Global CO_2 fluxes inferred from surface air-sample measurements and from TCCON retrievals of the CO_2 total column

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[1] We present the first estimate of the global distribution of CO_2 surface fluxes from 14 stations of the Total Carbon Column Observing Network (TCCON). The evaluation of this inversion is based on 1) comparison with the fluxes from a classical inversion of surface air-sample-measurements, and 2) comparison of CO_2 mixing ratios calculated from the inverted fluxes with independent aircraft measurements made during the two years analyzed here, 2009 and 2010. The former test shows similar seasonal cycles in the northern hemisphere and consistent regional carbon budgets between inversions from the two datasets, even though the TCCON inversion appears to be less precise than the classical inversion. The latter test confirms that the TCCON inversion has improved the quality (i.e., reduced the uncertainty) of the surface fluxes compared to the assumed or prior fluxes. The consistency between the surface-air-sample-based and the TCCON-based inversions despite remaining flaws in transport models opens the possibility of increased accuracy and robustness of flux inversions based on the combination of both data sources and confirms the usefulness of space-borne monitoring of the CO_2 column. **Citation:** Chevallier, F., et al. (2011), Global CO_2 fluxes inferred from surface air-sample measurements and from TCCON retrievals of the CO_2 total column, *Geophys. Res. Lett.*, *38*, L24810, doi:10.1029/2011GL049899.

1. Introduction

[2] The space-time gradients of CO_2 mixing ratios in the atmosphere are determined by CO_2 exchange with the Earth's surface and by atmospheric transport. Thus the joint availability of atmospheric measurements of CO_2 mixing ratios and of numerical models of the atmospheric transport allows the inference of CO_2 surface fluxes over the globe. This technique has been well established based on surface air-sample measurements [e.g., *Gurney et al.*, 2002], even though the sparse observation network only provides a blurred picture of the surface fluxes. Its extension to include retrievals of the total or partial columns of CO_2, such as those provided by satellite sounders [e.g., *Chevallier et al.*, 2005b], has been hampered by two difficulties. First, significant residual biases affect the satellite retrievals at the regional scale, while the variations in the column of a well-mixed gas like CO_2 are fairly small [*Chevallier et al.*, 2005a; *Wunch et al.*, 2011b]. Second, biases also affect the transport models that are needed to interpret the space-time gradients of columnar amounts in terms of surface fluxes [*Houweling et al.*, 2010].

[3] The best measurements of the CO_2 column are currently provided by the TCCON [*Wunch et al.*, 2011a], which consists of a series of sun-tracking Fourier transform spectrometers deployed at the Earth's surface. With 14 stations now operated throughout the world and several new sites planned, the wealth of data provided by TCCON is of growing interest to study the global carbon cycle [e.g., *Wunch et al.*, 2011a, and references therein; *Keppel-Aleks et al.*, 2011].

[4] This paper studies a first inference of global CO_2 surface fluxes from two years (2009–2010) of TCCON CO_2 column retrievals in order to investigate whether the current flaws of transport modelling prevent our inversion system to yield a realistic solution. The retrievals are deciphered in terms of grid-point surface fluxes by the variational

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inversion scheme of *Chevallier et al.* [2005b]. Our strategy for evaluation of the TCCON-inverted fluxes is twofold. First, we compare them with a classical inversion of surface air-sample measurements in terms of seasonal cycle and of annual sub-continental budgets. Second, the inverted fluxes are used as a boundary condition for a global atmospheric simulation of CO_2 to be compared with CO_2 mixing ratios measured from aircrafts during the HIAPER Pole-to-Pole Observations (HIPPO) campaigns [*Wofsy et al.*, 2011] and by the commercial aircraft–based observations of the Comprehensive Observation Network for TRace gases by AIr-Liner (CONTRAIL) database [*Machida et al.*, 2008].

[5] The inversion method and the various measurements are presented in Sections 2 and 3, respectively. Section 4 presents the results. Section 5 concludes the paper.

2. Inversion Method

[6] This study relies on the flux inversion scheme of *Chevallier et al.* [2005b] that implements Bayesian estimation in the form of the minimization of a cost function. The inversion system computes the Best Linear Unbiased Estimate (BLUE) of the CO_2 surface fluxes given the input information, their Bayesian uncertainty, and the number of degrees of freedom for signal [*Chevallier et al.*, 2007]. The BLUE fluxes are simply called *inverted fluxes* hereafter. The inversion system is operated twice here for the period 2009–2010, with the two computer runs differing by the input observations: one uses the TCCON measurements and the other one exploits surface air-sample measurements (see Section 3). In both cases, fluxes are estimated on a global longitude-latitude grid of regular mesh 3.75° × 2.5° at a temporal resolution of 8 days, with day-time and night-time separated. 3-hourly variations within the 8-day segments are prescribed *a priori* in the inversion. Prior fluxes come from the combination of a flux climatology, of emission inventories and of a model simulation as described by *Chevallier et al.* [2010]. The operator that links the flux space and the retrieval space in the inversion scheme is the general circulation model of the Laboratoire de Météorologie Dynamique (LMDZ) [*Hourdin et al.*, 2006], nudged to ECMWF winds. Tracer transport is simulated on the same 3.75° × 2.5° horizontal grid as the fluxes with 19 layers between the surface and the top of the atmosphere. The prior initial conditions (i.e., the 3D field of CO_2 at the start of the inversion window) are taken from the global analysis of surface air-sample measurements for the period 1988–2008 made by *Chevallier et al.* [2010]. They are adjusted within the two inversions. The present inversion configuration follows the one used by *Chevallier et al.* [2010] and will not be described here: only minor adaptations to process years 2009 and 2010 have been made.

3. Observations

[7] The TCCON monitors the column-averaged dry air mole fractions of CO_2 (hereafter X_{CO2}). The measurements for years 2009 and 2010 have been extracted from the TCCON database (http://tccon.ipac.caltech.edu/) on 1 October 2011 to serve as input to a first inversion. 14 stations provided some data during that period: Bialystok (Poland), Bremen (Germany), Darwin (Australia), Eureka (Canada), Garmisch (Germany), Izaña (Canary Islands, Spain), Karlsruhe (Germany), Lamont (OK, USA), Lauder (New Zealand), Orléans (France), Park Falls (WI, USA), Sodankylä (Finland), Tsukuba (Japan) and Wollongong (Australia). The TCCON X_{CO2} data correspond to daytime cloud-free conditions and are used here without any averaging. The data uncertainty, which is reported in the database in the form of standard deviations, range between 0.3 and 10 ppm, with a median of 0.8 ppm. It is quadratically summed here with estimates of the transport model error and of the representation error (set to 0.5 ppm standard deviation) to assign the observation errors in the inversion system. In addition, standard deviations are inflated (i.e., artificially increased) as follows, so that the system benefits from a diagonal observation covariance matrix even though the transport model errors likely make it dense [*Chevallier*, 2007]. For the time domain, it is hypothesized that correlated errors mainly operate at the sub-daily scale: noise cannot be damped by data density at this scale. The observation error variances are consequently multiplied each day and for each site by the number of data points for that day and for that site. In the spatial domain, the model correlated errors for the simulation of the total columns are assumed to be restricted to the five stations over France, Germany and Poland, where the TCCON is relatively dense: variances are empirically multiplied by 4 there. This study does not exploit the measurement averaging kernels, which induces errors of the order of 0.1 ppm that vary with the solar zenith angle, when simulating the X_{CO2} measurements [*Reuter et al.*, 2011].

[8] The surface air-sample measurements for the second inversion are mixing ratios of CO_2 (expressed as dry mole fractions) collected in flask air samples at various places in the world over land (from fixed sites) and over ocean (from commercial ships), or analysed *in situ* by continuous automated analyzers. From the station selection of *Chevallier et al.* [2010], 91 station records were found to have provided some data for years 2009 and 2010 by 6 July 2011, when they were extracted. Four databases are used: the NOAA Earth System Research Laboratory archive [*Conway et al.*, 2011; *Thoning et al.*, 2010; *Andrews et al.*, 2009], the CarboEurope atmospheric archive, the World Data Centre for Greenhouse Gases (WDCGG) archive and the Réseau Atmosphérique de Mesure des Composés à Effet de Serre (RAMCES) database. The station list is given in the auxiliary material.[1] The observed synoptic variability at each station is taken as a proxy of the observation uncertainty (which is driven by transport modeling errors) and is computed following *Chevallier et al.* [2010], with time-correlated errors for continuous measurements implicitly taken into account in the form of inflated variances, as for the first inversion.

[9] Aircraft measurements from two other databases provide additional observations of CO_2 mixing ratio that are kept out of the inversion system for independent evaluation of its results. The first archive comes from the CONTRAIL project [*Machida et al.*, 2008] that is being jointly conducted by NIES (National Institute for Environmental Studies), the MRI (Meteorological Research Institute), JAL (Japan Airlines International), JAMCO (JAMCO Corporation) and JAL-F (JAL Foundation). CONTRAIL gathers commercial aircraft-based observations for the period between 2005 and 2009. Data from take-off and landing at 28 airports in the world during year 2009 are used here. The second database

[1]Auxiliary materials are available in the HTML. doi:10.1029/2011GL049899.

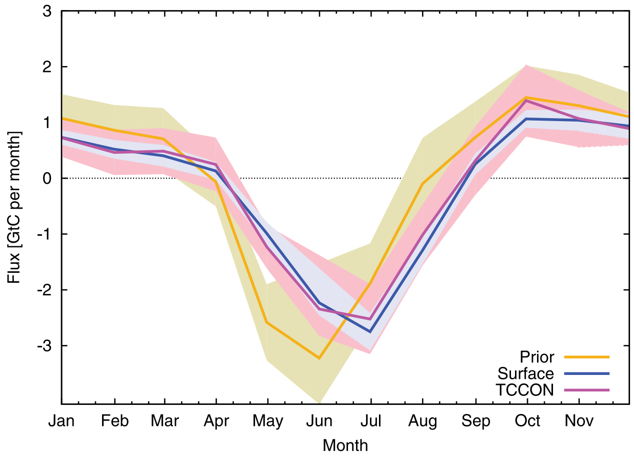

Figure 1. Seasonal cycle of the natural CO_2 fluxes over the terrestrial lands north of 20°N during 2009. The prior fluxes, fluxes from the TCCON-based inversion and the surface-air-sample-based inversion appear in gold, pink and blue, respectively. The shaded areas show the spread of the monthly flux uncertainty, as one standard deviation around the fluxes (overlaid). In the sign convention, positive fluxes correspond to a net carbon source into the atmosphere.

contains the HIPPO flights that sampled the atmosphere from pole to pole over the Pacific three times during 2009 (two transects) and 2010 (one transect), as described by *Wofsy et al.* [2011]. Each transect probes the atmosphere approximately between latitudes 67°S and 80°N with vertical profiles from 300 to 8500 m separated by 2.2° of latitude.

4. Results

[10] The inversion quality is assessed in the following by looking at simple large-scale quantities: the global annual mean CO_2 growth rate, the seasonal cycle over land in the northern hemisphere, and sub-continental annual budgets.

[11] The global annual mean CO_2 growth rate in the prior fluxes amounts to 2.8 and 3.2 ppm·yr^{-1}, for 2009 and 2010. The TCCON inversion reduces this growth rate to 1.8 and 2.2 ppm·yr^{-1}, for 2009 and 2010 respectively. The air-sample inversion yields similar numbers (1.7 and 2.2 ppm·yr^{-1}). Those inverted values are consistent with the more precise value inferred by NOAA with the method of *Conway et al.* [1994]: 1.6 ppm·yr^{-1} and 2.4 ppm·yr^{-1} for 2009 and 2010, respectively (ftp://ftp.cmdl.noaa.gov/ccg/co2/trends/co2_gr_gl.txt, accessed 8 September 2011).

[12] The 2009 mean seasonal cycle of the natural CO_2 flux over the lands north of 20°N is plotted in Figure 1 for the two inversions and for the prior fluxes. The natural CO_2 flux is obtained by subtracting a prescribed fossil fuel emission flux field (i.e., EDGAR version 4.1, http://edgar.jrc.ec.europa.eu/, scaled to 8.7 GtC·yr^{-1} as by *Friedlingstein et al.* [2010]) to the inverted net flux. The two inversions both reduce the global peak-to-peak amplitude by about one fourth compared to the prior and lengthen the uptake season by a few weeks. They converge towards a similar seasonal cycle, but the estimated uncertainty of the one deduced from TCCON is twice as large as the one constrained by the air samples. A comparison between the seasonal cycles at regional scale is shown in Figure S1 in the auxiliary material.

[13] Figure 2 displays the estimated annual land-to-atmosphere net CO_2 fluxes at the scale of the usual 11 large TransCom3 land regions [*Gurney et al.*, 2002], together with their uncertainty. Net fluxes correspond to the sum of all sources and sinks including fossil fuel emissions. The air-sea fluxes are not shown because the 14 TCCON stations mostly inform about the fluxes over land. The increments to the prior annual fluxes that have been generated by the two inversions are less spectacular than for the monthly fluxes: they only modestly change the flux interannual variations. In particular, the TCCON inverted fluxes include the prior fluxes and

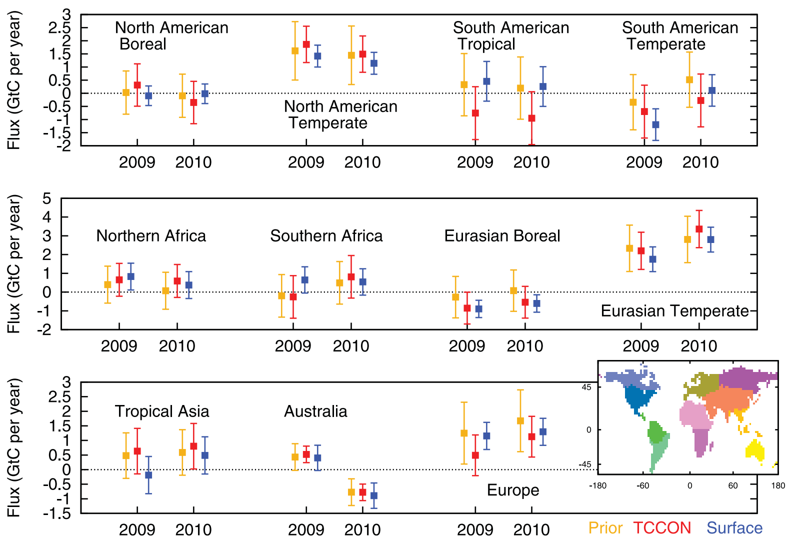

Figure 2. Net land-to-atmosphere CO_2 annual flux (including fossil fuel emissions) averaged over 11 land regions used in the TransCom3 program (see map in inset). For each region, the prior fluxes, the TCCON-inverted fluxes and the surface-air-sample-inverted fluxes are shown in gold, red and blue for 2009 and 2010. The 68% (1 standard deviation) confidence interval is reported around the mean values. In the sign convention, positive fluxes correspond to a net carbon source into the atmosphere.

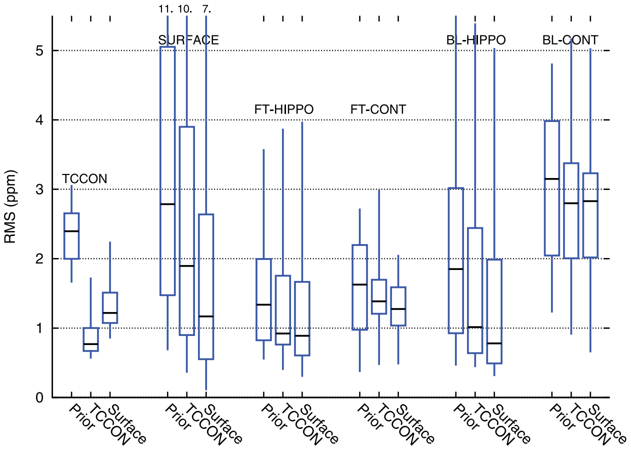

Figure 3. This box-and-whisker diagram shows the distribution of the RMS differences between the transport model simulations and six sets of atmospheric measurements for the 2009–2010 period, as indicated by the labels above the boxes. The six sets are the 14 TCCON stations, the 91 surface-air-sample records (SURFACE), the HIPPO flights in the free troposphere (FT-HIPPO), the CONTRAIL flights in the free troposphere (FT-CONT), the HIPPO flights in the boundary layer (BL-HIPPO) and the CONTRAIL flights in the boundary layer (BL-HIPPO). The simulations use the prior fluxes or the inverted fluxes as boundary conditions, as indicated on the horizontal axis. The spread of the statistics among the ensemble of surface stations (air sample and TCCON) or flights is represented by the whiskers. The boxes have horizontal lines at the lower quartile, the median and the upper quartile values for the measurement ensembles.

the air-sample-inverted fluxes within their one-sigma uncertainty in all regions for both years. The TCCON inversion reduces the flux uncertainty by more than 10% in all land regions except in regions North American Boreal, South American Temperate, Southern Africa and Tropical Asia, which are not sampled by the 14 TCCON stations. The air-sample inversion has systematically narrower uncertainty, except for Australia, where there are two TCCON sites. For Australia, a large decrease of the flux between 2009 and 2010 is seen, probably linked to heavy precipitation in 2010 in this region.

[14] The skill of the two inversions is indirectly evaluated by studying the evolution of the fit of the LMDZ simulation to the various measurements described in Section 3: the TCCON X_{CO2} retrievals, the surface air-sample measurements, the CONTRAIL measurements and the HIPPO measurements. An example of the measurement and of the simulations is shown in Figure S2 in the auxiliary material. Root mean square (RMS) differences are computed here on all individual measurements of a given site or flight. Distinction is made between the flight segment in the free troposphere, defined as the layer between 300 and 850 hPa, and the one in the boundary layer, defined by vertical pressures between 990 and 850 hPa. The distribution of the RMS statistics among the six ensembles is represented in the box-and-whisker plot of Figure 3. Each inversion is seen to improve the overall fit to the measurements, whether or not independent, compared to the prior. This improvement can still be seen when the prior fluxes are corrected with the global atmospheric growth rate provided by NOAA (following the *poor man's approach* detailed by *Chevallier et al.* [2010]), except for the HIPPO measurements in the free troposphere where the inversion impact is rather neutral in both cases (not shown). As expected, the best statistics against TCCON (respectively against the surface air-sample measurements) are obtained from the TCCON inversion (respectively the surface-air-sample inversion). Looking at the aircraft measurement statistics, which are independent of both inversions, the surface-air-sample inversion appears to provide slightly better RMS distributions than the TCCON inversion for the four datasets: the extreme RMS and the three RMS quartiles are usually better by a few tenths of ppm for the former inversion than for the latter one.

5. Conclusions

[15] Measurements of the CO_2 total column constrain the mass and distribution of carbon in the atmosphere. Provided that they are made with sufficient accuracy, they therefore allow monitoring of the variations of the carbon fluxes at the Earth's surface. The TCCON provides the first experimental opportunity to verify this concept. We find that the TCCON-inverted mean seasonal cycle north of 20°N, where 11 of the 14 stations are located, is strikingly similar to the one obtained from the surface air-sample inversion. Both inversions consistently show a large change in the amplitude and the phase of the cycle with respect to the prior. This result is remarkable given the inconsistency noted by *Yang et al.* [2007] between modelled and observed CO_2 vertical transport within the atmospheric column, for several atmospheric transport models, and given the large sensitivity of inversion systems to small biases in the X_{CO2} modelling [*Houweling et al.*, 2010]. The TCCON-inverted carbon budgets at sub-continental and annual scale are seen to be statistically consistent with the fluxes obtained from the surface air-sample measurements, but they generally have larger uncertainty. Finally, we show that, when used in a forward simulation of the atmospheric transport, the TCCON-inverted fluxes improve the fit to independent air-sample measurements made at a series of surface stations over the globe or made from aircraft by about a few tenths of a ppm (RMS). This exercise is particularly challenging because the air-sample measurements typically have a much smaller flux footprint than the total column. These results highlight the potential of space-borne monitoring of X_{CO2} from dedicated space-borne instruments, which started with the Japanese Greenhouse gases Observing SATellite (GOSAT) in January 2009.

[16] **Acknowledgments.** TCCON data were obtained from the TCCON Data Archive, operated by the California Institute of Technology from the website at http://tccon.ipac.caltech.edu/. This work was performed using HPC resources of DSM-CCRT and of [CCRT/CINES/IDRIS] under the allocation 2011-t2011012201 made by GENCI (Grand Equipement National de Calcul Intensif). It was co-funded by the European Commission under the EU Seventh Research Framework Programme (grants agreements 218793, MACC, and 212196, COCOS). Support for TCCON is provided by many national research support organizations that are listed on the TCCON web site. The authors are very grateful to the many people involved in the surface and aircraft measurement and in the archiving of these data.

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