

# **Elevated uptake of CO<sub>2</sub> over Europe inferred from GOSAT X<sub>CO2</sub> retrievals: a real phenomenon or an artefact of the analysis?**

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Estimates of the natural CO<sub>2</sub> flux over Europe inferred from in situ measurements of atmospheric CO<sub>2</sub> mole fraction have been used previously to check top-down flux estimates inferred from space-borne dry-air CO<sub>2</sub> column (X<sub>CO2</sub>) retrievals. Several recent researches have shown that CO<sub>2</sub> fluxes inferred from X<sub>CO2</sub> data from the Japanese Greenhouse gases Observing SATellite (GOSAT) and the Scanning Imaging Absorption Spectrometer for Atmospheric CHartography (SCIAMACHY) have a larger seasonal amplitude and a more negative annual net CO<sub>2</sub> balance than those inferred from the in situ data. The cause of this enhanced European uptake of CO<sub>2</sub> is still unclear, with some groups clearly arguing that this is a genuine scientific phenomenon. Here, we put forward an alternative hypothesis and show that realistic levels of bias in the GOSAT data can result in an erroneous elevated uptake over Europe. We use a global flux inversion system to examine the relationship between observations biases and estimates of European CO<sub>2</sub> uptake. We establish a reference in situ inversion that uses an Ensemble Kalman Filter (EnKF) to assimilate conventional surface mole fraction observations and the X<sub>CO2</sub> retrievals from the surface-based Total Carbon Column Observing Network (TCCON). The same EnKF system is also used to assimilate two independent versions of GOSAT X<sub>CO2</sub> data. We find that the GOSAT-inferred European terrestrial biosphere uptake peaks during the summer, similar to the reference inversion, but the net annual flux is 1.40±0.19 GtC/a compared to a value of 0.58±0.14 GtC/a for our control inversion that uses only in situ data. To reconcile these two estimates, we have performed a series of numerical experiments that assimilate observations with added biases or assimilate synthetic observations for which part or all of the GOSAT XCO<sub>2</sub> data are replaced with model data. We find that for our global flux inversions, a large portion (60-90%) of the elevated European uptake inferred from GOSAT data in 2010 is due to retrievals outside the immediate European region, while most of the remainder can be explained by a sub-ppm retrieval bias over Europe. We have used a data assimilation approach to estimate monthly GOSAT X<sub>CO2</sub> biases from the joint assimilation of in situ observations and GOSAT X<sub>CO2</sub>

retrievals. We find a monthly varying bias of up to 0.5 ppm can explain an overestimate of the annual sink of up to 0.20 GtC/a. Our results highlight the sensitivity of the flux estimates to regional observation biases, which have not been fully characterized by the current observation network. Without further dedicated measurements we cannot prove or disprove that European ecosystems are taking up a larger than expected amount of CO<sub>2</sub>.

## 1. Introduction

Observed atmospheric variations of carbon dioxide (CO<sub>2</sub>) are due to atmospheric transport and surface flux processes. Using prior knowledge of the spatial and temporal distribution of these fluxes and atmospheric transport it is possible to infer (or invert for) the a posteriori estimate of surface fluxes from atmospheric concentration data. The geographical scarcity of such observations precludes robust flux estimates for some regions due to large uncertainties associated with meteorology and a priori fluxes. Arguably, top-down estimate of regional CO<sub>2</sub> fluxes, particularly at tropical and high northern latitudes, has not been significantly improved for more than a decade [Gurney et al., 2002; Peylin et al., 2013], reflecting the difficulty of maintaining a surface measurement programme over vulnerable and inhospitable ecosystems. Atmospheric transport model errors compound errors introduced by poor observation coverage, resulting in significant differences between flux estimates on spatial scales < O(10,000 km) [e.g. Law et al., 2003; Yuen et al., 2005; Stephens et al., 2007]

The Greenhouse gases Observing SATellite (GOSAT), a space-borne mission launched in a sun-synchronous orbit in early 2009, was purposefully designed to measure CO<sub>2</sub> columns using short-wave IR wavelengths. Validation of current X<sub>CO2</sub> column retrievals using co-located upward-looking FTS measurements of the Total Carbon Column Observing Network (TCCON) [Wunch et al., 2011] show a standard deviation of 1.6-2.0 ppm (e.g., Parker et al., 2012). Their global biases are typically smaller than 0.5 ppm [Oshchepkov et al., 2013]. The disadvantage of using the TCCON is that sites are mainly at northern extra-tropical latitudes with little or no coverage where our knowledge of the carbon cycle is weakest. Many surface flux estimation algorithms are particularly sensitive to systematic errors so that sub-ppm biases can still significantly change the patterns of regional flux estimates [Chevallier et al., 2010]. This is further complicated by the seasonal coverage of GOSAT data at high latitudes during winter months when solar zenith angles are too large to retrieve reliable values for X<sub>CO2</sub> [Liu et al., 2014].

Several independent studies have shown that regional flux distributions inferred from GOSAT X<sub>CO2</sub> retrievals are significantly different from those inferred from in situ data [Basu et al., 2013; Deng et al., 2013; Chevallier et al., 2014]. In particular, these studies report a larger-than-expected annual net emission over tropical continents and a larger-than-expected net annual uptake over Europe. While the GOSAT inversions suffer from larger observation errors, atmospheric transport errors and issues from the seasonal coverage of higher latitudes, the in situ inversions are also unreliable over many regions due to poor coverage and atmospheric transport errors. Inter-comparisons revealed significant inconsistency in regional flux estimates inferred from in-situ observations by using different inversion systems, over many regions important for global carbon cycle, including Europe [Peylin et al., 2013]. Consequently, there is an ongoing debate about whether a recent study that shows a large European uptake of CO<sub>2</sub> [Reuter et al., 2014] reflects a real phenomenon or is an artefact due to deficiencies both in the observations and in the inverse modelling.

We report the results from a small set of experiments that show systematic bias can introduce a large difference between European fluxes inferred from GOSAT and those inferred from in situ data by using a global flux inversion approach. In the next section we provide an overview of the inverse model framework used to interpret data from the in-situ observation network (including both the conventional surface observation network and the relatively new TCCON network), and from the space-based GOSAT  $X_{CO_2}$  data. In section 3, we describe and present results from two groups of global inversion experiments that characterize the role of systematic bias in regional flux estimates. Further experiments for quasi-regional flux inversions are presented in Appendix A. In section 4, we use a modified version of the inverse model framework to estimate monthly biases by jointly assimilating all data. We conclude the paper in section 5.

## 2. Description and Evaluation of Control In-situ and GOSAT Experiments

We use the GEOS-Chem global chemistry transport model to relate surface fluxes to the observed variations of atmospheric  $CO_2$  concentrations [Feng et al., 2009] at a horizontal resolution of  $4^\circ \times 5^\circ$ , driven by GEOS-5 meteorological analyses from the Global Modeling and Assimilation Office Global Circulation Model based at NASA Goddard Space Flight Centre. We use an Ensemble Kalman Filter (EnKF) [Feng et al., 2009; 2011] to estimate regional fluxes from in situ or GOSAT observations for three years from 2009-2011, but we focus on 2010 to minimize error due to spin-up and edge effects. We estimate monthly fluxes on a spatial distribution that is based on TransCom-3 [Gurney et al., 2002] with each continental region further divided equally into 12 sub-regions and each ocean region further divided equally into 6 sub-regions. As a result, we estimate fluxes for 199 regions, compared to 144 regions we have used in previous studies [Feng et al, 2009; Chevallier et al., 2014].

In all global inversion experiments we assume the same set of a priori flux inventories, including: (1) monthly fossil fuel emissions [Oda and Maksyutov, 2011]; (2) weekly biomass burning emissions (GFED v3.0) [van der Werf et al., 2010]; (3) monthly oceanic surface  $CO_2$  fluxes [Takahashi et al., 2009]; and (4) 3-hourly terrestrial biosphere-atmosphere  $CO_2$  exchange [Olsen and Randerson, 2004]. We assume that the a priori uncertainty for each land sub-region is proportional to a combination of the net biospheric emission (70%) at the current month, and its annual variation (30%). We also assume that the a priori errors are correlated with each other with a spatial correlation length of 800 km, and a temporal correlation of 1 month [Chevallier et al., 2014]. We then determine the coefficient for the assumed a priori uncertainty by scaling the aggregated annual uncertainty over all 133 land sub-regions to 1.9 GtC/a. In particular, the resulting annual a priori uncertainty for European region is about 0.52 GtC/a, with the monthly uncertainty varying from 2.0 GtC/a for the summer months to about 0.8 GtC/a for winter months, which is generally larger than the a priori monthly uncertainty used by Deng et al. (2014). Prior uncertainties over oceans are determined under similar assumption but with a longer spatial correlation (1500 km), and a smaller aggregated annual error (0.6 Gt/a). Experiments show that doubling the a priori uncertainty increases the European uptake inferred from GOSAT data by about 0.21 GtC/a (from 1.40 GtC/a to 1.61 GtC/a), compared to a smaller increase of 0.09 GtC/a for the in situ inversion (from 0.58 GtC/a to 0.67 GtC/a).

The control inversion experiment (INV\_TCCON in table 1 and the red solid line in Figure 1) assimilates in situ observations, including the conventional surface observations at 76 sites [Feng et al., 2011] and, in particular, the total column  $X_{CO_2}$  retrievals from all the TCCON sites of the GGG2014

dataset (see Wennberg et al., 2014, and <https://tcccon-wiki.caltech.edu> for more details) to improve observation constraints. We use daytime (09:00 to 15:00 local time) mean TCCON retrievals, with the observation errors determined by the standard deviation about their daytime mean. To account for the inter-site biases as well as the model representation errors, we enlarge the TCCON observation errors by 0.5 ppm. Including TCCON observations increases the annual net uptake over Europe in 2010 from 0.49 GtC/a, as inferred from surface observations only, to 0.58 GtC/a. The increase is mainly due to a larger summer uptake. TCCON data also reduce the a posteriori uncertainty by about 15% from 0.16 GtC/a to 0.14 Gt/a. However considering the limited spatial resolution (only 12 sub regions for the whole TransCom European region), and unquantified model transport and representation errors, we anticipate that the complete a posteriori uncertainty is larger than the value estimated by the inversion system itself, as suggested by large inter-model variations found for in situ inversions [e.g., Peylin et al., 2013].

Evaluation shows that the INV\_TCCON a posteriori model concentrations agrees well with the independent HIAPER Pole-to-Pole Observations (HIPPO-3) aircraft measurements below 5 km in 2010 [Wofsy et al., 2010], with a small bias of 0.05 ppm, and a sub-ppm standard deviation of 0.87 ppm (Figure 2).

For the two control GOSAT inversions (Figure 1), we use two independent data sets: (1)  $X_{CO_2}$  retrievals from JPL ACOS team (v3.3) [Osterman et al., 2013] (INV\_ACOS); and (2) the full-physics  $X_{CO_2}$  retrievals (v4.0) from the University of Leicester [Cogan et al., 2012] (INV\_UOL). For both data sets, we assimilate only the H-gain data over land regions, and apply the bias corrections recommended by the data providers. We double the reported observation errors, as suggested by the retrieval groups.

As a performance indicator for our ability to fit fluxes to observed  $X_{CO_2}$  concentrations, we compare a posteriori model concentrations with GOSAT  $X_{CO_2}$  retrievals and show that INV\_ACOS and INV\_UOL agree much better than INV\_TCCON. For example, the bias against ACOS  $X_{CO_2}$  retrievals is -0.45 ppm for INV\_TCCON and 0.02 ppm for INV\_ACOS with a corresponding reduction in the global standard deviation from 1.69 ppm to 1.57 ppm. However comparison of GOSAT a posteriori concentrations against independent HIPPO-3 measurements is worse than INV\_TCCON with a positive bias of 0.47 ppm and 0.66 ppm for INV\_ACOS and INV\_UOL, respectively, which are mainly caused by the overestimation of  $CO_2$  concentrations (~1.5-2.0 ppm) at low latitudes (Figure 2).

### 3. Results

Figure 1 (and Table 1) compares the monthly and annual natural fluxes in 2010 over Europe for the three inversion experiments (INV\_TCCON, INV\_ACOS, and INV\_UOL). The three inversions show similar summer uptake values in June 2010 (0.69 GtC/m for INV\_TCCON and ~0.72 GtC/m for GOSAT inversions), which are generally consistent with other GOSAT inversion experiments (e.g., Deng et al., 2014; Chevallier et al., 2014). But the GOSAT inversions have an annual net uptake of about  $1.40 \pm 0.19$  GtC/a compared to the in situ inversion of  $0.58 \pm 0.14$  GtC/a. Figure 1 also shows significant differences between their monthly flux estimates in early spring and winter when there is only sparse GOSAT observation coverage, particularly over northern Europe. Both INV\_UOL and INV\_ACOS have a cumulative total of about 0.51 GtC more uptake than INV\_TCCON during February-April of 2010, with a further 0.37 GtC uptake accumulated over the following summer and autumn. This larger uptake is partially cancelled out by larger emissions (0.17-0.08 GtC) at the end of 2010.

As mentioned above, Figure 2 shows our evaluation of the associated a posteriori atmospheric mole fractions against HIPPO-3 measurements [Wofsy et al., 2010], which are mainly over the Pacific. As we are focused on European fluxes, Figure 2 presents further evaluation using descending and ascending profile observations over two European airports from the CONTRAIL experiment [Machida et al., 2008]. We calculate monthly mean CONTRAIL measurements during 2010 using data below 3 km, where there is greater sensitivity to local surface fluxes. Our current model resolution precludes small-scale sources (or sinks) so we expect model bias. We find that INV\_TCCON agrees best with CONTRAIL observations, in particular at the beginning of the 2010, partially reflecting the poor GOSAT  $X_{CO_2}$  coverage over Europe during the winter and early spring. However, we cannot conclude from the slightly degraded agreement with CONTRAIL (as well as with HIPPO-3) that the European uptake inferred from GOSAT data is incorrect, because unaccounted small local emissions/sinks, and model transport errors can affect the comparison against aircraft observations.

Figure 3 also presents an additional model simulation forced by a hybrid flux (denoted by the magenta broken line) where the INV\_TCCON a posteriori fluxes outside Europe are replaced by the results from INV\_ACOS. The resulting  $CO_2$  concentrations from these hybrid fluxes are, as expected, higher than the a posteriori model concentrations for INV\_ACOS because of the larger European emissions (i.e., less uptake) inferred by INV\_TCCON. But they are also systematically higher than the INV\_TCCON simulation, in particular during spring months, despite the same European fluxes being used to force these two simulations. This suggests an overestimate of  $CO_2$  transported into the European region by the GOSAT inversions. Further comparison of the INV\_TCCON simulation and the hybrid run reveals that systematic differences in the inflow into the European domain can affect the atmospheric  $X_{CO_2}$  gradient across this region. In the INV\_TCCON simulation, the mean  $X_{CO_2}$  difference between east (east of  $20^\circ E$ ) and west (west of  $20^\circ E$ ) Europe is  $\sim 0.04$  ppm for May, 2010, which is increased to 0.16 ppm in the hybrid run (cf. E-W  $X_{CO_2}$  gradient of -0.20 ppm for GOSAT ACOS data).

To understand the differences between the INV\_TCCON and GOSAT inversions, we conducted two groups of sensitivity tests (see Table 1 and Figure 4). First, we replaced all or part of the GOSAT  $X_{CO_2}$  retrievals assimilated in INV\_ACOS with those from a model simulation forced by the a posteriori fluxes from INV\_TCCON. In experiment INV\_ACOS\_MOD\_ALL (Figure 4), where we replace all GOSAT data with  $CO_2$  concentrations inferred from INV\_TCCON, we reproduce INV\_TCCON with small exceptions at beginning of 2010, reflecting the seasonal variation in GOSAT coverage. In a related experiment INV\_ACOS\_MOD\_NOEU for which we replace  $X_{CO_2}$  retrievals outside Europe with the model simulation, the differences between the GOSAT and in situ inversions are significantly reduced, particularly over the period with limited observation coverage, although the actual  $X_{CO_2}$  retrievals are still assimilated over Europe. The simulated GOSAT data outside Europe reduces the estimate of European uptake from 1.40 GtC/a to 0.88 GtC/a. In other words, the GOSAT observations outside the European region are responsible for about 60% (0.52 GtC/a) of the total enhanced European sink (0.82 GtC/a) with the remainder (0.30 GtC/a) due to observations taken directly over Europe. For INV\_UOL, when we replace the  $X_{CO_2}$  data outside Europe by the a posteriori INV\_TCCON model simulations, European uptake is reduced to 0.67 GtC/a (INV\_UOL\_MOD\_NOEU, Table 1), indicating an external contribution of nearly 90% to the enhanced uptake of 0.82 GtC/a. Together with Figure 2, these results suggest that GOSAT inversions appear to produce an overestimated  $CO_2$  inflow, which will make the fitted European flux have to compensate via mass balance by being erroneously too low even when un-biased GOSAT  $X_{CO_2}$  data are assimilated over the immediate European region. We find similar effects in the quasi-regional

inversions (Figure A1 in Appendix A), where only observations within European region are assimilated, with flux estimates from INV\_TCCON or from INV\_ACOS being used to provide lateral boundary conditions around Europe.

Second, we crudely demonstrate how regional bias could explain the remaining discrepancy of up to 0.30 GtC/a between GOSAT and in situ inversions over Europe. In our experiment INV\_ACOS\_SPR\_0.5ppm, we add a bias of +0.5 ppm to the GOSAT ACOS retrievals within Europe taken in February-April, inclusively, which effectively reduces the uptake by 0.1 GtC/a from 1.40 GtC/a to 1.30 GtC/a. Similarly, when the bias of +0.5 ppm is added to the GOSAT data taken in June-August we find a larger reduction of 0.15 GtC/a for the summer peak uptake (INV\_ACOS\_SUM\_0.5ppm), partially due to a larger a priori uncertainty and denser GOSAT coverage during the summer. These results emphasize the importance of characterizing sub-ppm regional bias to avoid erroneous flux estimates.

#### 4. Bias estimation.

Here we demonstrate a simple approach to quantify systematic bias in  $X_{CO_2}$  retrievals based on a simple on-line bias correction scheme. We assimilate the GOSAT  $X_{CO_2}$  retrievals together with the in situ and TCCON observations in two experiments: INV\_ACOS\_INS and INV\_UOL\_INS (Table 1). We also include monthly GOSAT  $X_{CO_2}$  regional biases over 11 TransCom land regions [Gurney et al., 2002] as parameters to be inferred together with surface fluxes from the joint assimilation of in-situ and satellite observations. To investigate the spatial pattern of the  $X_{CO_2}$  biases within Europe, we split Europe into West Europe (west of 20°E) and East Europe (east of 20°E). We assume that a priori for monthly biases is  $0.0 \pm 0.5$  ppm. For simplicity, we have assumed the a priori errors for regional  $X_{CO_2}$  biases are not correlated. Compared to the off-line comparisons between GOSAT  $X_{CO_2}$  retrieval and model concentrations, the main advantage of the on-line bias estimation is that the uncertainties associated with errors in flux estimates can be partially taken into account. However, biases derived by this approach reflect the systematic difference between the model simulation and GOSAT data over large (continental) regions, which also contains systematic model (such as transport and representation) errors. In addition, the inversion results are affected by the relative weights assigned to different data sets, as well as by the relative prior uncertainty assumed for surface fluxes and for the observation bias. The seasonal variation of the mean  $CO_2$  concentration is an important sign of the underlined biosphere seasonal cycle. We show in Appendix A that when we inflate the a priori uncertainty for the assumed observation bias, the observation constraints on flux estimate will become weaker. Also, the on-line bias correction is only effective for detecting and correcting bias at specified patterns, which may increase the sensitivity to other uncharacterized systematic errors. Despite these weaknesses, a joint data assimilation approach can exploit complementary constraints from in-situ and satellite  $X_{CO_2}$  data: for example there are a few GOSAT observations over northern Europe during autumn and winter months, while eastern Europe have few in situ observations. We have also limited the a priori uncertainty for the monthly observation biases to 0.5 ppm.

In the joint inversions INV\_ACOS\_INS and INV\_UOL\_INS, the annual European uptake is estimated to be 0.62 GtC/a, and 0.67 GtC/a, respectively (Table 1), which are close to the reference value of 0.58 GtC/a inferred from the in situ observations. To test the impact of the on-line bias correction, we set the a priori uncertainty of regional  $X_{CO_2}$  bias to be 0.01 ppm so that on-line bias correction is effectively turned off. As a result, the annual European uptake for INV\_ACOS\_INS is increased by

0.15 GtC to 0.77 GtC/a, which is close to INV\_ACOS\_MOD\_NOEU, but about 55% of the GOSAT only inversions (1.40 GtC/a).

Figure 5 shows the estimated monthly biases in ACOS and UOL  $X_{CO_2}$  retrievals over East and West Europe during 2010. Monthly biases are typically smaller than 0.5 ppm over the two regions, but have different seasonal cycles. Additional experiment shows that after ACOS  $X_{CO_2}$  data over Europe have been corrected for the inferred biases, the European annual uptake by INV\_ACOS is reduced by 0.20 GtC/a, representing more than half of the contribution from GOSAT observations within Europe. This result is consistent with our sensitivity tests. The effect of bias correction is much smaller for INV\_UOL (about 0.07 GtC/a), because of the different bias patterns. Differences in GOSAT  $X_{CO_2}$  retrievals and their effects on regional flux estimates have also been investigated in previous studies (e.g., Takagi et al., 2014).

## 5. Discussion and Conclusions

We used an ensemble Kalman Filter to infer regional  $CO_2$  fluxes from three different data sets: 1) surface observations and TCCON  $X_{CO_2}$  retrievals; 2) GOSAT  $X_{CO_2}$  retrievals by JPL ACOS team; and 3) GOSAT  $X_{CO_2}$  retrievals by University of Leicester. Our results, consistent with previous studies, show that these GOSAT data in a global flux inversion context result in a significantly larger European uptake than inferred from in situ data during 2010.

We showed using sensitivity experiments that a large portion (60-90%) of the enhanced European uptake of  $CO_2$  appears to be related to the systematically higher model  $CO_2$  mass being transported into Europe, due to the assimilation of GOSAT  $X_{CO_2}$  data outside the European region. We find some evidence using aircraft observations over the Pacific that GOSAT a posteriori fluxes result in higher  $CO_2$  concentration over lower latitudes. But limited observation coverage and unaccounted model errors prevent us from confidently concluding that GOSAT  $X_{CO_2}$  data are biased high or low. Our global and quasi-regional (Appendix A) flux inversion experiments show that the main consequence of the elevated  $CO_2$  inflow to the European domain is that the European uptake must increase because of mass balance, even when GOSAT  $X_{CO_2}$  retrievals within the European domain are not biased. A crude sensitivity test (INV\_ACOS\_OUT\_0.5ppm) shows that reducing ACOS  $X_{CO_2}$  data outside the European region by 0.5 ppm will reduce European annual uptake from 1.40 GtC/a to 0.98 GtC/a. Erroneous interpretation of  $X_{CO_2}$  data can result from analyses if biased boundary conditions are not addressed. However, as shown in Appendix A, a gross mis-characterization and correction of bias may weaken observation constraints, which can also lead to erroneous flux estimates.

We showed using sensitivity tests that sub-ppm bias can explain the remaining 0.30 GtC/a flux difference between the in situ inversion and INV\_ACOS after accounting for biased boundary conditions. By simultaneously assimilating the in situ and GOSAT observations to estimate surface fluxes and monthly  $X_{CO_2}$  biases, we infer a monthly observation bias that is typically less than 0.5 ppm over East and West Europe, but is able to cause an elevated sink of up to 0.20 GtC/a. The inferred monthly biases for UOL  $X_{CO_2}$  are also not the same as the ACOS  $X_{CO_2}$  data, particularly over West Europe during the summer months. This level of sensitivity of regional flux estimate to time-varying sub-ppm observation bias highlights the challenges we face as a community when evaluating  $X_{CO_2}$  retrievals using current observation networks.

Flux estimates are sensitive to a priori assumptions, idiosyncrasies of applied inversion algorithms, and the underlying model atmospheric transport [Chevallier et al., 2014; Peylin et al., 2014; Reuter et al., 2014]. Possible presence of regional observation biases further complicates the inter-comparisons of flux estimates based on different inversion approaches, as they may have different sensitivities to certain observation biases. In our assimilation of ACOS  $X_{CO_2}$  retrievals, we find that doubling the a priori flux error (INV\_ACOS\_DBL\_ERR) increases the estimated European uptake from 1.40 GtC/a to 1.61 GtC/a, consistent with the hypothesis on the increased vulnerability to the observation biases both within and outside Europe when using weak a priori constraints. In contrast, doubling the a priori flux errors only increases the uptake by 0.05 GtC/a to 0.67 GtC/a for the joint data assimilation (INV\_ACOS\_INS\_DBL\_ERR), with very little changes in the estimated biases (not shown). Examples in Appendix A also demonstrate different responses to regional and sub-regional biases before and after an on-line scheme is used to correct the systematic error across Europe. These differences emphasize the need for a closer examination of the responses of the inversion systems to the assimilated observations, as well as to their possible biases, to help understand the inter-model variations in estimated regional fluxes.

Complicated interactions between observations and the assimilation system also mean that our present study does not exclude other possible causes for the elevated European uptake reported by previous research from assimilation of GOSAT data. Instead, it highlights the adverse effects of possibly uncharacterized regional biases in current GOSAT  $X_{CO_2}$  retrievals that can attract erroneous interpretation of resulting regional flux estimates. A more thorough evaluation of the  $X_{CO_2}$  retrievals using independent and sufficiently accurate/precise observations is urgently required to increase the confidence of regional  $CO_2$  flux estimates inferred from space-based observations. Without additional observations, we cannot rule out either the lower European uptake estimate of around 0.6 GtC/a (by the in-situ inversion INV\_TCCON and the joint inversion INV\_ACOS\_INS and INV\_UOL\_INS) or the higher European uptake estimate of around 1.40 GtC/a (by GOSAT only inversions). There is also no sufficient reason to believe that the mean value among these diverse estimates is more reliable, because our study suggests that small systematic errors can result in significant differences in the estimated fluxes, and the influences of random errors have also not been fully quantified. The observational density required to infer flux estimates over a limited spatial domain such as Europe is crucial. For the time frame of this analysis, the TCCON network provided good coverage for Europe, North America, South East Asia and Australia/New Zealand. Great efforts were also taken to reduce inter-station biases. However, tropical stations that might provide better constraints for Southern Hemisphere air entering the European domain only came online after 2012. In the future, the TCCON observations may be supported by smaller and more mobile FTIR instruments that could be easier to set up in remote locations - at least on a campaign basis.

In the future, to achieve consistent flux estimates from assimilating multiple observation sources using different inversion approaches, we need to better quantify observation and model errors, and need to better understand the sensitivity of each inversion system to the assimilated observations as well as to their possible biases. It is difficult to develop a robust bias correction scheme before properly characterizing observation biases and the responses by the inversion system.

## Acknowledgements

LF and PIP designed the experiments and wrote the paper, RP provided the GOSAT XCO<sub>2</sub> data and comments on the paper, and NMD, DGF, RK, IM, and RS provided access to TCCON XCO<sub>2</sub> data and comments on the paper. Work at the University of Edinburgh was partly funded by the NERC National Centre for Earth Observation (NCEO). PIP gratefully acknowledges funding from the NCEO and his Royal Society Wolfson Research Merit Award. Work at the University of Leicester was funded by NCEO and the European Space Agency Climate Change Initiative (ESA-CCI). The TCCON Network is supported by NASA's Carbon Cycle Science Program through a grant to the California Institute of Technology. The TCCON stations from Bialystok, Orleans and Bremen are supported by the EU projects InGOS and ICOS-INWIRE, and by the Senate of Bremen. TCCON measurements at Eureka were made by the Canadian Network for Detection of Atmospheric Composition Change (CANDAC) with additional support from the Canadian Space Agency. The authors thank the NASA JPL ACOS team for providing their X<sub>CO<sub>2</sub></sub> retrievals. We also thank the CONTRAIL and HIPPO team for their observations used in our validations. We thank G. J. Collatz and S. R. Kawa for providing NASA Carbon Monitoring System Land Surface Carbon Flux Products: <http://nacp-files.nacarbon.org/nacp-kawa-01/>. We are grateful for Dr. Hartmut Bösch, Dr. Chris O'Dell and Dr. Thorsten Warneke for their helpful comments on the manuscript.

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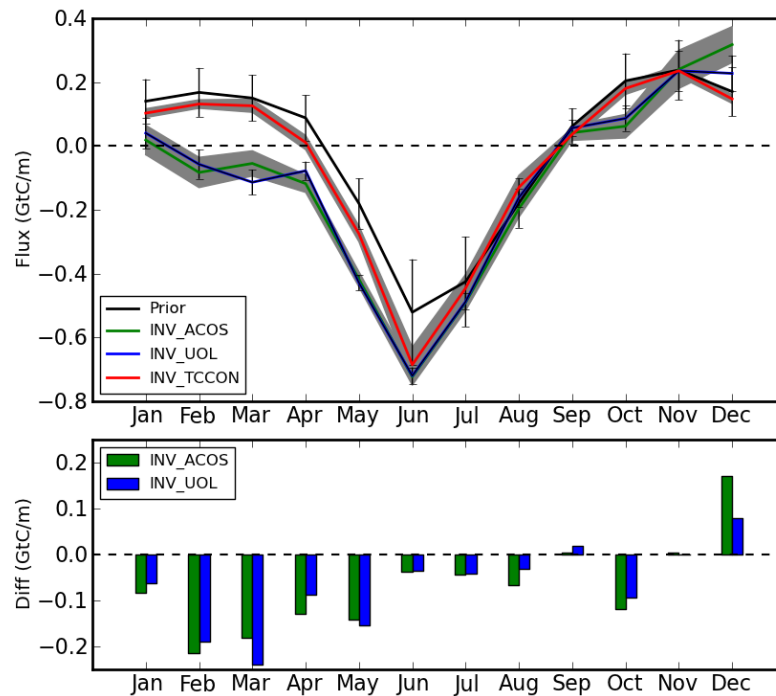


## Tables

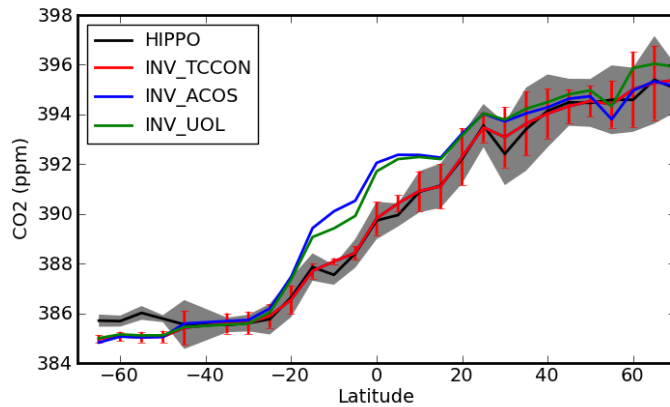
Name	Data	Flux (GtC/a)	Uncertainty (GtC/a)
INV_TCCON	In-situ Flask and TCCON $X_{CO_2}$	-0.58	0.14
INV_ACOS	ACOS $X_{CO_2}$ retrievals	-1.40	0.19
INV_UOL	UOL $X_{CO_2}$ retrievals	-1.4	0.20
INV_ACOS_MOD_ALL	Model simulation of ACOS $X_{CO_2}$ by using INV_TCCON posterior fluxes	-0.64	0.19
INV_ACOS_MOD_NOEU	As INV_ACOS_MOD_ALL but the real ACOS $X_{CO_2}$ retrievals are assimilated within Europe.	-0.88	0.19
INV_UOL_MOD_NOEU	As INV_UOL, but outside the Europe, UOL $X_{CO_2}$ retrievals are replaced with INV_TCCON simulations.	-0.67	0.19
INV_ACOS_MOD_ONLYEU	As INV_ACOS, but $X_{CO_2}$ retrievals within EU are replaced by INV_TCCON simulations	-1.17	0.19
INV_ACOS_OUT_0.5ppm	As INV_ACOS, but a bias of -0.5 ppm has been added to $X_{CO_2}$ retrievals outside Europe.	-0.98	0.19
INV_ACOS_SPR_0.5ppm	As INV_ACOS, but 0.5 ppm bias has been added to the European data in February, March, and April.	-1.30	0.19
INV_ACOS_SUM_0.5ppm	As INV_ACOS, but 0.5 ppm bias has been added to the European data in June, July, and August.	-1.25	0.19
INV_ACOS_INS	ACOS $X_{CO_2}$ retrievals and In-situ flask and TCCON data	-0.62	0.13
INV_UOL_INS	UOL $X_{CO_2}$ retrievals and in-situ flask and TCCON data	-0.67	0.13
INV_ACOS_DBL_ERR	ACOS $X_{CO_2}$ retrievals, but the a priori uncertainties have been doubled	-1.61	0.27
INV_ACOS_INS_DBL_ERR	GOSAT ACOS $X_{CO_2}$ retrievals and In-situ flask and TCCON data but the a priori flux uncertainties have been doubled	-0.67	0.16

**Table 1:** The magnitude and uncertainty of the European annual CO<sub>2</sub> biosphere flux (GtC/a) from 14 global flux inversion experiments. Except INV\_ACOS\_INS\_DBL\_ERR and INV\_ACOS\_DBL\_ERR, the aggregated European annual uptake of the a priori fluxes is  $-0.1 \pm 0.52$  GtC/a.

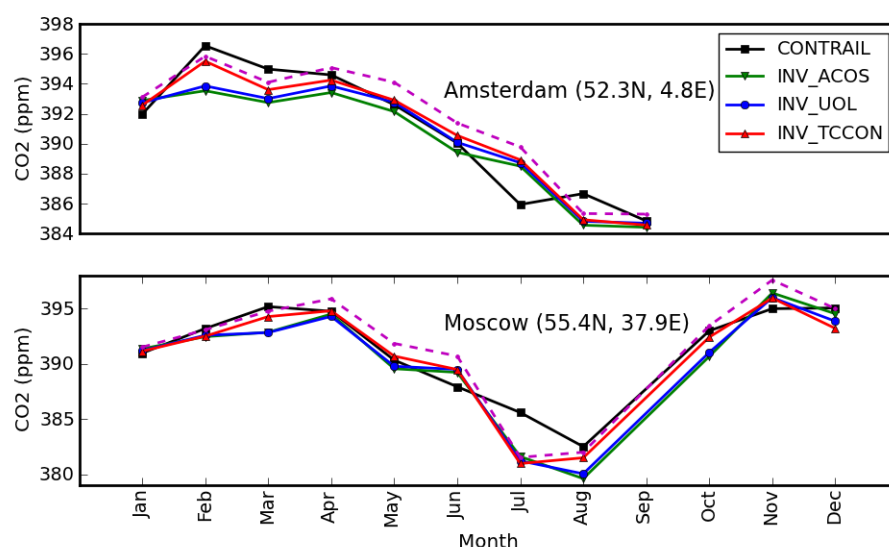
## Figures



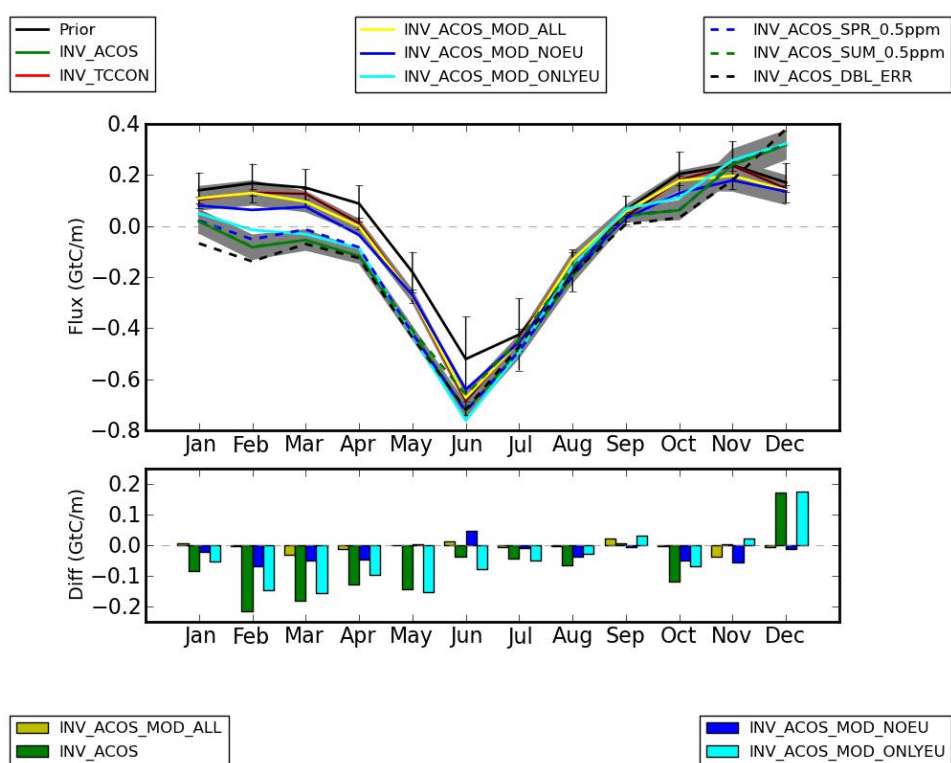
**Figure 1.** Monthly a posteriori estimates (GtC/m) for European biospheric fluxes in 2010 using three inversion experiments (top panel): 1) INV\_TCCON (red line), 2) INV\_ACOS (green line), and INV\_UOL (blue line). The black line denotes a priori values. The vertical black lines and grey shading denotes the uncertainties of the corresponding a priori or a posteriori flux estimates, respectively. Differences in monthly CO<sub>2</sub> uptake (GtC/m) between INV\_TCCON and two GOSAT inversions (bottom panel): INV\_ACOS (green bars) and INV\_UOL (blue bars).



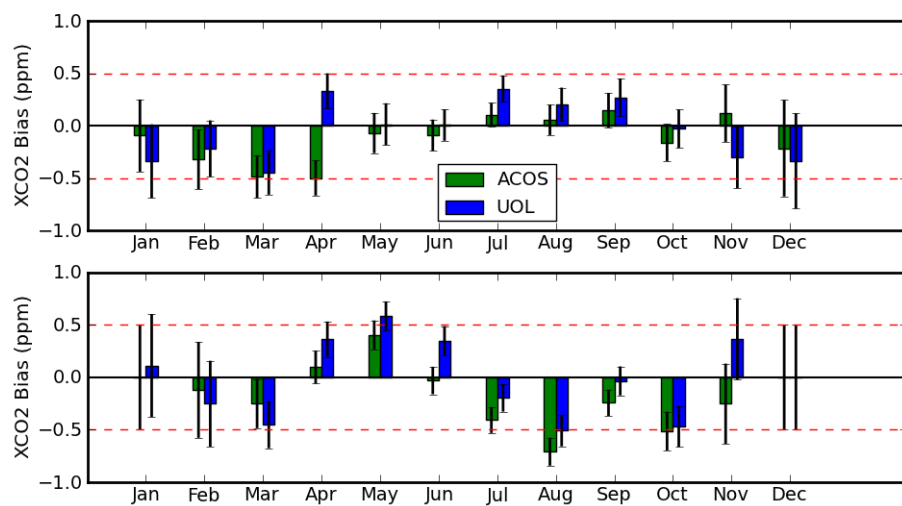
**Figure 2:** Comparison of HIPPO-3 measurements below 5 km (black) against co-located GEOS-Chem model simulations driven by different a posteriori flux estimates: 1) INV\_TCCON (red), 2) INV\_ACOS (blue), and 3) INV\_UOL (green). HIPPO-3 measurements and co-located model concentrations are binned into  $5^\circ$  latitude boxes. We calculate the mass-weighted average over these latitude boxes by assigning each HIPPO-3 measurement and its co-located model value a weighting factor according to the observation altitude (air pressure). The grey envelope (red vertical lines) indicates the one standard deviation of HIPPO-3 measurements (INV\_TCCON model values) within each latitude box.



495 **Figure 3:** Monthly means of the observed and a posteriori model CO<sub>2</sub> concentrations below 3km  
above Amsterdam (the top panel) and Moscow (the bottom panel) airports, respectively. They are  
the only descending and ascending CONTRAIL measurements over Europe available for more other 6  
months in 2010 [Machida et al., 2008]. The three sets of a posteriori model concentrations are  
inferred from three inversion experiments: INV\_TCCON (red line), INV\_ACOS (green line), and  
500 INV\_UOL (blue line). The broken magenta line represents a model simulation where the European  
fluxes from INV\_ACOS inversion are replaced by INV\_TCCON estimates.



**Figure 4:** Monthly European biospheric flux estimates from two groups of sensitivity experiments (top panel, Table 1). Black, green and red solid lines denote the a priori and the INV\_ACOS and INV\_TCCON inversions, respectively. Differences between INV\_TCCON inversion and sensitivity inversions (bottom panel): 1) INV\_ACOS\_MOD\_ALL (yellow), where all GOSAT retrievals are replaced by the model simulations forced by INV\_TCCON a posteriori fluxes; 2) INV\_ACOS (green), where original GOSAT ACOS retrievals are assimilated; 3) INV\_ACOS\_NOEU (blue) where all the GOSAT retrievals outside the European region are replaced by the INV\_TCCON simulations; and 4) INV\_ACOS\_MOD\_ONLYEU (cyan) where only GOSAT retrievals within the European region are replaced by the INV\_TCCON simulations.



530 **Figure 5:** Estimates of monthly biases in GOSAT ACOS (green) and UOL (blue)  $X_{\text{CO}_2}$  retrievals over (top) West (West of 20°E) and (bottom) East (East of 20°E) Europe. The black vertical lines represent the uncertainty.

## 535 **Appendix A: Quasi-regional flux inversion.**

To further study the contributions from  $X_{CO_2}$  retrievals within and outside Europe we have performed quasi-regional flux inversions to infer European uptake in 2010, based on the same EnKF approach as the global flux inversions. In contrast to the global experiments (Table 1), in the quasi-regional inversions we assimilate observations only over Europe, and assign a small a priori flux uncertainty to any region outside Europe in order to minimize the influence of observations taken over Europe on other regions. Consequently, a posteriori flux estimates outside of Europe are close to their a priori values. We use the a posteriori fluxes from INV\_TCCON as the a priori estimates for 12 sub-regions in Europe, and assume their uncertainty is two thirds of that we use for the global flux inversions. This is because the a posteriori estimates from INV\_TCCON have already been refined by in situ data.

To investigate the influence of lateral boundary conditions on the quasi-regional flux inversions, we use two different sets of a posteriori estimates to define fluxes outside Europe: 1) INV\_TCCON (INV\_BD\_TCCON) and 2) INV\_ACOS (INV\_BD\_ACOS). Figure A1 shows that INV\_BD\_ACOS has a higher annual uptake of 1.58 GtC/a than INV\_BD\_TCCON with an uptake of 0.79 GtC/a (Table A1), with differences larger during the first half of 2010. The estimate for INV\_BD\_ACOS is similar to its global inversion counterpart INV\_ACOS. Large differences between INV\_BD\_ACOS and INV\_BD\_TCCON highlight the importance of accurate lateral boundary conditions to a regional European inversion.

We use on-line bias correction schemes to reduce the adverse impacts from improper boundary conditions around Europe. Similar to Reuter et al. (2014), we estimate monthly observation biases across Europe using our quasi-regional flux inversion system. Here, we introduce a monthly bias to remove the systematic difference between model and GOSAT observations across the whole European region, and assume an associated a priori uncertainty of 100 pm (Reuter et al, 2014). This is different from our previous bias assumption of 0.5 ppm over East and West Europe for INV\_ACOS\_INS. Compared to INV\_ACOS\_INS, we also do not assimilate any in situ observations as additional constraints. Figure A1 shows that such a bias correction scheme (INV\_BD\_ACOS\_BC) successfully reduces European uptake of  $CO_2$  during 2010 to 0.96 GtC/a from 1.58 GtC/a for INV\_BD\_ACOS. Table A1 shows that after the applying bias correction scheme, INV\_BD\_ACOS\_BC and INV\_BD\_TCCON\_BC are consistent (0.94 GtC/a vs 0.96 GtC/a) despite different lateral boundary conditions provided by INV\_ACOS and from INV\_TCCON. But INV\_BD\_TCCON\_BC (0.94 GtC/a) has 0.15 GtC/a more uptake than INV\_BD\_TCCON (0.79 GtC/a). We find a similar difference using UOL data (not shown), which infer an annual uptake of 0.71 GtC/a (0.56 GtC/a) with (without) the on-line bias correction.

We next examine the effectiveness of the inversion system that uses an on-line bias correction with large a priori uncertainty. Generally, large a priori uncertainty for biases will lead to the eventual loss of constraint by the observed mean  $CO_2$  concentration across Europe. The weakened constraint can be seen by the enlarged a posteriori error (by 0.04 GtC/a) for INV\_BD\_TCCON\_BC. In additional OSSEs (Table A2) we find that the loss of such a constraint can result in large systematic errors in estimated fluxes.

In these OSSEs, we assume the a priori estimates for 12 European sub-regions to be the same as the a priori used by INV\_TCCON. Similar to INV\_BD\_TCCON, we set the fluxes outside European region to

be the a posteriori estimates by INV\_TCCON. We assimilate the INV\_TCCON model ACOS  $X_{CO_2}$  retrievals over Europe, to test the ability of the system to recover the “true” European flux (defined by INV\_TCCON) from the assumed a priori that we define as the CASA model. Without the on-line bias correction, the quasi-regional inversion INV\_REG\_ENKF reproduces the truth for most months (Figure A2), and the associated annual uptake of 0.55 GtC/a compared to the true value of 0.58 GtC/a. If we also estimate monthly  $X_{CO_2}$  bias with a large a priori uncertainty of 100 ppm (INV\_REG\_BC), the a posteriori European uptake is systematically underestimated for almost all months in 2010 (Figure A2). Consequently, the a posteriori annual uptake is about 0.38 GtC/a, which is 35% smaller than the truth (Table A2). Weakening the observation constraint also enlarges the a posteriori uncertainty from 0.22 GtC/a for INV\_REG\_ENKF to 0.27 for INV\_REG\_BC. But we find that increases in the estimated a posteriori uncertainty (by 0.05 GtC/a) are smaller than the increase in the systematic deviation from the true annual uptake (by 0.19 GtC/a).

More importantly, we find that the derived annual uptake is not linearly correlated to the assumed true fluxes. In experiment INV\_REG\_BC\_SP (Table A2) we replace the true fluxes (defined by INV\_TCCON) over the first 3 of 12 European sub-regions, which are at the south part of Europe (roughly south of 47° N), with values from CASA model. As a result, the new true fluxes have an annual uptake of about 0.48 GtC/a across Europe, which is about 18% (0.1 GtC/a) lower than the original one defined by INV\_TCCON for INV\_REG\_BC. We then re-generate model ACOS  $X_{CO_2}$  data by running GEOS-Chem driven by the new hybrid true fluxes. However, after assimilating the new model  $X_{CO_2}$  data, INV\_REG\_BC\_SP infers an annual uptake of 0.37 GtC/a, which is almost the same as the posterior estimate (0.38 GtC/a) of INV\_REG\_BC, failing to reproduce the 18% decrease from the true value of 0.58 GtC/a assumed for INV\_REG\_BC to the 0.48 GtC/a assumed for INV\_REG\_BC\_SP. In contrast, the quasi-inversion without on-line bias correction (INV\_REG\_ENKF\_SP) well reproduces such decrease.

The bias correction across Europe can also increase the sensitivity to sub-regional biases. To illustrate this we added 1 ppm bias to the simulated observations during June to August of 2010 over south-west Europe between 35°N to 42°N and 15°W to 20°E (mostly over Spain and Italy). Without an on-line bias correction, adding the 1 ppm bias over the south-west strip leads to a small change (0.01GtC/a) in the annual uptake: a (slightly) reduced uptake in the first half of 2010 is largely compensated by a slightly enhanced uptake in the second half of 2010. Conversely, when we use an on-line bias correction with large prior errors (INV\_REG\_BC\_1ppm), the 1 ppm positive bias increases the uptake by about 0.24 GtC in June, July and August. This implies that without the constraint from the mean concentration across the whole European region, the inversion system is free to interpret the higher concentrations over the small south-west strip as the signal of more uptakes over other larger part of Europe. As a result, the annual uptake changes from an underestimation of 35% by INV\_REG\_BC to an overestimation of 15% by INV\_REG\_BC\_1ppm (0.65 GtC/a) (Table A2).

In summary, our quasi-regional inversion experiments highlight the sensitivity of regional flux inversions to the accurate description of the boundary conditions around the domain. Using an on-line bias correction can be helpful when the bias has been properly characterized. Over-correcting the bias can weaken the observation constraints, and possibly increase sensitivity to other small-scale unknown biases. We have also tested bias correction schemes using a different inversion algorithm (the Maximum A Posteriori (MAP) approach, Fraser et al., 2014), and found similar

620 deficiencies when the a priori uncertainty of the regional observation bias is assumed to be very  
large. Our studies cannot prove or disprove Reuter et al. (2014), but it does highlight previously  
unrecognized limitation to the approach. The diversity of results reached under different  
assumptions associated with observation biases and emission spatial patterns highlight the  
importance for us as a community to investigate the interaction between observation and the  
625 inversion system for achieving consistent flux estimates in the future from assimilation of the up-  
coming observations from OCO-2 satellite as well as from the improved in situ networks.

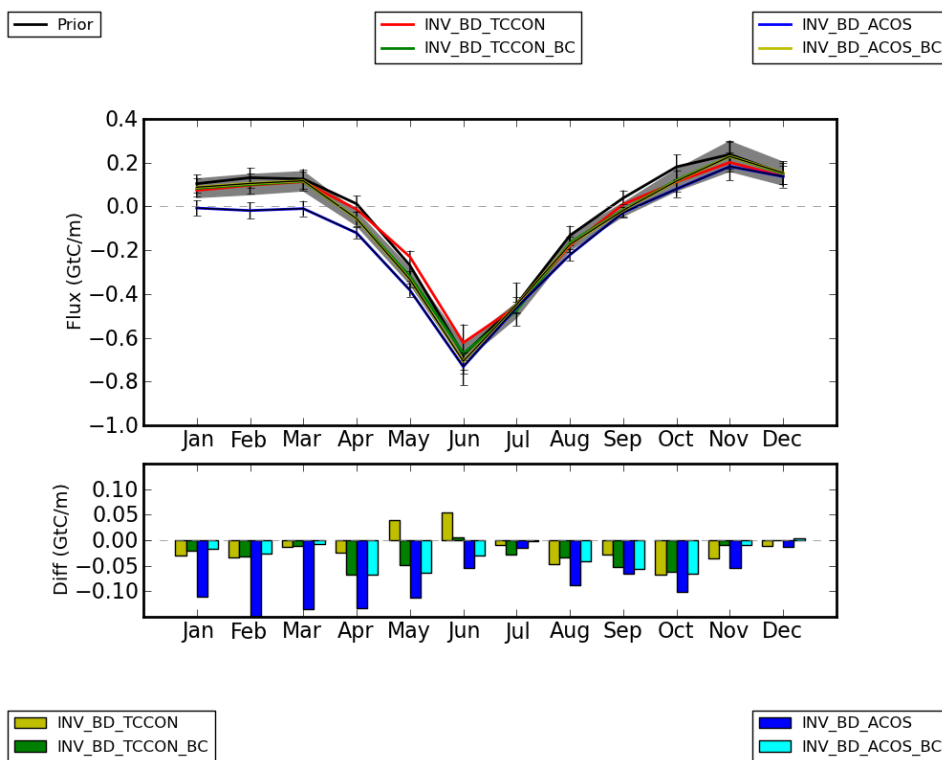


Name	Description	Flux (GtC/a)	Uncertainty (GtC/a)
INV_BD_TCCON	Only ACOS data over Europe are assimilated to infer monthly fluxes over 12 European sub-regions. Fluxes outside EU are fixed to INV_TCCON inversion.	-0.79	0.18
INV_BD_TCCON_BC	The same as INV_BD_TCCON, but monthly bias with an assumed prior uncertainty of 100 ppm are included as additional parameters to be estimated.	-0.94	0.22
INV_BD_ACOS	The same as INV_BD_TCCON, but external regional fluxes are fixed to INV_ACOS.	-1.58	0.18
INV_BD_ACOS_BC	The same as INV_BD_ACOS, but estimates for monthly observation bias included.	-0.96	0.22

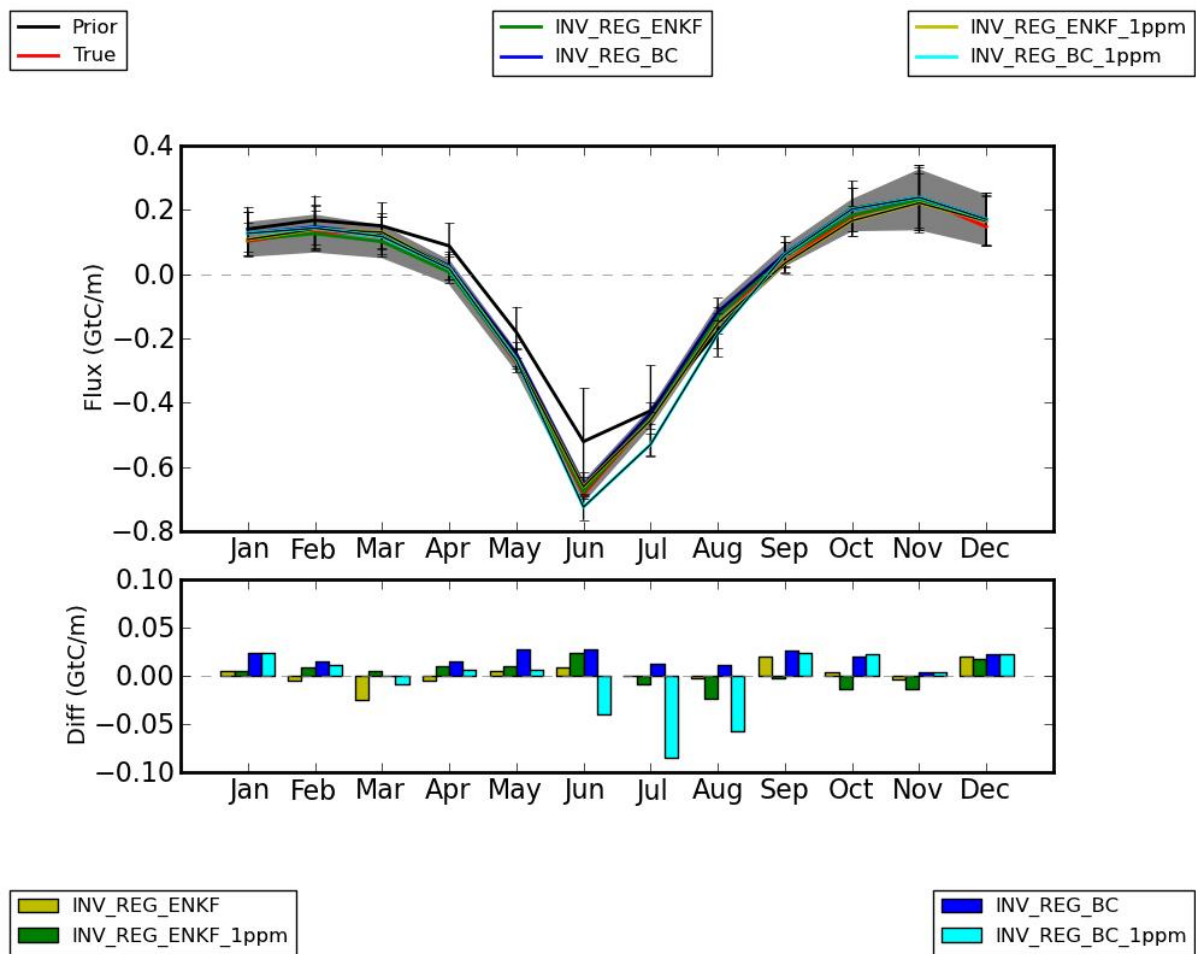
**Table A1:** The same as Table 1 but for quasi-regional inversions where only ACOS  $X_{CO_2}$  are assimilated.

Name	Description	Flux (GtC/a)	Uncertainty (GtC/a)
INV_REG_ENKF	Synthetic ACOS data over Europe are assimilated to infer monthly fluxes over 12 European sub-regions, which prior estimates are assumed to be same as INV_ACOS (i.e., CASA model). Here we assume the true fluxes be a posteriori of INV_TCCON inversion.	-0.55	0.22
INV_REG_BC	The same as INV_REG_ENKF, but estimates for monthly bias are included as additional parameters.	-0.38	0.25
INV_REG_ENKF_1ppm	The same as INV_REG_ENKF, but 1ppm bias is added to the synthetic observations over a strip at south-west Europe for three months from June to August in 2010.	-0.54	0.22
INV_REG_BC_1ppm	The same as INV_REG_BC, 1ppm bias is added to the synthetic observations over a strip at south-west Europe for three months from June to August in 2010.	-0.65	0.25
INV_REG_ENKF_SP	The same as INV_REG_ENKF, but the 'true fluxes' over the first 3 of the 12 European sub-regions are replaced by CASA model values.	-0.47	0.22
INV_REG_BC_SP	The same as INV_REG_ENKF_SP, but with on-line bias correction with assumed prior uncertainty of 100 pm.	-0.37	0.25

**Table A2:** The same as Table A1 but for Observation System Simulation Experiments, where we assimilate synthetic ACOS  $X_{CO_2}$  from model simulations forced by the assumed the 'True fluxes'.



**Figure A1:** As Figure 4, but for the comparisons between the quasi-regional inversions. All the inversion experiments assimilate the same ACOS data set over Europe, with the a priori for 12 European sub-regions taken from posterior estimates from INV\_TCCON. Fluxes outside Europe are fixed to the posterior estimates of INV\_TCCON (INV\_BD\_TCCON and INV\_BD\_TCCON\_BC) or to the estimates of INV\_ACOS (INV\_BD\_ACOS and INV\_BD\_ACOS\_BC). INV\_BD\_TCCON\_BC and INV\_BD\_ACOS\_BC also include the estimate for the monthly bias across Europe as additional parameter with an assumed prior uncertainty of 100 pm to be estimated from ACOS data.



**Figure A2:** As Figure 4, but for comparisons of the quasi-regional inversions for assimilation of synthetic ACOS retrievals against ‘True’ fluxes (INV\_TCCON). All the quasi-regional inversions have assumed the same a priori fluxes. But INV\_REG\_BC and INV\_REG\_BC\_1ppm also include the monthly observation bias across Europe, with a prior uncertainty of 100 ppm, as additional parameters to be estimated from the synthetic observations. In INV\_REG\_ENKF\_1ppm and INV\_REG\_BC\_1ppm, 1ppm observation bias is added to the (synthetic) observations over a small south-west strip of Europe during the summer of 2010.