

Impact of Prior Terrestrial Carbon Fluxes on Simulations of Atmospheric CO₂ Concentrations

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Key Points:

- Sensitivities of atmospheric CO₂ to the changes in terrestrial fluxes have been quantified using GEOS-Chem and ensemble product from MsTMIP
- Wide range of prior terrestrial flux in model leads to considerable variations in simulated CO₂ concentration at different time scales
- The uncertainties of ensemble optimized terrestrial fluxes may alter simulated CO₂ concentration by ±2.0 ppmv in all seasons

Supporting Information:

Supporting Information may be found in the online version of this article.

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Abstract Terrestrial ecosystems have a significant role in shaping the distribution of atmospheric CO₂, but it is uncertain how much they affect CO₂ concentrations. We assessed the impact of terrestrial fluxes derived from different biosphere models in the Multi-scale Synthesis and Terrestrial Model Intercomparison Project (MsTMIP) on atmospheric CO₂ estimations based on a global chemical transport model (GEOS-Chem) when these fluxes were applied as prior information. We examined the spatiotemporal uncertainty in terrestrial flux estimations from 15 MsTMIP terrestrial fluxes. We found high uncertainties in the terrestrial fluxes for temperate North America, tropical and temperate South America, southern Africa, Europe and tropical Asia. Sensitivity simulations showed that the annual mean CO₂ concentration changed by 6.0–8.0 ppmv with the spread of the terrestrial fluxes. The interannual trend in the terrestrial fluxes could significantly affect the simulated trend of atmospheric CO₂ concentrations. As a result of the spread in the prior terrestrial fluxes, large differences in the daily mean CO₂ anomalies changed with an interquartile range of –1.0 to +1.0 ppmv and the magnitude of change in the sub-daily CO₂ concentrations was in the range 4.0–6.0 ppmv for China, the USA and Europe. Our results suggest an urgent need to increase the reliability of terrestrial flux estimates in CO₂ simulations. Surface seasonal CO₂ concentrations were simulated to change by ±2.0 ppmv in most regions of the world due to the differences in the ensemble mean fluxes, reflecting the impact of the uncertainties in the existing optimized terrestrial fluxes on CO₂ simulations.

Plain Language Summary The variations in atmospheric CO₂ concentration are closely related to the changes in anthropogenic and natural carbon sources and sinks. Since the preindustrial time, the increases in atmospheric CO₂ concentration are mainly caused by fossil fuel burning and land use change. Terrestrial ecosystems play an important role in modulating the atmospheric CO₂ concentrations since they can uptake CO₂ emitted from human activities. Many efforts have been made to quantify the magnitude and distribution of CO₂ uptake by terrestrial ecosystems, but current estimations of terrestrial fluxes still have large uncertainties. It is of great interest to quantify how the uncertainties in terrestrial flux can influence simulated CO₂ concentrations. Our study shows that the simulated atmospheric CO₂ concentrations at different time scales are sensitive to the uncertainties in terrestrial flux. The simulated annual mean CO₂ concentrations differ by 6.0–8.0 ppmv when different terrestrial flux estimations are used. Daily CO₂ anomalies are simulated to change within –1.0 to +1.0 ppmv with the spread of terrestrial fluxes. Hourly CO₂ concentrations could be altered by 4.0–6.0 ppmv due to the differences in terrestrial fluxes. Results from this study have important implications for predicting future atmospheric CO₂ concentrations and for making plans of CO₂ emission control measures.

1. Introduction

Atmospheric carbon dioxide (CO₂) is thought to cause the largest radiative forcing responsible for the current global climate crisis. The annual mean atmospheric CO₂ concentrations at the Mauna Loa Observatory, Hawaii, has increased from about 370 ppm in the year 2000 to about 413 ppm in the year 2020, with a relatively high growth rate every year (www.esrl.noaa.gov/gmd/ccgg/trends/).

Atmospheric CO₂ levels are governed by the global budget of the carbon cycle, which includes two major components: (a) anthropogenic emissions from fossil fuel combustion and changes in land use (mainly land clearance); and (b) carbon fluxes from the natural carbon cycle, such as exchanges between the atmosphere and the terrestrial biosphere or oceans (Friedlingstein et al., 2020). Large variations in predictions of atmospheric CO₂ concentrations are related to biases in these terrestrial carbon sources and sinks (Bauska et al., 2015; Keenan et al., 2016; Schimel et al., 2001, 2015). An improved understanding of the magnitude of, and uncertainties in, current estimates of carbon exchange between the atmosphere and biosphere is crucial in determining the environmental and climate actions required to regulate anthropogenic CO₂ emissions on global and regional scales.

There have been many attempts to estimate the terrestrial carbon budget accurately. Previous studies have suggested that the terrestrial biosphere could take up carbon at a rate of about 1.0–4.0 Pg C yr⁻¹ and offset about 20%–60% of the carbon emitted from fossil fuels (IPCC, 2013). Although it is agreed that the terrestrial ecosystem acts as a key mitigating pathway of current climate change, the global and regional patterns of the terrestrial carbon flux and its estimation are still unclear and unconstrained. Some researchers have produced terrestrial carbon fluxes via process-based dynamic vegetation models or data-driven biosphere models (a bottom-up approach) (Le Quéré et al., 2018; Piao et al., 2009; Sitch et al., 2015; H. Tian et al., 2015). However, there are large spatiotemporal discrepancies in the existing terrestrial carbon inventories. The underlying reasons for such differences are differences in the data for environmental drivers and the physical, chemical and biological processes and parameters used in biospheric models (Huntzinger et al., 2013; Wei et al., 2014)—for example, the complex mechanisms of ecosystem functions, the large heterogeneity of vegetation and soils, and the effects of human and other disturbances (e.g., changes in land use and fires) (S. Liu et al., 2011; Piao et al., 2009; Schimel, 1995; Sitch et al., 2015; Zeng et al., 2005).

Other researchers have estimated carbon budgets through a top-down method—for instance, atmospheric inversions, which infer the posterior CO₂ flux by using the prior flux and an inverse modeling system based on atmospheric transport models with atmospheric CO₂ observations and/or satellite observations (D. F. Baker et al., 2006; Chevallier et al., 2005; Gurney et al., 2002, 2003, 2004; J. Liu et al., 2017; Peters et al., 2007; Peylin et al., 2013; Wang, Feng, et al., 2020). Top-down methods can help reduce the uncertainty in terrestrial fluxes, but the uncertainties in the terrestrial carbon budget from this approach probably arise from the differences in the inversion methods (Crowell et al., 2019; Houweling et al., 2015), atmospheric transport uncertainties (Basu et al., 2018; Houweling et al., 2010; Schuh et al., 2019) and the accuracy and precision of the atmospheric CO₂ observations used. These observations include in situ data, aircraft observations, the Greenhouse Gases Observing Satellite (GOSAT) and the Orbiting Carbon Observatory 2 (OCO-2) XCO₂ retrievals (J. Liu et al., 2017; Thompson et al., 2016; X. Tian et al., 2014; Zhang et al., 2014; H. Wang et al., 2019). Uncertainties also arise from prior flux estimates and their assumed uncertainty (D. F. Baker et al., 2010; Chevallier et al., 2005; Gurney et al., 2003; Philip et al., 2019). There are currently large differences in the terrestrial estimates calculated by the bottom-up biosphere models and the top-down inverse atmospheric models (Wang, Feng, et al., 2020).

Global or regional chemical transport models (CTMs) are often used in forward simulations of atmospheric CO₂ concentrations and the inverse model of terrestrial carbon flux estimates. Atmospheric CO₂ simulations in CTMs are driven and constrained by CO₂ emission inventories and fluxes and are mediated by atmospheric transport (Krol et al., 2005; Nassar et al., 2010). However, the estimates of CO₂ terrestrial fluxes in most CTM models are often treated as known quantities in each grid of the CTM, with spatiotemporal variations at different resolutions. Other sources (e.g., emissions from fossil fuels) are often presented in this way using emission inventories. The uncertainties in the terrestrial CO₂ fluxes are set aside when running the CO₂ simulations. This means that errors in the prior CO₂ sources and sinks may be translated into biases in the atmospheric CO₂ concentrations, as well as errors in estimates of the terrestrial biosphere fluxes in

inversion models (e.g., Gurney et al., 2003; Philip et al., 2019). For instance, Gurney et al. (2003) examined the sensitivity of CO₂ flux inversions to different transport models and the uncertainty in prior fluxes. They showed that the posterior estimates of terrestrial fluxes generated the largest variation because the prior fluxes changed over regions in the northern hemisphere with limited in situ observational data.

Philip et al. (2019) assessed the impact of different prior biospheric fluxes on inverse model estimates of terrestrial CO₂ fluxes using a series of observing system simulation experiments with OCO-2 observations. They found a larger spread among posterior net ecosystem exchange (NEE) estimates in regions and seasons that had limited coverage from satellite observations and large uncertainties in the prior NEE fluxes. The standard deviations of the seasonally averaged posterior NEE estimates for TransCom-3 land regions were about 10%–50% of the multi-model mean NEE when using different prior NEE fluxes.

The spatiotemporal characteristics of atmospheric CO₂ concentrations are a key target on global and regional scales. CTMs provide an opportunity to investigate the changes in CO₂ concentrations in detail and have the advantage of continuity and large-scale operations, which is important in modeling the Earth's climate and environment. Few studies have evaluated the impacts of different biosphere fluxes and associated uncertainties on the variations in forward CO₂ simulations. Biases in terrestrial fluxes could lead to discrepancies of about 5.0 ppm in seasonal concentrations of CO₂ (Chen et al., 2013; Messerschmidt et al., 2013). However, previous studies have focused on the impacts of only two or three terrestrial fluxes on sub-annual CO₂ cycles (monthly or seasonal). Increasing numbers of studies have shown that the variations in CO₂ on finer spatiotemporal scales (e.g., hourly or daily) are important in estimating carbon emissions from cities with high levels of human activity (Duren & Miller, 2012; Gurney et al., 2015). The uncertain influence of terrestrial fluxes on variations in CO₂ simulations on finer timescales (e.g., sub-daily or diurnal) has not been well studied. A thorough exploration of the influence of terrestrial fluxes on the variation in CO₂ simulations on different timescales is essential and this knowledge would provide valuable information to help our understanding of the sources and sinks of CO₂ and the carbon cycle on different spatial scales.

Philip et al. (2019) studied the impact of four different biosphere models and the ensemble mean NEE from the Multi-scale Synthesis and Terrestrial Model Intercomparison Project (MsTMIP) on estimates of the terrestrial NEE flux inferred from the “top-down” GEOS-Chem adjoint model with OCO-2 retrievals. We aim to quantitatively understand the implications for forward simulations of atmospheric CO₂ concentrations resulting from terrestrial fluxes and their uncertainties. This will help to gain a better understanding of the roles of the prior terrestrial carbon flux and its uncertainty on atmospheric CO₂ inversion and assimilation from a different perspective. To achieve this goal, we examined the sensitivity of atmospheric CO₂ concentrations, including annual and sub-annual cycles, to the variations in the terrestrial flux using a global CTM (GEOS-Chem) and an ensemble product of the multi-model ensemble results of terrestrial fluxes (NEE) from MsTMIP. Section 2 describes the model, methods and data and Section 3 presents the results of the sensitivity experiments. Our conclusions and the implications of this study are summarized in Section 4.

2. Materials and Methods

2.1. Model Description

We used the GEOS-Chem version 11-01 global CTM (<http://acmg.seas.harvard.edu/geos/>) driven by the MERRA-2 meteorological fields (Gelaro et al., 2017) to simulate atmospheric CO₂ concentrations. The model grid was run at (2.5° longitude × 2.0° latitude) with 47 reduced vertical layers up to 0.01 hPa. The current CO₂ simulation in GEOS-Chem was developed by Suntharalingam et al. (2004) and updated by Nassar et al. (2010, 2013). Simulation of CO₂ in the model is conducted as a separate tracer simulation with the prescribed CO₂ sources and sinks, including fossil fuel combustion and cement production, biomass burning, biofuel burning, atmosphere–terrestrial ecosystem exchanges, atmosphere–ocean exchanges, and shipping and aviation, in addition to the production of chemicals from the oxidation of carbon monoxide, methane and non-methane volatile organic compounds.

Table 1 describes the specific emission inventories of CO₂ used in this work. In the original GEOS-Chem CO₂ simulation, the NEE of the CO₂ flux for the years 2006–2010 from the Simple Biospheric Model version 3 (Sib3) was applied to estimate the atmosphere–terrestrial biosphere carbon exchange (Messerschmidt et al., 2013). The NEE is the net difference between the gross uptake of CO₂ by plants through

Table 1
Summary of the Simulations Conducted in This Study

Simulation	MERRA2 Meteorology	Terrestrial fluxes	Fossil fuel emission	Biomass burning	Ocean flux	Biofuel burning	Shipping emission	Aviation emission	Chemical Source
S_BGC	2004–2010	BIOME_BGC (2004–2010)	Open source Data Inventory for Atmospheric CO ₂ emissions (ODIAC) with monthly and interannual variation at 1° × 1° resolution (2004–2010) Oda and Maksyutov. (2011)	Global Fire Database (GFED) v4 Emission with daily and interannual variation at 0.25° × 0.25° spatial resolution (2004–2010) van der Werf et al. (2010)	Scaled monthly ocean exchange with interannual variation (2004–2010) Takahashi et al. (2009)	Yevich and Logan annually biofuel burning (fixed in 1995)	International Comprehensive Ocean-Atmosphere Data set (ICOADS) with monthly variation (fixed in 2004) Corbett and Koehler (2003, 2004)	AEIC scaled with global annual CO ₂ emission totals calculated from the IEA (fixed in 2005) Simone et al. (2013); Olsen et al. (2013)	CO ₂ chemical Source with monthly variation (fixed in year 2004) Nassar et al. (2010)
S_CLM		CLM (2004–2010)							
S_CLM4VIC		CLM4VIC (2004–2010)							
S_CTEM		CLASS_CTEM (2004–2010)							
S_DLEM		DLEM (2004–2010)							
S_GTEC		GTEC (2004–2010)							
S_ISAM		ISAM (2004–2010)							
S_LPJ		LPJ (2004–2010)							
S_ORCH		ORCHIDEE (2004–2010)							
S_SIB3		SIB3 (2004–2010)							
S_SIBCASA		SIBCASA (2004–2010)							
S_TEM6		TEM6 (2004–2010)							
S_TRIP		TRIPLEX_GHG (2004–2010)							
S_VEGAS		VEGAS (2004–2010)							
S_VISIT		VISIT (2004–2010)							

Table 1
Continued

Simulation	MERRA2 Meteorology	Terrestrial fluxes	Fossil fuel emission	Biomass burning	Ocean flux	Biofuel burning	Shipping emission	Aviation emission	Chemical Source
S_EOM		Weighted ensemble mean from MsTMIP (2004–2010)							
S_ENM		Un-weighted ensemble mean from MsTMIP (2004–2010)							

Note. All the simulations were run after a spin-up from the initialized state (January 1, 2003) with a globally uniform CO₂ field of 373.71 ppmv based on the marine surface monthly mean CO₂ concentration at Mauna Loa Observatory, Hawaii from NOAA-ESRL.

photosynthesis (gross primary production) and the total ecosystem respiration (the sum of autotrophic and heterotrophic respiration) (Philip et al., 2019). A positive value of the NEE commonly represents a net CO₂ flux from the ecosystem to the atmosphere (referred to as a land carbon source) and a negative value of the NEE represents the net removal of CO₂ from the atmosphere to the ecosystem (referred to as a land sink) (Hayes & Turner, 2012).

2.2. Terrestrial Carbon Flux Data

To investigate the impact of the terrestrial CO₂ flux on CO₂ concentrations, we replaced the terrestrial CO₂ flux in the original model with a suite of global NEE products with three-hourly time steps from terrestrial biosphere models under the MsTMIP project (Huntzinger et al., 2013, 2018) (version 1). These products were obtained from the NASA Carbon Monitoring System (https://daac.ornl.gov/CMS/guides/CMS_CO2_Fluxes_TBMO.html). This data set was created based on the monthly output of terrestrial biosphere model (TBM) data from MsTMIP by Fisher et al. (2016) using a temporal downscaling method. They evaluated these NEE data reproduced by temporal downscaling from the LPJ model by comparison with the observed NEE at Tonzi Ranch AmeriFlux/FIUXNET sites (Baldocchi et al., 2001; Baldocchi & Ma, 2013) and showed that the reproduced NEE agreed well with the observational data.

As described by Fisher et al. (2016), the NEE of 15 TBMs are included in this product: (a) BIOME_BGC (Thornton et al., 2002); (b) CLM (Mao et al., 2012); (c) CLM4VIC (Lei, Huang, et al., 2014); (d) CLASS_CTEM (Huang et al., 2011); (e) DLEM (H. Tian et al., 2012); (f) GTEC (Ricciuto et al., 2011); (g) ISAM (Jain & Yang, 2005); (h) LPJ-wsl (Sitch et al., 2003); (i) ORCHIDEE (Krinner et al., 2005); (j) SIB3 (I. T. Baker et al., 2008); (k) SIBCASA (Schaefer et al., 2008); (l) TEM6 (Hayes et al., 2011); (m) TRIPLEX-GHG (Peng et al., 2002); (n) VE-GAS2.1 (Zeng et al., 2005); and (o) VISIT (Ito, 2010). The multi-model ensemble NEE mean from MsTMIP is also provided in this product. The ensemble NEE means from MsTMIP include two forms: the unweighted ensemble NEE (referred to as ENM) and the weighted ensemble NEE (referred to as EOM). Following the method of Schwalm et al. (2015), the ENM represents the result of combining ensemble models to a single integrated mean value in which each model is weighted equally, whereas the EOM represents the result of combining ensemble models into a single integrated mean value in which each model's weight (the reliability factor) is derived using reliability ensemble averaging. For example, the reliability factors for each model are calculated using seven reference factors related to the gross primary production and vegetation biomass in each grid cell. Each model's weight therefore varies at the grid cell level. Detailed descriptions of the ensemble products and the MsTMIP model output data can be found in Fisher et al. (2016).

2.3. Satellite Observations

The column-averaged dry air mole fraction of CO₂ (X_{CO_2}) retrieved from satellite data offers a way to investigate the spatiotemporal variations in CO₂ concentrations. We used the GOSAT/ACOS X_{CO_2} level 2 products (GOSAT/ACOS_L2_Lite_FP.7.3) (Osterman et al., 2017) for the year 2010 for a comparative analysis of simulated CO₂ concentrations. Validations of GOSAT X_{CO_2} against the Total Carbon Column Observing Network data have shown that the mean bias between ACOS and the Total Carbon Column Observing Network observations is <1.5 ppm (GES DISC, 2017; Wunch et al., 2011). In this product, we use “xco2_quality_flag” as an indicator to filter the soundings based on quality; this applies several quality filters based on auxiliary variables (e.g., aerosols, clouds and pressure differences) that correlate with excessive X_{CO_2} scatter or bias (GES DISC, 2017). “xco2_quality_flag” is simply a byte array of zeros (for good soundings) and ones (for bad soundings) and therefore only good soundings were used in our study. We only considered GOSAT retrievals with solar zenith angles within 10° and 80° and latitude limits within 80°S and 80°N. De-

tails of the products are available at https://docserver.gesdisc.eosdis.nasa.gov/public/project/OCO/ACOS_v7.3_DataUsersGuide-RevF.pdf (last accessed June 28, 2021).

To examine the performance of GEOS-Chem, the X_{CO_2} values derived from model simulations ($X_{\text{CO}_2\text{-mod}}$) were compared with the GOSAT observations ($X_{\text{CO}_2\text{-obs}}$). The simulated CO_2 concentrations were converted into column-averaged values by first calculating the CO_2 profiles that were at the same levels as the GOSAT data profiles and then calculating $X_{\text{CO}_2\text{-mod}}$ at the corresponding times and locations of the satellite measurements following Equation 1 (Connor et al., 2008; Feng et al., 2009).

$$X_{\text{CO}_2\text{-mod}} = X_{\text{CO}_2}^a + h^T A (x - x_a) \quad (1)$$

where x_a and $X_{\text{CO}_2}^a$ are the prior CO_2 profile of GOSAT and the associated prior column-averaged amount, respectively. h is a pressure weighting function, A is the full averaging kernel matrix and x is the model-calculated CO_2 profile.

2.4. In Situ CO_2 Observations

Atmospheric surface CO_2 observations were obtained from a global network maintained by the Carbon Cycle Greenhouse Gases Group of the National Oceanic and Atmospheric Administration Earth System Research Laboratory (NOAA-ESRL), where CO_2 is measured either continuously in situ or by flask air samples. Most of the sites are currently incorporated into the Observation Package data products (GLOBALVIEW CO_2) (Masarie et al., 2014; <https://gml.noaa.gov/ccgg/obspack/>) and are also available from the World Data Center for Greenhouse Gases (<https://gaw.kishou.go.jp/>). These surface CO_2 observations have been widely used to support many previous global and regional CO_2 simulation studies (Chen et al., 2013; Fu et al., 2020; Li et al., 2017; Nassar et al., 2010; Peylin et al., 2013; H. F. Zhang et al., 2014).

Based on a coherent set of NEE data from MsTMIP, the simulations in our study accounted for the impacts of changes in terrestrial carbon flux on atmospheric CO_2 concentrations in the time period 2004–2010. We therefore collected in situ monthly CO_2 flask observations at about 38 global sites from NOAA-ESRL's Global Monitoring Laboratory (https://gml.noaa.gov/aftp/data/trace_gases/co2/flask/surface/), for which measurements are continuously available from 2004 to 2010 (Dlugokencky et al., 2021). Table S1 gives information about the surface sites used in this work.

2.5. Simulations and Methods of Analysis

We performed a series of model experiments (Table 1) to simulate atmospheric CO_2 concentrations using alternative terrestrial CO_2 fluxes from the MsTMIP project with the same meteorological fields and other sources of CO_2 fluxes (e.g., fossil fuel, biomass, and biofuel emissions). The resulting discrepancies in the simulated atmospheric CO_2 concentrations were induced by the differences in the terrestrial CO_2 fluxes alone. Following the approach of Nassar et al. (2010, 2011), all the simulations were initialized on January 1, 2003 with a globally uniform CO_2 field of 373.71 ppmv based on the monthly mean sea surface CO_2 concentration at Mauna Loa Observatory, Hawaii from the NOAA-ESRL. For each experiment, seven-year simulations from 2004 to 2010 were performed for consistency with the time range of the NEE data. For model evaluation, the simulations were conducted with an ensemble mean NEE of the MsTMIP models. We compared the results for eastern China (20° – 40°N , 105° – 122°E), western Europe (35° – 60°N , 10°W – 20°E) and the eastern USA (30° – 45°N , 75° – 90°W).

We used the similarity index (Ω) applied by Koster et al. (2000), which measures the phase and shape similarity among members of an ensemble forecast, to quantify the sensitivity of the atmospheric CO_2 simulations to the multi-model terrestrial flux uncertainties:

$$\Omega_x = \frac{m\sigma_b^2 - \sigma^2}{(m-1)\sigma^2} \quad (2)$$

$$\psi_x = \frac{1}{\Omega_x} \quad (3)$$

$$\gamma = \frac{\psi_{\text{CO}_2}}{\psi_{\text{flx}}} \quad (4)$$

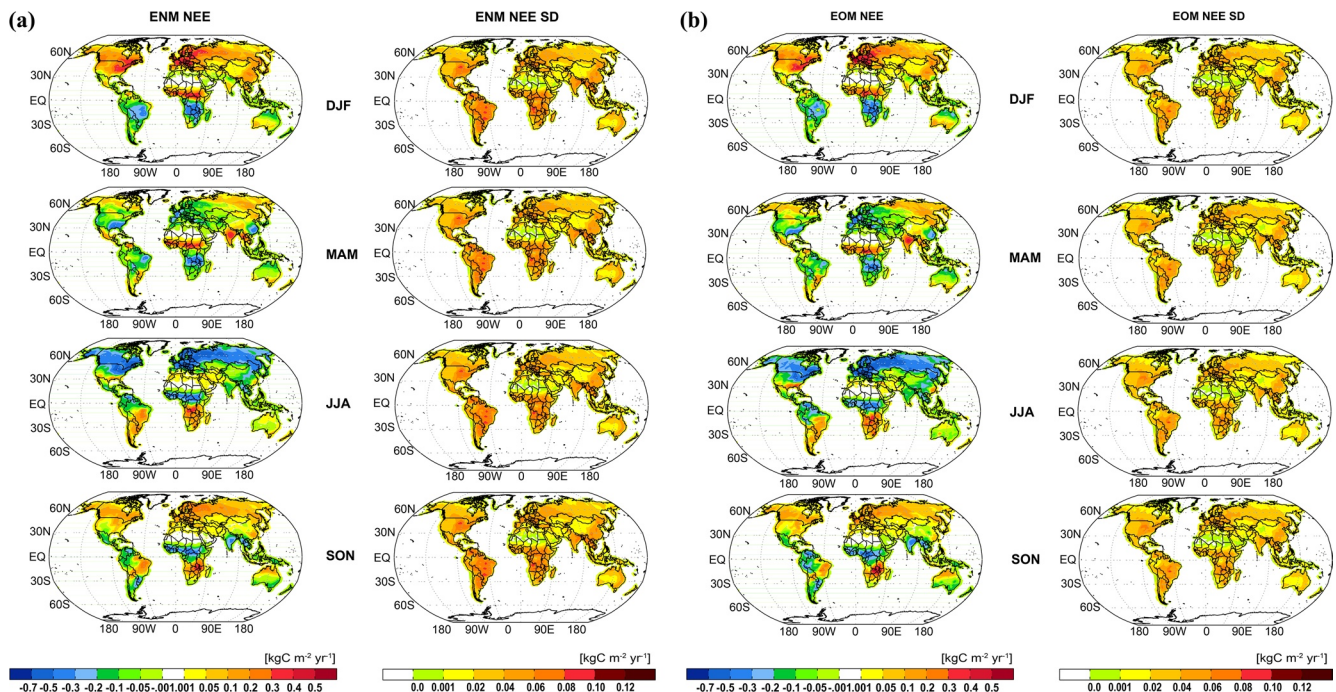


Figure 1. Multi-model seasonal averaged net ecosystem exchange (NEE) ($\text{kg C m}^{-2} \text{ yr}^{-1}$) and the standard deviation of the NEE from (a) the unweighted ensemble mean (ENM) and (b) the weighted ensemble mean (EOM) of MSTMIP over the time period 2004–2010.

where ψ_x represents an uncertainty index derived from the similarity index Ω and γ represents the sensitivity index. σ_b^2 and σ^2 are the temporal variance of the ensemble mean and the full-sample variance, respectively:

$$\sigma_b^2 = \frac{1}{n} \sum_{j=1}^n (b_j - \bar{x})^2 \quad (5)$$

$$\sigma^2 = \frac{1}{mn} \sum_{i=1}^m \sum_{j=1}^n (x_{ij} - \bar{x})^2 \quad (6)$$

where $b_j = \frac{1}{m} \sum_{i=1}^m x_{ij}$, $\bar{x} = \frac{1}{n} \sum_{j=1}^n b_j$, n denotes the time period and m is the number of ensemble members. If each ensemble member produces an identical time series, then σ_b^2 will equal σ^2 and Ω will equal 1. If the time series are completely uncorrelated, then σ_b^2 is expected to be approximately σ^2/m . Therefore the value of Ω will vary from approximately 0 to 1, with values closer to 1 suggesting a greater degree of similarity. This approach was used to quantify the sensitivity of snow simulations to different atmospheric forcing data in previous studies (e.g., Wang, Xie, et al., 2020). In this work, the sensitivity index measures the uncertainty in the ensemble of model simulations for CO_2 concentrations to the uncertainties in the corresponding terrestrial NEE fluxes from MSTMIP. High values indicate high sensitivity and low values indicate low sensitivity.

3. Results and Discussion

3.1. Prior Terrestrial Flux Estimates

Figures 1a and 1b show the distribution of the seasonal mean terrestrial carbon fluxes based on the ensemble results of MSTMIP averaged over the time period 2004–2010. Based on the seasons in the northern hemisphere, large terrestrial CO_2 sinks are found in the tropical areas of South America and South Africa, as well as the eastern USA, eastern China and Europe in summer and spring. Small terrestrial CO_2 sources are found in the boreal and temperate climate regions, such as Boreal North America, Boreal Eurasia, and Western Australia in winter and autumn. Such seasonal cycles of the terrestrial flux are related to the seasonality of photosynthesis and respiration of vegetation. Figures 1a and 1b also show the standard deviation across

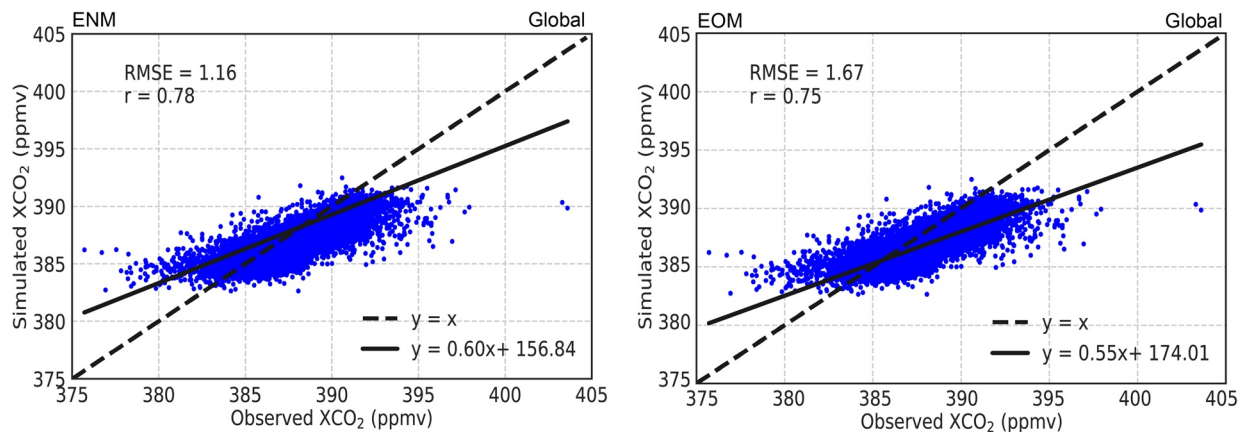


Figure 2. Scatter plots of the simulated $X_{\text{CO}_2_mod}$ based on simulations with the ensemble mean net ecosystem exchange and observed $X_{\text{CO}_2_sat}$ for the year 2010. The root-mean-square error and correlation coefficient between the simulation and observations are also shown.

the 15 prior model fluxes, indicating a large uncertainty in both the magnitude and sign of terrestrial carbon fluxes during all seasons. This is typically highest in certain regions (e.g., South America at mid- and low latitudes, North America at mid- and high latitudes, Europe and eastern China). The range of uncertainties in terrestrial fluxes is locally up to $0.12 \text{ kg C m}^{-2} \text{ yr}^{-1}$.

3.2. Comparison Between Simulated X_{CO_2} and GOSAT X_{CO_2}

The simulated atmospheric CO_2 concentrations based on the GEOS-Chem model have been widely used in previous studies (Cogan et al., 2012; Feng et al., 2011; Fu et al., 2020; Lei, Guan, et al., 2014). Large variations in the total terrestrial flux are found among the 15 models in M_sTMIP. Here, the ensemble means (ENM and EOM) of the terrestrial fluxes from M_sTMIP are taken as “reasonable” NEE fluxes because the ensemble fluxes are close to the optimized fluxes from CarbonTracker (e.g., CT2016; see supplementary materials), which is a widely used inverse model of atmospheric CO_2 (Peters et al., 2007, with updates documented at <http://carbontracker.noaa.gov>). The resulting CO_2 concentrations from simulations S_ENM and S_EOM were used for model evaluation and comparison with the GOSAT X_{CO_2} , respectively.

Figure 2 shows the correlations between the simulated and observed X_{CO_2} for the year 2010. Globally, there was general agreement between the simulated $X_{\text{CO}_2_mod}$ and GOSAT $X_{\text{CO}_2_obs}$ with correlation coefficients of 0.78 ($P < 0.05$) and 0.75 ($P < 0.05$) for simulations S_ENM and S_EOM, respectively, suggesting that the GEOS-Chem model can reasonably capture the spatiotemporal variations in the observed X_{CO_2} . The overall correlation between X_{CO_2} in the S_ENM and GOSAT simulations is a little higher than those between X_{CO_2} in the S_EOM and GOSAT simulations, indicating the uncertainty in “optimal” integrations of TBMs that depend on different ensemble methods and the uncertainty associated with other carbon sources and sinks in the model (e.g., fossil fuel emissions, biomass emissions, and ocean fluxes).

The distribution of the biases between the simulations and observations shows large spatial discrepancies based on simulations S_ENM and S_EOM, especially on a regional scale (Figure 3). For example, the simulated values of X_{CO_2} from S_ENM are generally higher than those from GOSAT over China, Europe and the USA, whereas the simulated values of X_{CO_2} from S_EOM are broadly underestimated over the same regions. This is because the estimates of the land biosphere sink over these regions are substantially larger (i.e., more uptake) in the EOM simulation than the estimates of ENM.

The ensemble NEE mean from M_sTMIP shows higher annual global sinks of $-4.37 \text{ Pg C yr}^{-1}$ in the EOM simulation than in the ENM simulation ($-3.89 \text{ Pg C yr}^{-1}$) in the year 2010. The different model performances with ENM or EOM fluxes in the comparison with the GOSAT X_{CO_2} indicated that the uncertainty in the multi-model ensemble mean fluxes is dependent on the region. Overall, the difference between the GEOS-Chem and GOSAT values of X_{CO_2} varied from -4.0 to $+4.0$ ppmv in most places in the world. For seasonal concentrations, the mean point-by-point biases varied from -0.08 to -2.3 ppmv, with large

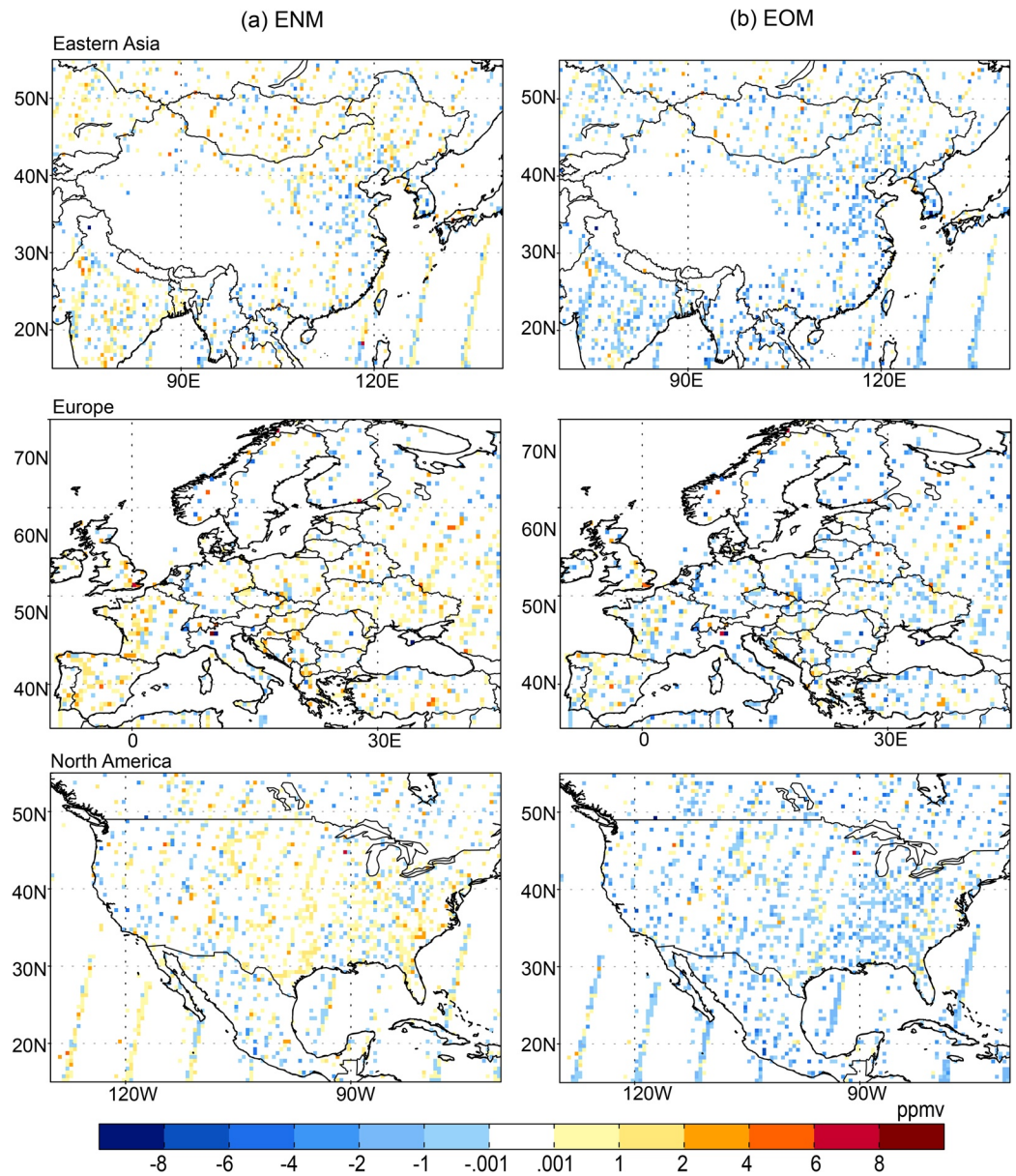


Figure 3. Comparisons of the biases ($X_{\text{CO}_2_mod}$ minus $X_{\text{CO}_2_sat}$) in Eastern Asia, Europe, and North America for the year 2010. Simulated $X_{\text{CO}_2_mod}$ from (a) the S_ENM simulations and (b) the S_EOM simulations.

biases occurring in December–January–February and March–April–May and small biases in June–July–August and September–October–November for both simulations S_ENM and S_EOM (Table 2). This suggests that the GEOS-Chem CO_2 simulation performs better in summer than in other seasons. This finding is consistent with the results from previous studies (Lei, Guan, et al., 2014; Li et al., 2017; H. Zhang et al., 2015).

Such discrepancies in the comparison of the CTM and GOSAT X_{CO_2} at the seasonal scale is probably due to the uncertainties in the terrestrial flux estimations and other emission inventories in different seasons. Huntzinger et al. (2013) showed that the uncertainty in terrestrial flux estimations is dependent on season by comparing different regions. In addition, GOSAT may capture the spatial heterogeneity and temporal variations in CO_2 caused by changes in carbon sources and sinks better than the CTM model, which is constrained by emissions inventories (Li et al., 2017). The GOSAT measurements have a constant sensitivity to

Table 2

Statistical Characteristics of the Simulated and Observed CO₂ Column-Averaged Dry Air Mole Fractions (X_{CO_2} ; Units: ppmv) Averaged Globally for the Year 2010

		DJF	MAM	JJA	SON
Average	A_{obs}	389.86	389.31	387.95	388.20
	$A_{\text{S_EOM}}$	387.60	387.07	386.86	387.04
	$A_{\text{S_ENM}}$	388.90	388.30	387.95	388.12
Standard deviation	σ_{obs}	3.01	2.04	2.59	1.82
	$\sigma_{\text{S_EOM}}$	2.12	1.46	1.01	0.70
	$\sigma_{\text{S_ENM}}$	2.23	1.47	1.20	0.79
Bias (Sim-Obs)	$b_{\text{S_EOM}}$	-2.26	-2.24	-1.09	-1.17
	$b_{\text{S_ENM}}$	-0.96	-1.01	0.00	-0.08
Root mean square error	$\text{RMSE}_{\text{S_EOM}}$	2.90	2.81	2.31	2.03
	$\text{RMSE}_{\text{S_ENM}}$	2.03	1.97	1.87	1.61
Correlation coefficient	$R_{\text{S_EOM}}$	0.80	0.57	0.68	0.41
	$R_{\text{S_ENM}}$	0.80	0.58	0.75	0.47

the entire atmospheric column and show the maximum sensitivity to changes in CO₂ concentrations near the surface (H. Zhang et al., 2015).

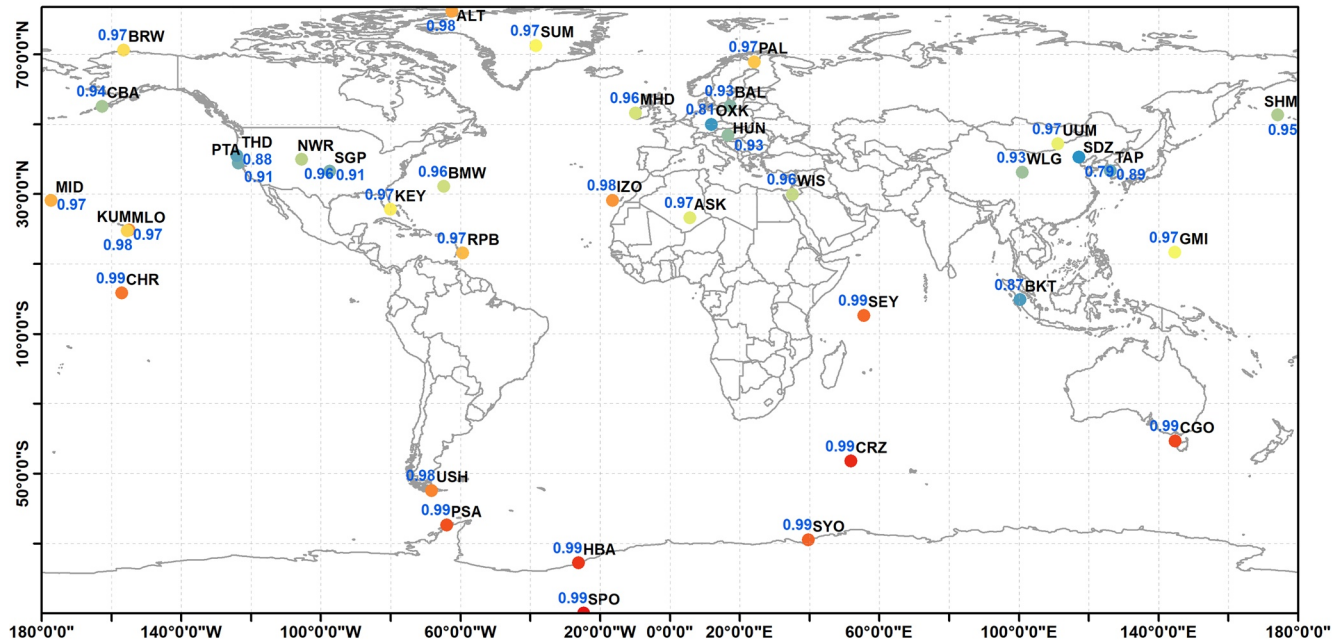
Some studies have indicated that the exchange of CO₂ between the upper troposphere and lower stratosphere at high latitudes and in the mid- and upper troposphere at subtropical and mid-latitudes affect CO₂ concentrations in December–January–February and March–April–May, but the model is prone to underestimate CO₂ concentrations in the upper troposphere of the tropics and subtropics and overestimate CO₂ concentrations in the lower stratosphere of the extratropical regions because the model does not correctly simulate the impacts of stratospheric intrusion (Miyazaki et al., 2008; Deng et al., 2015).

3.3. Comparison Between Simulated CO₂ and In Situ CO₂ Observations

To evaluate the ability of GEOS-Chem to simulate the seasonal and interannual variations in surface CO₂ concentrations, we compared the monthly simulated surface CO₂ concentrations from the simulations for different terrestrial fluxes with surface measurements at 38 sites around the world in the time period 2004–2010. The simulations with the ENM and EOM fluxes performed best when compared with observations at all sites. The correlations between the S_ENM simulations and the observations were very close to the correlations between the S_EOM simulations and observations at all 38 sites, ranging from 0.8 (SDZ) to almost 1.0 (e.g., CGO, CRZ, HBA, and SPO) (Figures 4 and S2). However, the absolute value of the mean bias between the observations and the S_ENM simulations averaged over 2004–2010 are generally smaller than those between the observations and S_EOM simulations at 80% of the sites (in which 60% of the mean bias was <1.0 ppm), suggesting that the GEO-Chem simulation may perform best when using the ENM flux. The average simulated CO₂ concentrations from both the S_ENM and S_EOM models at all sites were 0.44 ppm and 1.2 ppmv lower than the observations, respectively, whereas the simulated CO₂ concentrations were overestimated at some sites (e.g., BKT, HUN, and WIS). Among all sites, large biases were found at sites BKT, PTA, OXK, and THD, which were within the range -4.0 to +4.0 ppm based on the S_ENM and S_EOM simulations. Overall, GEOS-Chem can capture the seasonal and interannual variations in surface CO₂ well at large scales.

When we compared the results from other sensitivity simulations using different terrestrial fluxes with the observations, we found that the performance of models varied greatly with different prior terrestrial fluxes. Although the simulations with most terrestrial fluxes of the MsTMIP model can capture the overall seasonal and interannual variations in surface CO₂, the mean biases between the observations and simulations showed a large discrepancy among all the simulations, ranging from -7.6 to +9.5 ppm. The simulated CO₂ concentrations from GEOS-Chem with the TRIPLEX-GHG NEE flux showed large inconsistencies with the

(a) correlation coefficient



(b) mean bias

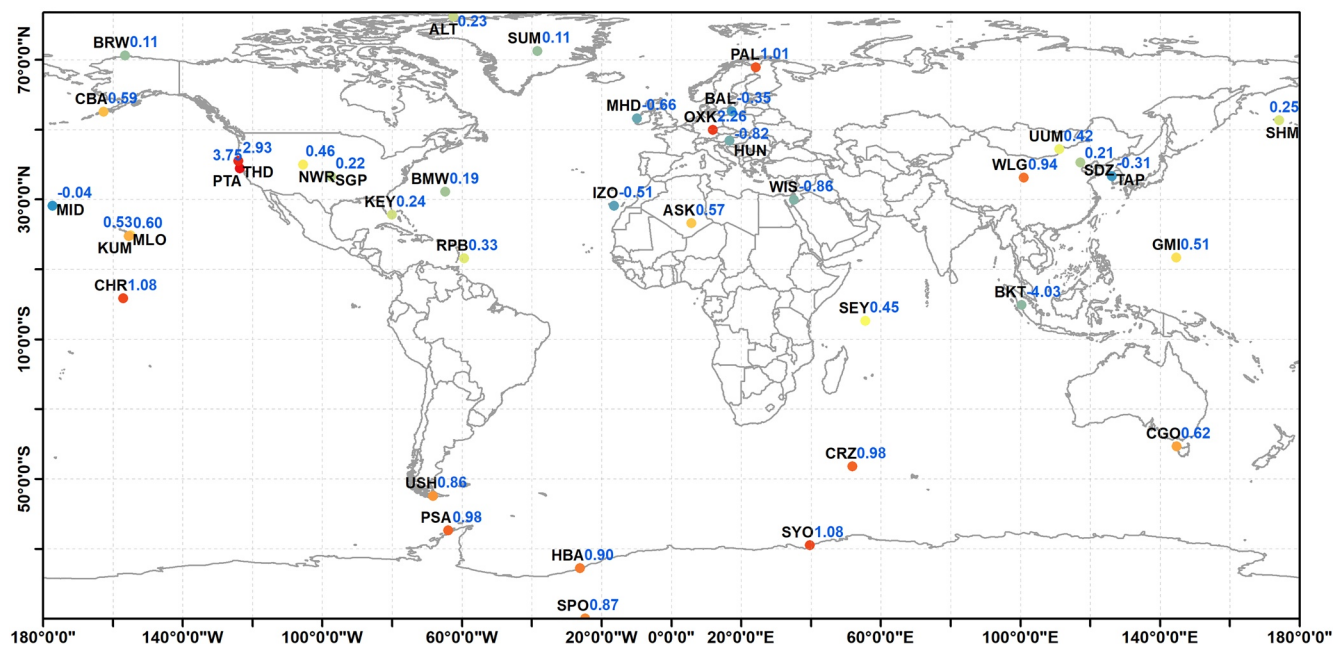


Figure 4. (a) Correlation coefficient between the simulated surface CO₂ concentrations from S₂ENM and the observed CO₂ concentrations at the in situ sites. (b) Mean bias between the observed surface CO₂ concentrations and the simulated surface CO₂ concentrations from S₂ENM (units: ppmv).

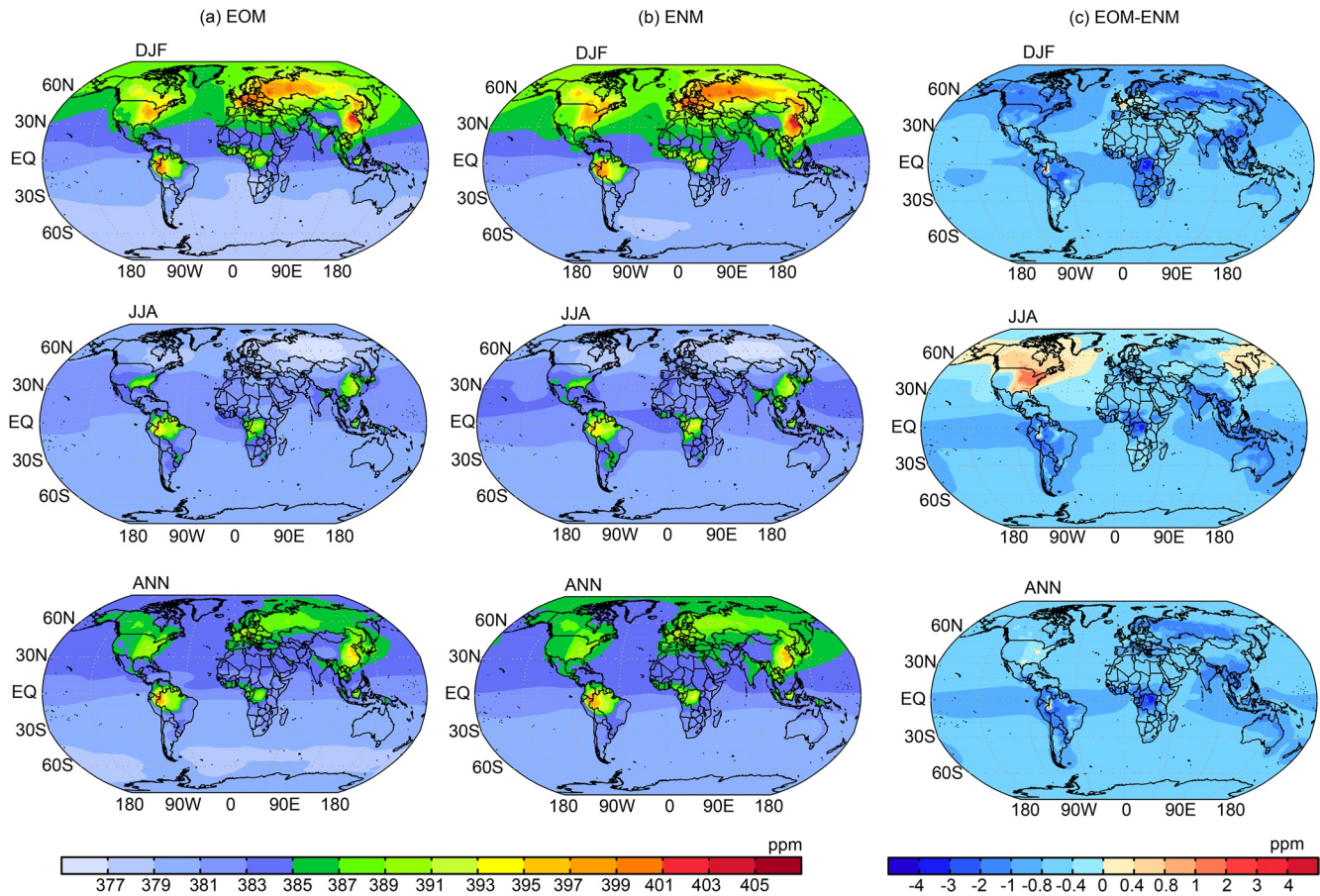


Figure 5. Distribution of the winter (DJF), summer (JJA) and annual mean (ANN) surface layer CO₂ concentrations in the time period 2004–2010 from the (a) S_EOM and (b) S_ENM simulations. (c) Differences in surface CO₂ concentrations between the S_EOM and S_ENM simulations.

observations, in contrast with the observed increase in CO₂. This is because the terrestrial carbon sinks are estimated to increase in TRIPLEX-GHG. The bias between the observed and simulated atmospheric CO₂ from S_TRIP is therefore excluded in the ranges given here.

3.4. Modeling the Response of Atmospheric CO₂ to Changes in Terrestrial Fluxes

Although the simulated CO₂ concentrations derived from the ensemble fluxes generally converged with the observations, there were some differences between the ENM and EOM fluxes in terms of magnitude and distribution (Figure S1). In the time period 2004–2010, the annual mean terrestrial fluxes for the ENM and EOM simulations were -3.92 and -4.31 Pg C yr⁻¹, respectively. Using model sensitivity simulations, we found that the differences between the EOM and ENM NEE could lead to changes in the annual and seasonal mean CO₂ concentrations by -2.0 to $+2.0$ ppmv globally (Figure 5). This suggests that there is a range in the bias for atmospheric CO₂ among simulations using different ensemble mean fluxes obtained from different methods. Large differences in the CO₂ concentrations between the S_EOM and S_ENM simulations were found in the southeastern USA and eastern Asia, where the highest CO₂ concentrations were seen, indicating that the NEE estimates in these regions have a large uncertainty. More information can be found in the supplementary materials, which compare the fluxes in the TransCom regions from the ensemble mean of MsTMIP with the optimized flux from CarbonTracker (Table S2). Table S3 gives the discrepancies in the regional CO₂ concentrations induced by the differences in the ensemble product.

The large diversity in the annual mean NEE was predicted by the biosphere models in MsTMIP. The estimates of the global annual NEE were within the range -8.6 to -0.33 Pg C yr⁻¹ averaged over 2004–2010, except for models TRIPLEX-GHG and SIB3. The magnitude of the CO₂ sinks (-19.2 Pg C yr⁻¹) from TRI-

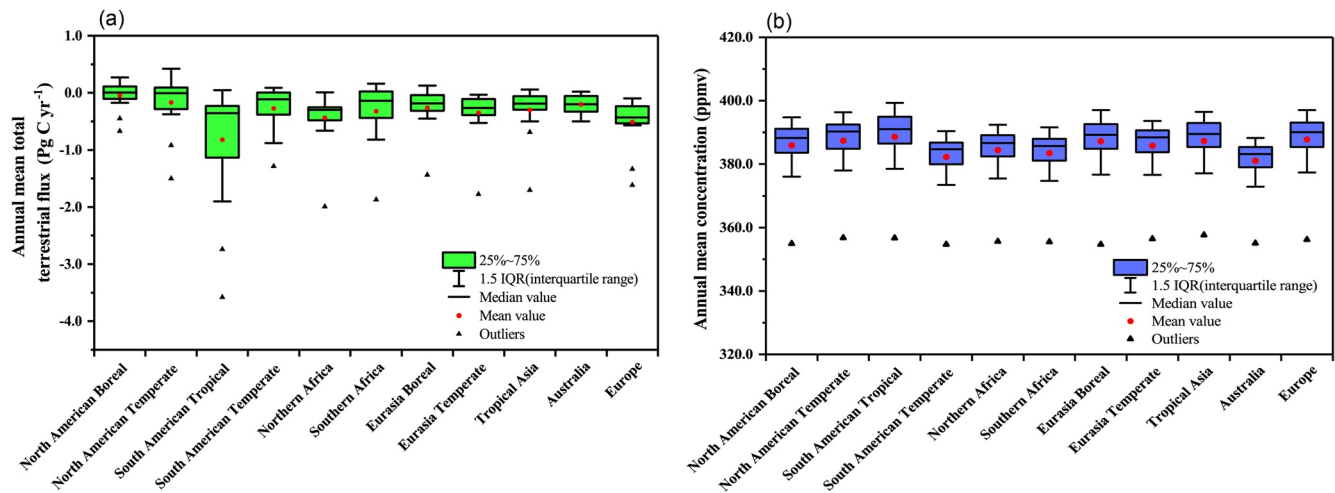


Figure 6. (a) Boxplot of the annual terrestrial fluxes over 11 land regions based on the net ecosystem exchange results of 15 biosphere models from MsTMIP. (b) Boxplot of the annual mean surface CO₂ concentrations averaged over 11 land regions from the sensitivity simulations with different terrestrial fluxes of MsTMIP. The boxes enclose the 25th, 50th, and 75th percentiles; the red dots represent the average values and the triangles represent the outliers.

PLEX was more than twice that of the CO₂ sinks from other models, whereas the estimate of the CO₂ flux from vegetation (+0.38 Pg C yr⁻¹) might be a CO₂ source based on the SIB3 model. The deviation from the mean terrestrial flux based on MsTMIP ranged from -6% to 380% (Figure S3) and these discrepancies in the terrestrial fluxes among different models were clear at the scale of terrestrial ecosystems. A comprehensive assessment of the differences between the current in situ CO₂ flux measurements in different ecosystems and estimates of the terrestrial flux based on multi-model ensembles are needed to give a clearer picture of the model performance and uncertainties in estimating terrestrial fluxes over large spatial and temporal scales. This is still a challenge for many countries (Jones et al., 2016; Piao et al., 2013; Sitch et al., 2015).

Figure 6 shows the spread in the annual mean CO₂ concentrations simulated using different prior terrestrial fluxes. The simulated annual mean surface CO₂ concentrations in different terrestrial regions could be altered by 6.0–8.0 ppmv (25%–75% interquartile range) as a result of deviations in the terrestrial fluxes. In addition, large differences were found among the simulated interannual trends of surface CO₂ concentrations when using different terrestrial fluxes as prior estimates (Figure 7). The simulated interannual variations in surface CO₂ concentrations showed an overall increasing trend driven by most of the predicted terrestrial fluxes, but the interannual variations showed different behaviors, with a decreasing trend in two simulations of MsTMIP fluxes (CTEM and TRIPLEX-GHG). This decreasing trend was attributable to the large increase in the terrestrial uptake of CO₂ in these biosphere models.

We examined the interannual trends of five representative background sites using linear regression—including Mauna Loa and the Southern Great Plains in the USA, Waliguan in China, Pallas-Sammaltunturi in Finland and Cape Grim, Tasmania in Australia—based on the continuous measurements from NOAA-ERSL. The simulated interannual variations in surface CO₂ from the S_EOM simulation agree well with the observed trend over 2004–2010 at Mauna Loa, Waliguan, Pallas-Sammaltunturi and Cape Grim (about 1.8–1.9 ppm yr⁻¹), but tends to underestimate the interannual trend in the Southern Great Plains (2.64 ppm yr⁻¹). The simulated interannual trends from the S_ENM model (about 2.0–2.1 ppm yr⁻¹) are a little higher than the observations at these five sites.

Figure 7 shows that the trends in the simulated global mean surface CO₂ concentration varied from -5.5 to 4.0 ppmv yr⁻¹ during the time period 2004–2010 driven by changes in the terrestrial flux, which are consistent with the findings at in situ sites. This finding shows that an accurate prior terrestrial flux is crucial for the correct simulation of the seasonal and interannual cycles of atmospheric CO₂, indicating the upper limits of the effects of uncertainties in terrestrial fluxes on the interannual variations in atmospheric CO₂ concentrations. Evaluation of the spatiotemporal variation in CO₂ concentrations using both in situ measurements and satellite observations might also provide some limited insights into the quality of the GEOS-Chem CO₂ simulation using different prior terrestrial fluxes. For instance, the choice of ensemble

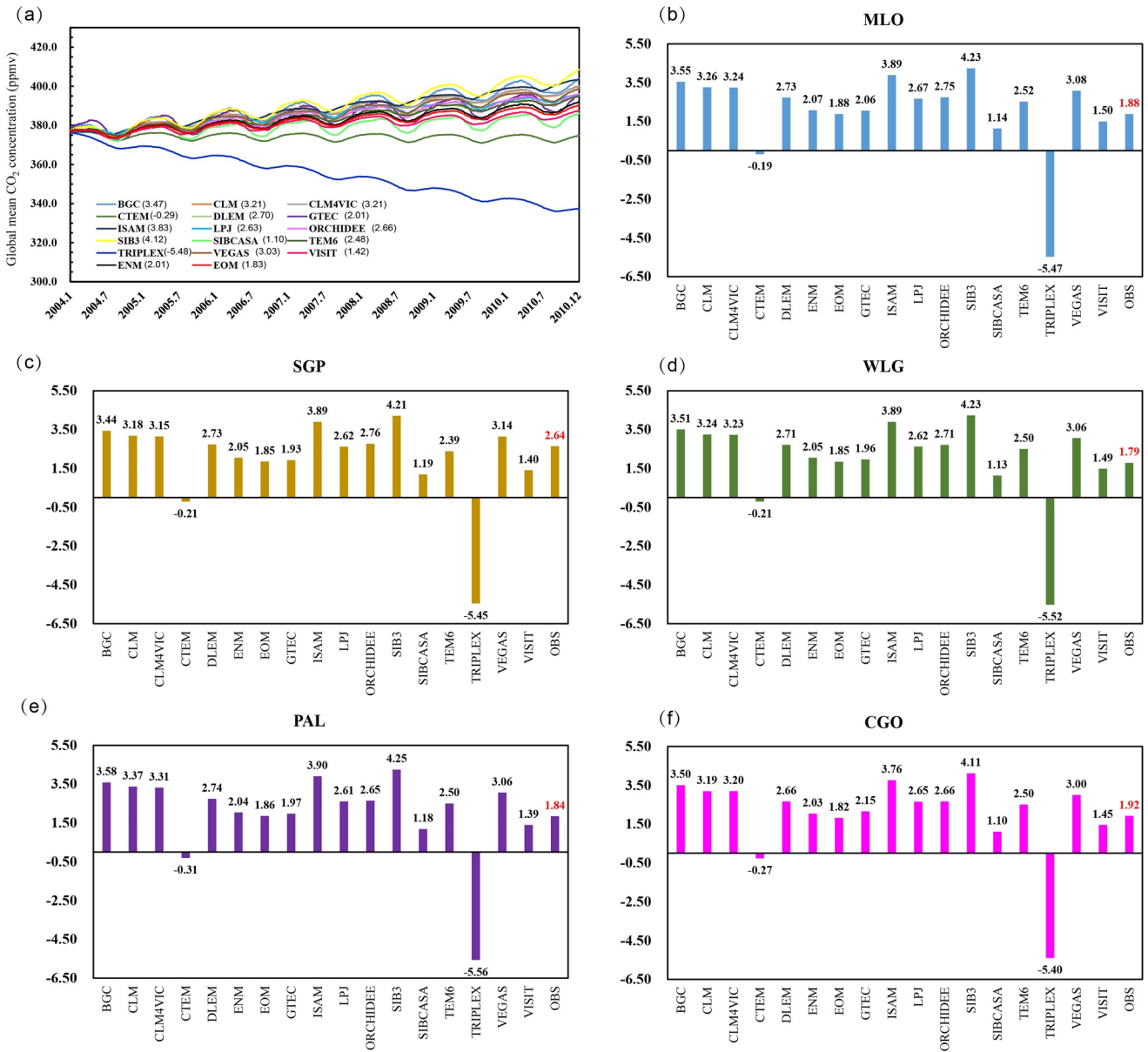


Figure 7. (a) Interannual variations in the global mean surface CO₂ concentrations (ppmv) derived from the sensitivity simulations with ensemble terrestrial fluxes product from MstMIP. The values in parentheses represent the simulated linear trends of surface CO₂ during the time period 2004–2010 (units: ppmv yr⁻¹). (b–f) The observed interannual trends (red numbers) and simulated interannual trends from the simulations with individual terrestrial fluxes from MstMIP at Mauna Loa (MLO), Southern Great Plains (SGP), Waliguan (WLG), Pallas-Sammaltunturi (PAL) and Cape Grim (CGO) sites (units: ppmv yr⁻¹).

mean products in MstMIP is made before the choice of the individual model flux when carrying out CO₂ simulations and the NEE fluxes from the TRIPLEX-GHG, CTEM, and SIB3 models are not recommended as the prior terrestrial flux for GEOS-Chem CO₂ simulations if there are no further constraints and corrections in these models.

To estimate the uncertainties in the simulated CO₂ concentrations among the 15 simulations, we calculated the similarity index of the simulated CO₂ concentrations, the similarity index of the corresponding terrestrial NEE fluxes of the MstMIP ensemble and the sensitivity index of the uncertainty in the ensemble of model simulations for CO₂ concentrations to the uncertainties in the corresponding terrestrial NEE fluxes (Figure 8). Low values of the similarity index of the simulated CO₂ concentrations/terrestrial fluxes showed

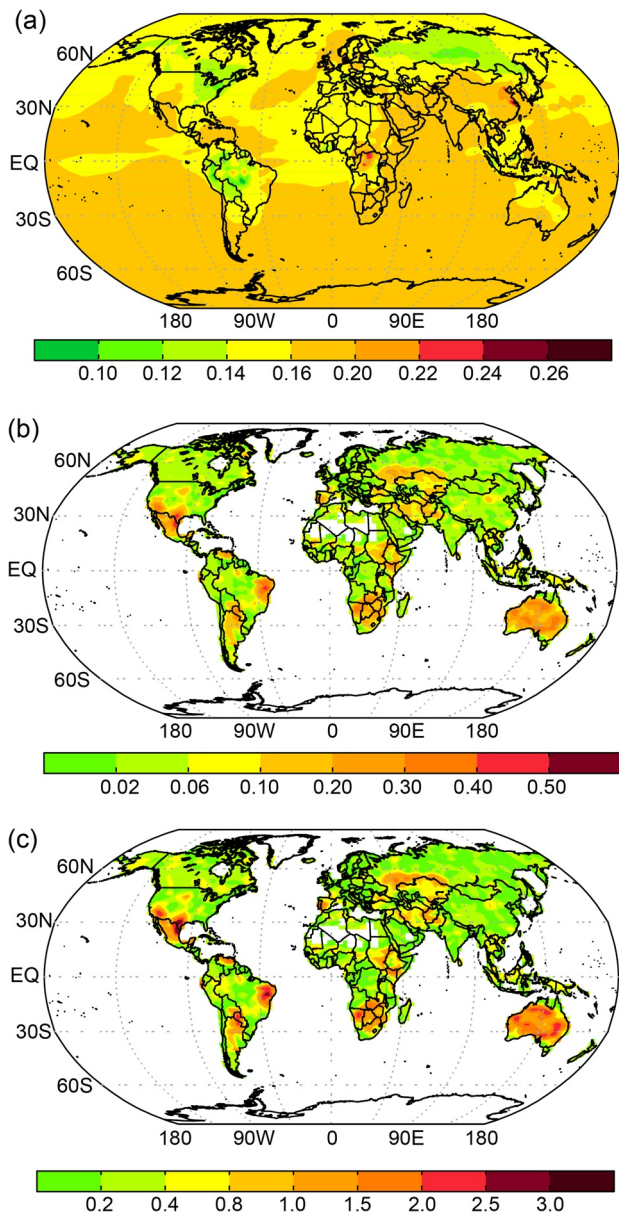


Figure 8. (a) Similarity index of the annual mean surface CO₂ concentration. (b) Similarity index of terrestrial biospheric flux (net ecosystem exchange) for the time period 2004–2010. (c) Sensitivity index of the annual mean CO₂ concentration to the uncertainties in the terrestrial biospheric flux.

large differences among the simulated CO₂ concentration/terrestrial flux time series from the 15 simulations.

Figure 8 shows that the sensitivity of the simulated CO₂ concentrations to the uncertainties in the terrestrial flux varied in different regions. The simulated CO₂ concentrations were highly sensitive to the uncertainties in the terrestrial flux in the steppe zones or in desert–grassland biome transitional regions where vegetation is relatively sparse, although there was a relatively high similarity of the terrestrial flux among multi-models over these regions. The values of the sensitivity index in eastern China, the eastern USA and many parts of Europe were relatively low compared with the biome transition regions, indicating that the uncertainties in the terrestrial flux had a relatively small effect on the simulated CO₂ concentrations. Despite this, there was a relatively strong sensitivity in parts of southeastern China and the eastern USA, where there are major areas broadleaf vegetation, suggesting the important effects of terrestrial flux uncertainties on the simulation of CO₂ at high CO₂ concentrations.

To quantify the response of the daily mean CO₂ concentrations to variations in terrestrial fluxes, we removed the long-term trends and monthly variations of CO₂ concentrations during the study period. Figure S4 shows the detrended and deseasonalized daily mean CO₂ concentration averaged over three study regions during the time period 2004–2010 from simulations S_ENM and S_EOM. The variations in the daily mean CO₂ concentration from simulation S_ENM were mostly consistent with those from simulation S_EOM, with changes averaged over China, the USA and Europe in the range -2.5 to $+3.0$ ppmv. Such variations in the daily mean CO₂ concentration are caused by changes in the CO₂ sources and sinks on a daily scale, which are ultimately affected by atmospheric transport. Among the three regions, the fluctuation in the daily mean CO₂ concentrations was a little larger in the USA than in China and Europe.

We calculated the sub-annual anomalies in the daily CO₂ concentrations relative to the mean fluctuation in CO₂ derived from the S_ENM simulation (Figure 9). Large differences in the daily CO₂ anomalies are expected as a result of the spread in the prior terrestrial fluxes. The interquartile range of the daily CO₂ anomalies was within the range -1.0 to $+1.0$ ppmv and the median value was about ± 0.5 ppmv for China, the USA and Europe. These anomalies induced by the spread of the terrestrial fluxes were significant when compared with the observed natural atmospheric CO₂ variabilities (about 1–3 ppmv) driven by climatic variability (e.g., the El Niño Southern Oscillation) (Zeng et al., 2020), reflecting the inescapable contribution of the uncertainties in the terrestrial flux to current CO₂ simulations.

The impacts of the uncertainties in the terrestrial flux can be further understood through an examination of the simulated CO₂ concentrations in the diurnal cycle (Figure 10). Figure 10 shows that the variations in the hourly CO₂ concentrations have distinct characteristics in different regions, mostly influenced by the diurnal variation in CO₂ emissions and the biosphere flux. Except for a few outliers, the magnitude of change in the sub-daily CO₂ concentrations was within the range 4–6 ppmv (standard deviation) for China, the USA and Europe. Our results quantitatively demonstrate the significant impacts of accurate estimates of the terrestrial flux on annual and sub-annual modeling of CO₂ variations. One implication of this work is the urgent need to enhance the reliability of the prior terrestrial flux in the CTMs, which are the basis of atmospheric CO₂ forward simulations and inverse modeling of CO₂ fluxes (Philip et al., 2019).

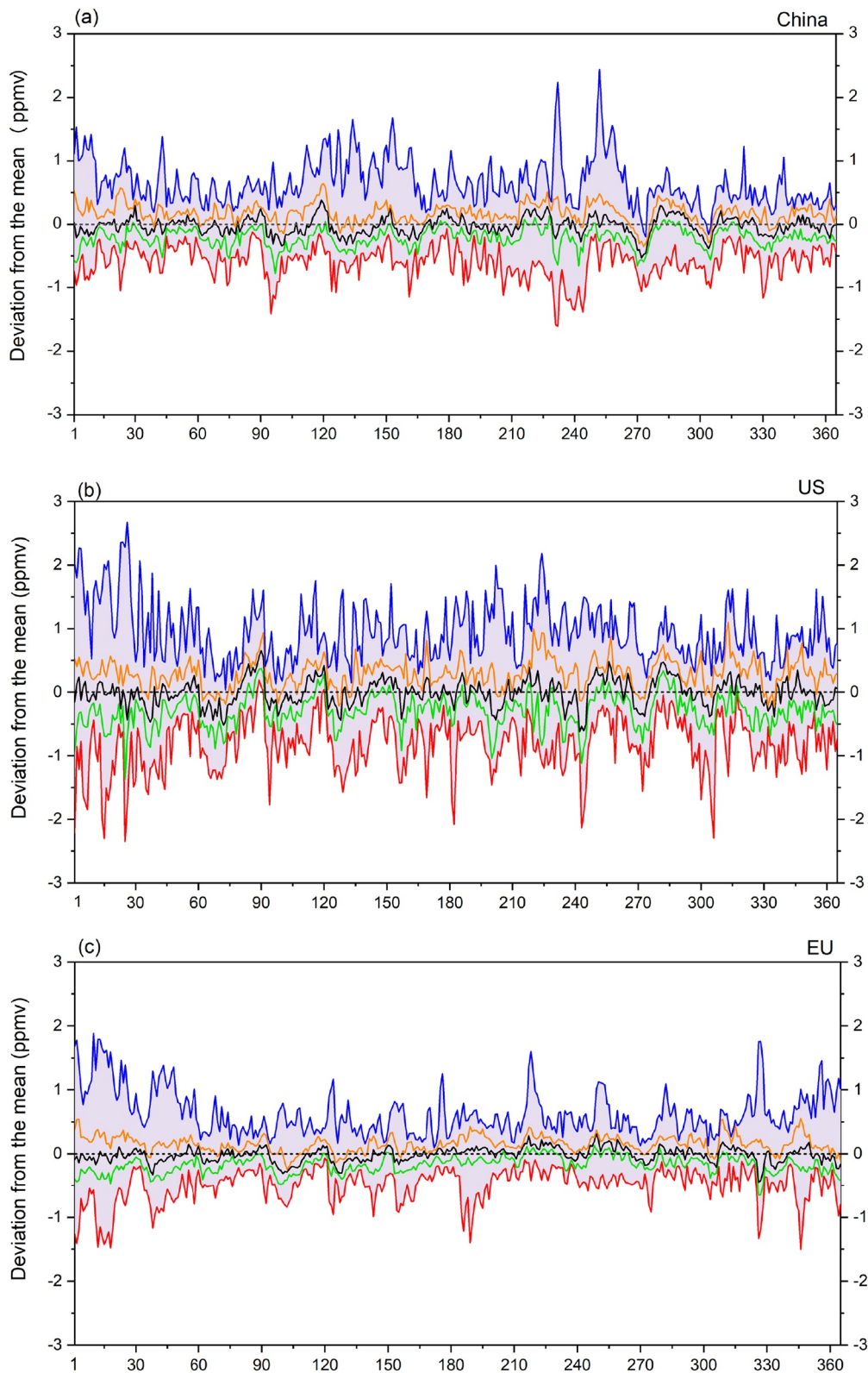


Figure 9. Deviations in the regional surface daily mean CO₂ concentrations (ppmv) between the simulations with the individual net ecosystem exchange of 15 MsTMIP models and simulations with the ensemble mean net ecosystem exchange for (a) China, (b) the USA and (c) the EU. The green, black and orange lines represent the 25th, 50th, and 75th percentiles. The shading between the blue (maximum deviation) and the red (minimum deviation) line represents the range of deviation of the daily CO₂ concentrations resulting from differences in terrestrial fluxes.

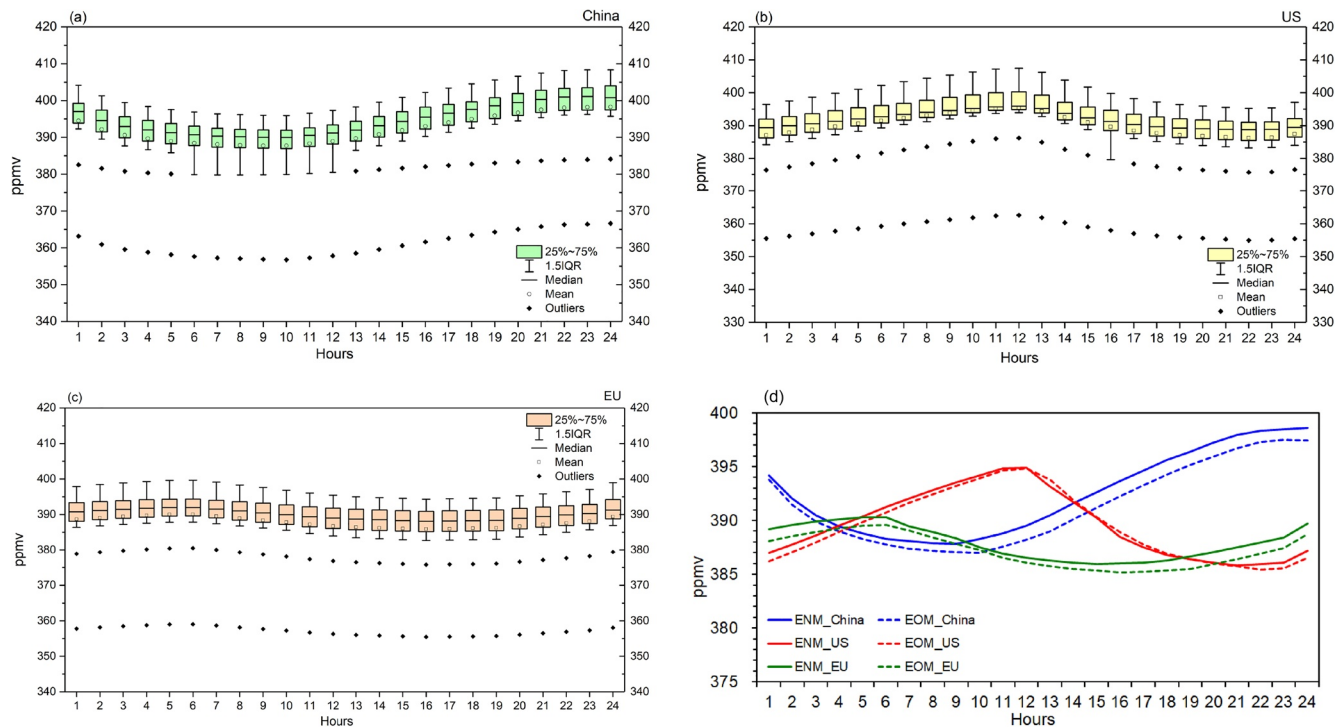


Figure 10. Boxplots of the simulated regional mean hourly CO₂ concentration averaged over the time period 2004–2010 for (a) China, (b) the USA, and (c) the EU. (d) Variations in the regional mean hourly CO₂ concentrations from the S_ENM and S_EOM simulations averaged over the time period 2004–2010. The boxes enclose the 25th, 50th, and 75th percentiles, the circles represent the average values and the diamonds represent the outliers.

4. Discussion and Conclusions

We examined the impact of terrestrial fluxes on the simulated atmospheric concentrations of CO₂ on different timescales (yearly, monthly, daily, and hourly) through a series of global CTM (GEOS-Chem) simulations with multiple prior terrestrial fluxes from the MsTMIP TBM. We determined the effects of uncertainties in the terrestrial carbon flux on the forward simulation of CO₂ concentrations. This work will help our understanding of the impact of different prior terrestrial carbon fluxes on atmospheric CO₂ simulations and provide useful information for reconciling the differences between CO₂ model simulations and observations.

The great diversity in the NEE predicted by current biosphere models from MsTMIP and for each TBM resulted in the deviation from the mean NEE flux averaged over the time period 2004–2010 ranging from –6% to 380% on the global scale. We used the results of CO₂ forward simulations applying the unweighted (ENM) and weighted (EOM) ensemble mean terrestrial carbon fluxes from MsTMIP to evaluate the performance of the models in simulating atmospheric CO₂ concentrations. Comparison of the model column CO₂ concentrations with GOSAT observations indicated that GEOS-Chem reproduces the spatiotemporal variations in atmospheric CO₂ reasonably well by using the ensemble mean NEE fluxes as the prior terrestrial flux. However, the simulated CO₂ concentrations showed substantial seasonal and annual differences globally when applying different ensemble mean flux datasets from MsTMIP, reflecting the effects of the remaining uncertainties in different sources of ensemble terrestrial flux data based on different optimized approaches. For a further quantitative assessment, monthly simulated surface CO₂ concentrations with different prior terrestrial fluxes were compared with surface measurements at 38 sites around the world. The results showed that the simulations with ENM and EOM fluxes had the best performance compared with the observations at all sites. We also found large differences among the simulated interannual trends of surface CO₂ concentrations when using different terrestrial fluxes as prior estimates.

The results of the sensitivity experiments showed that the spread of terrestrial fluxes can cause a simulated CO₂ concentration amplitude of 6.0–8.0 ppmv on annual timescales. The simulated trend of atmospheric

CO₂ might also be significantly affected. The simulated daily mean CO₂ concentration anomalies over the time period 2004–2010 may change within the range -2.0 to $+2.0$ ppmv as a result of the spread in the prior terrestrial fluxes. The magnitude of the change in the sub-daily CO₂ concentrations varied in the range 4.0–6.0 ppmv on a regional scale as a result of the discrepancies among prior terrestrial fluxes.

This work implies that the uncertainties among current estimates of the terrestrial flux are important in CO₂ simulations and that CO₂ simulations on different timescales are closely related to the choice of the prior terrestrial flux. There are some sources of uncertainty in this study. The uncertainties associated with other sources and sinks, such as anthropogenic emissions, emissions from biomass burning and ocean exchange, were not taken into account, which may affect the simulation of atmospheric CO₂ concentrations. Some studies have assessed anthropogenic CO₂ emissions and uncertainties as prior information for modeling and data assimilation on regional to global scales (Choulga et al., 2020; Han et al., 2020). Some studies have suggested that the variability among transport models may be a large source of uncertainties in forward and inverse modeling of the CO₂ flux of biospheres (e.g., Basu et al., 2018; Schuh et al., 2019), indicating that the impact of prior terrestrial fluxes on CO₂ simulations found here needs further examination using model ensembles. The impact of the initial conditions of the model on the simulation of atmospheric CO₂ remains unclear and should be investigated further.

Accurate estimates of terrestrial carbon sinks in particular regions could be important because anthropogenic carbon emissions are expected to decrease in the future, especially in regions where the terrestrial carbon flux is still less constrained by observations. Many studies have tried to provide optimized estimates of terrestrial carbon fluxes (D. F. Baker et al., 2010; Chevallier et al., 2005; Gurney et al., 2003) and J. Liu et al. (2017) tried to quantify the response of tropical terrestrial carbon fluxes to the El Niño Southern Oscillation by the assimilation of multiple chemical species. Crowell et al. (2019) estimated surface CO₂ fluxes from multiple inversion systems in the Intercomparison Project ensemble model framework using OCO-2 observations. Philip et al. (2019) investigated the effects of multiple sources of a prior terrestrial CO₂ flux on inversion modeling of terrestrial flux estimates.

All these studies have greatly improved our understanding of the estimates of terrestrial carbon sources and sinks. However, our study is important because the effects of uncertainties in the terrestrial carbon flux on the forward simulation of CO₂ concentrations, quantitatively obtained in this work, may help our understanding of the impact of different prior terrestrial carbon fluxes on atmospheric CO₂ simulations and may be useful in reconciling the differences between CO₂ simulations and observations.

Acknowledgments

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Data Availability Statement

Carbon Monitoring System: The modeled Net Ecosystem Exchange at 3-hourly time steps over 2004–2010 from the Multi-scale Synthesis and Terrestrial Model Intercomparison Project (MsTMIP) is available at <http://dx.doi.org/10.3334/ORNLDAAC/1315>. CarbonTracker optimized terrestrial flux (CT2016) at three-hourly resolution is publicly available at <https://gml.noaa.gov/aftp/products/carbontracker/co2/CT2016/fluxes/three-hourly/>. GOSAT XCO₂ retrieval (ACOS GOSAT/TANSO-FTS Level 2 bias-corrected XCO₂, ACOS_L2_Lite_FP V7.3) is obtained from https://disc.gsfc.nasa.gov/datacollection/ACOS_L2_Lite_FP_7.3.html. The in situ monthly CO₂ flask observations from Atmospheric Carbon Dioxide Dry Air Mole Fractions from the NOAA GML Carbon Cycle Cooperative Global Air Sampling Network are downloaded from https://gml.noaa.gov/aftp/data/trace_gases/co2/flask/surface/. Model results can be accessed at <https://zenodo.org/record/5081676#.YOarhnJ61oU>.

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