



Separation of biospheric and fossil fuel fluxes of CO₂ by atmospheric inversion of CO₂ and ¹⁴CO₂ measurements: Observation System Simulations

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Abstract. National annual total CO₂ emissions from combustion of fossil fuels are likely known to within 5–10% for most developed countries. However, uncertainties are inevitably larger (by unknown amounts) for emission estimates at regional and monthly scales, or for developing countries. Given recent international efforts to establish emission reduction targets, independent determination and verification of regional and national scale fossil fuel CO₂ emissions are likely to become increasingly important. Here, we take advantage of the fact that precise measurements of ¹⁴C in CO₂ provide a largely unbiased tracer for recently added fossil-fuel-derived CO₂ in the atmosphere and present an atmospheric inversion technique to jointly assimilate observations of CO₂ and ¹⁴CO₂ in order to simultaneously estimate fossil fuel emissions and biospheric exchange fluxes of CO₂. Using this method in a set of Observation System Simulation Experiments (OSSEs), we show that given the coverage of ¹⁴CO₂ measurements available in 2010 (969 over North America, 1063 globally), we can recover the US national total fossil fuel emission to better than 1% for the year and to within 5% for most months. Increasing the number of ¹⁴CO₂ observations to ~5000 per year over North America, as recently recommended by the National Academy of Science (NAS) (Pacala et al., 2010), we recover monthly emissions to within 5% for all months for the US as a whole and also for smaller, highly emissive regions over which the specified data coverage is relatively dense, such as for the New England states or the NY-NJ-PA tri-state area. This result suggests that, given continued improvement in state-of-the-art

transport models, a measurement program similar in scale to that recommended by the NAS can provide for independent verification of bottom-up inventories of fossil fuel CO₂ at the regional and national scale. In addition, we show that the dual tracer inversion framework can detect and minimize biases in estimates of the biospheric flux that would otherwise arise in a traditional CO₂-only inversion when prescribing fixed but inaccurate fossil fuel fluxes.

1 Introduction

The terrestrial biosphere and the oceans have taken up roughly half the anthropogenic emissions of CO₂, with the remainder contributing to the observed increase in atmospheric CO₂ concentration from ~280 ppm in the early 1800s to ~395 ppm in 2013 (Ballantyne et al., 2012). However, while CO₂ observations from sampling networks over large, industrialized land areas will be influenced by emissions from combustion of fossil fuels, they are often dominated by seasonally and diurnally varying fluxes of the terrestrial biosphere. Thus, it is nearly impossible to make use of the atmospheric CO₂ observations alone as an independent constraint on the space–time patterns of fossil fuel CO₂ emissions (Shiga et al., 2014). In addition, conventional inversion schemes (Rödenbeck et al., 2003; Peters et al., 2007; Gurney et al., 2008; Mueller et al., 2008; Chevallier et al., 2010a; Basu et al., 2013; Takagi et al., 2014) typically prescribe fossil fuel CO₂ fluxes from inventories based on economic

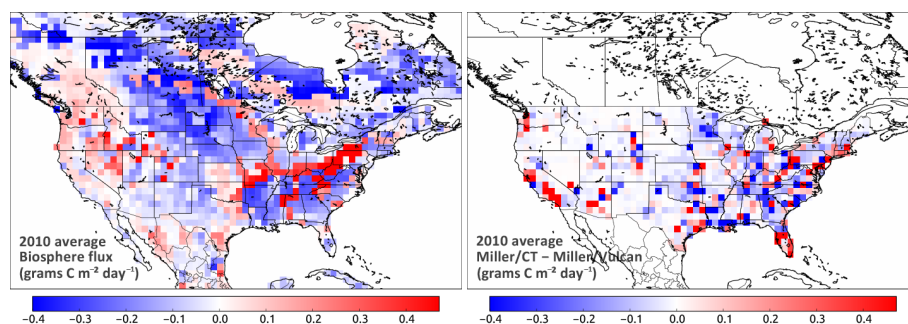


Figure 1. Comparative magnitudes of the annual average NEE estimated by CarbonTracker 2013B (left) and the difference between two fossil fuel inventories, Miller/CT and Miller/Vulcan (right). CarbonTracker is an atmospheric inversion which estimates CO₂ surface fluxes given atmospheric CO₂ measurements and “perfectly known” fossil fuel emissions. Miller/CT is the fossil fuel emission map prescribed in CarbonTracker 2013B, while Miller/Vulcan is a redistribution of the Miller/CT annual total fossil fuel CO₂ emission over the conterminous United States according to the spatiotemporal pattern of the Vulcan fossil fuel inventory (Gurney et al., 2009). While annual total emissions over the conterminous US for the two inventories are the same (i.e., the reds and blues in the right figure sum to zero), over individual 1° × 1° grid cells their difference can be comparable to the NEE estimated at the same location.

statistics on fossil fuel consumption and assumed combustion efficiencies (cf. Andres et al., 2012) with an assigned uncertainty of zero. Under these conditions, any deviation of the prescribed fossil fuel CO₂ fluxes from their true values can be expected to result in errors in the retrieved estimates of the terrestrial biospheric exchange fluxes. In areas over which the total carbon budget is well constrained by large number of observations, such as the conterminous US, these “carry-over biases” may be comparable in magnitude to the errors in the specified fossil fuel CO₂ fluxes themselves.

The assumption of perfectly well-known fossil fuel fluxes has been a reasonable starting point since annual total fossil fuel CO₂ emissions from most developed (i.e., UNFCCC Annex I and Annex II) countries are likely known to within 5–10 % (Andres et al., 2012), a level of certainty that greatly surpasses our knowledge of the annual net terrestrial biosphere CO₂ flux over those areas. However, for developing (non-Annex) countries, fossil fuel uncertainties are likely to be much larger. For example, estimates of Chinese emissions from fossil fuel combustion and cement production have been revised by +17 % (Guan et al., 2012) and –14 % (Liu et al., 2015) over the past 5 years alone. Moreover, uncertainties in fossil fuel CO₂ emissions are likely to be larger (by unknown amounts) at subnational and subannual scales, even in developed countries. To illustrate this, we show maps of the difference between two widely used inventories of the annual fossil fuel CO₂ flux over North America along with an estimate of annual average net ecosystem exchange (NEE) in Fig. 1. The inventory differences are in some cases similar in magnitude to estimates of NEE for individual grid cells (at a resolution of 1° × 1° in this example). Making matters worse, it is frequently necessary to extrapolate emissions inventories forward in time to correspond with the times of atmospheric observations. Such extrapolations might reasonably account for changes in population but will not capture changes in

fossil fuel use associated with, for example, protracted regional heat and cold waves. At the time of this writing, both the Vulcan (<http://vulcan.project.asu.edu/research.php>) and EDGAR (<http://edgar.jrc.ec.europa.eu/overview.php?v=42>) inventories provide emissions estimates only up to 2008, and even the “fast track” version of EDGAR (EDGAR v4.2 FT2010) has yet to be updated beyond 2010.

Here we take the initial steps at determining fossil fuel emissions and evaluating our ability to reduce carry-over bias using an atmospheric top-down method and the existing and anticipated array of precise measurements of atmospheric ¹⁴CO₂, which provide for direct, precise (~ 1 ppm) and largely unbiased observational constraints on fossil-fuel-derived CO₂ in the same samples that provide the primary CO₂ observations (Turnbull et al., 2009; Levin et al., 2011; Miller et al., 2012; Lehman et al., 2013). Below we first describe a new inversion framework that assimilates both CO₂ and ¹⁴CO₂ in a system that simultaneously optimizes both fossil fuel and biospheric exchange fluxes of CO₂. We then outline a set of Observation System Simulation Experiments (OSSEs) designed to evaluate the ability of the dual tracer inversion framework to separately estimate these fluxes over the conterminous US using synthetic observations corresponding in space and time to (a) actual observations in the NOAA ESRL Global Greenhouse Gas Reference Network (GGRN) in 2010 (1063 ¹⁴CO₂ measurements globally, of which 969 were in North America) and (b) an enhanced observational network with 6448 ¹⁴CO₂ measurements globally in 2010 (5304 in North America), similar to the annual ¹⁴CO₂ coverage recently recommended by the US NAS (Pacala et al., 2010). We use the observational network of (b) in three additional experiments. First, we perform an ensemble of inversions with and without ¹⁴CO₂ data in order to evaluate the degree to which the inclusion of ¹⁴CO₂ observations allows us to distinguish between biospheric and

fossil fuel CO₂ fluxes. We also repeat (b) without ¹⁴CO₂ data in order to quantify (by contrast to the dual tracer results) the degree to which the dual tracer system is able to detect and minimize potential carry-over bias in NEE that might otherwise arise from a biased fossil fuel prior. Finally we repeat (b) but with different models of atmospheric transport to generate and assimilate the synthetic observations, in order to evaluate the potential impact of transport model error on our emissions estimates.

2 The inversion framework

Our inversion framework builds on the TM5 4DVAR system (Meirink et al., 2008), which has been used for estimating sources and sinks of CH₄ (Bergamaschi et al., 2013; Houweling et al., 2014), CO (Hooghiemstra et al., 2011), CO₂ (Basu et al., 2013), and N₂O (Corazza et al., 2011). Here we describe modifications to the TM5 4DVAR system that permit us to jointly assimilate the measurements of two tracers, CO₂ and ¹⁴CO₂.

The atmospheric mass balances of CO₂ and ¹⁴CO₂ have been presented previously by Miller et al. (2012). Following those equations, we rewrite the isotopic mass balance (Eq. 1b and c of Miller et al., 2012) in terms of the transported and conserved quantity $C \Delta_{\text{atm}}$, while the carbon balance (1a) remains the same, such that

$$\frac{d}{dt}C = F_{\text{bio}} + F_{\text{oce}} + F_{\text{fos}} \quad (1a)$$

$$\frac{d}{dt}(C \Delta_{\text{atm}}) = \frac{N}{r_{\text{std}}}(F_{\text{nuc}} + F_{\text{cosmo}}) + \Delta_{\text{fos}}F_{\text{fos}} \quad (1b)$$

$$\begin{aligned} &+ \Delta_{\text{atm}}(F_{\text{oce}} + F_{\text{bio}}) + (\Delta_{\text{oce}} - \Delta_{\text{atm}})F_{\text{oceatm}} \\ &+ (\Delta_{\text{bio}} - \Delta_{\text{atm}})F_{\text{bioatm}} \\ &= \frac{N}{r_{\text{std}}}(F_{\text{nuc}} + F_{\text{cosmo}}) + \Delta_{\text{fos}}F_{\text{fos}} \quad (1c) \\ &+ \Delta_{\text{atm}}(F_{\text{oce}} + F_{\text{bio}}) + F_{\text{ocedis}} + F_{\text{biodis}}, \end{aligned}$$

where C is the atmospheric burden of CO₂ and Δ_{atm} is the isotope signature of ¹⁴CO₂ in the atmosphere expressed in Δ notation, which includes corrections for mass-dependent isotopic fractionation between reservoirs and radioactive decay between the times of sample collection and measurement, such that the quantity $\Delta^{14}\text{CO}_2$ is conserved in time (Stuiver and Polach, 1977, where $\Delta^{14}\text{C}$ is equivalent to $\Delta^{14}\text{CO}_2$ here). F_{bio} , F_{oce} , and F_{fos} are net CO₂ surface fluxes to the atmosphere from the terrestrial biosphere, oceans, and fossil fuel burning respectively, and we set Δ_{fos} to -1000% , corresponding to a fossil fuel source devoid of ¹⁴C as a result of radioactive decay. F_{nuc} is the ¹⁴CO₂ flux from nuclear power and reprocessing plants, and F_{cosmo} is the cosmogenic production of ¹⁴CO₂, corresponding to the terms $\text{iso}F_{\text{nuc}}$ and $\text{iso}F_{\text{cosmo}}$ respectively of Miller et al. (2012). To convert these pure ¹⁴CO₂ fluxes into units of CO₂ flux $\times \Delta$ (e.g., PgC % yr⁻¹), as in the other terms on the right-hand

side of Eq. (1c), we divide by the ¹⁴C : C standard ratio, $r_{\text{std}} = 1.176 \times 10^{-12}$, and account for mass-dependent fractionation by multiplying by $N = (975/(\delta^{13}\text{C} + 1000))^2$ (Stuiver and Polach, 1977), where $\delta^{13}\text{C}$ has an assumed atmospheric value of -8% . Δ_{oce} and Δ_{atm} are the isotope signatures of the ocean and the atmosphere respectively. In Eq. (1c) we assume that in converting from ¹⁴C : ¹²C to $\Delta^{14}\text{C}$ all isotopic fractionation between reservoirs “drop out” of the equations, such that we can equate the isotopic signature $\Delta_{\text{atm} \rightarrow x}$ to Δ_{atm} , and $\Delta_{x \rightarrow \text{atm}}$ to Δ_x . F_{oceatm} and F_{bioatm} are the one-way gross ocean to atmosphere and biosphere to atmosphere CO₂ fluxes. The terms $F_{\text{ocedis}} = (\Delta_{\text{oce}} - \Delta_{\text{atm}})F_{\text{oceatm}}$ and $F_{\text{biodis}} = (\Delta_{\text{bio}} - \Delta_{\text{atm}})F_{\text{bioatm}}$ are so-called disequilibrium fluxes (where $\Delta_{\text{bio}} - \Delta_{\text{atm}} = \Delta_{\text{biodis}}$ in Miller et al., 2012). Note, finally, that an extra term involving the net ocean and terrestrial fluxes (F_{oce} and F_{bio}) appears in Eq. (1c), compared to Eq. (1b) of Miller et al. (2012), due to the slightly different left-hand sides ($d(C \Delta_{\text{atm}})/dt$ vs $C d\Delta_{\text{atm}}/dt$) of the two equations. Their magnitudes are only $\sim 100 \text{ PgC } \% \text{ yr}^{-1}$ which is relatively small compared to, for example, the fossil fuel flux of $\sim 10\,000 \text{ PgC } \% \text{ yr}^{-1}$.

To solve Eq. (1) in an inversion, we further separate terms in Eq. (1a) into the sum of oceanic and terrestrial biospheric (hereafter referred to as “natural”) components and fossil fuel components, where CO₂^{ff} denotes the CO₂ in the atmosphere accumulated due to fossil fuel burning since the beginning of the simulation period (t_0). We note here that natural in this context is a mathematical definition of convenience, not to be confused with pre-anthropogenic. What we refer to as the natural flux has been influenced by historical anthropogenic emissions, land use changes and climate change.

$$\frac{d}{dt}\text{CO}_2^{\text{nat}} = F_{\text{oce}} + F_{\text{bio}} \quad (2a)$$

$$\frac{d}{dt}\text{CO}_2^{\text{ff}} = F_{\text{fos}} \quad (2b)$$

$$\text{CO}_2^{\text{ff}}(t = t_0) = 0 \quad (2c)$$

Our system is primarily designed to estimate fossil fuel CO₂ fluxes and NEE. However, we also solve for F_{ocedis} and F_{biodis} at a coarser temporal resolution, as explained in Sect. 2.1.2. Note that Eq. (1c) contains Δ_{atm} on both sides. However, we do not solve for a Δ_{atm} field self-consistently within the inversion framework. On the left-hand side, we treat $C \Delta_{\text{atm}}$ as a single tracer. Accordingly, we convert all measured ¹⁴CO₂ values to “measurements” of $C \Delta_{\text{atm}}$ for the flux estimation. On the right-hand side, for the term $\Delta_{\text{atm}}(F_{\text{oce}} + F_{\text{bio}})$, we specify a Δ_{atm} that is spatially uniform and has a smooth temporal variation based on observations from the well mixed free troposphere at Niwot Ridge, Colorado (NWR: 40.0531° N, 105.5864° W, <http://www.esrl.noaa.gov/gmd/dv/iadv/graph.php?code=NWR&program=cgg&type=ts>), filtered to remove possible local urban influences from the Denver-Boulder area to the east (Lehman et al., 2013). The error

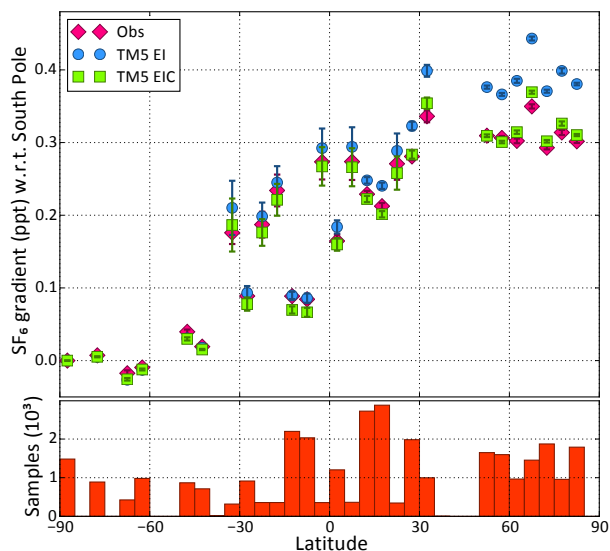


Figure 2. The observed and modeled latitudinal gradients of SF₆, estimated as the difference between SF₆ concentrations at marine boundary layer sites of the NOAA ESRL GGRN (<http://www.esrl.noaa.gov/gmd/ccgg/ggrn.php>) and the South Pole. Observations and models span 10 years from 2002 to 2011. For each site, we account for time-dependent changes by calculating a linear trend from observed SF₆ and removing that from all three time series (observed, TM5 EI, TM5 EIC). All observations were binned by latitude in 5° increments and averaged. The bottom panel shows the number of samples averaged per latitude bin. The error bars denote ±2σ intervals, where σ is the standard error of the mean difference with respect to the South Pole. Measured SF₆ mole fractions used here are available in the Supplement.

made in the inversion by using this smoothed approximation of Δ_{atm} on the right-hand side of Eq. (1c) is small, since it will in practice be very close to Δ_{atm} in $C\Delta_{\text{atm}}$ and, as noted above, the term $\Delta_{\text{atm}}(F_{\text{oce}} + F_{\text{bio}})$ is small compared to others in the overall budget. For the disequilibrium fluxes on the right-hand side, we solve for F_{ocedis} and F_{biadis} but do not attempt to separate those into the one-way gross CO₂ fluxes and their respective isotopic disequilibria.

2.1 Modeling framework

2.1.1 Atmospheric transport

We use the TM5 atmospheric tracer transport model (Krol et al., 2005) to simulate atmospheric tracer concentrations from surface fluxes. TM5 can be run with convective entrainment and detrainment fluxes determined directly from the ERA-Interim reanalysis from the European Centre for Medium range Weather Forecasts (henceforth called TM5 EIC) or with those fluxes computed within TM5 according to the convective scheme of Tiedtke (1989) (henceforth, TM5 EI), which was the standard scheme prior to 2014. The largest difference between TM5 EI and TM5 EIC is in the

vertical transport into the free troposphere over temperate latitudes. For tracers with surface sources and sinks and negligible atmospheric chemical production and loss – such as CO₂ and SF₆ – this difference creates markedly different north–south (N–S) gradients at the surface, even though the advective winds are the same. As an illustration, in Fig. 2 we show the average simulated N–S gradient of SF₆ within the marine boundary layer for both TM5 EIC and TM5 EI, compared to average observations from 2002 to 2011.

The 0.3 ppt N–S gradient in SF₆ of TM5 EIC is very close to the observed gradient of 0.295 ppt, whereas the 0.38 ppt N–S gradient of TM5 EI is the farthest outlier among 16 global transport models considered by Patra et al. (2011). Moreover, in the analysis of Patra et al. (2011), most modeled N–S gradients were between 0.27 and 0.32 ppt. Thus, the difference of 0.08 ppt in the N–S gradients simulated by TM5 EI and TM5 EIC is larger than typical inter-model differences, indicating that these two schemes represent very different realizations of transport, at least at the hemispheric and global scales. Since TM5 EIC delivers markedly better agreement with the observed SF₆ N–S gradient, we use TM5 EIC for both forward simulation and inversion in all experiments, except when evaluating the impact of transport error on estimated fluxes (for which we use TM5 EI to assimilate synthetic observations produced by TM5 EIC, as outlined in Sect. 3.4).

To better resolve atmospheric transport over the domain of interest, we run the atmospheric transport model at 1° × 1° resolution over North America (20–64° N, 132–60° W), and at 3° × 2° resolution elsewhere. This is the same nested zoom configuration employed in NOAA’s CarbonTracker North America (carbontracker.noaa.gov).

2.1.2 TM5 4DVAR

The TM5 4DVAR inversion system estimates fluxes \mathbf{x} given observations \mathbf{y} by minimizing the so-called cost function J (Meirink et al., 2008):

$$J = \frac{1}{2}(\mathbf{H}\mathbf{x} - \mathbf{y})^T \mathbf{R}^{-1}(\mathbf{H}\mathbf{x} - \mathbf{y}) + \frac{1}{2}(\mathbf{x} - \mathbf{x}_0)^T \mathbf{B}^{-1}(\mathbf{x} - \mathbf{x}_0), \quad (3)$$

where H is an atmospheric transport operator, \mathbf{x}_0 is the prior flux before doing a data assimilation, and \mathbf{R} and \mathbf{B} are the respective error covariance matrices of the model–data mismatch and the prior flux. The TM5 variational framework for atmospheric inversion of a single species has been described in detail previously (Meirink et al., 2008; Hooghiemstra et al., 2011; Basu et al., 2013). In this work, \mathbf{x} contains the surface fluxes of the three species CO₂^{ff} (F_{fos}), CO₂^{nat} (F_{oce} and F_{bio}), and $C\Delta_{\text{atm}}$ (F_{ocedis} and F_{biadis}). We solve for F_{bio} , F_{oce} , and F_{fos} weekly, and for F_{biadis} and F_{ocedis} monthly. The prior flux error covariance matrix is assumed to be separable in time and space, as in

Table 1. Spatial flux error covariance parameters of Eq. (4) for different categories.

Category	Optimized	C_r type	L (km)	σ
F_{bio}	yes	e	200	$0.5 \times \text{respiration}$
F_{oce}	yes	e	1000	$1.57 \times \text{absolute flux}$
F_{fos}	yes	h	500	$2.5 \times \text{inter-prior spread}^{\text{a}}$
F_{ocedis}	yes	r	–	$0.2 \times \text{absolute flux}^{\text{b}}$
F_{biodis}	yes	r	–	$0.5 \times \text{absolute flux}^{\text{c}}$
F_{nuc}	no	–	–	–
F_{cosmo}	no	–	–	–

^a For fossil fuel CO₂ flux, the “inter-prior spread” denotes the spread between three fossil fuel inventories, CarbonTracker/Miller, CarbonTracker/Vulcan, and ODIAC (Oda and Maksyutov, 2011). For defining the region boundaries across which the prior flux correlation goes to zero, we used nine divisions of the continental United States defined by the US Census Division (www.eia.gov/forecasts/aeo/pdf/f1.pdf), shaded in Fig. 5. The rest of North America falls into a single region, while other continents, namely South America, Europe, Africa, Asia, and Australia form five separate regions. All ocean pixels fall in one single region, while non-optimized pixels (Greenland and Antarctica) fall into one region.

^b The world’s oceans are divided into the 11 TRANSCOM ocean regions (Law et al., 2000).

^c Outside North America, the land is divided up into nine TRANSCOM land regions. Inside North America, the North American temperate region is by itself, while the North American boreal region is further subdivided into 11 regions used by CarbonTracker 2013b (http://www.esrl.noaa.gov/gmd/ccgg/carbontracker/CT2013B_doc.php, Sect. 8.1.1).

$$\mathbf{B}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) = \text{cov}(x_{r_1, t_1}, x_{r_2, t_2}) \quad (4)$$

$$= \sigma_{r_1, t_1} \sigma_{r_2, t_2} C_r(\mathbf{r}_1, \mathbf{r}_2) C_t(t_1, t_2),$$

where \mathbf{r} and t are space and time coordinates respectively, $\sigma_{r, t}$ is the uncertainty of the prior flux at location \mathbf{r} at time t , and $C_r(\mathbf{r}_1, \mathbf{r}_2)$ (or $C_t(t_1, t_2)$) is the correlation between flux errors at locations \mathbf{r}_1 and \mathbf{r}_2 (or times t_1 and t_2). No prior correlation is assumed between the five flux categories being optimized. For each category, the temporal error correlation C_t is assumed to be exponential, $C_t(t_1, t_2) = e^{-|t_1 - t_2|/T}$, with T being 3 months for F_{oce} , F_{fos} , and F_{ocedis} , and 1 month for F_{bio} and F_{biodis} . The spatial error correlation is either

e: exponential, $C_r(\mathbf{r}_1, \mathbf{r}_2) = e^{-|\mathbf{r}_1 - \mathbf{r}_2|/L}$, or

r: regional, where the globe is subdivided into regions, and grid cells within one region are perfectly correlated, whereas grid cells from different regions are completely uncorrelated, or

h: a hybrid of the first two, where the grid cell to grid cell correlation decays exponentially within each defined region, but is zero between regions.

The parameters of spatial correlation for the five categories, as well as the prior errors per grid cell (i.e., $\sigma_{r, t}$ of Eq. 4) are listed in Table 1.

Surface fluxes are solved for at the same lateral resolution as the transport ($3^\circ \times 2^\circ$ globally, $1^\circ \times 1^\circ$ over North America), to provide the inversion with flexibility to change surface fluxes where there are more observations, and to reduce aggregation error (Kaminski et al., 2001). This transport/flux configuration is similar to NOAA’s CarbonTracker North America, except that we solve for additive corrections

to surface fluxes per grid cell instead of multiplicative corrections to regional surface fluxes. We focus on the year 2010, and our inversions run from 4 July 2009 to 1 April 2011, to allow for sufficient spin up time at the beginning and sufficient time for the fluxes at the end of 2010 to be captured by subsequent observations.

2.2 ¹⁴CO₂ flux terms

Equation (1a) and (1c) contain seven different flux terms on the right-hand side. In the OSSE we create synthetic observations of $C \Delta_{\text{atm}}$ by specifying and transporting “true” flux fields for all seven terms. For the inversions, we specify prior fluxes associated with fossil fuel CO₂ emissions (F_{fos}) and net oceanic and biospheric fluxes (F_{oce} and F_{bio}) that differ from those used to produce the simulated observations, and evaluate our ability to recover true fluxes using the synthetic observations. The two different sets (true vs. a priori) of fossil fuel CO₂ and net CO₂ flux terms are described in Sect. 3.3. The construction of the isofluxes for the remaining terms is described below and is consistent with the recent tropospheric $\Delta^{14}\text{CO}_2$ budget and distribution based on observations.

Gridded estimates of the ¹⁴C production flux from nuclear reactors and fuel reprocessing plants, F_{nuc} , were taken from Graven and Gruber (2011) and did not vary with time. Only the portion of this flux estimated to be directly emitted as ¹⁴CO₂ was included. The production of ¹⁴C in the atmosphere, F_{cosmo} , and the sensitivity of this production to geomagnetic latitude depend on the solar modulation parameter Φ , a scalar which varies with time. Annual values of Φ were calculated through 2012 based on a global array of neutron monitor data obtained from <http://nmdb.eu/> (all amplitude normalized to count rates at Deep River, Canada, http://neutronm.bartol.udel.edu/~pyle/bri_table.html) and the slope of a linear regression between annual average Deep River Neutron Monitor count rate and estimates of Φ between 1955 and 1995 from Masarik and Beer (1999). Then, for each year of our simulation period, we calculated the ¹⁴C production as a function of geomagnetic latitude given the annual average Φ of that year (Masarik and Beer, 2009). This resulted in annually varying production fields dependent on geomagnetic latitude. These production fields were then distributed vertically over the TM5 model layers corresponding to the stratosphere (between 150 and 3 hPa), with the mass of ¹⁴CO₂ in each layer proportional to the total mass of air in that layer. To better match the observed ¹⁴CO₂ trend at NWR, the global total cosmogenic production was scaled by 0.9 in all years.

To calculate the terrestrial disequilibrium flux term ($\Delta_{\text{bio}} - \Delta_{\text{atm}}) F_{\text{bioatm}}$, we first constructed the historical time series of atmospheric ¹⁴CO₂ by compositing overlapping time series from tree ring measurements (Stuiver, 1982), atmospheric records from Vermont, Austria (Levin et al., 1994), Schauinsland, Germany ¹⁴C (Levin and Kromer, 1997), Jungfrau-

Table 2. Inversions and model runs performed in this work.

Section (Sect.) Figure (Fig.)	Experiment	Model run	Obs. network	Transport	Initial fluxes	Optimized fluxes*
Sect. 3.1, Sect. 3.2	2010 (simul)	forward	2010 coverage	TM5 EIC	true	None
Sect. 3.1, Sect. 3.2	NRC 5000 (simul)	forward	NRC 5000	TM5 EIC	true	None
Sect. 4, Fig. 6, Fig. 7, Fig. 8	2010 (assim)	inverse	2010 coverage	TM5 EIC	prior	all except F_{nuc} , F_{cosmo}
Sect. 4, Fig. 6, Fig. 7, Fig. 8, Fig. 11	NRC 5000 (assim)	inverse	NRC 5000	TM5 EIC	prior	all except F_{nuc} , F_{cosmo}
Sect. 4.3, Fig. 8, Fig. 11	NRC 5000 (EI)	inverse	NRC 5000	TM5 EI	prior	all except F_{nuc} , F_{cosmo}
Sect. 4.1, Fig. 9	NRC 5000	inverse	NRC 5000	TM5 EIC	true + random perturbations	all except F_{nuc} , F_{cosmo}
Sect. 4.1, Fig. 9	NRC 5000 (no ¹⁴ CO ₂)	inverse	NRC 5000 (only CO ₂)	TM5 EIC	true + random perturbations	all except F_{nuc} , F_{cosmo}
Sect. 4.2, Fig. 10	NRC 5000 (traditional)	inverse	NRC 5000 (only CO ₂)	TM5 EIC	prior	only F_{bio} , F_{oce}

* Of the seven flux terms in Eq. (1).

joch, Switzerland ¹⁴C (Levin et al., 2013), and more recently NWR, United States (Lehman et al., 2013). This historical time series was convolved with the age distribution of respired carbon derived from pulse response functions from the Carnegie Ames Stanford Approach (CASA) biosphere model (Thompson and Randerson, 1999), to obtain a monthly $\Delta^{14}\text{CO}_2$ of respired carbon, Δ_{bio} , for each $1^\circ \times 1^\circ$ CASA grid cell. Δ_{atm} was derived from filtered, monthly average observations at NWR, to obtain $(\Delta_{\text{bio}} - \Delta_{\text{atm}})$, and F_{bioatm} was determined from the monthly total heterotrophic respiration flux for each CASA grid cell. Monthly F_{bioatm} did not vary from year to year, while Δ_{bio} and Δ_{atm} were updated monthly and from year to year based on observed changes in atmospheric $\Delta^{14}\text{CO}_2$.

The oceanic isotopic disequilibrium ($\Delta_{\text{oce}} - \Delta_{\text{atm}}$) was estimated from observations of the $\Delta^{14}\text{C}$ of surface ocean dissolved inorganic carbon field available from World Ocean Circulation Experiment (WOCE) for the 1980's–1990's and updated yearly through 2012 using rates of change for different ocean regions based on subsequent observations from the Climate and Ocean Variability (CLIVAR) measurement program (<http://cdiac.ornl.gov/oceans/datmet.html>) as in Lindsay (2016). The gridded annual estimates of Δ_{oce} were differenced from a zonally uniform surface layer Δ_{atm} field based on filtered and seasonally smoothed observations from NWR, but with a specified increase of +10‰ between 20° N and 20° S. The disequilibrium flux was then calculated by multiplying the isotopic disequilibrium by the one-way ocean to atmosphere CO₂ flux for each grid cell, which was derived from a climatology of surface ocean pCO₂ from Takahashi (2009) and a quadratic wind-speed-dependent pis-

ton velocity (Wanninkhof, 1992) scaled to a more recent analysis of the oceanic ¹⁴C inventory (Sweeney et al., 2007).

2.3 Initial atmospheric CO₂ and ¹⁴CO₂ fields

Initial concentration fields of CO₂ and ¹⁴CO₂ for the inversions were obtained by specifying realistic troposphere–stratosphere and latitude gradients of $\Delta^{14}\text{CO}_2$ and CO₂ and then propagating time-varying flux terms in Eq. (1) through the atmosphere using TM5 EIC, starting on 1 January 2000. The three-dimensional atmospheric mole fractions of CO₂ × $\Delta^{14}\text{CO}_2$ and CO₂ on 4 July 2009 were used as initial fields for the inversions. The relatively long forward run was implemented to ensure that the simulated large-scale atmospheric gradients were consistent with the prior fluxes.

3 Experimental design

Our OSSEs (Table 2) are designed to evaluate the ability of a network of ¹⁴CO₂ observations – in conjunction with more widely available CO₂ observations – to constrain regional fossil fuel CO₂ and net biosphere exchange fluxes within our inversion framework. To do this, we first create synthetic atmospheric ¹⁴CO₂ and CO₂ concentrations at real and projected measurement locations based on transport of a set of true fluxes in TM5 EIC (this step is sometimes referred to as the “nature run” for an OSSE). By true we do not suggest that these fluxes are accurate but that they are consistent with the synthetic observations for the purpose of conducting the OSSE. We then assimilate the synthetic measurements in an atmospheric inversion using prior flux estimates which differ

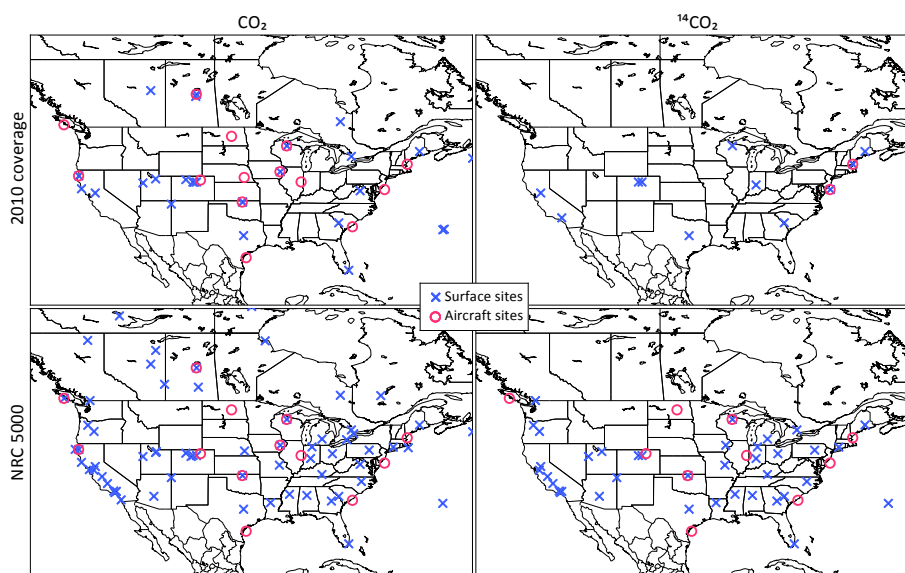


Figure 3. The sites for which CO₂ and ¹⁴CO₂ measurements were simulated and then assimilated in our OSSEs for two different coverage scenarios, 2010 and NRC 5000, as described in the text.

substantially from the true fluxes. The extent to which fluxes estimated by the inversion match the true fluxes is a measure of the performance of our inversion framework and the network of (synthetic) observations. An additional metric of performance is the degree to which ¹⁴CO₂ data can distinguish between NEE and fossil fuel CO₂ fluxes, measured by the posterior correlation between the two. This metric is further discussed in Sects. 3.5 and 4.1.

3.1 True fluxes

True fluxes used to simulate the observations are those for ¹⁴CO₂ described in Sect. 2.2 along with those for fossil fuel CO₂ and net ocean and biosphere exchange. For fossil fuel CO₂, we use fossil fuel fluxes from CarbonTracker 2013, redistributed within the continental US according to the Vulcan spatiotemporal pattern. In addition, we impose scaling factors of Nassar et al. (2013) in order to represent the diurnal variability. True ocean fluxes were taken from posterior fluxes of CarbonTracker 2013b, specifically the variant which used the ocean interior inversion of Jacobson et al. (2007b) to construct prior ocean fluxes. True terrestrial fluxes were based on the CASA Global Fire Emissions Database (GFED) 3 model (van der Werf et al., 2003). CASA GFED 3 provided only monthly NEE fluxes; in order to represent variability at higher frequencies we imposed daily and 3 hourly variations from SiBCASA GFED4 (van der Velde et al., 2014) on the monthly fluxes.

3.2 Synthetic observations

We simulated two sets of observations, with distributions as shown in Fig. 3 and Table 3. The first set, which we refer to as 2010 coverage, placed a ¹⁴CO₂ (or CO₂) observation at each spatiotemporal point where there was an actual ¹⁴CO₂ (or CO₂) measurement between 4 July 2009 and 1 April 2011. This resulted in a total of 1639 ¹⁴CO₂ and 45 330 CO₂ observations over the 21 month period (1475 and 18 008 over North America, respectively). The accuracy of the estimated surface fluxes with respect to the true fluxes is expected to provide a measure of the performance of the real observational network in 2010.

For the second set, which we refer to as NRC 5000, we simulated ~ 5000 ¹⁴CO₂ measurements per year over North America. In constructing the expanded, hypothetical observational network (Fig. 3) we first sought to increase measurements at existing NOAA and NOAA-partner monitoring locations, including tall towers and airborne and surface flask sampling locations, adding six new tall tower sites to fill gaps in the sampling network. For CO₂ we also added shipboard samples from two monthly cruises in the Pacific Ocean. Table 3 lists the sampling frequencies for CO₂ and ¹⁴CO₂ at the different sites.

Sampling within the NRC 5000 network conformed as closely as possible to the actual sampling protocols and periodicities at tower, flask, aircraft, and cruise locations maintained by NOAA and its partner networks. At tower sites, we sampled the true CO₂ field twice a day at the highest intake height, at 00:30 and 03:30 local solar time (LST) for mountaintop sites and at 12:30 and 15:30 LST otherwise. The true ¹⁴CO₂ field was sampled on Mondays and Thursdays follow-

Table 3. Sampling frequency of CO₂ and ¹⁴CO₂ measurements at the sites of our hypothetical NRC 5000 network. Even though Fig. 3 only displays sites over the conterminous US and part of Canada, our NRC 5000 network also contains some background sites such as the South Pole and Mauna Loa. The numbers below include all sites, globally.

Site type	CO ₂		¹⁴ CO ₂	
	No. of sites	Sampling freq.	No. of sites	Sampling freq.
Tower	62	2 day ⁻¹	35	2 week ⁻¹
Flask	76	1 week ⁻¹	21	1 week ⁻¹
Aircraft	19	1 week ⁻¹ , up to 16 altitudes	11	1 week ⁻¹ , 3 altitudes
Cruise	2	1 transec month ⁻¹ , every 5° latitude	–	–

ing the same protocol for intake height and LST. Flask sites were sampled on Wednesdays at 13:30 LST (01:30 LST for mountaintop sites) for both tracers. Some NOAA flask sites – such as Ascension Island, Cold Bay (Alaska), and Guam – collect CO₂ samples less frequently. At those sites, our sampling followed the protocol for CO₂ at the other flask sites, but with sampling only every other week. At aircraft sites, we sampled simulated CO₂ at 13:30 LST, at altitudes where actual CO₂ samples are obtained (typically every 1000 to 2000 feet, to a site-dependent maximum altitude). This resulted in between nine and twelve samples per profile, depending on the site. For ¹⁴CO₂, three samples were taken per aircraft profile, distributed between the boundary layer and the free troposphere, reflecting the actual ongoing aircraft sampling strategy for ¹⁴CO₂ (cf. Miller et al., 2012). Shipboard samples for CO₂ were simulated as samples along a transect once every 5° latitude, successive samples being separated by 1 day, along NOAA Pacific Ocean and Western Pacific cruises, which go back and forth once a month.

3.3 Prior flux specifications for OSSEs

For the inversion of synthetic observations, we specified a set of prior fossil fuel CO₂ and net biospheric and oceanic fluxes that differed from those used to create the data. Prior fossil fuel CO₂ fluxes were taken from the EDGAR 4.2 FT2010 global inventory (<http://edgar.jrc.ec.europa.eu/overview.php?v=42FT2010>). EDGAR fluxes were available at 1° × 1° resolution, but had no subannual variability and were available only through 2010. For 2011, country totals for 2010 were scaled up according to the growth rate between 2010 and 2011 for each country from statistics compiled by BP (<http://www.bp.com/en/global/corporate/about-bp/energy-economics/statistical-review-of-world-energy/statistical-review-downloads.html>). The fossil fuel flux was optimized over weekly time steps. We imposed – but did not optimize – an hour-of-day variability on the fossil fuel fluxes using the diurnal (but not day of week) scaling factors of Nassar et al. (2013). Prior terrestrial fluxes were

from SiBCASA/GFED4, which included NEE, fires, and biomass burning (van der Velde et al., 2014). The fluxes were specified globally on a 1° × 1° grid at 3 hour time steps. The inversion optimized weekly terrestrial fluxes at the lateral resolution of the TM5 transport model. Within each week, the prior 3 hourly variations were imposed as additive temporal patterns, but not optimized; i.e., only the mean NEE over a week was adjusted. Prior oceanic CO₂ fluxes, also at 1° × 1° and 3 hourly resolution, were taken from the ocean prior of CarbonTracker 2013b (the variant based on Jacobson et al., 2007a), and optimized weekly. The prior errors assumed for the different fluxes are listed in Table 1. Of the remaining four ¹⁴CO₂ flux terms described in Sect. 2.2, only the two disequilibrium terms were optimized during the inversion, while the nuclear and cosmogenic terms were held fixed.

3.4 Transport errors

The OSSEs described above allow for an accurate assessment of our ability to calculate fossil and biosphere fluxes given different sets of ¹⁴CO₂ and CO₂ observations, in the limit of perfectly known atmospheric transport (note, however, that the elements of the model–data mismatch matrix **R** are inflated to account for expected transport uncertainty). The performance of an inversion of real ¹⁴CO₂ data will be limited not only by the observations ingested, but also by errors in simulated atmospheric transport not adequately represented by **R** (e.g., Nassar et al., 2014; Liu et al., 2014; Hungershofer et al., 2010; Chevallier et al., 2009). Thus, a more comprehensive way to estimate the impact of transport model error is to use different transport models for the simulation and assimilation steps (Chevallier et al., 2010b). To that end, we conducted a controlled experiment where TM5 EI was used to assimilate synthetic data simulated by TM5 EIC. As described in Sect. 2.1.1, these two model variants differ substantially in their representations of vertical transport, which is an especially important component of the atmospheric transport with regard to flux estimation, since vertical transport directly influences the residence time of air

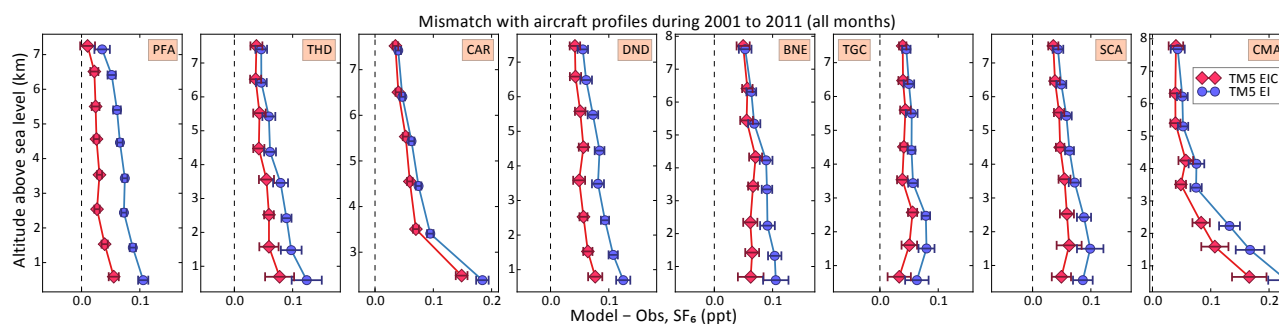


Figure 4. Average difference between observed and modeled aircraft profiles of SF₆ at eight different sites over the continental United States for the period 2001 to 2011 (inclusive). The error bars denote $\pm 2\sigma$ intervals, where σ is the standard error of the mean difference between each model and observations. The locations of the profiles are identified by three-letter site codes. Details for each site can be found at http://www.esrl.noaa.gov/gmd/dv/site/site_table2.php. Atmospheric mole fractions of SF₆ were simulated using the EI and EIC variants of the TM5 transport model. Three-dimensional initial conditions on 1 January 2000 were based on (a) vertical gradients from the end (1 January 2006) of a previous 6-year TM5 run (with initial conditions which included a specified latitude gradient but no vertical gradient) and (b) the 1 January 2000 smoothed marine boundary layer latitudinal gradient (Masarie and Tans, 1995) derived from SF₆ observations from the NOAA ESRL GGRN. The 1 January 2006 vertical gradients were zonally averaged, scaled back to 1 January 2000 and then added to the observed latitude gradient to create a zonally uniform but vertically and meridionally variable field. SF₆ emissions for the run were based on the spatial emission pattern from EDGAR v4.2 scaled to match the annual increases in SF₆ emissions derived from the observed SF₆ growth rate (assuming no atmospheric SF₆ loss). Measured SF₆ mole fractions used here are available in the Supplement.

within the continental boundary layer (CBL) and therefore the relationship between tracer flux and simulated concentrations in the CBL.

To illustrate this for our case, Fig. 4 shows the mismatch between modeled and measured vertical profiles of SF₆ over the continental US (Sweeney et al., 2015) for both TM5 EI and EIC. We once again consider SF₆ because it is a nearly inert gas (lifetime ~ 2000 years) and, like CO₂^{ff}, it has purely continental sources linked to industrial activity overwhelmingly in the northern midlatitudes (http://edgar.jrc.ec.europa.eu/part_SF6.php), but without a substantial seasonal cycle, Miller et al., 2012). Thus, we expect the vertical gradient of SF₆ over the continental US to depend on the strength of vertical mixing between the boundary layer and the free troposphere, and any systematic differences between simulated and observed gradients to provide an observational constraint on the representation of vertical transport processes in the different models. As shown in Fig. 4, both models display a mean offset from observations of ~ 0.04 ppt in the free troposphere, even at Trinidad Head (THD), which is upwind of the continent. This uniform free tropospheric offset is consistent with a 2002–2011 average offset of ~ 0.04 ppt between both models and the observed SF₆ at the South Pole. Apart from the upper level offset, the SF₆ gradient of TM5 EIC is consistently and significantly closer to the observations, suggesting that the EIC vertical transport scheme better represents the real atmosphere. Moreover, the vertical gradient of SF₆ between 850 and 400 hPa (i.e., between ~ 1.5 and ~ 7.1 km above sea level) for the two models differs by an average of 0.025 ppt across all sites, which is larger than the 0.018 ppt 1σ spread across 16 modern global transport models over midlatitude continents found by Patra et al. (2011).

This suggests that TM5 EI and TM5 EIC provide substantially different realizations of the transport not just at hemispheric scale (Fig. 2), but also at a location and scale most relevant to an “imperfect transport” OSSE for the conterminous US (Fig. 4).

3.5 OSSE evaluation

Flux inversion OSSEs are often evaluated according to the so-called “uncertainty reduction”, defined as the fractional reduction between the prior and posterior flux uncertainty (e.g., Rayner and O’Brien, 2001; Hungershofer et al., 2010). That metric, however, depends heavily on the prior uncertainties prescribed, and a large uncertainty reduction could easily arise from insufficient weighting of the prior during flux estimation. Moreover, iterative schemes such as the variational scheme used in TM5 4DVAR cannot estimate the full-rank posterior error covariance matrix (Meirink et al., 2008; Basu et al., 2013). Because of this limitation, we focus here instead on the mismatch between the inversion-estimated fluxes and true fluxes. Since prior and true fluxes differ significantly in both space and time, the ability of our inversion to recover the true fluxes (also referred to hereafter as the truth) should serve as a rigorous test of our observational and inversion framework.

For the “perfect transport” OSSEs, we also evaluate the posterior correlation between F_{fos} and F_{bio} to assess the degree to which these fluxes can be retrieved independently using (a) CO₂ data only and (b) using ¹⁴CO₂ and CO₂ data together. Conventional CO₂-only inversions solve Eq. (1a), but F_{fos} is prescribed and not optimized. However, if we were to solve Eq. (1a) for both $F_{\text{bio}} + F_{\text{oce}}$ and F_{fos} , in a CO₂-only

system we would expect a large negative correlation between the natural and fossil fuel fluxes, since under most circumstances CO₂ observations constrain the total flux and not its components. The magnitude of this correlation would be limited in part by how well the CO₂ observations constrained the total CO₂ budget for the domain of interest; in the limiting case of a perfectly constrained total CO₂ budget, this correlation would be -1 . Assimilating ¹⁴CO₂ observations in order to solve both Eq. (1a) and (1c) simultaneously, we should expect a reduction in the magnitude of negative correlation due to the independent information ¹⁴CO₂ provides about F_{fos} . The amount of reduction in the correlation between $F_{\text{bio}} + F_{\text{oce}}$ and F_{fos} thus serves as an objective metric of the ability of ¹⁴CO₂ observations to separate natural and fossil fuel CO₂ fluxes within our observational framework. In the case of the conterminous US, the natural CO₂ flux is largely equivalent to F_{bio} , or NEE.

The evaluation of this posterior flux correlation, however, is imprecise in a variational approach because $F_{\text{fos}} : F_{\text{bio}}$ correlations are derived from an approximate posterior covariance matrix as mentioned above. To obtain a more accurate estimate of the posterior covariance (and hence correlation) matrix, we follow the prescription of Chevallier et al. (2007). The posterior covariance between any two elements x_i and x_j of the state vector \mathbf{x} being estimated is

$$C_{ij}^{\text{apos}} = \left\langle (x_i^{\text{apos}} - \bar{x}_i^{\text{apos}})(x_j^{\text{apos}} - \bar{x}_j^{\text{apos}}) \right\rangle,$$

where the ensemble average is taken over an ensemble of variational inversions, each of which starts from a different prior and assimilates a different set of measurements, such that the probability distribution of all the priors follows the prior covariance matrix \mathbf{B} of Eq. (3), and the probability distribution of all the measurements follows the model–data mismatch (covariance) matrix \mathbf{R} of Eq. (3).

Choosing the number of inversions in the ensemble is a balancing act between statistical robustness and computer resources. Bousserez et al. (2015) recommended at least 50 inversions to estimate the posterior covariance matrix to within 10%. A key assumption in their recommendation was that the mean of the posterior estimates \mathbf{x}^{apos} corresponded to the analytical solution; i.e., each individual inversion had already “reached convergence” to within the analytical posterior error. In our case, due to the limited number of iterations performed (40 out of the theoretically required $n_{\text{state}} = 4\,095\,000$), we cannot be sure that within the ensemble, the \mathbf{x}^{apos} estimates are distributed with the analytical solution as their mean. However, in the case of our OSSEs, we know the analytical solution, which is the true flux of Sect. 3.1. Therefore, for evaluating the posterior covariance between F_{fos} and F_{bio} , we perform an ensemble of inversions where the prior fluxes are perturbations from the true flux following the statistics of \mathbf{B} . This approach of perturbing around a known truth to better estimate the posterior covariance is similar to that used by, e.g., Liu et al. (2014).

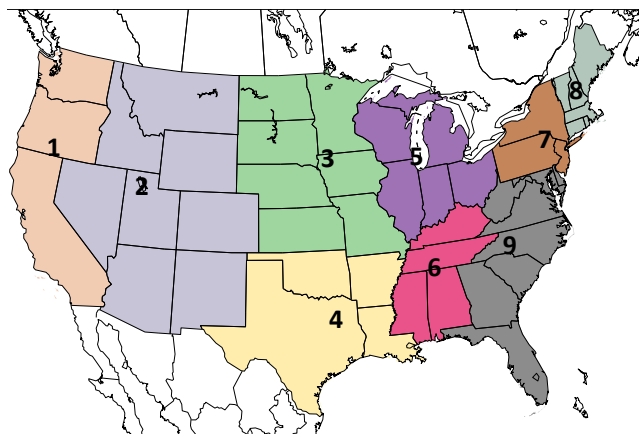


Figure 5. Nine regions defined by the US Census Division, over which we aggregate our fossil fuel CO₂ flux estimates (www.eia.gov/forecasts/aeo/pdf/f1.pdf).

To be on the safe side of the recommendation of Bousserez et al. (2015), our ensembles contain 100 inversions each.

Performing 100 independent inversions is computationally expensive. Therefore, we only evaluate the posterior correlation between CO₂^{ff} and CO₂^{nat} for two scenarios, (a) the NRC 5000 scenario, and (b) the NRC 5000 scenario without ¹⁴CO₂ observations. In an ideal system, for scenario (b) we expect to see large negative correlations between the posterior natural and fossil fuel CO₂ flux, at least over large areas where the total CO₂ flux is well constrained, and in scenario (a) we expect the negative correlations to be measurably smaller.

4 Results

OSSE results are considered at scales ranging from monthly national totals, monthly totals for regions specified in Fig. 5, and for groups of neighboring regions. Figures 6 and 7 compare monthly totals of the estimated fossil fuel CO₂ flux to specified true fluxes used to create the observations and the prior fluxes used in the inversions, for both 2010 and NRC 5000 measurement coverage. At the national scale, the monthly fossil fuel flux over the contiguous United States is recovered to within 5% (orange shaded region in Fig. 6) for all but 1 month for the 2010 measurement coverage, while the national, annual total is recovered to better than 1% (true flux = 1497.5 TgC, estimated flux = 1497.2 TgC). For the considerably denser measurement coverage of NRC 5000, the monthly US fossil fuel flux is recovered to within 5% (and usually to within 3%) for all months, while the national, annual total is again recovered to better than 1% (true flux = 1497.5 TgC, estimated flux = 1506.5 TgC). The impact of the increased coverage is more obvious when we consider smaller regions. Over the eastern and central US, the NRC 5000 scenario always yields monthly flux estimates

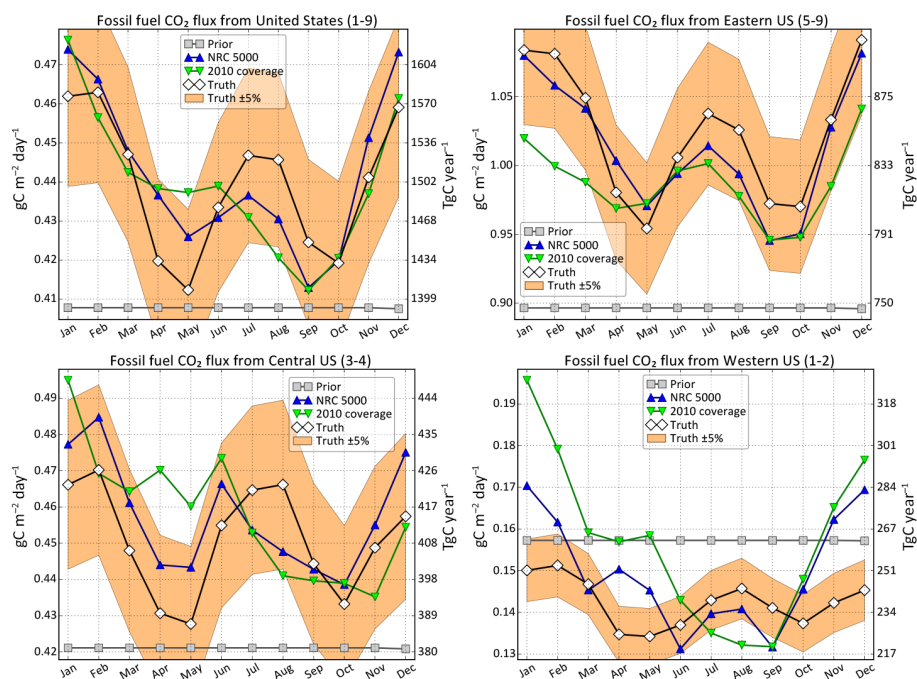


Figure 6. Monthly total emissions estimates for 2010 and NRC 5000 network scenarios, along with prior and true fluxes, aggregated for the conterminous US and neighboring groups of regions identified in Fig. 5. The orange band depicts the $\pm 5\%$ margin around the true fluxes, and the numbers next to region names refer to the region labels in Fig. 5.

that are within 5 % of the truth, and over the central US the phasing of the NRC 5000 estimate is much closer to the truth than that for the 2010 coverage. Estimates for the western US frequently deviate by more than 5 % from truth, even for the NRC 5000 scenario. This is likely due to the combination of the relatively small regional emissions and the fact that even for our case of effectively perfect transport, the elements of the transport that carry emissions from upwind regions to the sampling sites may be biased; indeed it appears that both 2010 and NRC 5000 observation networks are detecting a transported signal from a region with a larger emission signal and greater seasonality than the western US (compared to the truth). In addition, unlike other US regions, the western US tends to lack constraints from upwind observations (i.e., over the Pacific), which are relatively sparse in both measurement scenarios.

Over smaller regions (i.e. those of Fig. 5), monthly flux estimates deviate more significantly from the truth under both coverage scenarios (Fig. 7). This is expected, since the number and distribution of observations and the information content of the prior ultimately limit the spatiotemporal scale at which independent flux estimates can be reliably obtained. NRC 5000 monthly flux estimates are as good as or better than 2010 coverage estimates over almost all regions. Over regions 1, 4, 7, and 9, the NRC 5000 monthly flux estimates are almost always within 5 % of the true fluxes, whereas over regions 3, 5, 6, and 8 the NRC 5000 estimates sometimes fall outside the 5 % interval, but are always within 10 % of the

truth. Over region 2 (Mountain US), even though the NRC 5000 flux estimates do not follow the truth closely (likely for reasons discussed with respect to the western US above), they are closer to the truth on average than the estimates from 2010 coverage. By contrast, the estimates from 2010 coverage consistently fall within the 5 % error range only over region 9 (South Atlantic US), whereas over several regions (e.g., 3, 6, 7, and 8) its performance is significantly worse than for NRC 5000. The good performance of the 2010 coverage over the Southern Atlantic states, compared to other regions, may be due to the presence of a surface (tower) sampling site at Beech Island, SC (SCT), and aircraft profiles and surface measurements at Cape May, NJ (CMA), which are typically downwind of that region.

Figure 8 shows the accuracy of estimated annual total fossil fuel fluxes over the United States and several subregions. For all the regions, the prior annual emission estimate is outside a 5 % margin around the true emissions (orange rectangles). For the relatively sparse 2010 coverage scenario, the true fluxes are recovered to within 5 % for the US, the eastern US, the central US, and two out of the nine regions of Fig. 5. Under the augmented NRC 5000 coverage scenario, annual total fossil fuel flux estimates are within 5 % of the truth for the conterminous US and all of its subregions except one (Mountain US).

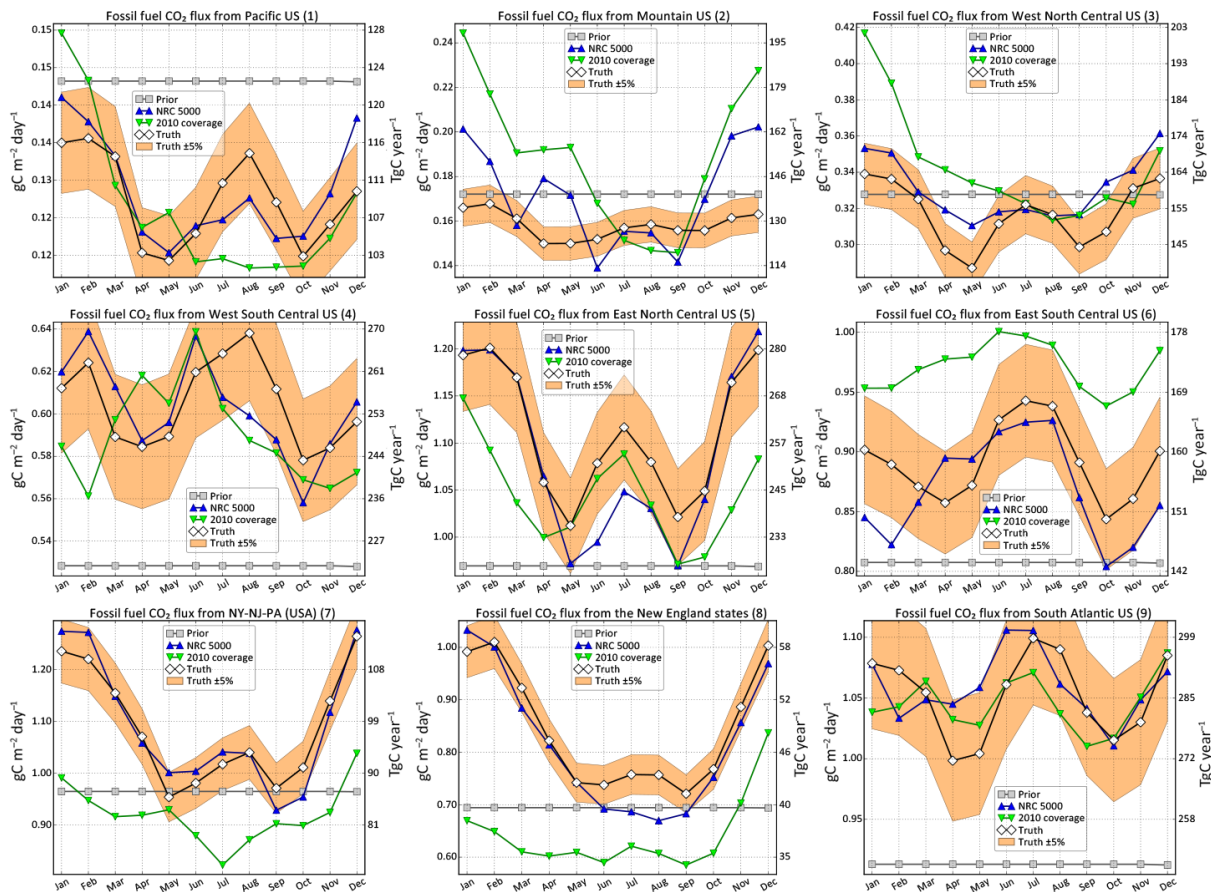


Figure 7. Monthly total emissions estimates for 2010 and NRC 5000 network scenarios, along with prior and true fluxes for individual regions identified in Fig. 5. The orange band depicts the $\pm 5\%$ margin around the true fluxes, and the numbers next to region names refer to the region labels in Fig. 5.

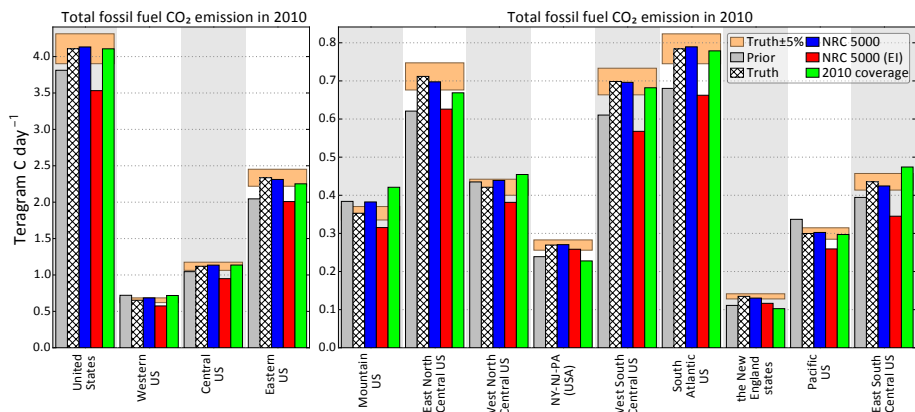


Figure 8. Annual total fossil fuel CO₂ emissions estimates for 2010 and NRC 5000 network scenarios along with true and prior fluxes aggregated for the conterminous US, individual regions and neighboring groups of regions identified in Fig. 5. The orange rectangles denote the $\pm 5\%$ range around the true emission each region.

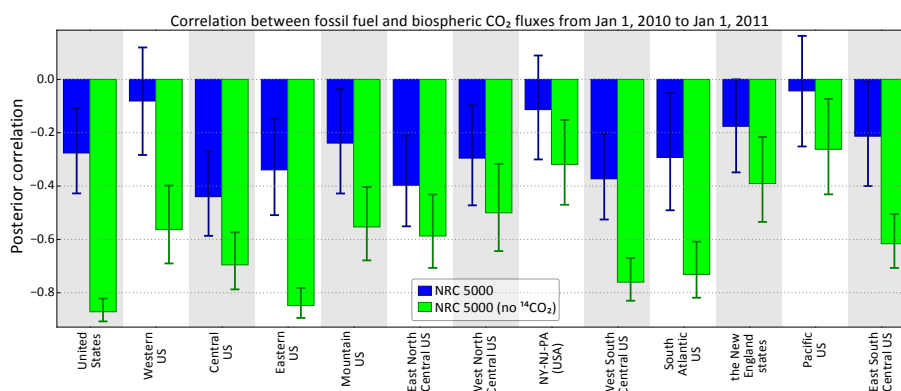


Figure 9. Posterior correlation between fossil fuel and biospheric CO₂ fluxes obtained with and without ¹⁴CO₂ measurements for the NRC 5000 network scenario.

4.1 Correlation between F_{fos} and F_{bio} with and without ¹⁴CO₂ observations

Over large land areas, CO₂ observations constrain only the sum of biospheric and fossil fuel CO₂ fluxes; thus any attempt to separately estimate the two based on CO₂ observations alone should lead to large negative correlations between the two flux types. Any independent information on fossil fuel fluxes from ¹⁴CO₂ observations can be expected to result in a reduction in this negative correlation. To evaluate this, we calculate the posterior correlation between fossil fuel and biospheric fluxes for two scenarios, (a) an inversion using only CO₂ data to estimate both fossil fuel and biospheric CO₂ fluxes, and (b) an inversion using both CO₂ and ¹⁴CO₂ data for the same purpose. The synthetic data sets in both cases are drawn from the NRC 5000 coverage scenario. The method used to calculate the posterior correlation matrix was outlined in Sect. 3.5. If y^{ff} and y^{nat} denote the fossil fuel and natural CO₂ flux aggregates over some spatiotemporal extent (e.g., North America over 2010), then the correlation between fossil fuel and natural fluxes over that extent is

$$r = \frac{\sum_{i=1}^N (y_i^{\text{ff}} - \langle y^{\text{ff}} \rangle) (y_i^{\text{nat}} - \langle y^{\text{nat}} \rangle)}{\sqrt{\sum_{i=1}^N (y_i^{\text{ff}} - \langle y^{\text{ff}} \rangle)^2} \sqrt{\sum_{i=1}^N (y_i^{\text{nat}} - \langle y^{\text{nat}} \rangle)^2}}, \quad (5)$$

where y_i is the estimate of the spatiotemporal flux aggregate from the i th inversion, and $\langle y \rangle$ is the mean y across all N inversions.

Characterizing an error for r is not straightforward since r is bounded within ± 1 and does not have a normal distribution. We therefore estimate a confidence interval of r using a bootstrap method (Efron and Tibshirani, 1994) in which we randomly resample the 100 inversions with replacement and calculate the correlation coefficient from that random drawing. We repeat this 50 000 times to produce a distribution of r . We report the median value of r , and call the range between percentiles 2.5 and 97.5 the error in r (i.e., covering 95 % of the values, analogous to $\pm 2\sigma$ limits for a normal distribution).

The median value of the posterior correlation r and its error range (95 % confidence interval) for the NRC 5000 scenario with and without ¹⁴CO₂ observations is plotted in Fig. 9 for the conterminous US and several subregions. For the inversion with only CO₂ data, we expect the correlation to be strongly negative (i.e., close to -1) over regions for which the total carbon budget is well constrained by the CO₂ observations, and less negative (i.e., closer to 0) over regions with fewer observational constraints. In Fig. 9 this is seen, for example, for the conterminous US (called the United States) due to the strong observational constraint posed by the large number of CO₂ observations (37 884 for the year 2010 in the NRC 5000 coverage). Results for the eastern US also show a strong negative correlation because of the dense coverage in the NRC 5000 network (Fig. 3) for that area compared to the central and western US. The observational constraint on the total CO₂ budget is less stringent, and hence the negative correlation weaker, over smaller regions (such as the NY-NJ-PA tri-state area or the New England states) or for regions for which the upwind influence is less well characterized and the downwind area is not well sampled (such as the Pacific coast and the western US).

Over all regions in Fig. 9 the addition of ¹⁴CO₂ data weakens the negative correlation between fossil fuel and biospheric CO₂ flux, indicating that ¹⁴CO₂ provides information needed to partition CO₂ flux components. Over all the large regions, this reduction is significant; the 95th percentile error bars barely overlap for the central US, and for the eastern, western, and the conterminous US, the error bars are well separated. These represent areas where fossil fuel and biospheric flux estimates can be separated based on CO₂ and ¹⁴CO₂ observations from the NRC 5000 network.

4.2 Carry-over bias in NEE

As discussed in Sect. 1, errors in fossil fuel fluxes specified in traditional CO₂-only inversions (usually with zero prior uncertainty) may be expected to result in spatial and tem-

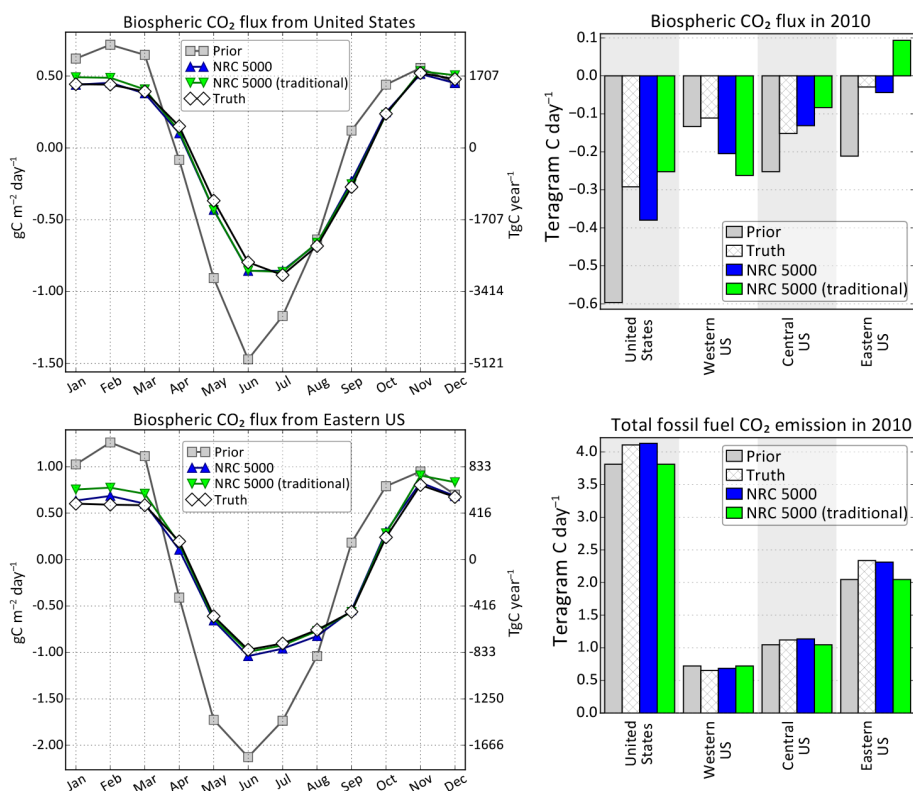


Figure 10. Monthly net biospheric CO₂ flux estimates for the NRC 5000 network scenario with and without ¹⁴CO₂ observations along with prior and true fluxes aggregated for the conterminous and eastern US (left) and annual net biospheric and fossil fuel fluxes for the conterminous US and groups of neighboring regions (right). As discussed in the text, the NRC 5000 (traditional) inversion does not optimize fossil fuel fluxes and does not assimilate ¹⁴CO₂ observations. For both the inversions above, large numbers of CO₂ observations in the NRC 5000 scenario drive the biosphere flux estimates toward true fluxes, while adding ¹⁴CO₂ helps to address carry-over bias arising from erroneous specification of the fossil fuel prior.

poral biases in estimated NEE, which we refer to as carry-over bias. To evaluate the magnitude of potential carry-over bias, and the extent to which it may be reduced by assimilating ¹⁴CO₂ observations, we compare two inversions in which the prior fossil fuel CO₂ flux fields are deliberately biased. The first is the NRC 5000 dual CO₂ + ¹⁴CO₂ inversion already discussed. The second, referred to as NRC 5000 (traditional), is a CO₂-only inversion in which we attempt to estimate both biospheric and oceanic fluxes of CO₂ by assimilating synthetic CO₂ observations from the NRC 5000 network, but not ¹⁴CO₂ observations. For both inversions, the prior fossil fuel flux is from EDGARv4.2 FT2010 and the prior biospheric flux is from SiBCASA, as described in Sect. 3.3. As can be seen in Figs. 6, 7, and 8, both the annual and monthly totals for the prior fossil fuel fluxes differ markedly from the true fossil fuel fluxes for the US and all subregions. Using the entire conterminous US as an example, and assuming stringent total carbon constraint based on the large number of CO₂ observations in the NRC 5000 scenario, we may anticipate monthly carry-over biases as large as 100–200 TgC yr⁻¹ based on differences between true and prior fossil fuel CO₂ fluxes in winter and midsummer (e.g.,

185 TgC yr⁻¹ in January 2010, 133 TgC yr⁻¹ in July 2010, and 176 TgC yr⁻¹ in December 2010).

Estimated biospheric fluxes for the two inversions are given along with true and prior biospheric fluxes as both monthly and annual net totals in Fig. 10 for the conterminous US and several subregions. In all cases, both inversion estimates (those with and without ¹⁴CO₂ observations) migrate away from the specified prior biospheric fluxes and lie close to true biospheric fluxes. This is due to the observational constraints provided by the very large number of synthetic CO₂ measurements and the fact that even the largest potential carry-over bias (e.g., 188 TgC yr⁻¹ in February 2010 for the US) is small relative to either prior or true monthly NEE, which is typically at least an order of magnitude larger. However, we note that for regions that are rich in both CO₂ and ¹⁴CO₂ observations, such as the eastern US, we resolve differences between the cases with and without ¹⁴CO₂ assimilation that are directly comparable to differences in the underlying fossil fuel inventories. For example, the fossil fuel prior in February 2010 over the eastern US is biased low by 154 TgC yr⁻¹, which results in an NEE estimate 153 TgC yr⁻¹ higher than the truth if ¹⁴CO₂

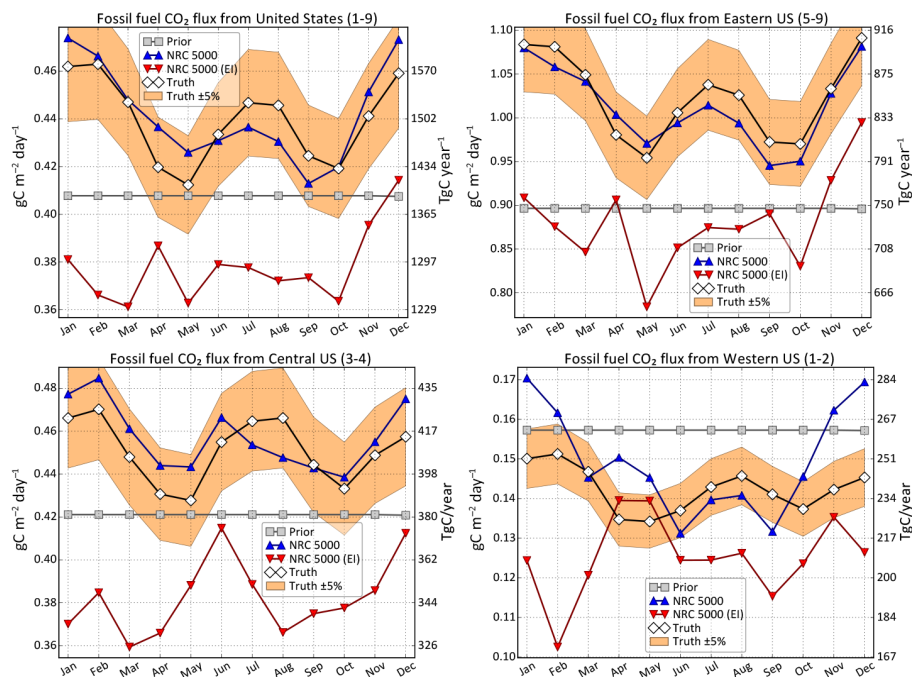


Figure 11. Monthly total fossil fuel CO₂ emission estimates along with prior and true fluxes aggregated for the conterminous US and neighboring groups of regions identified in Figure 5, using perfect (NRC 5000) and intentionally biased transport (NRC 5000 (EI)). As discussed in the text, estimates for biased transport are in this case uniformly low because of systematic differences in the vertical transport between the two model variants.

data are not assimilated, but only 78 TgC yr⁻¹ higher than the truth if ¹⁴CO₂ data are assimilated. Similarly, in December 2010, the fossil fuel prior over the eastern US is biased low by 163 TgC yr⁻¹, resulting in a bias in the estimated NEE of 133 TgC yr⁻¹ without assimilation of ¹⁴CO₂ and only 9 TgC yr⁻¹ with ¹⁴CO₂ observations. These results indicate that carry-over biases that would otherwise go unresolved can in large part be overcome by adding observational constraints from ¹⁴CO₂.

For the three US subregions in Fig. 10 (right panel), the annual NEE estimate with ¹⁴CO₂ is closer to truth than without. However, the reverse is true for annual NEE aggregated over the conterminous US (i.e. the sum of the three subregions). This is due to a cancellation between the western US (where the CO₂-only NEE estimate is too negative) and the other two regions (where the CO₂-only estimate is too positive compared to the truth).

4.3 Imperfect transport OSSE

As mentioned in Sect. 3.4, we performed an inversion with intentionally biased transport. That is, we simulated CO₂ and ¹⁴CO₂ measurements with true fluxes in TM5 EIC, and assimilated those observations using TM5 EI. As noted in Sect. 3.4, forward simulations of an inert tracer sourced largely from the northern continents (SF₆, which is in this respect similar to fossil fuel CO₂) produce substantially dif-

ferent vertical profiles over the conterminous US for the two model versions (Fig. 4), indicating that the two models represent meaningfully different realizations of atmospheric transport.

Figure 11 shows the monthly fossil fuel fluxes estimated over the United States and three of its subregions for both biased transport (NRC 5000 (EI)) and what is effectively perfect transport (NRC 5000). For assimilation of observations using TM5 EI, the monthly flux estimates over the conterminous United States (and over its three large-scale subregions) no longer lie within 5% of the true fluxes. The flux estimates with biased transport are in this case uniformly low, consistent with our understanding of the primary difference between EI and EIC transport schemes involving vertical entrainment and detrainment fluxes over the northern temperate latitudes. As seen for forward simulations in Fig. 4, EIC tends to better ventilate the CBL such that the surface signal is more efficiently transferred to the well mixed free troposphere compared to EI (which allows more signal to build up within the CBL). Thus, TM5 EI requires smaller surface fluxes in order to recover the surface layer signal simulated by TM5 EIC; annual fossil fuel flux estimates from EI transport are thus in all cases lower than the estimates from EIC transport (Fig. 8).

As outlined in Sects. 2.1.1 and 3.4, TM5 EI and TM5 EIC differ significantly in terms of their respective vertical transport schemes, giving rise to large differences in trans-

ported tracer distributions at the global scale and, importantly, over the northern midlatitude continents. TM5 EI is in particular demonstrably biased compared to the ensemble of transport models used in most state-of-the-art global inversions according to several metrics considered by Patra et al. (2011). Thus, while the differences between our fossil fuel CO₂ flux estimates serve as a demonstration of the potential biases that can arise from poor or differing representations of the real transport, they almost certainly exaggerate flux biases likely to be seen amongst models that are well validated against observations. Conversely, our results with effectively perfect transport serve to demonstrate that assimilation of ¹⁴CO₂ along with CO₂ observations has the potential to yield direct, independent top-down observational constraints on fossil fuel emission at subcontinental, regional scales (in our case, corresponding to ~ 250 000 km²) with uncertainties comparable to those estimated for bottom-up inventories. Ongoing improvements in tracer transport models along with rigorous evaluation of transported tracer distributions against a growing network of observations, of the kind we show for SF₆ in Figs. 2 and 4, provide a clear path towards a more complete realization of the full potential of the dual ¹⁴CO₂ and CO₂ assimilation capability described in this work.

5 Conclusions

In this work we develop and present a new dual tracer inversion framework that makes use of the present and anticipated networks of precise atmospheric ¹⁴CO₂ measurements to simultaneously estimate fossil-fuel-derived and biospheric fluxes of CO₂. Using a set of OSSEs, we demonstrate the ability of atmospheric CO₂ and ¹⁴CO₂ measurements to recover previously specified true fossil fuel CO₂ emissions over North America. As expected, the accuracy of the flux estimates depends both on the coverage of the measurement network and the spatiotemporal scale of analysis. We simulated two coverage scenarios, namely the coverage of the NOAA GGRN network in 2010 (969 ¹⁴CO₂ measurements over North America), along with an augmented coverage of ~ 5000 ¹⁴CO₂ measurements over North America (NRC 5000), as recently recommended by the US NAS (Pacala et al., 2010). With the 2010 coverage, we recover true annual total fossil fuel emissions over the conterminous US to better than 1 % and over several highly emissive subregions to within 5 %. For NRC 5000 coverage, we also recover monthly emissions to within 5 % for the United States. For all but one of nine subregions, we also recover the monthly emission to within 5 % for at least 9 months of the year with the NRC 5000 coverage (where, for the subregion which is the exception, emissions are small and upwind observations are sparse). For regions with a strong constraint on the total CO₂ flux based on large numbers of CO₂ observations in the NRC 5000 scenario, the anticipated ¹⁴CO₂

coverage allows for detection of and substantial reduction in biases in regional NEE that would otherwise arise from erroneous specification of the fixed fossil fuel CO₂ emission in a traditional CO₂-only inversion. Additionally, we evaluate biases in fossil fuel CO₂ flux estimates that can arise from poor representation of atmospheric transport and suggest that the growing network of other tracer measurements may be used to select and improve the best transport models. For the best models, our ability to recover fossil fuel emissions over the US should approach that of our idealized OSSEs and be comparable to that for most bottom up fossil fuel emission inventories with estimated annual and monthly regional uncertainties of 5–10 %. It is likely that even an inverse model with less than optimal transport would still be able to resolve trends in fossil fuel emissions precisely. In a future world with anticipated national commitments to reduce CO₂ emissions (e.g. Intended Nationally Determined Contributions, or INDCs, http://unfccc.int/focus/indc_portal/items/8766.php), such a capability could provide for independent top-down verification of such commitments for the US and other areas where atmospheric observing networks are or can be established.

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