

Estimates of European uptake of CO₂ inferred from GOSAT X_{CO2} retrievals: sensitivity to measurement bias inside and outside Europe

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Estimates of the natural CO₂ flux over Europe inferred from in situ measurements of atmospheric CO₂ mole fraction have been used previously to check top-down flux estimates inferred from space-borne dry-air CO₂ column (X_{CO2}) retrievals. Several recent studies have shown that CO₂ fluxes inferred from X_{CO2} data from the Japanese Greenhouse gases Observing SATellite (GOSAT) and the Scanning Imaging Absorption Spectrometer for Atmospheric CHartography (SCIAMACHY) have larger seasonal amplitudes and a more negative annual net CO₂ balance than those inferred from the in situ data. The cause of this elevated European uptake of CO₂ is still unclear, but some recent studies have suggested that this is a genuine scientific phenomenon. Here, we put forward an alternative hypothesis and show that realistic levels of bias in GOSAT data can result in an erroneous estimate of elevated uptake over Europe. We use a global flux inversion system to examine the relationship between measurement biases and estimates of CO₂ uptake from Europe. We establish a reference in situ inversion that uses an Ensemble Kalman Filter (EnKF) to assimilate conventional surface mole fraction observations and X_{CO2} retrievals from the surface-based Total Carbon Column Observing Network (TCCON). We use the same EnKF system to assimilate two independent versions of GOSAT X_{CO2} 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 perform a series of numerical experiments that assimilate observations with added biases or assimilate synthetic observations for which part or all of the GOSAT X_{CO2} 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 the remainder can largely be explained by a sub-ppm retrieval bias over Europe. We use a data assimilation approach to estimate monthly

GOSAT X_{CO_2} biases from the joint assimilation of in situ observations and GOSAT X_{CO_2} retrievals. The inferred biases represent an estimate of systematic differences between GOSAT X_{CO_2} retrievals and the inversion system at regional or sub-regional scales. We find that 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 CO_2 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_2 . More robust inversion systems are also needed to infer consistent fluxes from multiple observation types.

1. Introduction

Observed atmospheric variations of carbon dioxide (CO_2) 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, our knowledge of top-down estimates of regional CO_2 fluxes, particularly at tropical and high northern latitudes, have not significantly improved for over 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 $< O(10,000 \text{ km})$ [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_2 columns using short-wave IR wavelengths. Validation of current X_{CO_2} 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_{CO_2} [Liu et al., 2014].

Several independent studies have shown that regional flux distributions inferred from GOSAT X_{CO_2} 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 CO₂ [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_{CO2} data. In section 3, we 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 CO₂ concentrations [Feng et al., 2009] at a horizontal resolution of 4°x5°, 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 insitu 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 CO₂ fluxes [Takahashi et al., 2009]; and (4) 3-hourly terrestrial biosphere-atmosphere CO₂ 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). Our 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).

Our control inversion experiment (INV_TCCON, Table 1 and 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. In some studies, TCCON data was used to evaluate posterior fluxes. However TCCON data has been used to derive bias corrections for GOSAT X_{CO_2} retrievals [Cogan et al., 2012], and also the nature of total column measurements means that they are sensitive to air mass transported from other regions, which complicate the assessment of European flux estimates.

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 GtC/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].

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 shows the three inversion experiments, INV_TCCON, INV_ACOS, and INV_UOL, have similar European uptake values in June 2010 (0.69 GtC/m for INV_TCCON and ~0.72 GtC/m for GOSAT inversions), and 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 ± 0.19 GtC/a compared to the in-situ inversion of 0.58 ± 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.

Figure 2 shows that INV_TCCON a posteriori CO₂ mole fractions agree well with the independent HIPPER Pole-to-Pole Observations (HIPPO-3) aircraft measurements below 5 km over the Pacific Ocean 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 3 shows further evaluation of a posteriori CO₂ mole fractions 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_{CO2} 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 CO₂ 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 CO₂ 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_{CO2} gradient across this region. In the INV_TCCON simulation, the mean X_{CO2} difference between east (east of 20°E) and west (west of 20°E) Europe is ~0.04 ppm for May, 2010, which is increased to 0.16 ppm in the hybrid run (cf. E-W X_{CO2} 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 (Table 1 and Figure 4). First, we replaced all or part of the GOSAT X_{CO2} 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 CO₂ 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 only replace X_{CO2} 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_{CO2} 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. The large contribution from GOSAT retrievals outside Europe has also been confirmed by the high uptake (1.17 Gt/a) in a counterpart experiment (INV_ACOS_MOD_ONLYEU) where only GOSAT retrievals within Europe are replaced by the model simulations. We show in Appendix B that theoretically the difference between INV_ACOS and INV_ACOS_MOD_ALL is equal to the sum of the individual uptake increases in the paired synthetic inversions of INV_ACOS_MOD_NOEU and INV_ACOS_MOD_ONLYEU.

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 3, these results suggest that GOSAT inversions result in an overestimated CO_2 inflow. This will subsequently lead to the fitted European flux having to compensate, via mass balance, by being erroneously 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), 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 surface 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 ± 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 error 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 contain systematic model errors (such as the atmospheric 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 few GOSAT observations over northern Europe during autumn and winter months, while Eastern Europe has few in-situ observations. We have also limited the a priori uncertainty for the monthly observation biases to 0.5 ppm. Figure C1 (Appendix C) shows, for example, the inferred monthly mean bias for March 2010.

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 is 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 CO_2 data sets: 1) surface in situ mole fraction observations and TCCON X_{CO_2} retrievals; 2) GOSAT X_{CO_2} retrievals from the JPL ACOS team; and 3) GOSAT X_{CO_2} retrievals from the 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 elevated European uptake of CO_2 is 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 also 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]. The 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 (inferred from 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 (inferred from GOSAT data). 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. In future the TCCON measurement network may be supported by smaller, more mobile FTIR instruments, which can be established, at least on a campaign basis, in tropical and high latitude locations where observational gaps are greatest.

Our joint data assimilation approach assimilates in-situ and space-borne observations. It also provides estimates of systematic differences between XCO₂ retrievals and the inversion system at regional /sub-regional scales. However the resulting differences will include the observation biases and deficiencies in the underlying inversion approach. To achieve consistent flux estimates inferred from assimilating multiple data sets 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.

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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 CO₂ 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 ± 0.52 GtC/a.

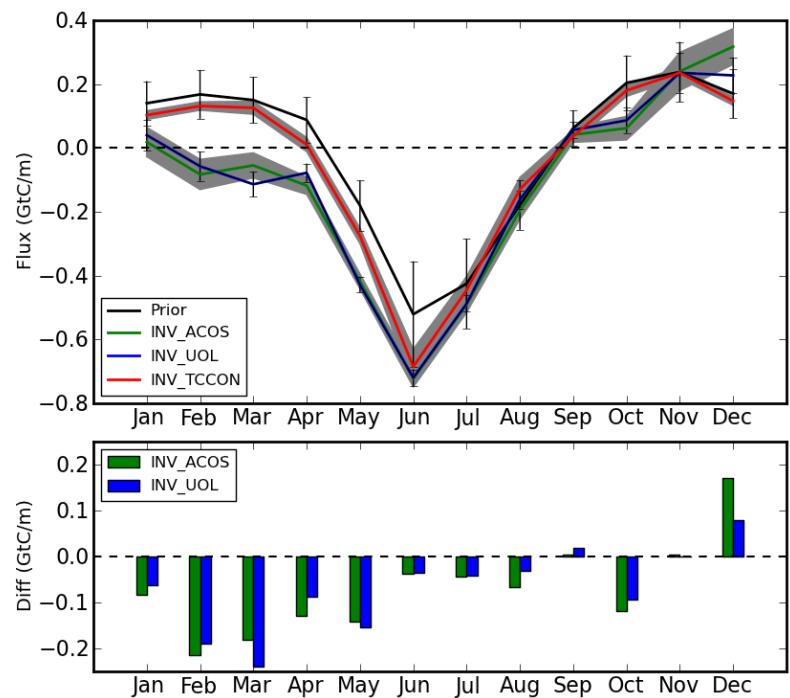


Figure 1. Monthly a posteriori estimates (GtC/m) for European biospheric CO₂ fluxes in 2010 using three inversion experiments (top panel): 1) INV_TCCON (red line), 2) INV_ACOS (green line), and INV_UOL (blue line). The black line denotes a priori values. The vertical black lines and grey shading denotes the uncertainties of the corresponding a priori or a posteriori flux estimates, respectively. Differences in monthly CO₂ uptake (GtC/m) between INV_TCCON and two GOSAT inversions (bottom panel): INV_ACOS (green bars) and INV_UOL (blue bars).

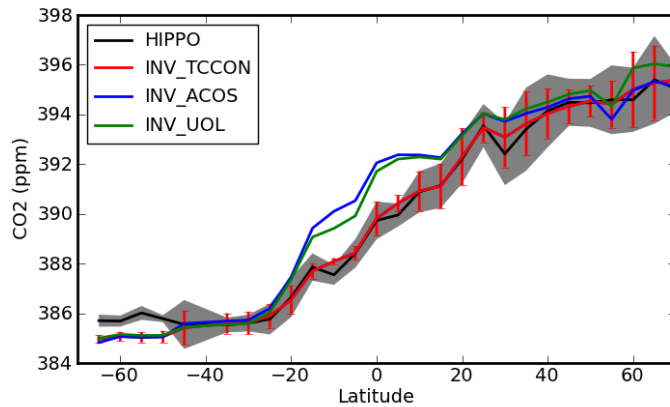


Figure 2: HIPPO-3 and GEOS-Chem model atmospheric CO₂ mole fractions (ppm) over the Pacific Ocean below 5 km (black). GEOS-Chem is driven by different a posteriori flux estimates: 1) INV_TCCON (red), 2) INV_ACOS (blue), and 3) INV_UOL (green). HIPPO-3 and model CO₂ mole fractions are binned into 5° latitude boxes. We calculate the mass-weighted average over these latitude boxes by assigning each HIPPO-3 and GEOS-Chem 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.

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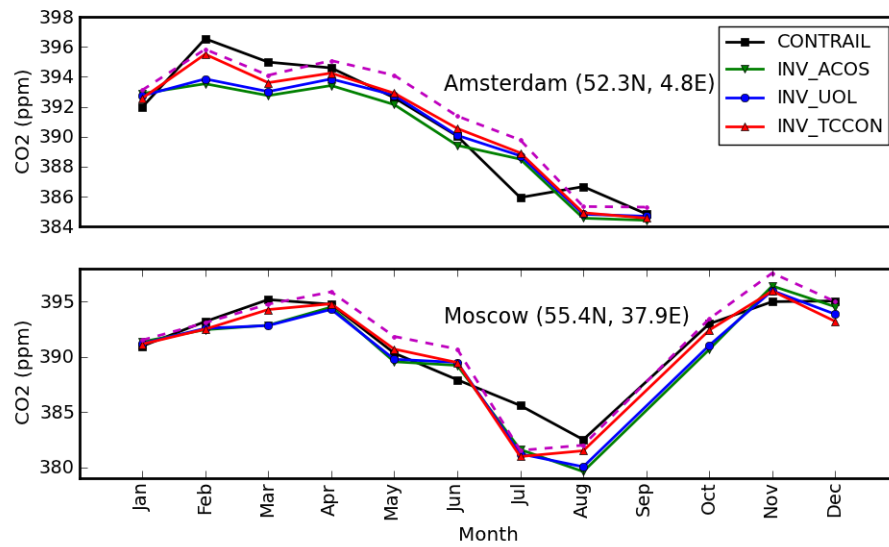


Figure 3: Monthly mean observed and model a posteriori model CO₂ mole fractions (ppm) below 3km above Amsterdam (the top panel) and Moscow (the bottom panel) airports during 2010, respectively [Machida et al., 2008]. The three sets of a posteriori model concentrations are inferred from three inversion experiments: INV_TCCON (red line), INV_ACOS (green line), and 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.

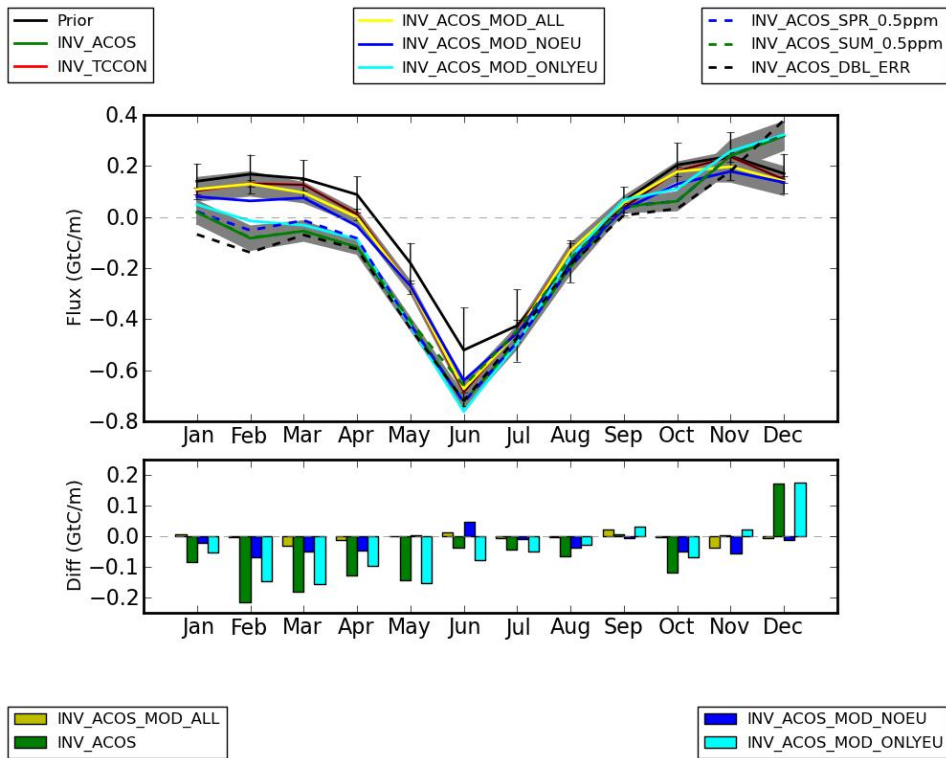


Figure 4: Monthly European biospheric flux estimates (GtC/m) from two groups of sensitivity experiments (top panel, Table 1). Black, green and red solid lines denote the a priori and the INV_ACOS and INV_TCCON inversions, respectively. Differences between INV_TCCON inversion and sensitivity inversions (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.

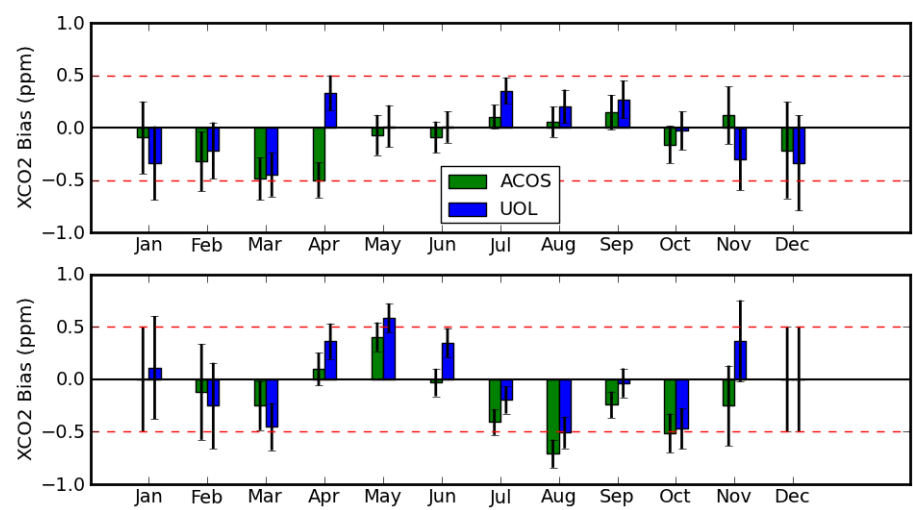


Figure 5: Estimates of monthly CO₂ biases (ppm) in GOSAT ACOS (green) and UOL (blue) X_{CO2} retrievals over (top) West (West of 20°E) and (bottom) East (East of 20°E) Europe. The black vertical lines represent the uncertainty.

Appendix A: Quasi-regional flux inversion.

To further study the contributions from X_{CO_2} retrievals within and outwith Europe we have performed quasi-regional flux inversions to infer the European uptake of CO_2 in 2010, based on the same EnKF approach as the global flux inversions. In contrast to the global experiments (Table 1), for 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 incorrect 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

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 inversion system for achieving consistent flux estimates in the future from assimilation of the upcoming 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

655 **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'.

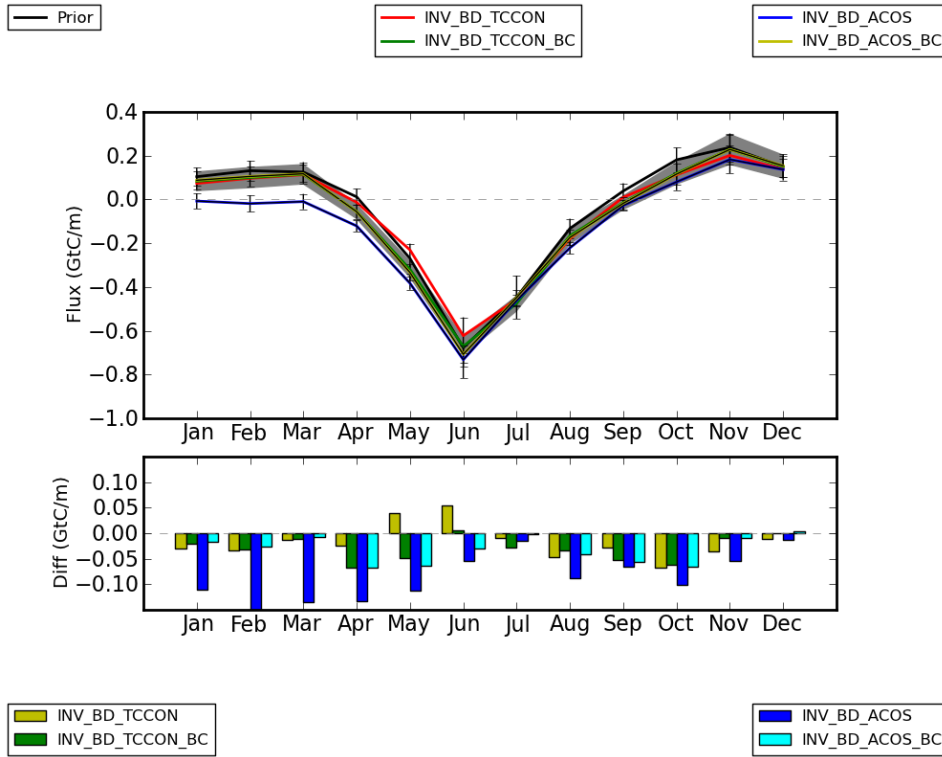


Figure A1: As Figure 4, but for the comparisons between the quasi-regional inversions. All the inversion experiments assimilate the same ACOS data set over Europe, with the a priori for 12 European sub-regions taken from posterior estimates from INV_TCCON. Fluxes outside Europe are fixed to the posterior 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 estimate the monthly bias across Europe as an additional parameter with an assumed a priori uncertainty of 100 pm estimated from ACOS data.

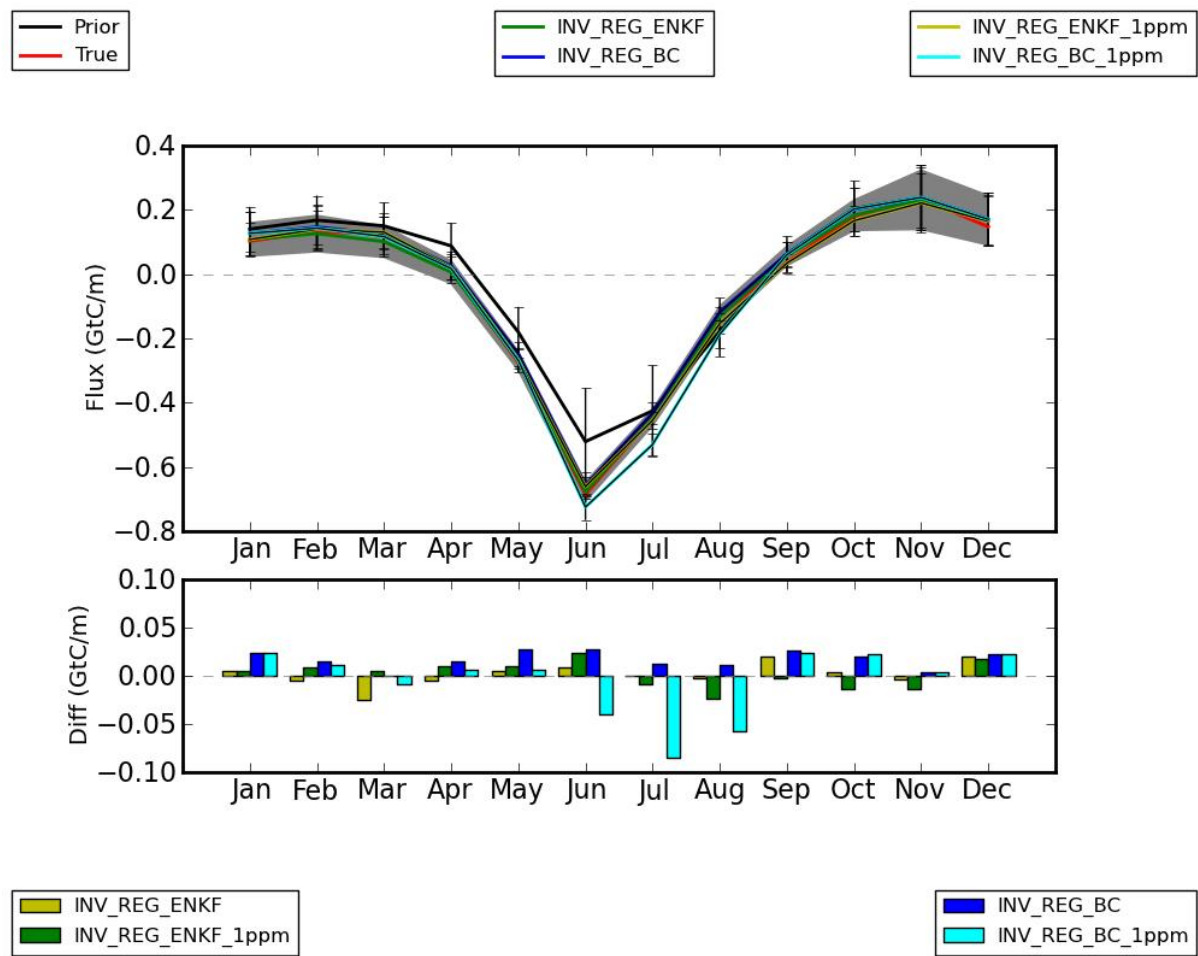


Figure A2: As Figure 4, but for comparisons of the quasi-regional inversions for assimilation of
680 synthetic ACOS retrievals against ‘True’ fluxes (INV_TCCON). All the quasi-regional inversions have
assumed the same a priori fluxes. But INV_REG_BC and INV_REG_BC_1ppm also include the
monthly observation 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
685 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.

Appendix B: Additivity of the increased European uptake estimates.

In the framework of Kalman Filter data assimilation [Feng et al., 2009], posterior flux estimates are determined by:

$$\mathbf{F}^a = \mathbf{F}^f + \mathbf{K}(\mathbf{y}_{obs} - H(\mathbf{F}^f)), \quad (\text{B1})$$

where $\mathbf{F}^a, \mathbf{F}^f$ are the prior and posterior estimates of monthly regional surface CO_2 fluxes, respectively; \mathbf{y}_{obs} represents the GOSAT (real or simulated) X_{CO_2} retrievals. H is the observation operator for relating the surface fluxes to the observed GOSAT X_{CO_2} , which includes complicated atmospheric transporting as well as convolving of co-located model profiles with GOSAT averaging kernels [Feng et al., 2009; Chevallier et al. 2010]. Here, the Kalman gain matrix \mathbf{K} is given by

$$\mathbf{K} = \mathbf{B}\mathbf{H}^T[\mathbf{H}\mathbf{B}\mathbf{H}^T + \mathbf{R}]^{-1}, \quad (\text{B2})$$

where \mathbf{B} is the a priori flux error covariance, \mathbf{R} is the observation error covariance, and \mathbf{H} is the Jacobian defined by

$$\mathbf{H} = \frac{\partial H(\mathbf{F}^f)}{\partial \mathbf{F}^f}. \quad (\text{B3})$$

Although the atmospheric transport is non-linear, the dependence of model concentrations (such as the column mixing ratios X_{CO_2}) on the surface fluxes is nearly linear if we do not take into account any feedback of varying CO_2 concentrations on atmospheric dynamics (for example, Chevallier et al., 2010; Baker et al., 2006). As a result, the gain matrix is eventually independent of actual observation values, but will still be affected by the location and uncertainty of observations.

As described in the main text, we split the actual (or simulated) X_{CO_2} observations into two parts: Part A for observations within Europe; and Part B for observations outside Europe. For the GOSAT inversions (such as INV_ACOS), we denote the observation vector as:

$$\mathbf{y}_{obs} = \begin{bmatrix} \mathbf{G}^A \\ \mathbf{G}^B \end{bmatrix}. \quad (\text{B4})$$

The corresponding posterior flux estimate is given as:

$$\mathbf{F}_G^a = \mathbf{F}^f + \mathbf{K} \left(\begin{bmatrix} \mathbf{G}^A \\ \mathbf{G}^B \end{bmatrix} - H(\mathbf{F}^f) \right). \quad (\text{B5})$$

In experiment INV_MOD_ALL, we replace the retrieved X_{CO_2} values by the reference model simulation (from INV_TCCON), so that the observation vector becomes

$$\mathbf{y}_{obs} = \begin{bmatrix} \mathbf{M}^A \\ \mathbf{M}^B \end{bmatrix}, \quad (\text{B6})$$

so that the resulting flux estimates are:

$$\mathbf{F}_M^a = \mathbf{F}^f + \mathbf{K} \left(\begin{bmatrix} \mathbf{M}^A \\ \mathbf{M}^B \end{bmatrix} - H(\mathbf{F}^f) \right). \quad (\text{B7})$$

720 The gain matrix in Eq. B7 is the same as Eq. B5. Similarly, for INV_MOD_ONLYEU where GOSAT X_{CO2} retrievals over Europe are replaced by model simulations, we have

$$\mathbf{F}_{MG}^a = \mathbf{F}^f + \mathbf{K} \left(\begin{bmatrix} \mathbf{M}^A \\ \mathbf{G}^B \end{bmatrix} - H(\mathbf{F}^f) \right). \quad (\text{B8})$$

And for INV_MOD_NOEU where GOSAT XCO2 retrievals outside Europe are replaced by model simulations, we have

$$\mathbf{F}_{GM}^a = \mathbf{F}^f + \mathbf{K} \left(\begin{bmatrix} \mathbf{G}^A \\ \mathbf{M}^B \end{bmatrix} - H(\mathbf{F}^f) \right). \quad (\text{B9})$$

725 From equations B5, B7, B8, and B9, we can directly obtain:

$$\mathbf{F}_G^a - \mathbf{F}_M^a = (\mathbf{F}_{MG}^a - \mathbf{F}_M^a) + (\mathbf{F}_{GM}^a - \mathbf{F}_M^a). \quad (\text{B10})$$

730 Equation B10 demonstrates that elevated European uptake is the sum of the individual contributions from INV_MOD_NOEU and INV_MOD_ONLYEU. As discussed in Section 3, such additivity has also been found in our inversion results (Table 1), despite approximations in numerically solving posterior fluxes [Feng et al., 2009].

Appendix C: Regional and Sub-regional systematic errors inferred in joint data assimilation.

In the joint data assimilation, we attempt to estimate and remove systematic errors at the regional and sub-regional scales from GOSAT XCO₂ retrievals. The assimilated XCO₂ retrieval can be described as :

$$y^c = y - \text{bias}(m, i), \text{ (C1)}$$

where y represents GOSAT retrievals before the (extra) bias correction, and y^c is the bias-corrected XCO₂ data that we assimilate in our joint data assimilation experiments. For simplicity, we have assumed the regional (sub-regional) bias, $\text{bias}(m, i)$, is a function only of month (m) and geographical region (i).

In the joint data assimilation experiments, we consider $\text{bias}(m, i)$ as part of the state vector that we infer from assimilating in situ and satellite observations. Figure C1 shows the resulting bias (in ppm) for March 2010. Like other model and GOSAT inter-comparisons (see for example, Lindqvist et al., 2015), our results demonstrate a strong spatial dependence of the derived systematic errors. As discussed in Section 4, our results reflect the mean differences between the inversion system and XCO₂ retrievals at (sub) regional scales, which does not necessarily suggest that the GOSAT XCO₂ bias (as well as the coverage) within these (sub) regions is homogeneous.

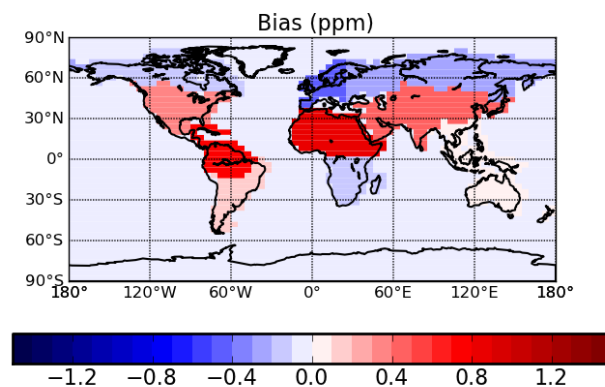


Figure C1: Inferred regional bias (in ppm) for March 2010 over TransCom regions and 2 European (West and North) sub-regions.