act as sinks for atmospheric CO_2 and absorb approximately half of the anthropogenic emissions (IPCC, 2007). Inverse models have been used to infer the geographical distribution of the sinks from atmospheric measurements. Until recently inverse modelling studies were solely based on a network of surface in-situ measurement stations. This approach is limited by the sensitivity of the sink estimates to vertical transport and by the sparse spatial coverage of the sampling sites. Remote sensing measurements overcome some of the limitations of the in situ network. Remote sensing measurements provide a column integral, a different kind of information than the in situ measurements. The CO_2 column is not sensitive to vertical transport and space-borne sensors provide global coverage. Column measurements have not contributed to carbon cycle studies in the past because their precision was not sufficient. This situation has changed in recent years and column measurements of precision better than 0.25% are now available for CO_2 from ground-based solar absorption measurements using FTIR-spectrometers. Spaceborne sensors show promising results and it is expected that data of sufficient precision will become available in the near future.

The tropics are among the key regions for the understanding of the atmospheric CO_2 budgets, but the tropics are not well constrained by measurements. Reasons for the lack of observations in the tropics include the inaccessibility of the tropical forests, the lower priorities given to these observations by developing countries as well as the difficulty

Table 1. Dry seasons in Paramaribo.

Short dry season (SDS)	Meteorological Northern Hemisphere	March–April	Northeast trade winds
Long dry season (LDS)	Meteorological Southern Hemisphere	August–November	Southeast trade winds

to long-term observations due to political reasons. The lack of measurements results in large uncertainties in the emissions and biosphere-atmosphere exchange fluxes in the tropics. Deforestation in the tropics has a significant contribution to the global CO_2 emissions, accounting for almost 20% of anthropogenic greenhouse gas emissions during the 1990s (Gullison et al., 2007). Another important issue is the uptake of CO_2 in the tropics by carbon sinks. Due to the lack of measurements the uncertainties about the sinks are high in the tropics (Guerney et al., 2002). Recent studies showed that a large set of atmospheric inverse model results, using only in-situ boundary layer measurements, were inconsistent with total column measurements and vertical aircraft profiles as a result of incorrect vertical transport in the models (Stephens et al., 2007; Yang et al., 2007). In contrast to previous studies Stephens et al. (2007) suggests that the tropics are not a net source, but that the uptake of CO_2 in the tropics balances deforestation.

Ground-based column CO_2 measurements in the tropics are performed at only two sites, at Darwin (Australia) in the outer tropics as part of the Total Carbon Column Observing Network (TCCON) and at the inner-tropical site at Paramaribo (Suriname), which is presented in this work. Ground-based column measurements will play a vital role for the validation of current and upcoming satellite measurements. For satellite validation it is highly important to have sufficient ground truth in the tropics. The high abundance of water vapour as well as the frequent occurrence of (subvisual) cirrus clouds have previously caused problems in tropical satellite retrievals of greenhouse gases (Frankenberg et al., 2008; Schneising et al., 2008).

In this paper we present co-located surface and column measurements of CO_2 from the inner-tropical measurement site at Paramaribo (Suriname), discuss improvements needed for the measurements and compare the obtained measurements to TM3 model simulations.

2 Measurements and data analysis

Solar absorption Fourier-transform near infrared measurements were performed at the Meteorological Service (MDS) in the south-western outskirts of Paramaribo, Suriname (5.8 N, 55.2 W) during six dry seasons between September 2004 and November 2007. Paramaribo is the capital and largest city of Suriname, with a population of about 250 000 people. Paramaribo is located in the northern, lowland coastal area. The southern part of Suriname consists of

tropical rainforest, covering about 80% of Suriname's land surface. The Intertropical Convergence Zone (ITCZ) migrates twice a year over the measurement site resulting in two dry and two rainy seasons. During the short dry season from February to March the measurement site belongs to the meteorological Northern Hemisphere and during the long dry season from August to November to the meteorological Southern Hemisphere (Table 1).

Solar absorption spectra were recorded by a Bruker 120M FTIR spectrometer using an 18 cm optical path difference. An InSb-detector, a CaF_2 beamsplitter and an optical infrasilglass filter were used for the measurements. The recorded spectra were analysed using the line-by-line code GFIT, developed at NASA/JPL (e.g. Toon et al., 1992). Profiles of pressure, temperature and relative humidity up to the 10 mbar level are taken from NCEP re-analysis data. Above 10 mbar a typical stratospheric profile was attached. The initial vmr-profiles are taken from the GFIT-package and are based on balloon observations at Ft Sumner (35° N, 104° W) using the JPL MkIV Interferometer. The initial vmr-profiles of CO_2 and O_2 are constant in time.

O_2 was analysed between $7765\,\mathrm{cm}^{-1}$–$8005\,\mathrm{cm}^{-1}$ (O_2 0–0 $^1\Delta g$–$^3\Sigma g^-$ band) and CO_2 between $6297\,\mathrm{cm}^{-1}$–$6382\,\mathrm{cm}^{-1}$ ($2\nu_1+2\nu_2+\nu_3$ band centered at $6348\,\mathrm{cm}^{-1}$). Spectral line parameters for the O_2 retrieval were taken from an updated version (December 2006) of the ATMOS database (Brown et al., 1996). For the CO_2 retrieval spectroscopic parameters from Toth et al. (2008) and for H_2O parameters from Jenouvrier et al. (2007) have been used. The pressure-weighted dry vmr of CO_2 was calculated by scaling the CO_2/O_2 column ratio with the mean O_2 vmr (0.2095), which is, to the degree required, constant in the atmosphere.

Surface air was collected with a portable sampling system consisting of a pump and glass flasks from October 2005– November 2007. The air samples were collected at the site of the FTIR measurements. For the campaigns in 2007 additional samples were taken at a coastal location north-west of Paramaribo, about 10 km away from the Meteorological Service (MDS). The flask samples were analyzed for CO_2, CH_4, CO, N_2O, SF_6, and H_2. The isotopic composition of CO_2 was also determined, except for the measurements during the LDS in 2007. Average relative precisions are 0.07% for CH_4 (1.3 ppb at atmospheric mixing ratio levels), 0.02% for CO_2 (0.08 ppm), 0.04% for N_2O (0.13 ppb), 0.3% for CO (0.5 ppb), 0.5% for SF_6 (0.03 ppt) and 0.4% for H_2 (2 ppb). The precision is 0.013‰ for $\delta^{13}C$ and 0.025‰ for $\delta^{18}O$.

3 Results

3.1 CO_2 in surface air

The air is transported from the Atlantic Ocean to the measurement site by the easterly tradewinds. During the short dry season (SDS) from February to March the air passes directly over the Surinamese coastal region to the measurement site. During the long dry season (LDS) from August to November the air passes frequently over French Guiana resulting in a longer passage of the air over the South American continent, which can also be seen in stronger impact by biomass burning during the LDS (Petersen et al., 2008). During both dry seasons the air might be impacted by emissions from the City of Paramaribo.

We have measured $\delta^{13}C$ in the samples to distinguish between different sources. For each of the measurement campaigns a very high linear correlation between the $\delta^{13}C$ and the inverse of CO_2 concentration has been observed (Fig. 1). This shows that the measurements can be described by mixing of background CO_2 with CO_2 from one local source (e.g. Pataki et al., 2003), assuming the $\delta^{13}C$ of the background CO_2 and the CO_2 from the local source are constant over the time of measurement campaigns. The $\delta^{13}C$ is not strongly changed by ocean exchange, but by the biosphere. Therefore it can be assumed that the local source is either the terrestrial biosphere or fossil fuel combustion. The intercept of the mixing line with the vertical axis corresponds to the isotopic composition of the local-source component. The $\delta^{13}C$ of the local source component is around $-25.3‰$ during the LDS and approximately $3‰$ higher during the SDS. Plant CO_2 uptake via photosynthesis discriminates against ^{13}C resulting in a smaller $\delta^{13}C$ in plant tissues compared to atmospheric CO_2. Plant respiration does not change the isotopic composition of CO_2, hence the CO_2 emitted to the atmosphere via plant respiration is also depleted in ^{13}C. The depletion depends on the type of carbon fixation during photosynthesis (C3, C4, CAM) and is strong for C3 plants and small for C4 plants and also depends on plant physiological parameters (Flanagan et al., 2005). Due to the biological origin of fossil energy sources, fossil fuel emissions are also depleted in ^{13}C. Depending on the type of fuel and its region of origin, the $\delta^{13}C$ ranges from $\delta^{13}C=-30‰$ to $-6.4‰$ for oil, natural gas has a signature of about $\delta^{13}C=-44‰$ and coal of about $\delta^{13}C=-24.1‰$ (Andres et al., 1999). The emissions in the City of Paramaribo are predominantly from cars and natural gas. The isotopic signature of the local source component as well as that the calculated CO_2 for the local source does not correlate with the measured CO in the flasks (not shown) suggests that the measurements are not strongly influenced by urban pollution and the local source component is the terrestrial biosphere. This is also supported by three samples from a remote location in the coastal region at the border to French Guiana, which are on the mixing line.
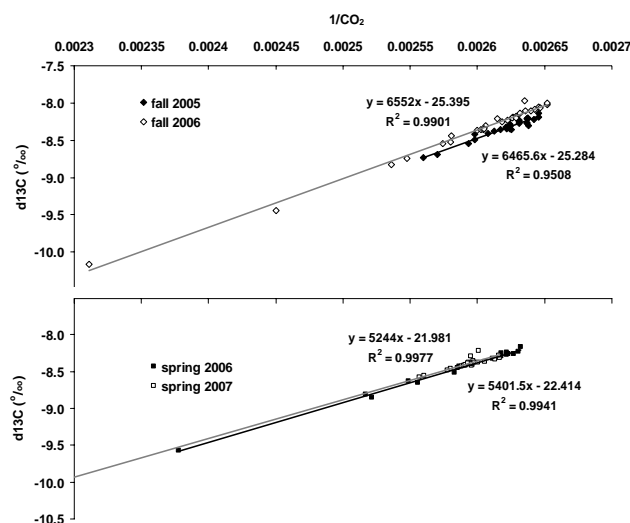
Fig. 1. $\delta^{13}C$ vs. $1/CO_2$ ("Keeling plot" e.g. Keeling, 1961) in flask samples collected at Paramaribo for the different measurement campaigns. The equation of the linear regression and its R^2 value is written next to the regression line.

Measured surface air mixing ratios of CO_2 vary between 377 ppm and 433 ppm. As inferred from the $\delta^{13}C$ the strong variation is most likely arising from emissions from the terrestrial biosphere during the passage of the air from the ocean to the measurement site. The background CO_2 mixing ratio has been calculated using the linear mixing lines (Fig. 1) as described in the following.

The linear relation in Fig. 1 can be described by

$$\delta^{13}C^{meas}=m\frac{1}{CO_2^{meas}}+\delta^{13}C^{source}, \tag{1}$$

where the values measured in the flask samples are CO_2^{meas} and $\delta^{13}C^{meas}$ and the intercept of the mixing line with the vertical axis is $\delta^{13}C^{source}$. The slope m can be calculated from the CO_2 and $\delta^{13}C$ in the two mixing components, namely background and local source:

$$m=\frac{\delta^{13}C^{backgnd}-\delta^{13}C^{source}}{\frac{1}{CO_2^{backgnd}}-\frac{1}{CO_2^{source}}}, \quad \text{where} \quad \frac{1}{CO_2^{source}}=0 \tag{2}$$

From Eqs. (1) and (2) it follows

$$\delta^{13}C^{meas}=\frac{\delta^{13}C^{backgnd}-\delta^{13}C^{source}}{\frac{1}{CO_2^{backgnd}}}\frac{1}{CO_2^{meas}}+\delta^{13}C^{source} \tag{3}$$

and

$$CO_2^{backgnd}=\frac{\delta^{13}C^{meas}-\delta^{13}C^{source}}{\delta^{13}C^{backgnd}-\delta^{13}C^{source}}CO_2^{meas} \tag{4}$$

For this calculation the $\delta^{13}C$ for background air, $\delta^{13}C^{backgnd}$, has been taken from NOAA/ESRL flask measurements on

Ascension Is. (ASC) for the LDS and from Ragged Point (RPB) for the SDS (White et al., 2009). $\delta^{13}C$ is only weakly influenced by ocean exchange and therefore not changed during transport from the two NOAA/ESRL sites to the east coast of South America. For each year the mean $\delta^{13}C$ and its standard deviation for the NOAA/ESRL flask measurements has been calculated for the time period 15 September–15 December for the LDS and 15 January–15 April for the SDS. This time period corresponds to approximately one month before the start and one month after the end of the measurement campaigns. The mean $\delta^{13}C$ at the NOAA/ESRL stations are -8.16 ± 0.05 ‰ for the LDS 2005, -8.31 ± 0.04 ‰ for the SDS 2006, -8.16 ± 0.04 ‰ for the LDS 2006 and -8.36 ± 0.07 ‰ for the SDS 2007.

For comparison global spatio-temporal CO_2 concentration fields have been calculated by the atmospheric tracer transport model TM3 driven with re-analysed meteorological data (NCEP) (Roedenbeck, 2005). Surface CO_2 fluxes supplied to the model comprise detailed representations of fossil fuel emissions, land biosphere exchange, and oceanic exchange, as well as an inversely calculated correction flux ensuring consistency with measured atmospheric CO_2 concentrations at many sites around the globe. Details about the model are described by Roedenbeck (2005), and data are available online http://www.bgc-jena.mpg.de/~christian.roedenbeck/download-CO2-3D/.

The TM3 model does not capture the high variations observed in the surface air samples (Fig. 2, upper panel). The surface samples are likely be influenced by local sources, which are averaged out in the large grid of the model of 3.8×5.0 degrees. Therefore we have compared the surface data corrected for the local source component with an eastward model grid, which mostly contains ocean. Due to the trade winds the airmasses pass this model grid prior to the measurements. The corrected mixing ratios compare well with TM3 model simulations for the long dry seasons (Fig. 2, lower panel). During the SDS 2007 the variation of $\delta^{13}C$ at Ragged Point results in higher uncertainties in the corrected mixing ratios compared to the other campaigns and the modeled values are within the errors of the corrected vmrs. During the SDS 2006 the corrected mixing ratios are on the average about 1–2 ppm lower than the model. The reason for the slightly worse agreement during the short dry season might arise from the large grid size of the model, which comprises part of the ITCZ during the short dry seasons.

3.2 Column averaged volume mixing ratios of CO_2

Previous studies have shown that the spectroscopic data of CO_2 and O_2 have an offset and a scaling factor is required to compensate this (Warneke et al., 2005; Washenfelder et al., 2006). To determine this scaling factor aircraft in situ measurements have been performed at several TCCON sites, demonstrating that the scaling factor is instrument-independent for the instruments used within TCCON and
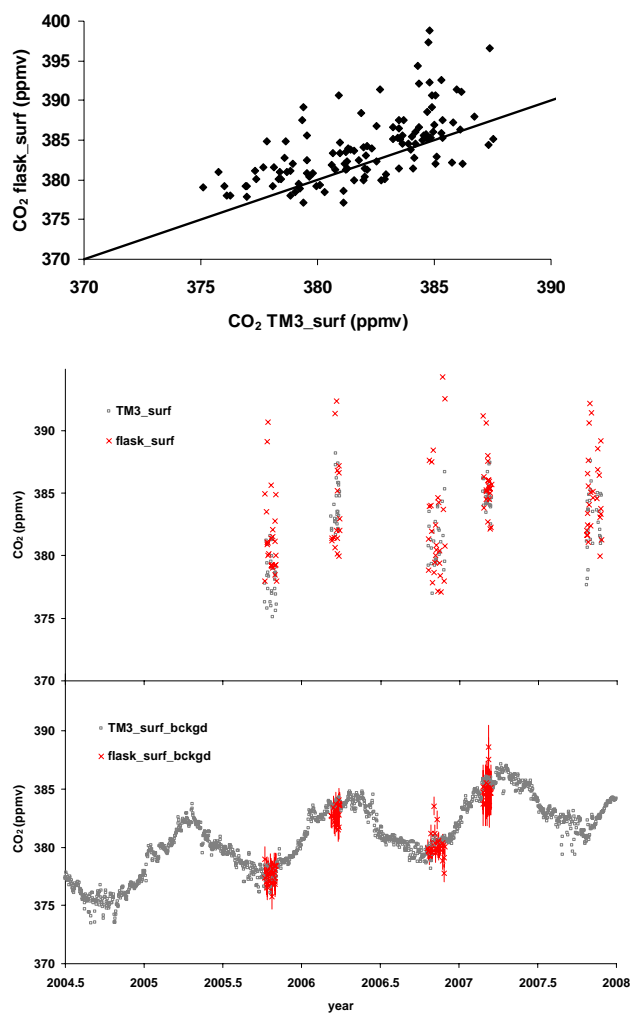

Fig. 2. Measured surface air mixing ratios of CO_2 compared to model simulations. CO_2 mixing ratios in surface air at Paramaribo (flask_surf) vary between 377 ppm and 433 ppm (values above 395 ppm not shown). In the upper and middle plots the flask data are compared with TM3 model simulations for the model grid containing Paramaribo (grid center 5.81° N 55.21° W). The lower panel shows model data for the adjacent model grid towards the east (grid center 5.81° N 50.21° W), mainly containing ocean compared with calculated background CO_2. The background CO_2 mixing ratio (flask_surf_bckgd) has been calculated using linear mixing lines (Fig. 1) and a background $\delta^{13}C$ from NOAA/ESRL flask measurements from Ascension Is. (ASC) and Ragged Point (RPB). $\delta^{13}C$ values from Ascension Is. are used for the LDS-campaigns, when the air at Paramaribo belongs to the Southern Hemisphere and $\delta^{13}C$ values from Ragged Point for the SDS campaigns, when the air at Paramaribo belongs to the Northern Hemisphere.

one scaling factor can be applied for the different TCCON-sites (Deutscher et al., 2010; Wunch et al., 2010). However, this single scaling factor can only be applied when using the Bruker 120/125HR instrument, which is the standard instrument type in TCCON. In Paramaribo we use the Bruker 120M spectrometer. Since this instrument type has

never been compared to the TCCON measurements. Instrumental artifacts might impact the measurements, which can be compensated by a different, unknown, scaling factor. Therefore these measurements cannot compared to the XCO_2 measured at the TCCON sites and cannot be used for inversions. Since the focus of this study is the comparison of the variabilities between measurements and model, we have choosen the scaling factor in the way that the comparison between measurement and model is most easily visible. For this comparison a scaling factor of 1.018 has been used to match the model simulations. This scaling factor results in 0.7% higher XCO_2 values compared to the scaling used in TCCON. It is important to assume that the scaling factor is constant throughout the measurement period, but this can be assured with our regular instrumental line shape measurements. In the future we plan to exchange the instrument and perform a side-by-side with a TCCON spectrometer prior the exchange.

The measurements of XCO_2 at Paramaribo show a higher scatter than the ones at the TCCON site at Spitsbergen (Warneke et al, 2005). Partly this can be attributed to a less stable instrumental lineshape (ILS) of the Bruker 120M spectrometer used at Paramaribo compared to the Bruker 120HR spectrometer used at Spitsbergen. However, the main reason is assumed to be the impact by clouds. Spectra at Paramaribo were recorded whenever possible and many spectra are impacted by the frequent occurrence of cirrus clouds. Keppel-Aleks et al. (2007) presented a method to correct for source brightness fluctuations caused by clouds which would help to overcome this problem. This method requires that the interferograms are recorded without AC coupling of the detector signal (DC recording), which is unfortunately not feasible with the instrument used at Paramaribo due to its analog-to-digital converter. To sort out the spectra, which are affected by clouds we use the column averaged vmr of O_2, which is, to the degree required, constant in the atmosphere. Only spectra with an O_2 vmr within 2.5% of the mean retrieved vmr of O_2 were used for this study.

The average of the XCO_2 was calculated for each measurement campaign (red diamonds in Fig. 3) at Paramaribo. For the southern hemishere (LDS) XCO_2 measurements are available for four consecutive years. The measurement campaigns in the LDS took place during the same months of each year. The mean annual increase of XCO_2 from 2004 to 2007 is about 2.2 ppm, which agrees well with the modeled annual increase. For the comparison with the model a representative averaging kernel and the retrieval a priori are taken into account. The measurements agree very well with the model simulations for the SDS and LDS in 2006. For the other LDS the mean measured XCO_2 are about 0.6 ppm higher and for the SDS 2007 about 1 ppmv lower than the model. Given the standard deviation of 0.9 ppm of the mean in the measurements the deviation from the model it is not significant and it can be concluded that the measurements agree with the model within the uncertainties.
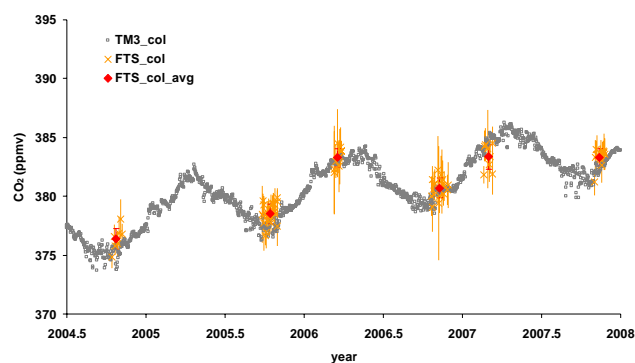
Fig. 3. Comparison of modeled and measured column averaged vmrs of CO_2. Daily averages (FTS_col) and averages for the measurement campaigns (FTS_col_avg) of column averaged vmrs of CO_2 have been calculated by scaling the column ratio CO_2/O_2 by the vmr of O_2 and a factor 1.018. The TM3 model simulations (TM3_col) have a grid size of $3.8° \times 5°$ and the center coordinate is $5.81°$ N $55.21°$ W. For XCO_2 the difference between the ocean grid cell used for the in situ comparison and the grid cell containing Paramaribo is small and therefore it is not relevant, which model grid is used for the XCO_2 comparison.

4 Conclusions

The first ground-based remote sensing measurements of XCO_2 for the inner tropics have been obtained at Paramaribo (Suriname). Due to the migration of the ITCZ over the measurement location the sampled air masses belong to the northern or Southern Hemisphere depending on the time of the year. The XCO_2 show a mean annual increase of 2.2 ppm between 2004 and 2007 for the Southern Hemisphere.

Co-located in-situ measurements of surface air exhibit a strong variation. Using the $\delta^{13}C$ determined in the air samples the strong variation has been attributed to local emissions from the terrestrial biosphere. $\delta^{13}C$ data from NOAA/ESRL measurements on Ascension Is. (ASC) and at Ragged Point (RPB) have been used to calculate the background volume mixing ratios of CO_2 from the surface measurements at Paramaribo.

Column averaged volume mixing ratios and background mixing ratios for surface air, derived from the in-situ measurements are compared with TM3 model simulations. Except for the surface background data for the SDS 2006 the background mixing ratios for the surface as well as the seasonality of the XCO_2 agree well with the model simulations. This demonstrates that the TM3 model is capable to simulate surface concentrations as well as the seasonality of column densities of CO2 correctly at this location in the tropics. The precision of the measurements is currently not high enough to decide if tropical uptake balances deforestation like suggested Stephens et al. (2007) or if the previous studies are correct that predict the tropics as a net source.

The current limitations of the solar absorption measurements at Paramaribo are the missing calibration and the high scatter of the data due to cirrus clouds. To overcome these limitations in the future we plan to exchange the Paramaribo FTS with a similar instrument that allows DC recording, resolving the problem of source brightness fluctuations due to cirrus clouds and to perform a side-by-side measurements with the TCCON spectrometer in Bremen prior shipment to Paramaribo.

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