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## **Enhanced regional terrestrial carbon uptake over Korea revealed by atmospheric CO<sub>2</sub> measurements from 1999 to 2017**

Running head: Terrestrial carbon cycle from atmosphere

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## Abstract

Understanding changes in terrestrial carbon balance is important to improve our knowledge of the regional carbon cycle and climate change. However, evaluating regional changes in the terrestrial carbon balance is challenging due to the lack of surface flux measurements. This study reveals that the terrestrial carbon uptake over the Republic of Korea has been enhanced from 1999 to 2017 by analyzing long-term atmospheric CO<sub>2</sub> concentration measurements at Anmyeondo Station (36.53°N, 126.32°E) located in the western coast. The influence of terrestrial carbon flux on atmospheric CO<sub>2</sub> concentrations ( $\Delta\text{CO}_2$ ) is estimated from the difference of CO<sub>2</sub> concentrations that were influenced by the land sector (through easterly winds) and the Yellow Sea sector (through westerly winds). We find a significant trend in  $\Delta\text{CO}_2$  of  $-4.75 \text{ ppmv decade}^{-1}$  ( $p < 0.05$ ) during the vegetation growing season (May through October), suggesting that the regional terrestrial carbon uptake has increased relative to the surrounding ocean areas. Combined analysis with satellite measured normalized difference vegetation index and gross primary production shows that the enhanced carbon uptake is associated with significant nationwide increases in vegetation and its production. Process-based terrestrial model and inverse model simulations estimate that regional terrestrial carbon uptake increases by up to 9.9 and 4.2 Tg C decade<sup>-1</sup>, accounting for 13.4 and 5.7% of annual domestic carbon emissions averaged for the study period, respectively. Atmospheric chemical transport model simulations indicate that the enhanced terrestrial carbon sink is the primary reason for the observed  $\Delta\text{CO}_2$  trend rather than anthropogenic emissions and atmospheric circulation changes. Our results highlight the fact that atmospheric CO<sub>2</sub> measurements could open up the possibility of detecting regional changes in the terrestrial carbon cycle even where anthropogenic emissions are not negligible.

## Introduction

Atmospheric CO<sub>2</sub> concentration has risen from 278 parts per million by volume (ppmv) in pre-industrial times (1750) (Joos & Spahni, 2008) to 405 ppmv in 2017 (Dlugokencky & Tans, 2019) owing to anthropogenic carbon emissions. Elevated atmospheric CO<sub>2</sub> increases the global mean temperature, which contributes to changes in the magnitudes and intensities of extreme weather events such as heat waves, droughts, and heavy rainfall (Min, Zhang, Zwiers, & Hegerl, 2011; IPCC, 2014; Seneviratne, Donat, Pitman, Knutti, & Wilby, 2016). Thus, a clear understanding of the carbon cycle is critical for the recognition of contemporary climate change as well as future climate projections. The Global Carbon Project (GCP) (<http://www.globalcarbonproject.org/>), an international research project, estimates the annual global carbon budget using the latest models and observations to identify processes contributing to atmospheric CO<sub>2</sub> changes (Le Quéré et al., 2018). As a part of the GCP, the REgional Carbon Cycle Assessment and Processes project (RECCAP; Canadell et al., 2011) quantifies carbon fluxes on subcontinental scales. GCP estimated an imbalance of approximately 5% in the recent global carbon budget (Le Quéré et al., 2018). In line with GCP estimations, RECCAP highlighted significant uncertainties in the regional carbon budget (Haverd et al., 2013; Patra et al., 2013; Piao et al., 2013; Sitch et al., 2015; Thompson et al., 2016), implying limitations in our knowledge about spatiotemporal variations in the regional carbon cycle. Therefore, comprehensive analysis and assessment of the regional carbon cycle, including anthropogenic carbon emissions and land surface carbon balance, could reduce uncertainties in the global carbon cycle.

Terrestrial ecosystems alleviated the increasing atmospheric CO<sub>2</sub> by absorbing approximately one third of emitted anthropogenic carbon from fossil fuel combustion and land use change in 2017 (Le Quéré et al., 2018). Due to the high capability for carbon assimilation, terrestrial ecosystems are considered one of the main components of the regional carbon cycle. However, lack of relevant information on its spatial distribution and temporal variation has made it difficult to assess changes in the terrestrial carbon sink. To estimate the changes, previous studies have utilized process-based land surface models (Piao et al., 2011; Cui et al., 2014; Lee et al., 2014; Yue, Unger, & Zheng, 2015), the inverse modeling approach (Enting & Mansbridge, 1989; Gurney et al., 2003; Nassar et al., 2011; Liu et al., 2014), forest inventories (Choi, Lee, & Chang, 2002; Pan et al., 2011; Jeong et al., 2013; Fang et al., 2014), and eddy covariance tower

measurements (Piao et al., 2008; Dragoni et al., 2011; Keenan et al., 2014; Yun & Chun, 2018). Process-based models are useful for estimating responses of terrestrial ecosystems to environmental change on regional to global scales, but they have inherent uncertainties. While inverse modeling frameworks help to reduce the uncertainties in flux estimation by using atmospheric CO<sub>2</sub> measurements, the inversion results are highly sensitive to the distribution of observation sites (Bruhwiler, Michalak, and Tans, 2011) and physical parameterizations in transport models (Patra et al., 2008; Peylin et al., 2013). National forest inventories provide reliable quantitative information on forest biomass changes, but parameter uncertainties also occur when estimating forest carbon stock. Flux tower measurements directly observe terrestrial carbon exchange with the atmosphere; however, the number of measurement sites is insufficient for detecting regional-scale changes due to its large spatial heterogeneity. Recently, new approaches have been introduced to overcome the limitations of ground-based observations and models (Parazoo et al., 2016; Commane et al., 2017, Jeong et al., 2018). For example, the study by Jeong et al. (2018) showed that the ecosystem in the northern Alaska region has become a carbon source during autumn by separating regional influence on atmospheric CO<sub>2</sub> at Barrow from the background. As such, previous studies have shown that long-term atmospheric CO<sub>2</sub> observations could help us to evaluate the role of specific components, such as terrestrial ecosystems, in determining atmospheric CO<sub>2</sub> concentrations in data-limited environments.

The Republic of Korea (Korea), located in East Asia, has been experiencing rapid economic development and land-use change over recent decades. The gross domestic product has more than tripled between 1999 and 2017 (World Bank, 2018) because of accelerated industrialization and urbanization since the 1970s. With this economic growth, there is an associated increase in energy consumption that is predominantly supplied by fossil fuels, making Korea the 9th largest fossil fuel CO<sub>2</sub> emitter in the world in 2017 (168 Tg C year<sup>-1</sup>; Global Carbon Project, 2018). Thus, Korea is likely a hot spot of carbon emissions. In contrast to the large emissions, there is the carbon sink across the country, which has changed considerably over the recent decades. A national reforestation and forest management plan was implemented in 1973, and sustained forest management by the national forestry department and the local governments has increased the forest volume in Korea from 60.3 m<sup>3</sup> ha<sup>-1</sup> in 1999 to 146.0 m<sup>3</sup> ha<sup>-1</sup> in 2015 (Korea Forest Service, 2018). The managed young forests, occupying around 65% of the country, had the highest carbon

sequestration rate over East Asia of 5–12 Tg C year<sup>-1</sup> in the 1990s and 2000s (Choi et al., 2002; Li, Yi, Son, Jin, & Han, 2010) and have potentially offset 8.9% of the domestic anthropogenic emissions from the 1970s to 2000s (Fang et al., 2014). Furthermore, a longer vegetation growing season related to regional warming could contribute to enhanced carbon sequestration over Korea (Jeong et al., 2013). Therefore, assessing the role of terrestrial ecosystem change in the regional carbon cycle is critical to understanding the carbon cycle dynamics.

Continuous monitoring of atmospheric CO<sub>2</sub> concentrations has been carried out in Korea at Anmyeondo (AMY) since 1999. The AMY site is located on the western tip of the Korean Peninsula, which is surrounded by the ocean on three sides and has the advantage of being able to monitor variations in atmospheric CO<sub>2</sub> both within and outside the mainland. Using the long-term measurements at the AMY site, we assessed the possibility of using atmospheric CO<sub>2</sub> measurements to understand the terrestrial carbon cycle at a regional scale (i.e. Korea). First, we estimated the influence of regional land-surface on atmospheric CO<sub>2</sub> from the difference between CO<sub>2</sub> concentrations measured when wind blows from the land sector and from the Yellow Sea sector, which can be considered a proxy for the net ecosystem exchange, as in Jeong et al. (2018). To further explain the observed regional changes in atmospheric CO<sub>2</sub>, we identified vegetation and terrestrial carbon flux changes using satellite measurement datasets and model simulation results, respectively. The influences of changes in regional land-surface carbon fluxes as well as atmospheric transport on the CO<sub>2</sub> concentration were also evaluated through chemical transport model simulations. Multiple lines of evidence provide reliable information on changes in the terrestrial carbon cycle over Korea during the last two decades in this study.

## **Materials and methods**

This study combines both observational analysis and model simulations to investigate changes in the terrestrial carbon cycle over Korea in the last two decades. We first detected the signal of changes in regional land-surface carbon fluxes from the difference between CO<sub>2</sub> concentrations measured at the AMY Station when wind blows from the land sector and from the Yellow Sea sector ( $\Delta\text{CO}_2$ ). We hypothesized that the observed  $\Delta\text{CO}_2$  trend could be associated with changes in the regional terrestrial carbon fluxes. To support our hypothesis, we identified changes in

vegetation and terrestrial carbon fluxes in the region. The changes in vegetation and its production were investigated using the GIMMS and MODIS NDVI datasets and MODIS GPP dataset, respectively. The changes in terrestrial carbon fluxes were estimated using the TRENDY model simulation results and the CT2017 dataset. GEOS-Chem model simulations were also conducted to evaluate the influences of changes in regional terrestrial carbon fluxes as well as fossil fuel carbon emissions and atmospheric circulation on the CO<sub>2</sub> concentration over Korea.

#### *Atmospheric CO<sub>2</sub> observation*

The Anmyeondo Observatory (36.53°N, 126.32°E) is one of 112 World Meteorological Organization (WMO) Global Atmosphere Watch (GAW) regional stations (Figure 1). The Korea Meteorological Administration (KMA) has continuously measured atmospheric CO<sub>2</sub> concentrations at the site from a 40 m tower (47 m above sea level) since January 1999 (Cho, Kim, & Yoo, 2007; Lee, Han, Ryoo, Lee, & Lee, 2019). The collected data are managed by the World Data Centre for Greenhouse Gases (WDCGG). We used hourly CO<sub>2</sub> concentration data from January 1999 to October 2017 that were calibrated through quality control processes reported by Lee et al. (2019). The reliability of the hourly data was evaluated by comparison with flask-air samples collected weekly at the AMY Station (Lee et al., 2019).

Because long-term CO<sub>2</sub> measurements have been performed at only one station in Korea, we sampled hourly CO<sub>2</sub> concentration data based on wind direction and built two different monthly datasets, which likely represent land-influenced CO<sub>2</sub> concentration (CO<sub>2</sub><sup>land</sup>) and background CO<sub>2</sub> concentration for the CO<sub>2</sub><sup>land</sup> (CO<sub>2</sub><sup>YellowSea</sup>), respectively. Due to absence of local wind data, we used hourly wind data from the Automatic Synoptic Observation System at the Seosan Station, which is the wind measurement site nearest to the AMY Station (around 30 km). To reduce the effect of local CO<sub>2</sub> sources and expand the area of influence, we only considered the hourly CO<sub>2</sub> data with wind speeds exceeding 3 m s<sup>-1</sup> during the daytime (from 12:00 p.m. to 17:00 p.m.) when vertical mixing is active. Our main findings are identical when the threshold is between 2.5 and 3.5 m s<sup>-1</sup>. Taking into account the coastline layout, CO<sub>2</sub><sup>land</sup> is defined as observations made when the wind direction was between 45° and 135° (land sector). CO<sub>2</sub><sup>YellowSea</sup> is defined as observations made when the wind direction was between 225° and 315° (Yellow Sea sector). Using wider



ranges of wind direction did not alter our results, but we aimed to discriminate between  $\text{CO}_2^{\text{land}}$  and  $\text{CO}_2^{\text{YellowSea}}$ . The separated  $\text{CO}_2^{\text{land}}$  and  $\text{CO}_2^{\text{YellowSea}}$  were smoothed by the curve-fitting method of Thoning et al. (1989) and aggregated into monthly values. Finally, the difference between the monthly  $\text{CO}_2^{\text{land}}$  and  $\text{CO}_2^{\text{YellowSea}}$  ( $\Delta\text{CO}_2 = \text{CO}_2^{\text{land}}$  minus  $\text{CO}_2^{\text{YellowSea}}$ ) was treated as a proxy for the atmospheric signal for regional land–surface carbon fluxes over Korea at a monthly scale. This approach has been applied to determine the changes in regional land-surface  $\text{CH}_4$  and  $\text{CO}_2$  exchange (Sweeney et al., 2016; Commane et al., 2017; Jeong et al., 2018).

The KMA has also been monitoring atmospheric  $\text{CO}_2$  concentrations at Ulleungdo (ULD; 37.48°N, 130.90°E) from a 10 m tower (221 m above sea level) with the same measurement system of Anmyeondo (AMY) Station since 2014 after two years of test operation (Lee et al., 2019). The ULD Station is located at Ulleungdo (72 km<sup>2</sup>), approximately 155 km east from the mainland (Figure 1), and data from this site represents the atmospheric  $\text{CO}_2$  flowing into the mainland from the East Sea. Hourly  $\text{CO}_2$  concentrations at the ULD Station were compared with the  $\text{CO}_2^{\text{land}}$  to validate whether the variations of  $\text{CO}_2^{\text{land}}$  are mainly attributed to the regional land–surface  $\text{CO}_2$  fluxes or the air coming from the East Sea. The hourly ULD  $\text{CO}_2$  concentration data were smoothed by the curve-fitting method of Thoning et al. (1989) and aggregated into monthly values from January 2014 to October 2017.

#### *Back-trajectory analysis*

To verify our classification between  $\text{CO}_2^{\text{land}}$  and  $\text{CO}_2^{\text{YellowSea}}$  based on locally measured wind data, we investigated inflow direction of air masses for winds from the land and Yellow Sea sectors using a back-trajectory model. We used the National Oceanic and Atmospheric Administration (NOAA) HYbrid Single-Particle Lagrangian Integrated (HYSPLIT, version 4; Stein et al., 2015) model to calculate 48-h back-trajectories of air masses arriving at the AMY Station. The trajectories started 500 m above sea level, which are within the daytime mixing layer height. Altering the initial altitude level from 500 m to 1000 m did not change results in this study. The National Centers for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) reanalysis meteorological dataset with a 6-h interval and spatial resolution of 2.5° was used as the model input.

Trajectory densities of the air masses were generated at a  $0.5^\circ \times 0.5^\circ$  spatial resolution to investigate the overall spatial patterns of the trajectories for each case; we calculated the percentage of the number of trajectories passing through each grid cell among the total number of back-trajectories. The percentage decreases farther away from the station where a value of 100% appears, which indicates that all trajectories pass the area (Figure 2). Trajectory distributions of the air masses for wind from the Yellow Sea sector and from the land sector explicitly differ. To gather quantitative information, we set a domain centered at the station ( $35.5\text{--}37.5^\circ\text{N}$ ,  $125.3\text{--}127.3^\circ\text{E}$ ; boxed area in Figure 2a) and noted the direction from which the air masses arrived at the domain. 88.3% of the air mass for wind from the Yellow Sea sector enters the domain from the ocean side (Figure 2a). The northwestern airflow, which crosses the west coast of the Korean Peninsula, is dominant. Conversely, 82.9% of the air mass for wind from the land sector enters the domain from the landside (Figure 2b). The main airflows arrive at the station directly from the eastern Korean Peninsula. The remaining 17.1% of the air mass enters the domain from the ocean side, especially northwest of the station. The mismatched air mass contributes to shifting the median of back-trajectories in the east coast of the Korean Peninsula toward the northwest between 36 and 48 hours prior. These trajectory patterns indicate that our wind direction-based classification is reasonable for separating the air mass into two groups: inflow from outside and inflow from inside the Korean Peninsula ( $\text{CO}_2^{\text{YellowSea}}$  and  $\text{CO}_2^{\text{land}}$ ).

#### *Satellite-measured NDVI dataset*

Remote sensing provides continuous observational data over vast areas. Satellite-measured normalized difference vegetation index (NDVI) datasets have been widely used to detect variations in vegetation growth (Jeong, Ho, & Jeong, 2009; Jeong et al., 2013; Zhang, Song, Band, Sun, & Li, 2017). Here, we used two different satellite-measured NDVI datasets, the Global Inventory Monitoring and Modeling System (GIMMS) 3g NDVI and Moderate Resolution Imaging Spectroradiometer (MODIS) NDVI, to gather more reliable information of the changes in vegetation over Korea during the last two decades.

The third generation of the GIMMS NDVI dataset was produced from advanced very high-resolution radiometers (AVHRRs) on the polar-orbiting meteorological satellites of NOAA and European organizations for the exploitation of meteorological satellites (Pinzon & Tucker, 2014). The dataset was calibrated using the Bayesian method and other satellite-measured NDVI data derived from the sea-viewing wide field-of-view sensor. The reported accuracy of the data is within  $\pm 0.005$  (Pinzon & Tucker, 2014). The GIMMS NDVI dataset covers the period from July 1981 to December 2015 with a 1/12 degree of spatial resolution and 15-day intervals. The data for the period overlapping with the AMY CO<sub>2</sub> data-collection period were used for the current study (from 1999 to 2015).

The MODIS NDVI dataset from MOD13C1 (Version 6) products was derived from MODIS Terra images (Didan, 2015). The MOD13C1 dataset is spatially aggregated from 1 km of cloud-free MOD13A2 data to a 0.05° geographic climate modeling grid with 16-day intervals and covers the period from February 2000 to the present. Reliability of the MODIS NDVI dataset was evaluated by comparisons with other existing satellite-measured NDVI data, and its accuracy is within  $\pm 0.025$  (MODIS Land Team, 2018). The data for the period overlapping with the AMY CO<sub>2</sub> data collection period was used for the current study (from 2000 to 2017).

#### *MODIS GPP*

The MODIS gross primary production (GPP) dataset from MOD17A2H (Version 6) products from MODIS Aqua and Terra images were used to estimate the changes in GPP in Korea from 2000 to 2017 (Running and Zhao, 2015). The algorithm based on light-use efficiency concepts was applied to generate the GPP dataset from the meteorological field of the Global Modeling and Assimilation Office supported by the National Aeronautics and Space Administration and satellite-derived fraction of photosynthetically active radiation from MOD15 (Running and Zhao, 2015). The MOD17A2H dataset is a cumulative 8-day composite of values with 500 m spatial resolution and covers the period from February 2000 to the present. The MODIS GPP dataset was spatially aggregated from 500 m to a 0.05° geographic climate modeling grid to match the spatial resolution of MODIS NDVI for the current study.

### *TRENDY models*

We used monthly averaged carbon fluxes (GPP, Ra; autotrophic respiration, Rh; heterotrophic respiration, and NBP; net biome production) simulated by 11 dynamic global vegetation models from the TRENDY project version 6 (simulation S3; Sitch et al., 2015) to estimate changes in terrestrial carbon fluxes over Korea from 2000 to 2016. The list of models used is as follows: CABLE (Haverd et al., 2018), CLM4.5 (Oleson et al., 2013), DLEM (Tian et al., 2015), ISAM (Jain, Meiyappan, Song, & House, 2013), LPJ (Sitch et al., 2003), LPX-Bern (Stocker, Feissli, Strassmann, Spahni, & Joos, 2014), OCN (Zaehle, Friedlingstein, & Friend, 2010), ORCHIDEE (Krinner et al., 2005), VEGAS (Zeng, Mariotti, & Wetzal, 2005), VISIT (Ito & Inatomi, 2012), and JULES (Clark et al., 2011). Models were forced by observed climate fields from the Climate Research Unit (CRU) dataset (CRU-TS 3.23 and 3.25) or CRU-National Centers for Environmental Prediction version 8 meteorological dataset, global atmospheric CO<sub>2</sub> concentration data from ice-cores and atmosphere measurements from NOAA Earth System Research Laboratory (ESRL) (Dlugokencky & Tans, 2019), and land use and land cover change data from the HYDE database (Hurtt et al., 2011). The model outputs from the TRENDY project have been applied in a number of studies to estimate the changes in terrestrial carbon balance and to understand the driving factors for the change at global as well as regional scales (Piao et al., 2012; Le Quéré et al., 2018).

### *GEOS-Chem model simulations*

We used the Goddard Earth Observing System (GEOS)-Chem model, a global 3-D chemical transport model (CTM) for atmospheric compositions (Bey et al., 2001), to evaluate the influences of changes in the regional land–surface carbon fluxes and atmospheric transport on the CO<sub>2</sub> concentrations over Korea. We conducted GEOS-Chem CO<sub>2</sub> simulations, which ignore the chemical reactions of CO<sub>2</sub> but consider surface CO<sub>2</sub> fluxes to the atmosphere and transport of atmospheric CO<sub>2</sub>. This version of the model has been widely used to investigate the response of atmospheric CO<sub>2</sub> to changes in fossil fuel emissions (Nassar et al., 2013) and terrestrial carbon fluxes (Barnes, Parazoo, Orbe, & Denning, 2016; Parazoo et al., 2016).

We used a nested-grid GEOS-Chem model (version 11.2) with the native  $0.5^\circ \times 0.667^\circ$

horizontal resolution and 47 vertical layers from surface pressure to 0.01 hPa over East Asia (11°S–55°N, 70–150°E) (Wang, McElroy, Jacob, & Yantosca, 2004). Boundary conditions for the nested version of the model were provided hourly from the global GEOS-Chem simulations with  $4^\circ \times 5^\circ$  horizontal resolution. All model simulations were performed by using observed meteorological data and land-surface carbon flux datasets. We used the hourly meteorological data from the Modern-Era Retrospective analysis for Research and Applications version 2 (MERRA-2) reanalysis datasets with the same spatial resolutions as each simulation (Gelaro et al., 2017). The carbon flux datasets include terrestrial biospheric exchange, ocean exchange (Takahashi et al., 2009), emissions from fossil fuels, biomass burning (Randerson, Van Der Werf, Giglio, Collatz, & Kasibhatla, 2018), biofuel burning (Yevich & Logan, 2003), shipping, and aviation (Simone, Stettker, & Barrett, 2013). Among the carbon flux dataset, monthly terrestrial carbon fluxes and fossil fuel carbon emissions were taken from CarbonTracker outputs (CT2017; Peters et al., 2007) and the Open-source Data Inventory for Anthropogenic CO<sub>2</sub> (ODIAC) dataset (Oda & Maksyutov, 2011), respectively.

GEOS-Chem was spun up from 1990 to 1999 using the meteorological data and fossil fuel carbon emission data of 1990–1999 and the other surface CO<sub>2</sub> flux datasets fixed at their values in 2000. Starting from the model state in 2000, we performed a set of sensitivity simulations from 2000 to 2016 (Table 1). In simulation E1, all variables used are transient. In simulations E2, E3, and E4, one or both monthly terrestrial carbon fluxes and fossil fuel carbon emissions in 2000 are repeatedly prescribed over Korea (34–38°N, 126–130°E) during the entire simulation period; however, other conditions are identical to those of simulation E1. Simulation E5 is identical to simulation E3, but the carbon emissions in 2000 are prescribed over eastern China (20–40°N, 100–125°E and 40–50°N, 100–140°E) not Korea. From the differences between simulation E1 and simulations E2 and E3 (e.g. simulation E1 minus simulation E2), influences of the changes in regional terrestrial carbon flux and anthropogenic carbon emissions on atmospheric CO<sub>2</sub> in Korea were estimated, respectively. The possible influences of changes in transported atmospheric CO<sub>2</sub> from other regions were estimated in simulation E4. The effect of changes in anthropogenic carbon emissions in eastern China, which are expected to account for the largest portion of the transported CO<sub>2</sub>, was estimated from the difference between simulations E1 and E5. The model bias related to the overestimation of the long-term trend in CO<sub>2</sub> concentrations was corrected by

comparing the AMY CO<sub>2</sub> concentration data. Simulated surface CO<sub>2</sub> concentrations were only used for analysis in the study.

In the case of terrestrial carbon fluxes, we conducted an additional set of model simulations using TRENDY model results. In simulation T1 (control simulation), monthly terrestrial carbon flux for a specific year are repeatedly prescribed during the entire simulation period; there are no annual changes in the seasonal variation of the terrestrial carbon flux. In simulations T2, T3, and T4 (sensitivity simulations), the monthly flux in Korea is set to linearly change from the fixed value by the average, maximum, and minimum trends of the estimated NBP in the TRENDY models during the entire simulation period. The range of influences of the estimated NBP changes on the CO<sub>2</sub> concentrations are obtained from the differences of simulations T2, T3, and T4 with simulation T1.

## Results

### *The signal of regional land-surface carbon flux changes on atmospheric CO<sub>2</sub>*

Both time series of CO<sub>2</sub><sup>land</sup> and CO<sub>2</sub><sup>YellowSea</sup> at the AMY Station show gradually increasing trends and clear seasonal variations, with the maximum in early spring and minimum in late summer from January 1999 to October 2017 (Figure 3a). However, the seasonal variability of CO<sub>2</sub><sup>land</sup> is greater than that of CO<sub>2</sub><sup>YellowSea</sup>. Compared with CO<sub>2</sub><sup>YellowSea</sup>, annually averaged CO<sub>2</sub><sup>land</sup> is lower by 1.9 ppmv on average during the growing season (from May to October) and higher by 3.6 ppmv on average during the non-growing season (from November to April) for the total study period (Figure 3b; black thick line). In recent years, notable decreases in annually averaged  $\Delta$ CO<sub>2</sub> during the growing season appear from  $0.5 \pm 2.1$  ppmv in 1999–2006 to  $-5.0 \pm 2.2$  ppmv in 2010–2017 (Figure 3b; bar). The annually averaged monthly  $\Delta$ CO<sub>2</sub> in the two periods are out of each other's error ranges as represented by the  $1\sigma$  in May, June, July, and September. For example, the annually averaged  $\Delta$ CO<sub>2</sub> in May is  $1.0 \pm 3.0$  ppmv in the first eight years and  $-4.8 \pm 3.8$  ppmv in the last eight years. The results suggest that the recent decreases in  $\Delta$ CO<sub>2</sub> for the growing season are not considered as noise. A statistically significant trend of the  $\Delta$ CO<sub>2</sub> is also observed for the growing season, on average, at  $-4.75$  ppmv decade<sup>-1</sup> ( $p < 0.05$ ; Figure 3c); it

corresponds to an increase of 250% decade<sup>-1</sup> of the magnitude of its climatological value since the climatological value is small. On the contrary, an increase in the  $\Delta\text{CO}_2$  is observed from November to March at 1.27 ppmv decade<sup>-1</sup>, but this is not significant. The results suggest that the atmospheric  $\text{CO}_2$  concentration over Korea rises slowly relative to the surrounding ocean regions during the growing season for the last two decades.

#### *Changes in vegetation and terrestrial carbon fluxes*

To understand the reasons for the observed negative  $\Delta\text{CO}_2$  trends during the growing season, we evaluated the changes in vegetation and terrestrial carbon fluxes over Korea for the last two decades. Korea, where two thirds of the country are forested (Korea Forest Service, 2018), has a climatological NDVI of 0.69 (GIMMS: 0.69, MODIS: 0.69; Figure 4a, b) and an accumulated GPP of 919 gC m<sup>-2</sup> during the growing season (Figure 4c). Both two different satellite observe nationwide positive trends in NDVI (Figure 4a, b). For the growing season, the NDVI from GIMMS increases by 0.030 decade<sup>-1</sup> over 92.1% of the region from 1999 to 2015; this accounts for a 4.3% decade<sup>-1</sup> increase. Similarly, the NDVI from MODIS increases by 0.025 decade<sup>-1</sup> over 93.7% of the entire study area from 2000 to 2017; this accounts for a 3.6% decade<sup>-1</sup> increase. The increasing trends of NDVI from GIMMS and MODIS are significant at the 95% confidence level in 34.4% and 75.1% of the study area, respectively. In line with the observed nationwide vegetation greening, accumulated GPP from MODIS increases by 68.6 gC m<sup>-2</sup> decade<sup>-1</sup> over 93.9% of the entire study area during the growing season from 2000 to 2017; this accounts for a 7.5% decade<sup>-1</sup> increase (Figure 4c). Some parts of central Korea show decreases or little changes in the GPP, but a significant increase in the GPP is observed in 45.1% of the analysis domain, especially over the southwestern part of Korea.

The TRENDY multi-model mean overestimates the climatological value of the area-averaged accumulated GPP over the study area during growing season as  $1269 \pm 277$  gC m<sup>-2</sup>, but it estimated an increase in GPP for the season by  $54.8 \pm 27.5$  gC m<sup>-2</sup> decade<sup>-1</sup>, similar to the GPP from MODIS ( $61.4$  gC m<sup>-2</sup> decade<sup>-1</sup>) for 2000–2016 (Figure 5a). Due to the relatively low increase in total respiration ( $36.0 \pm 29.0$  gC m<sup>-2</sup> decade<sup>-1</sup>) compared with the GPP, the net terrestrial carbon uptake (i.e. NBP) has been enhanced by  $18.5 \pm 24.0$  gC m<sup>-2</sup> decade<sup>-1</sup> during the growing season

(Figure 5b, c), corresponding to a  $14.8 \pm 19.2\%$  decade<sup>-1</sup> increase of its climatological value ( $125 \pm 105$  gC m<sup>-2</sup>). Even though large variations among the TRENDY models exist from  $-48.7$  to  $101.3$  gC m<sup>-2</sup> decade<sup>-1</sup> (this accounts for  $39.0\%$  decade<sup>-1</sup> decrease and  $81.0\%$  decade<sup>-1</sup> increase), the NBP from the multi-model mean has the same sign with the trends of the biospheric carbon flux of  $42.9$  gC m<sup>-2</sup> decade<sup>-1</sup> from CT2017, corresponding to  $45.5\%$  decade<sup>-1</sup> increase of its climatological value ( $94.2$  gC m<sup>-2</sup>). The estimated enhancement of terrestrial carbon uptake from TRENDY and CT2017 models accounts up to  $13.4$  and  $5.7\%$  of annual domestic carbon emissions averaged for the study period ( $141$  Tg C year<sup>-1</sup>; Global Carbon Project, 2018), respectively. Both models also estimate a relatively weak increasing trend of the carbon flux in July than adjacent months. This is due to the stagnant rain front, which blocks solar radiation and inhibits vegetation growth over Korea during the summer rainy season (Lee et al., 2017). These results derived from independent datasets and models suggest that the terrestrial carbon uptake in Korea has increased during the growing season due to the nationwide vegetation greening for the last two decades.

#### *Influences of changes in regional land–surface carbon fluxes on atmospheric CO<sub>2</sub>*

We performed CTM simulations to implement the influences of model-estimated changes in regional terrestrial carbon fluxes on the atmospheric CO<sub>2</sub> in Korea from 2000 to 2016. The enhanced terrestrial carbon uptake estimated in both CT2017 and TRENDY multi-model mean consistently induce decreases in CO<sub>2</sub> concentrations in Korea during the growing season, distinct from the non-growing season (Figure 6a). In the case of the CT2017, CO<sub>2</sub> concentration decreases by  $0.64$  ppmv decade<sup>-1</sup> during the growing season and shows little increase ( $0.06$  ppmv decade<sup>-1</sup>) during the non-growing season (Figure 6a; red line). The maximum decreasing trend appears in June at  $1.25$  ppmv decade<sup>-1</sup>, as do the observed  $\Delta$ CO<sub>2</sub> trend. In the case of the TRENDY models, CO<sub>2</sub> concentration decreases up to  $1.37$  ppmv decade<sup>-1</sup> during the growing season, even though the multi-model mean estimated a relatively small decrease ( $0.24$  ppmv decade<sup>-1</sup>) than CT2017 (Figure 6a). A possible maximum decreasing trend of the CO<sub>2</sub> concentration is also exhibited in June at  $2.39$  ppmv decade<sup>-1</sup>. The results suggest that the regional enhancement of terrestrial carbon uptake is the main reason for the observed negative trend in the  $\Delta$ CO<sub>2</sub> during the growing season for the study period.



In addition to the biosphere fluxes, anthropogenic carbon emissions increased by 33 Tg C decade<sup>-1</sup> in Korea for the analysis period. Seasonal differences in the monthly carbon emissions trends also appear; the trends of the non-growing season (2.9 Tg C month<sup>-1</sup> decade<sup>-1</sup>) are greater than those of the growing season (2.6 Tg C month<sup>-1</sup> decade<sup>-1</sup>) by 0.3 Tg C month<sup>-1</sup> decade<sup>-1</sup> on average; the difference accounts for around 11% of the annual mean trends. Our model estimates that the rise in domestic carbon emissions increases annual mean CO<sub>2</sub> concentrations in Korea by 1.11 ppmv decade<sup>-1</sup> (Figure 6b). However, notable seasonal differences are not observed in the increasing trends; the seasonally averaged trends are 1.04 and 1.17 ppmv decade<sup>-1</sup> during the growing season and non-growing season, respectively. The model simulation results suggest that domestic carbon emissions are not a main reason for the observed ΔCO<sub>2</sub> trends.

#### *Influences of atmospheric transport changes on atmospheric CO<sub>2</sub> over Korea*

Korea is located in the downstream area of China, the biggest CO<sub>2</sub> emitter in the world, and close to a strong summer uptake region (Siberia). Considering the geographical location, the influences of atmospheric transport changes on CO<sub>2</sub> concentrations over Korea are also evaluated through model simulations. Even without considering the land–surface carbon flux changes in Korea, annual mean CO<sub>2</sub> concentrations increased more than 21 ppmv decade<sup>-1</sup> from 2000 to 2016 over East Asia. The largest increase is found in eastern China at more than 31 ppmv decade<sup>-1</sup> and the increasing trend is weakened further away from the region (Figure 7a). The spatial distributions of the increasing trends are mostly associated with a steep rise of fossil fuel carbon emissions in eastern China at 1.4 Pg C decade<sup>-1</sup> for the analysis period. The increasing carbon emissions raises CO<sub>2</sub> concentrations in the main source regions such as the north China plain, Beijing, and Shanghai by more than 13 ppmv decade<sup>-1</sup>, as well as surrounding regions including Korea by more than 6 ppmv decade<sup>-1</sup> (Figure 7b). We compared the influences of atmospheric transport change on the monthly trends of atmospheric CO<sub>2</sub> in the surrounding area (Yellow Sea) and in the Korean Peninsula (boxed area in Figure 8): Region1 (35–38°N, 123.0–126.0°E) and Region2 (35–38°N, 126.5–129.5°E).

The atmospheric transport changes raise the annual mean CO<sub>2</sub> concentration in Region1 more than that in Region2 by 0.86 ppmv decade<sup>-1</sup> (Figure 8a). More than 1.3 ppmv decade<sup>-1</sup> of the

regional differences in CO<sub>2</sub> concentration trends (Region2 minus Region1) exhibits in April and November, and less than 0.5 ppmv decade<sup>-1</sup> of the regional differences are found in January and August. The annual mean trends of the regional differences mostly result from the increasing carbon emissions in eastern China (0.69 ppmv decade<sup>-1</sup>; Figure 8b). The monthly variations of the regional differences are associated with wind circulation changes. For example, reduced westerly flow and northwesterly flow over the Korean Peninsula intensify the CO<sub>2</sub> concentration gradient from eastern China to Korea and increase the regional differences in April and November, respectively. However, the two regions shared similar seasonal patterns in CO<sub>2</sub> concentration trends. Even seasonally averaged trends of the regional differences are smaller during the growing season (−0.76 ppmv decade<sup>-1</sup>) than they are during the non-growing season (−0.92 ppmv decade<sup>-1</sup>). These results suggest that the change in atmospheric transport is not a main reason for the observed negative trends in ΔCO<sub>2</sub> during the growing season, distinct during the non-growing season.

## Discussion

The best way to understand the terrestrial carbon cycle would be to use as many terrestrial carbon flux measurements as possible. However, the number of terrestrial carbon flux measurements is still limited in many parts of the globe (Schimel et al., 2015). Recent studies suggest the applicability of long-term measurements of atmospheric CO<sub>2</sub> concentrations to evaluate the regional terrestrial carbon flux changes (Commane et al., 2016; Jeong et al., 2018). This study assessed whether surface atmospheric CO<sub>2</sub> measurements can be used to understand the regional terrestrial carbon cycle in Korea. However, unlike the clean-air environment in Alaska where previous studies have been performed (Commane et al., 2016; Jeong et al., 2018), Korea is located at the center of the East Asia region where a rapid increase in anthropogenic carbon emissions have been reported over the recent decades (Global Carbon Project, 2018). The higher increasing trends of CO<sub>2</sub><sup>YellowSea</sup> and CO<sub>2</sub><sup>land</sup> than that of the globally averaged CO<sub>2</sub> concentration (2.07 ppmv year<sup>-1</sup>), derived from the NOAA/ESRL data (Dlugokencky & Tans, 2019), indicate that the influences of anthropogenic emissions on the spatiotemporal variations in CO<sub>2</sub> concentrations cannot be negligible over the region as highlighted in previous modeling (Ballav et al., 2012; Ballav et al., 2016) and observation studies (Umezawa et al., 2018).

The changes in regional land–surface carbon fluxes were estimated from the difference between  $\text{CO}_2^{\text{YellowSea}}$  and  $\text{CO}_2^{\text{land}}$  in this study. Although our back-trajectory analysis showed that  $\text{CO}_2^{\text{land}}$  could represent the inflow of air masses at the AMY Station from the Korea Peninsula, we further checked the possible influences of air coming from the East Sea on the  $\text{CO}_2^{\text{land}}$  by comparing with  $\text{CO}_2$  concentration at the ULD Station. On average, the  $\text{CO}_2$  concentration at the ULD Station is lower than both  $\text{CO}_2^{\text{land}}$  and  $\text{CO}_2^{\text{YellowSea}}$  by 2.2 and 3.1 ppm from January 2014 to October 2017 (Figure 9), respectively, because the ULD Station is located far from the  $\text{CO}_2$  source regions. However, the seasonal variability of  $\text{CO}_2^{\text{land}}$  is greater than that of the  $\text{CO}_2$  concentrations at the ULD Station and annual minimum value of the monthly  $\text{CO}_2^{\text{land}}$  is lower than that of the ULD Station in three of the past four years. For example, in 2014, the annual maximum and minimum values of monthly  $\text{CO}_2^{\text{land}}$  are 415.6 and 387.9 ppmv but those of the monthly ULD  $\text{CO}_2$  concentration are 407.7 and 396.2 ppmv, respectively. In addition, averaged  $\text{CO}_2^{\text{land}}$  from May to September (400.3 ppmv) is lower than that of the ULD  $\text{CO}_2$  concentration (403.9 ppmv) by 3.6 ppmv during the analysis period. These results indicate that the variations in  $\text{CO}_2^{\text{land}}$  are mainly attributed to the regional land–surface carbon fluxes rather than the air coming from the East Sea. Further, it is worth noting that Ballav et al. (2016) showed that the relatively high nocturnal  $\text{CO}_2$  concentration over land was sometimes observed over the open ocean region the next day, mostly when wind blows from land to sea; thus, higher  $\text{CO}_2^{\text{land}}$  could be observed in those cases. However, our results show that clear seasonal variations in  $\Delta\text{CO}_2$  appear (Figure 3b) and  $\text{CO}_2^{\text{land}}$  has greater seasonal variability than that of both  $\text{CO}_2^{\text{YellowSea}}$  (Figure 3a) and the ULD  $\text{CO}_2$  concentrations (Figure 9). This indicates that  $\Delta\text{CO}_2$  can capture the regional land–surface carbon flux signals that are distinct from the surrounding area (the Yellow Sea and East Sea).

Based on the observed  $\Delta\text{CO}_2$ , we found that regional land carbon uptake significantly increases relative to the surrounding ocean region during the growing season, as opposed to increase in land carbon emissions during the non-growing season. We first assumed that this was from an enhancement of terrestrial carbon uptake for the period and investigated the changes in vegetation and terrestrial carbon fluxes using available satellite datasets and model simulation results. Our analysis of the satellite datasets show that significant nationwide increasing trends in vegetation and its photosynthesis appear for the growing season. The results are in line with the

previous finding that the NDVI and vegetation production showed consistent long-term increasing trends in the region (Fang et al., 2014). The national forest inventory reporting that the forest volume of Korea increased by 142% from 1999 to 2015 is additional evidence for the increase in vegetation production (Korea Forest Service, 2018). Generally, major reasons for the vegetation greening are considered as fertilization effects of increasing ambient CO<sub>2</sub> and nitrogen deposition, warming-driven enhancement of photosynthesis, and lengthening of growing seasons (Jeong et al., 2013; Piao et al., 2015; Zhu et al., 2016). In addition to the environmental changes at a global scale, sustained national forest management campaigns during the last two decades seem to be another key driver. Contrasting NDVI trends between South Korea (Republic of Korea) and North Korea (Democratic People's Republic of Korea) are observed in Figure 4. While forest areas in South Korea have remained steady, massive deforestation has taken place in North Korea since the early 1990s (Cui et al., 2014; Engler, Tepyakov, & Adams, 2014). The different forest management history causes the vegetation browning trend in the southern area of North Korea despite the favorable environmental conditions for vegetation growth.

In accordance with the observed increasing trends of vegetation and its production, both TRENDY multi-model mean and CT2017 estimate the enhancement of terrestrial carbon uptake during the growing season, although the magnitudes differ by models. This study used the results of simulation S3, considering all changes to climate, CO<sub>2</sub>, and land use, but found that there are little differences with the results of simulation S2, ignoring land use change (not shown here) despite Korean forests at least doubling in volume during the study period. It seems that the TRENDY models underestimate influences of land use change on terrestrial carbon fluxes. In addition, the globally averaged annual CO<sub>2</sub> concentration was equally prescribed in all regions in the TRENDY models. Considering that the increasing rate of atmospheric CO<sub>2</sub> concentration at the AMY Station is higher than that of the global mean value, the CO<sub>2</sub> fertilization effect could also be underestimated over the region. Thus, the process-based models (TRENDY) seem to estimate the increasing trends less than that of the inverse model constrained by CO<sub>2</sub> measurements data (CT2017). Although our CTM model simulations have relatively weaker responses to the estimated terrestrial carbon flux changes, they show that the observed  $\Delta$ CO<sub>2</sub> decrease is mainly associated with the enhancement of terrestrial carbon uptake in Korea during the growing season rather than other factors (influences of other factors will be discussed in the

next paragraph). The results are consistent with previous studies estimating the strengthening of the carbon sink in Korean forests based on forest inventories (Pan et al., 2011; Jeong et al., 2013; Fang et al., 2014). Therefore, our findings provide evidence that atmospheric CO<sub>2</sub> measurements could be used to detect changes in the terrestrial carbon cycle at regional scales even where anthropogenic activities cannot be treated as minimal components.

This study suggests that enhanced terrestrial carbon uptake is a key process causing changes in temporal variations in  $\Delta\text{CO}_2$ . However,  $\Delta\text{CO}_2$  variations can also be modulated by fossil fuel carbon emissions and atmospheric circulation changes. From the CTM model simulations, we checked all the potential influences of the factors. The model estimates that the increasing domestic fossil fuel carbon emissions do not cause notable seasonal differences in the CO<sub>2</sub> concentration trends in Korea. That is because seasonal variability of the carbon emissions trends is too small relative to the terrestrial carbon fluxes, which change sign in the growing season and non-growing season. Previous studies also show that the fossil fuel carbon emissions in main source regions have around 20% of seasonal variability of biospheric carbon fluxes over the tropics and northern middle latitude (Zhang, Gurney, Rayner, Baker, & Liu, 2016). Even with the small seasonal variability, our results show that the increasing carbon emissions in China, which are more than 40 times of those in Korea, has the potential to induce around 1 ppmv decade<sup>-1</sup> increase of seasonal amplitude of atmospheric CO<sub>2</sub> over Korea. However, because the seasonal patterns of CO<sub>2</sub> concentration trends are similar around and in the Korean Peninsula, China's carbon emissions also seem to be a minor factor on the long-term trends in the seasonal variations of  $\Delta\text{CO}_2$ . One thing to keep in mind is that the greater increasing rate of CO<sub>2</sub> concentrations in the Yellow Sea than the Korean Peninsula could alleviate the  $\Delta\text{CO}_2$  increase caused by increasing domestic carbon emissions. Readers need to be cautious when interpreting our results (decrease in annual  $\Delta\text{CO}_2$  trends does not mean that Korea becomes a net carbon sink). We also found that the atmospheric circulation changes have possibilities to induce unequal temporal changes in CO<sub>2</sub> concentrations over Korea. Our model simulation suggests, however, that there are no notable changes in atmospheric circulation patterns leading to the observed  $\Delta\text{CO}_2$  trend during the study period. In addition, although this study treated the anthropogenic emissions and atmospheric circulation as main drivers of transport changes, the enhancement of terrestrial carbon uptake in boreal forests, highlighted in previous studies (Graven et al., 2013; Welp et al., 2016), can also be

another driver. Welp et al. (2016) showed that significant increases in amplitude of biospheric CO<sub>2</sub> flux appear in boreal regions at 0.31% year<sup>-1</sup> and in arctic regions at 0.85 % year<sup>-1</sup> for 1985–2012 from inversion results. We evaluated the possible effect through a set of ideal model simulations with and without a 1% year<sup>-1</sup> increase in seasonal amplitude of monthly terrestrial CO<sub>2</sub> fluxes in high-latitude regions (> 45°N) during the study period. The model estimated that the increasing biospheric carbon uptake increases the seasonal amplitude of CO<sub>2</sub> concentration around 1.5 ppmv decade<sup>-1</sup> over Korea, but it also did not induce remarkable regional differences in the CO<sub>2</sub> concentration trends around the AMY station (not shown here).

The CTM simulations provide valuable evidence to support our hypothesis in company with independent satellite measurement and model simulation results; however, a practical limitation remains in our simulations. Our sensitivity tests of atmospheric CO<sub>2</sub> were conducted at a monthly scale instead of using hourly composite like the observations. Current CTMs are not able to represent locally measured hourly variations in CO<sub>2</sub> concentration due to the absence of validated hourly datasets representing land–surface carbon fluxes in Korea as well as most other regions (Zhu et al., 2014; Ballav et al., 2016), hence the difficulty of performing the exact same model configuration that matches our observation analysis. Nevertheless, in the case of atmospheric transportation effect, we checked whether our results are different when the sampling method is applied to the results of simulation E4 based on the assumption that the changes in air sources into the AMY Station have more effect than sub-monthly variations in land–surface carbon fluxes in remote areas. However, the difference between simulated CO<sub>2</sub><sup>land</sup> and CO<sub>2</sub><sup>YellowSea</sup>, derived by sampling simulated hourly CO<sub>2</sub> concentration at the station based on the MERRA2 wind data, shows seasonally uniform decreasing trends consistent with our findings based on the monthly averaged values (not shown here).

This study takes advantage of the long-term continuous CO<sub>2</sub> measurement at the AMY Station. In addition to the AMY Station, numerous surface stations are continuously measuring atmospheric CO<sub>2</sub> concentration over the globe and provide hourly data. Because many of these observation sites are located in coastal areas, our approach has potential to provide wide-area information. Even if the station is located far from the coast, we could retrieve signals of terrestrial

carbon exchange by considering physical characteristics of local wind circulation. For example, the Mauna Loa Observatory (MLO), located at a high altitude (3397 m) in the center of the Hawaiian Islands, observes CO<sub>2</sub> concentrations above the planetary boundary layer, but CO<sub>2</sub> concentrations are affected by the airflow blowing from lower altitudes to the site during a few hours after sunset (Sharma and Barnes, 2016). The difference in observed CO<sub>2</sub> concentration between the two periods could play the role of  $\Delta\text{CO}_2$  of this study. Additionally, local variations in land carbon flux within the continent could be detected by comparing atmospheric CO<sub>2</sub> data from adjacent observation sites (Sargent et al., 2018).

In summary, this study showed that the terrestrial carbon uptake has been enhanced resulting from the nationwide vegetation greening over Korea during the growing season for the past two decades based on multiple lines of evidence from atmospheric measurements, satellite observations, and model simulations. Additional surface observation sites have been operated in the mainland (Seoul) and test operations for long-term aircraft measurements across the Korean Peninsula have been conducted since 2018. Satellites like the Greenhouse gases Observing SATellite (GOSAT), Orbiting Carbon Observatory 2 (OCO-2), and TanSat also have measured a large spatial extent of column-averaged CO<sub>2</sub> concentrations. Inverse modeling using the added in situ, airborne, and satellite data would provide more reliable quantitative information on the enhanced terrestrial carbon uptake by using the relationship between our results ( $\Delta\text{CO}_2$ ) and the estimated biosphere flux in future studies (Commane et al., 2016). Atmospheric CO<sub>2</sub> measurements could generate the possibility for a better understanding of the terrestrial carbon cycle where ground-based observation is limited, therefore, sustained efforts to expand the observation network are required.

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**Data Accessibility Statement:**

The atmospheric CO<sub>2</sub> measurement datasets at AMY and ULD Stations are available at <https://gaw.kishou.go.jp/> and from Haeyoung Lee ([leehy80@korea.kr](mailto:leehy80@korea.kr)) through e-mail request. The GIMMS NDVI is publicly available at <https://ecocast.arc.nasa.gov/data/pub/gimms/3g.v1>. MODIS NDVI and GPP are publicly available at <https://lpdaac.usgs.gov/products/mod13c1v006> and <https://lpdaac.usgs.gov/products/mod17a2hv006>. The TRENDY model simulation results are available from Stephen Sitch ([S.A.Sitch@exeter.ac.uk](mailto:S.A.Sitch@exeter.ac.uk)) or Pierre Friedlingstein ([p.friedlingstein@exeter.ac.uk](mailto:p.friedlingstein@exeter.ac.uk)) through e-mail request. CarbonTracker results are publicly available at <http://carbontracker.noaa.gov>. ODIAC dataset are publicly available at [doi:10.17595/20170411.001](https://doi.org/10.17595/20170411.001). The GEOS-Chem model simulation results that support the findings of this study are available from the corresponding author upon reasonable request.



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## Tables

**Table 1** A set of model simulations to estimate the influences of changes in regional land–surface carbon fluxes and atmospheric transport on atmospheric CO<sub>2</sub> over the Republic of Korea for the period 2000–2016.

Simulations	Descriptions	
	Terrestrial carbon flux	Fossil Fuel carbon emissions
E1	<b>T</b>	<b>T</b>
E2	Korea <b>FIX</b>	<b>T</b>
E3	<b>T</b>	Korea <b>FIX</b>
E4	Korea <b>FIX</b>	Korea <b>FIX</b>
E5	<b>T</b>	Eastern China <b>FIX</b>

**T**: All transient variables are used.

**FIX**: Fluxes in 2000 are repeatedly prescribed over the region.

## Figure legends

**Figure 1** Locations of Anmyeondo (AMY; open box) and Ulleungdo Observatories (ULD; open circle) and positions of the Yellow Sea and land sectors. The Yellow Sea sector is defined as the region where wind blows from 225–315° and the land sector is defined as the region where wind blows from 45–135° with respect to the AMY Station. The inset figure shows the location of the Republic of Korea over East Asia.

**Figure 2** Density plots for 48-h trajectories terminating at the AMY Station for wind from (a) the Yellow Sea sector and (b) the land sector for the period 1999–2017. Solid line represents the median trajectory. Markers on the trajectories represent the position of air masses at 6-hour intervals. Upper-right numbers of each plot signify the ratio of air masses that came into the domain (boxed area; 35.5–37.5°N, 125.3–127.3°E) from the ocean (land) side among the total air mass of wind from the Yellow Sea sector (land sector).

**Figure 3** Variations of (a) monthly CO<sub>2</sub> concentrations for wind from the Yellow Sea sector (CO<sub>2</sub><sup>YellowSea</sup>; blue line) and land sector (CO<sub>2</sub><sup>land</sup>; red line) for the period 1999–2017. (b) Annually averaged monthly  $\Delta$ CO<sub>2</sub> during the entire study period (black thick line), the first eight years (1999–2006; red bar), and the last eight years (2010–2017; blue bar). Error bar shows the monthly standard derivation of  $\Delta$ CO<sub>2</sub>. (c) Annual trends of monthly  $\Delta$ CO<sub>2</sub> for the same period. The asterisk and cross indicate statistical significance at the 95% and 90% confidence level, respectively.

**Figure 4** Spatial distributions of climatology and annual trend in averaged (a) GIMMS and (b) MODIS normalized difference vegetation index (NDVI) during the growing season (May through October) over the period 1999–2015 and 2000–2017, respectively. The stippled areas represent statistically significant correlations ( $p < 0.05$ ). (c) Same as (b) but for accumulated gross primary production (GPP) from MODIS.

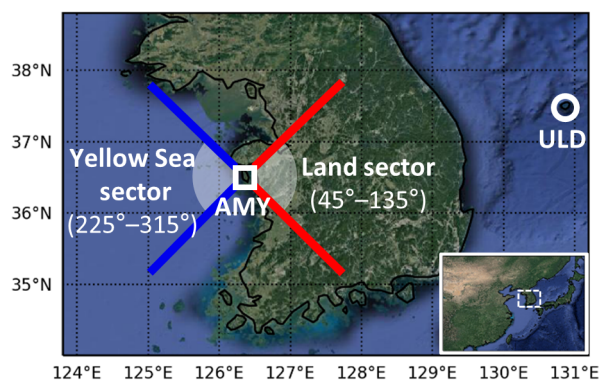
**Figure 5** Area-averaged climatology and annual trend in monthly (a) gross primary production (GPP), (b) total respiration (i.e. autotrophic respiration + heterotrophic respiration), and (c) net biome production (NBP) over the Republic of Korea simulated from TRENDY models (black line) over the period 2000–2016. The shading denotes one inter-model standard deviation range of each month. The dotted lines represent the monthly minimum and maximum trends of the estimated carbon fluxes from TRENDY models. Blue and red lines indicate GPP from MODIS and biosphere carbon flux from CT2017, respectively.

**Figure 6** Area-averaged changes in annual trends in monthly CO<sub>2</sub> concentrations due to (a) regional terrestrial carbon fluxes (red solid line: CT2017; simulation E1 minus simulation E3, black solid and dotted lines: average, maximum, and minimum of TRENDY models' estimates; simulations T2, T3, and T4 minus simulation T1, respectively) and (b) fossil fuel carbon emissions (i.e., simulation E1 minus simulation E2) at the Republic of Korea for the period 2000–2016.

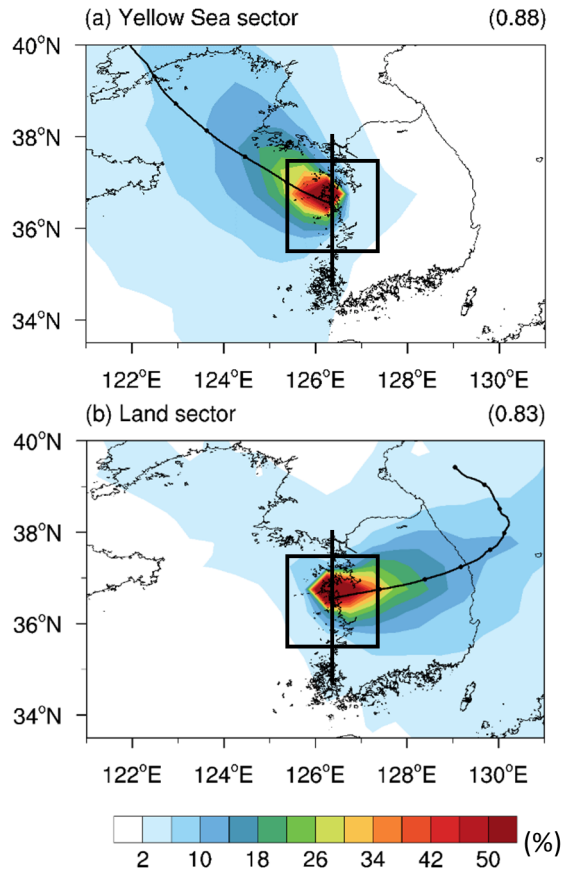
**Figure 7** Spatial distributions of changes in simulated annual mean CO<sub>2</sub> concentration trends due to (a) atmospheric transport (simulation E4) and (b) China's fossil fuel carbon emissions (simulation E1 minus simulation E5) for the period 2000–2016.

**Figure 8** Area-averaged changes in simulated monthly CO<sub>2</sub> concentration trends due to (a) atmospheric transport (i.e. simulation E4) and (b) China's fossil fuel carbon emissions (i.e., simulation E1 minus simulation E5) at the Region1 (35–38°N, 124.3–126.3°E), Region2 (35–38°N, 126.3–128.3°E), and difference between them for the period 2000–2016.

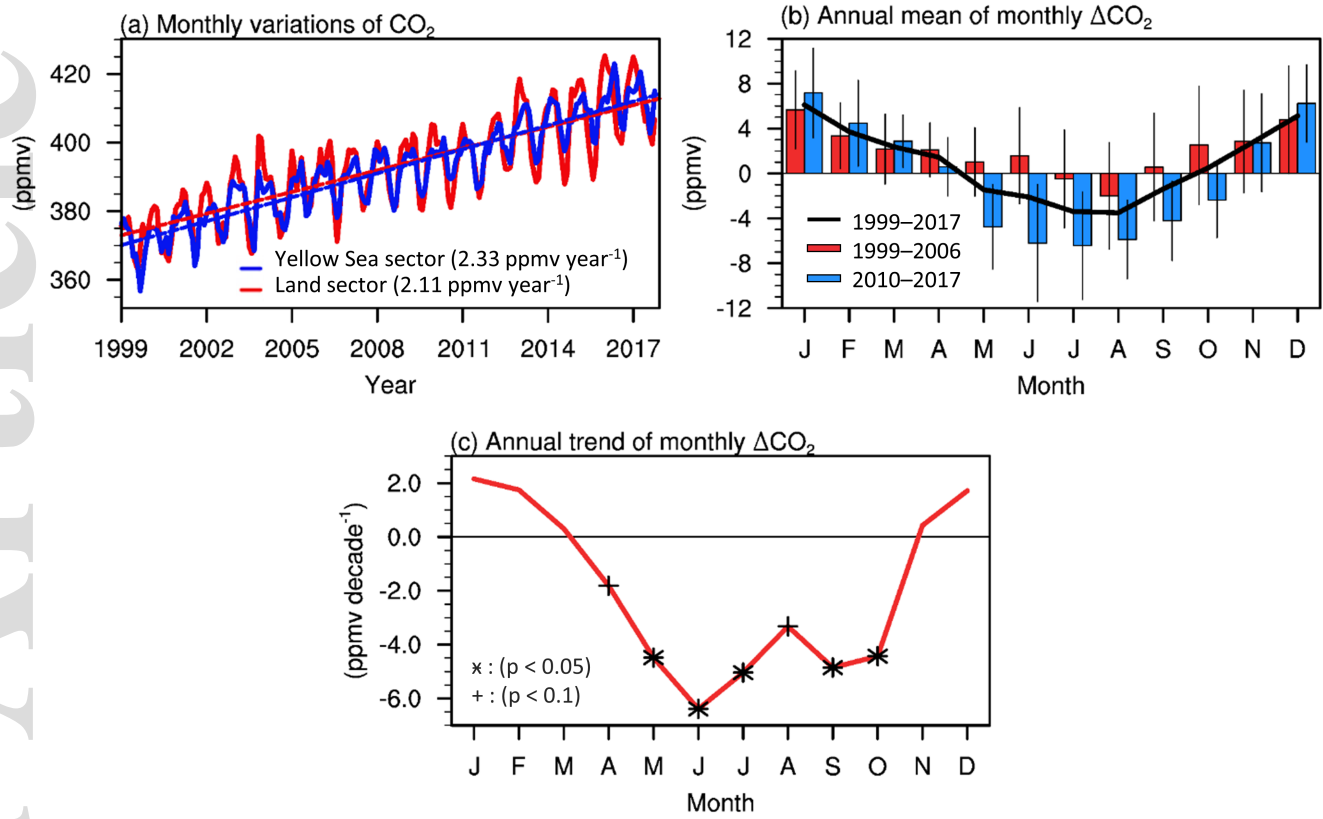
**Figure 9** Variations of monthly CO<sub>2</sub> concentration at Ulleungdo Station (ULD; black line) and CO<sub>2</sub> concentrations for wind from the Yellow Sea sector (CO<sub>2</sub><sup>YellowSea</sup>; blue line) and land sector (CO<sub>2</sub><sup>land</sup>; red line) at Anmyeondo Station (AMY) from January 2014 to October 2017. The shading denotes two standard deviation range of CO<sub>2</sub> concentration of each month.



gcb\_15061\_f1.png



gcb\_15061\_f2.png

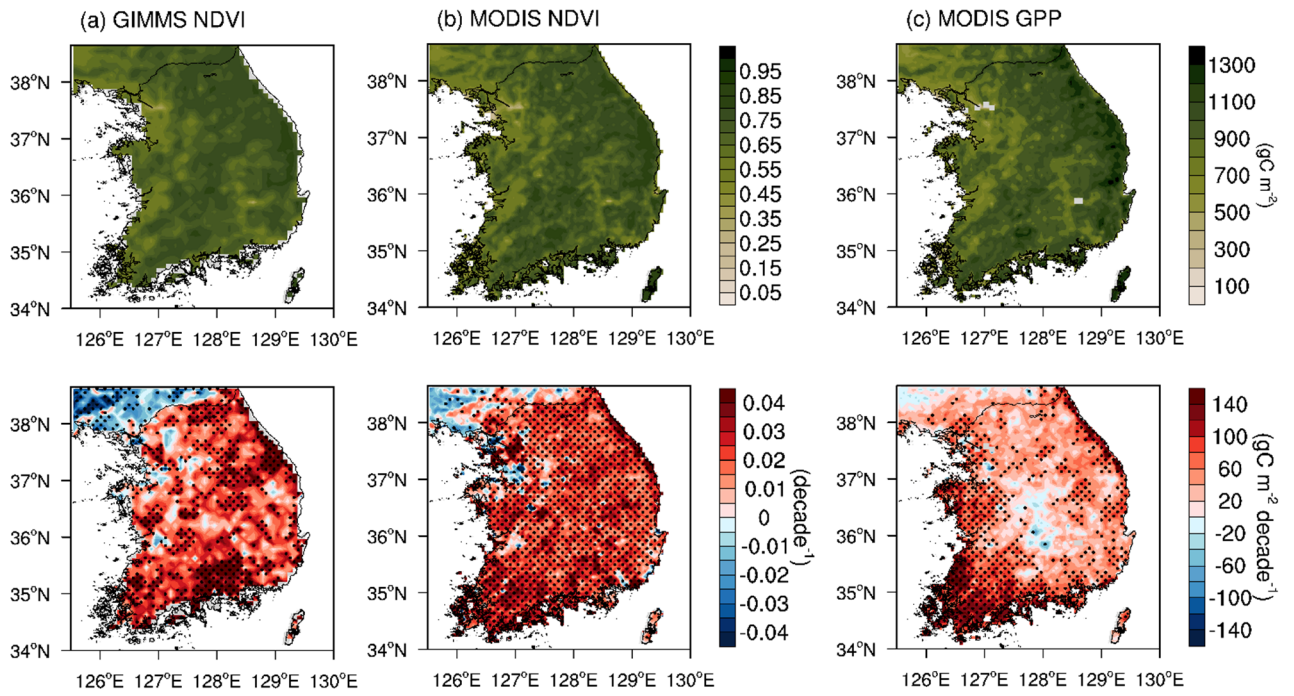


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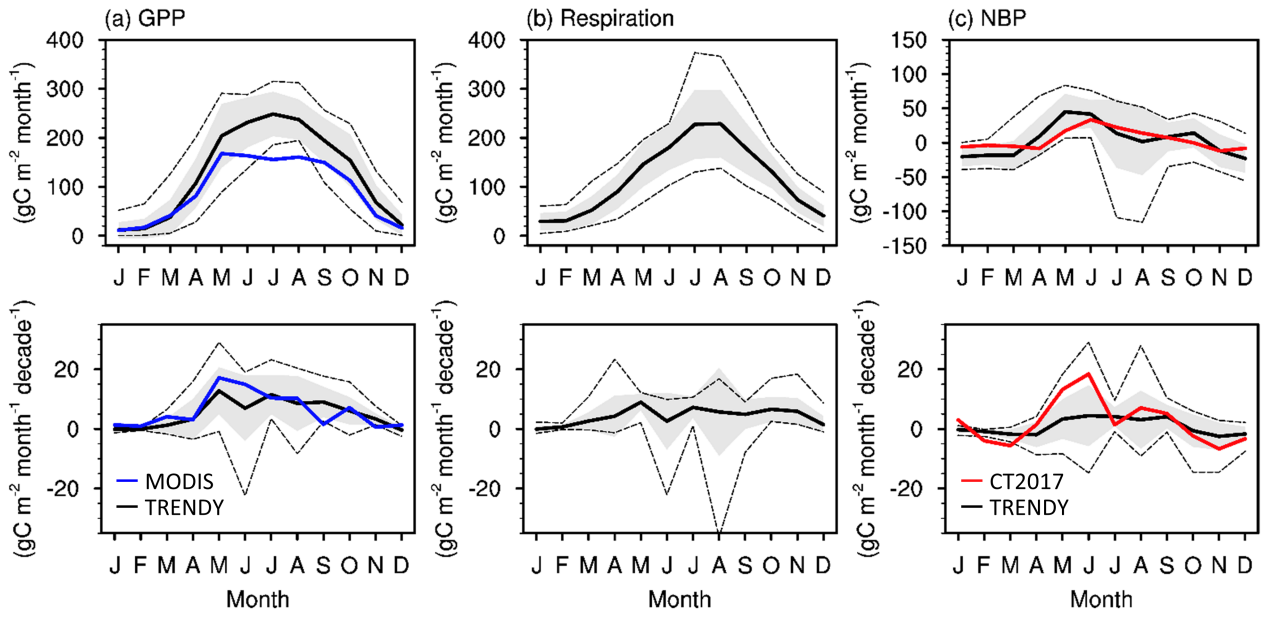
Climatology

Annual trend

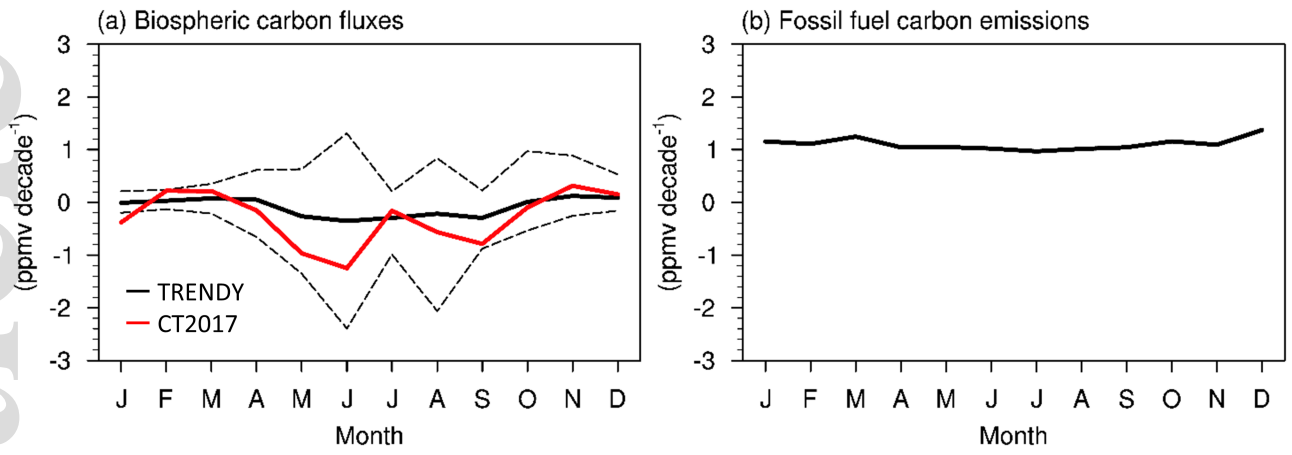


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Climatology

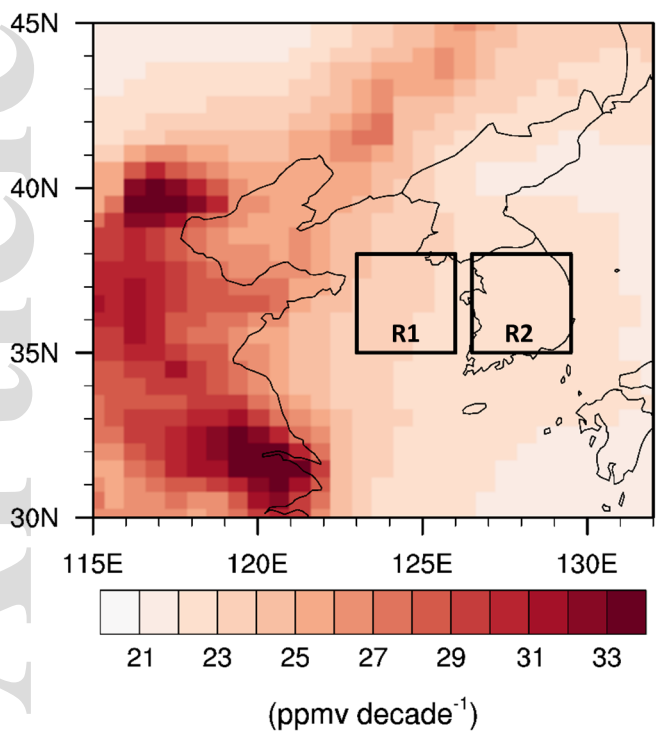


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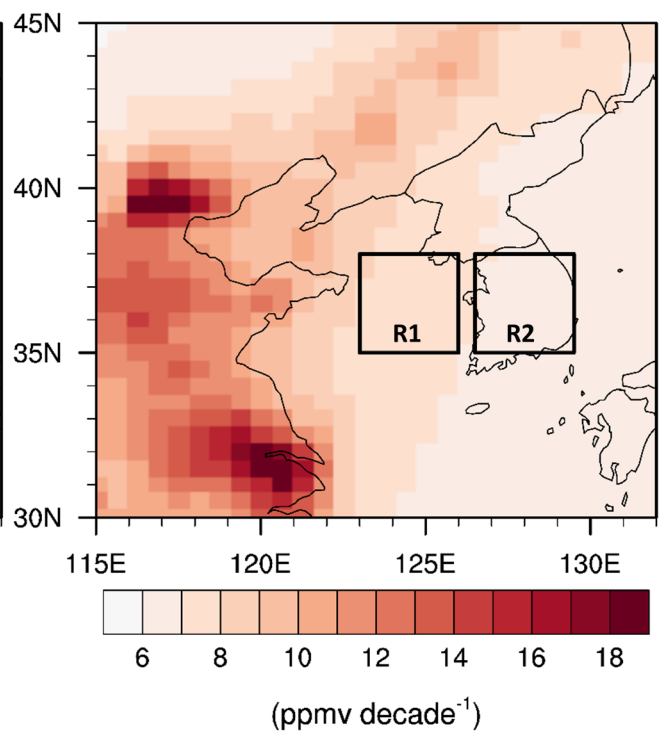


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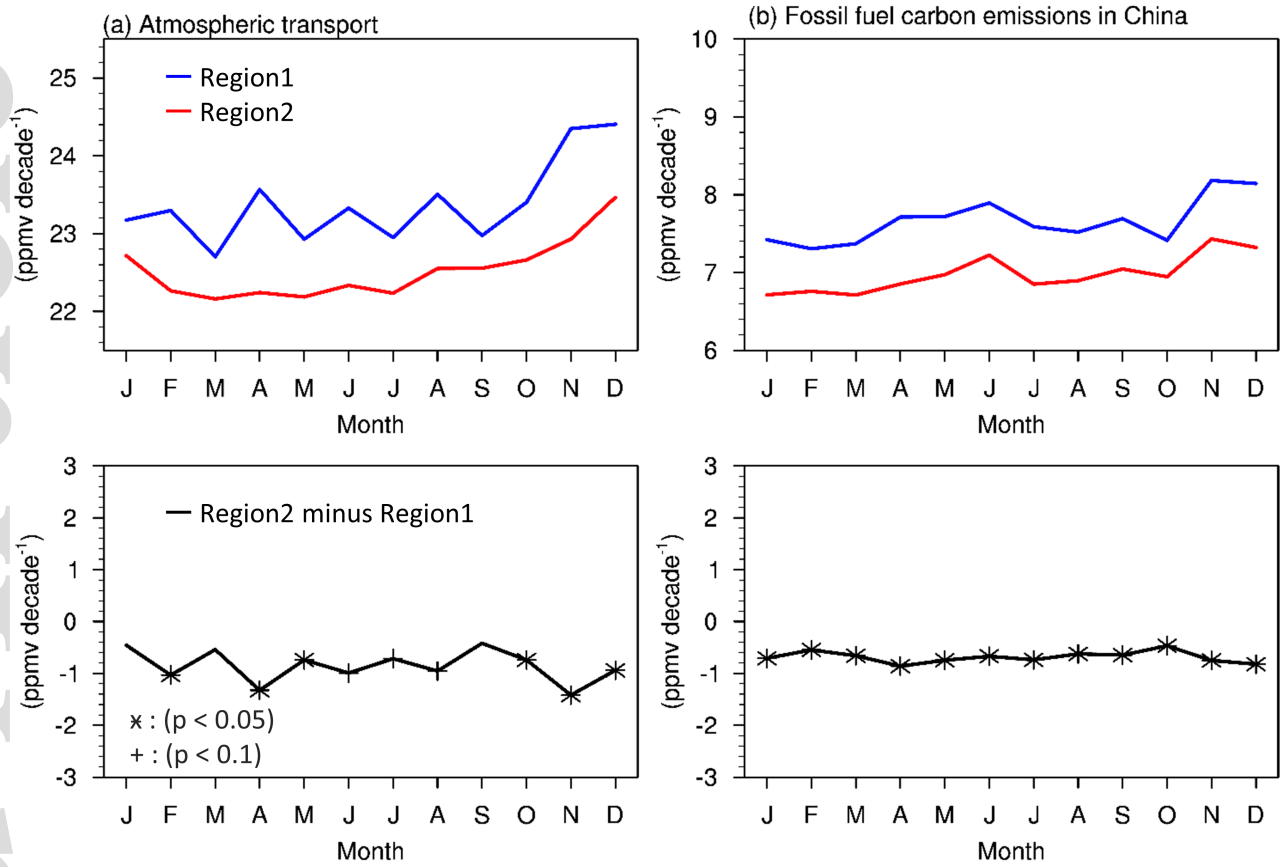
(a) Atmospheric transport



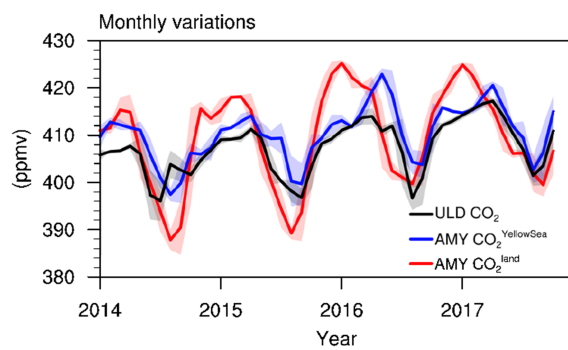
(b) Fossil fuel carbon emissions in China



gcb\_15061\_f7.png



gcb\_15061\_f8.png



gcb\_15061\_f9.png