1	Enhanced regional terrestrial carbon uptake over South Korea revealed by atmospheric		
2	CO <sub>2</sub> measurements from 1999 to 2017		
3	Running head: Terrestrial carbon cycle from atmosphere		
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7	Atual Jain <sup>9</sup> , Sönke Zaehle <sup>10</sup> , Etsushi Kato <sup>11</sup> , Hanqin Tian <sup>12</sup> , Nicolas Vuichard <sup>13</sup> , Andy		
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### 43 Abstract

The exact assessment of the role of terrestrial carbon balance in the regional carbon cycle is 44 critical for understanding climate change. However, the lack of local measurements limits our 45 knowledge of the regional-scale terrestrial carbon cycle. This study explored changes in the 46 regional-scale terrestrial carbon cycle over South Korea by utilizing long-term atmospheric 47 CO<sub>2</sub> measurements for the period 1999–2017. The influence of the regional land surface CO<sub>2</sub> 48 fluxes on atmospheric CO<sub>2</sub> was estimated from the difference between the CO<sub>2</sub> concentrations 49  $(\Delta CO_2)$  for two major wind directions: wind from the land sector and wind from the ocean 50 sector. We found a significant decreasing trend in  $\Delta CO_2$  of 4.75 ppmv decade<sup>-1</sup> (p < 0.05) 51 during the growing season (May through October) suggesting that the regional land carbon 52 uptake increased relative to the surrounding ocean areas. For the same period, the satellite-53 observed normalized difference vegetation index increased by 0.03 decade-1 across 90% of the 54 South Korean territory; this accounts for a 4% increase. In line with the observed nationwide 55 greening trend, satellite-measured gross primary production increased by 65.1 gC m<sup>-2</sup> decade 56 <sup>1</sup> over 93.9% of the region, accounting for a 8% increase. Both process-based terrestrial model 57 and inversion model simulations estimated that the vegetation increase would lead to an 58 enhancement of terrestrial carbon uptake by up to 101.3 gC m<sup>-2</sup> decade<sup>-1</sup>. Chemical transport 59 model simulations showed that the observed  $\Delta CO_2$  decrease is associated with the enhancement 60 of terrestrial carbon uptake rather than changes in anthropogenic emissions and atmospheric 61 circulations. Based on the multiple lines of evidence from surface observations, satellite data, 62 and modeling, our results suggest that the terrestrial carbon uptake over South Korea has been 63 enhanced during the growing season for the recent two decades. These findings highlight the 64 fact that atmospheric CO<sub>2</sub> measurements could open up the possibility of detecting regional 65 changes in the terrestrial carbon cycle in data-limited environments. 66

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### Introduction

Atmospheric CO<sub>2</sub> concentration has risen from 278 parts per million by volume (ppmv) in pre-69 industrial times (1750) (Joos & Spahni, 2008) to 405 ppmv in 2017 (Dlugokencky & Tans, 70 2019) due to anthropogenic carbon emissions. Elevated atmospheric CO<sub>2</sub> increases the global 71 mean temperature, which contributes to changes in the magnitudes and intensities of extreme 72 weather events such as heat waves, droughts, and heavy rainfall (Min, Zhang, Zwiers, & Hegerl, 73 2011; IPCC, 2014; Seneviratne, Donat, Pitman, Knutti, & Wilby, 2016). Thus, a clear 74 understanding of the carbon cycle is critical for recognition of contemporary climate change as 75 well future climate projections. The Global Carbon **Project** (GCP) 76 as (http://www.globalcarbonproject.org/), an international research project, estimates the annual 77 global carbon budget using the latest models and observations to identify the processes that 78 contribute to atmospheric CO<sub>2</sub> changes (Le Quéré et al., 2018). As a counterpart of GCP, the 79 REgional Carbon Cycle Assessment and Processes project (RECCAP; Canadell et al., 2011) 80 quantifies carbon fluxes on subcontinental scales. GCP estimated that around 5% of imbalance 81 existed in the recent global carbon budget (Le Quéré et al., 2018). In line with GCP estimations, 82 RECCAP highlighted significant uncertainties in the regional carbon budget (Huntzinger et al., 83 84 2012; Piao et al., 2013, Sitch et al., 2015; Thompson et al., 2016), implying limitations in our knowledge about spatiotemporal variations of the regional carbon cycle. Therefore, 85 comprehensive analysis and assessment of the regional carbon cycle, including anthropogenic 86 carbon emissions and land surface carbon balance, could reduce uncertainties in the global 87 carbon cycle. 88

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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 91 use change in 2017 (Le Quéré et al., 2018). Due to the high capability of carbon assimilation, 92 terrestrial ecosystems are considered one of the main components of the regional carbon cycle. 93 However, lack of relevant information on its spatial distribution and temporal variation has 94 made it difficult to assess changes in the terrestrial carbon sink. To estimate the changes, 95 previous studies have utilized process-based land surface models (e.g. Piao et al., 2011; Cui et 96 al., 2014; Lee et al., 2014; Yue, Unger, & Zheng, 2015), inverse modeling approach (e.g. Enting 97 and Mansbridge, 1989; Gurney et al., 2003; Nassar et al., 2011; Liu et al., 2014), forest 98 inventories (e.g. Choi, Lee, & Chang, 2002; Pan et al., 2011; Jeong et al., 2013; Fang et al., 99 100 2014), and eddy covariance tower measurements (e.g. Piao et al., 2008; Dragoni et al., 2011; Keenan et al., 2014; Yun & Chun, 2018). Process-based models are useful for estimating 101 responses of terrestrial ecosystems to environmental change on regional to global scales, but 102 103 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 104 are highly sensitive to the distribution of observation sites (Bruhwiler, Michalak, and Tans, 105 2011) and physical parameterizations in transport models (Patra et al., 2008; Peylin et al., 2013). 106 107 National forest inventories give reliable quantitative information on forest biomass changes, 108 but parameter uncertainties also occur when estimating forest carbon stock. Flux tower measurements directly observe terrestrial carbon exchange with the atmosphere and yet the 109 number of measurement sites is insufficient for detecting regional-scale changes due to the 110 large spatial heterogeneity. Recently, new approaches have been introduced to overcome the 111 limitations of ground-based observations and models (Parazoo et al., 2016; Commane et al., 112 2017, Jeong et al., 2018). For example, the study by Jeong et al. (2018) showed that the 113 114 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, 115

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.

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South Korea (Republic of Korea), located in East Asia, has been experiencing rapid economic development and land-use change over recent decades. Accelerated industrialization and urbanization since the 1970s has more than tripled the gross domestic product between 1999 and 2017 (World Bank, 2018). With this economic growth, there is an associated increase in the consumption of energy, that is predominantly supplied by fossil fuels, making South 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, South Korea is likely a hot-spot of carbon emissions. In contrast to the large emissions is the carbon sink across the country, which has changed considerably over the recent decades. A national plan of reforestation and forest management was implemented in 1973. Sustained forest management by the national forestry department and the local governments has increased the volume of forests 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  $5 \sim 12$ 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 enhancing carbon sequestration over South 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 ongoing in South 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, the present study assessed the possibility of using atmospheric CO<sub>2</sub> measurements to understand the terrestrial carbon cycle at a regional scale (i.e. South Korea). We first estimated the influence of the regional land surface on atmospheric CO<sub>2</sub> from the difference between CO<sub>2</sub> concentration values during wind from the land sector and wind from the ocean 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 changes in vegetation and terrestrial carbon fluxes 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 the chemical transport model simulations. Based on multiple lines of evidence, this study provides reliable information on changes in the terrestrial carbon cycle over South Korea during the last two decades.

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## Materials and methods

This study combines both observation analysis and model simulations to investigate changes in the terrestrial carbon cycle over South Korea in the last two decades. We first estimated the changes in regional land-surface  $CO_2$  fluxes from the difference between  $CO_2$  concentration measured at AMY Station when wind blows from the land sector and when wind blows from the ocean sector (i.e.  $\Delta CO_2$ ). We assumed that the observed  $\Delta CO_2$  trend is associated with changes in the regional terrestrial carbon fluxes and identified changes in vegetation and

terrestrial carbon fluxes in the region. The changes in vegetation and vegetation activity (i.e. photosynthesis) 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. To verify our assumption, we conducted GEOS-Chem model simulations 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 South Korea.

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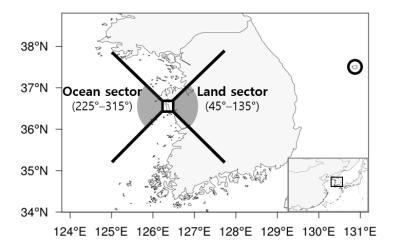
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#### Atmospheric CO<sub>2</sub> observation 172

173 The Anmyeondo Observatory (36.53°N, 126.32°E) is one of 111 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 is managed by the World Data Centre for Greenhouse Gases (WDCGG; https://gaw.kishou.go.jp/). We used hourly CO<sub>2</sub> concentration data from January 1999 to October 2017, which were calibrated through quality control processes reported in 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).



**Figure 1** Locations of Anmyeondo (AMY; open box) and Ulleungdo Observatories (ULD; open circle) and position of the ocean sector and the land sector with respect to AMY. The ocean 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 AMY. The inset figure shows the location of South Korea over East Asia.

Influences of regional land surface CO<sub>2</sub> fluxes on atmospheric CO<sub>2</sub> were distinguished based on wind speed and direction. Due to absence of local wind data, we used hourly wind data from the Automated Synoptic Observing System at Seosan Station, which is the wind measurement site nearest to the AMY Station (around 30 km). To reduce the effect of local 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>. The wind direction at Seosan Station has been recorded using 16 directions with 22.5° spacing. Taking into account the coastline layout, regionally influenced CO<sub>2</sub> concentration (CO<sub>2</sub>land) was defined as observations made when the wind direction is between 45° and 135° (land sector). Background CO<sub>2</sub> concentration for the CO<sub>2</sub>land (CO<sub>2</sub>ocean) was defined as

observations made when the wind direction is between 225° and 315° (ocean sector). The wider ranges of wind direction did not alter our results, but we aimed to discriminate between  $CO_2^{land}$  and  $CO_2^{ocean}$ . The separated  $CO_2^{land}$  and  $CO_2^{ocean}$  were smoothed by the curve-fitting method of Thoning et al. (1989) and aggregated into monthly values. We treat the difference between the monthly  $CO_2^{land}$  and  $CO_2^{ocean}$  ( $\Delta CO_2 = CO_2^{land}$  minus  $CO_2^{ocean}$ ) as a proxy for the atmospheric signal for regional land-surface  $CO_2$  fluxes over South Korea. This approach has been applied to determine the changes in regional  $CH_4$  and  $CO_2$  exchange (Sweeney et al., 2016, Commane et al., 2017, Jeong et al., 2018).

The KMA has also been monitoring atmospheric CO<sub>2</sub> 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 site is located at Ulleung-do (72 km²), around 155 km east from the mainland (Figure 1), and data from this site represents the atmospheric CO<sub>2</sub> flowing into the mainland from the East Sea. Hourly CO<sub>2</sub> concentrations at the ULD site is used to compare with the CO<sub>2</sub>land for validating whether the variations of CO<sub>2</sub>land are attributed to the regional land-surface CO<sub>2</sub> fluxes or the air coming from the East Sea. The hourly ULD CO<sub>2</sub> concentration data is smoothed by the curve-fitting method of Thoning et al. (1989) and aggregated into monthly values from January 2014 to October 2017 to compare with the CO<sub>2</sub>land.

## Back-trajectory analysis

To verify our classification between  $CO_2^{land}$  and  $CO_2^{ocean}$ , we investigated directions of air mass inflow for winds from the land sector and ocean sector 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 backtrajectories of air masses arriving at the AMY Station. The trajectories started at 500 m above sea level, which are within the height of the mixing layer for daytime. 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. To investigate the overall spatial patterns of the trajectories for each case, we generated trajectory densities of the air masses at a 0.5° x 0.5° spatial resolution. We then calculated the percentage of the number of trajectories passing through each grid cell among the total number of back-trajectories and the median trajectory at 6-h endpoints for each case. Altering the start altitude level from 500 m to 1000 m did not change results in this study (not shown here).

### 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 (e.g. 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 on the changes in vegetation over South 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; data available at https://ecocast.arc.nasa.gov/data/pub/gimms/3g.v1). 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 (i.e. from 1999 to 2015) with the AMY CO<sub>2</sub> data collection period was used for the current study.

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 present (data available at <a href="https://lpdaac.usgs.gov/products/mod13c1v006/">https://lpdaac.usgs.gov/products/mod13c1v006/</a>). Reliability of the MODIS NDVI dataset was evaluated by comparisons with other existing satellite-measured NDVI data and its accuracy is within ± 0.025 (MODIS Land Team, 2018). The data for the period overlapping (i.e. from 2000 to 2017) with the AMY CO2 data collection period was used for the current study.

## 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 South 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 500m spatial resolution, and covers the period from February 2000 to present (data available at <a href="https://lpdaac.usgs.gov/products/mod17a2hv006/">https://lpdaac.usgs.gov/products/mod17a2hv006/</a>). The MODIS GPP dataset was spatially aggregated from 500 m to a 0.05° geographic climate modeling grid to match 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 (S3 simulation; Sitch et al., 2015) to estimate changes in terrestrial carbon fluxes over South Korea between 2000-2016. The list of the models used are 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, & Wetzel, 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 (e.g. CRU-TS 3.23 and 3.25) or CRU-National Centers for Environmental Prediction version 8 meteorological dataset, global atmospheric CO2 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 driving factors for the change at global as well as regional scales (e.g. 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 South 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 (e.g. Nassar et al., 2013) and terrestrial carbon fluxes (e.g. Barnes, Parazoo, Orbe, & Denning, 2016, Parazoo et al., 2016).

We used a nested-grid GEOS-Chem model (version 11.2) with the native  $0.5^{\circ}$  x  $0.667^{\circ}$  horizontal resolution and 47 vertical layers from surface pressure to 0.01 hPa over East Asia  $(11^{\circ}\text{S}-55^{\circ}\text{N}, 70-150^{\circ}\text{E})$  (Wang, McElroy, Jacob, & Yantosca, 2004). Boundary conditions for the nested version of the model were provided hourly from the global GEOS-Chem simulations with  $4^{\circ}$  x  $5^{\circ}$  horizontal resolution. All model simulations were performed by using observed meteorological data and a surface  $CO_2$  flux dataset. We used the 3-hourly meteorological data from Modern-Era Retrospective analysis for Research and Applications version 2 (MERRA-2) datasets with the same spatial resolutions as each simulation (Gelaro, et al., 2017). The surface

carbon flux dataset includes 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, 2001), shipping, and aviation (Simone, Stettker, & Barrett, 2013). From the carbon flux dataset, monthly terrestrial carbon fluxes and fossil fuel CO<sub>2</sub> emissions were taken from CarbonTracker outputs (CT2017; Peters et al. 2007 with updates documented at http://carbontracker.noaa.gov) and the Open-source Data Inventory for Anthropogenic CO<sub>2</sub> (ODIAC) dataset (Oda & Maksyutov, 2011), respectively.

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

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Table 1 Set of model simulation to estimate the influences of changes in regional land-surface carbon fluxes and transport on CO<sub>2</sub> concentrations over South Korea for the period 2000–2016.

Simulations	Descriptions		
	Terrestrial carbon flux	Fossil Fuel carbon emissions	
E1	Т	Т	
E2	South Korea FIX	Т	
E3	Т	South Korea FIX	
E4	South Korea FIX	South Korea FIX	
E5	Т	Eastern China FIX	
T: all transien			

FIX: flux in 2000 were repeatedly prescribed over the region.

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In the case of terrestrial carbon fluxes, we conducted an additional set of model simulations using TRENDY model results. In the T1 simulation (i.e. control simulation), balanced monthly terrestrial carbon flux (from SiB3 model; Messerschmidt et al., 2013) with climatological value of annual terrestrial exchange (from TransCom models; Baker et al., 2006) were repeatedly prescribed during the entire simulation period; there are no annual changes in the seasonal cycle of the terrestrial carbon fluxes. In T2, T3, and T4 simulations (i.e. sensitivity simulations), the monthly flux in South Korea is set to linearly change from the balanced 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 change in NBP on the CO<sub>2</sub> concentrations, estimated by TRENDY models, are obtained from the differences of T2, T3, and T4 simulations

with the T1 simulation,

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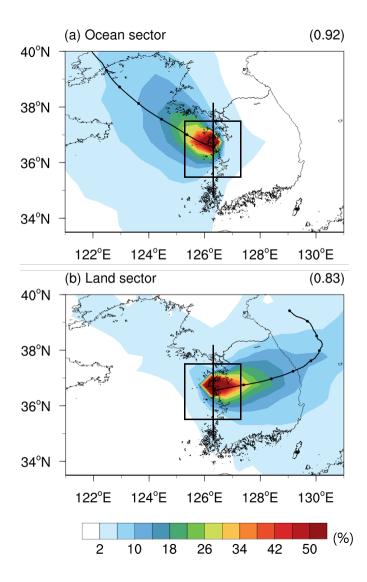
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### Results

Comparison of the back-trajectories of air masses between the land and ocean sectors

Our method for distinguishing the regional influence on atmospheric CO<sub>2</sub> was first verified by back trajectory analysis. Figure 2 shows probability densities of 48-h trajectories ending at the AMY Station at 500 m altitude. The percentage decreases farther away from the station with a value of 100%, which indicates that all trajectories pass the area. Trajectory distributions of the air masses for wind from the ocean sector and land sector explicitly differ. To gather quantitative information, we set a domain centered at the station (35.5–37.5°N, 125.3–127.3°E; boxed area in Figure 2a) and noted which direction the air masses arrived from at the domain. 88.3% of the air mass for wind from the ocean sector enters the AMY Station from the ocean side (Figure 2a). The northwestern airflow is dominant, and it crosses the west coast of the Korean Peninsula. 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 east of the Korean Peninsula. The rest 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 northeast between 36 and 48 hours prior. These trajectory patterns show that our wind direction-based classification is reasonable for separating of air mass into two groups: inflow from outside and inflow from inside the Korean Peninsula (i.e., CO<sub>2</sub>ocean and CO<sub>2</sub>land).



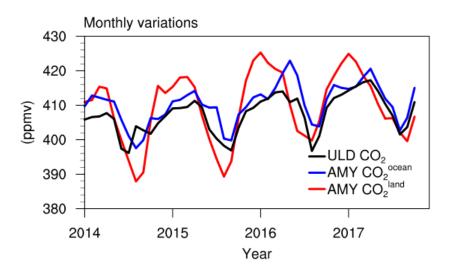
**Figure 2** Density plots for 48-h trajectories terminating at AMY at 500m altitude for wind from (a) the ocean sector and (b) the land sector for the period 1999–2017. Solid line represents the median trajectory. Markers on the trajectories represent the position of air masses at 6-hour intervals. 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 side among the total air mass of wind from the ocean sector and vice versa.

Comparison of the atmospheric CO<sub>2</sub> concentrations between Stations

The possible influences of air coming from the East Sea of Korea were also assessed from

the comparison between CO<sub>2</sub> concentrations at land sector and ULD Station. On average, the CO<sub>2</sub> concentration at the ULD site is lower than both CO<sub>2</sub><sup>land</sup> and CO<sub>2</sub><sup>ocean</sup> by 2.2 and 3.2 ppm from January 2014 to October 2017 (Figure 3), respectively, because the ULD site is located far from the CO<sub>2</sub> source regions. However, the seasonal variability of CO<sub>2</sub><sup>land</sup> is greater than that of the CO<sub>2</sub> concentrations at the ULD site and annual minimum value of the monthly CO<sub>2</sub><sup>land</sup> is lower than that of the ULD site in three of the past four years. For example, in 2014, the annual maximum and minimum value of CO<sub>2</sub><sup>land</sup> are 415.4 and 387.9 ppmv but those of the ULD CO<sub>2</sub> concentration are 407.4 and 396.2 ppmv, respectively. In addition, averaged CO<sub>2</sub><sup>land</sup> from May to September (400.3 ppmv) is lower than that of the ULD CO<sub>2</sub> concentration (403.9 ppmv) by 3.6 ppmv during the analysis period. These results indicate that the variations of CO<sub>2</sub><sup>land</sup> are mainly attributed to the regional land-surface CO<sub>2</sub> fluxes rather than the air coming from the East Sea.



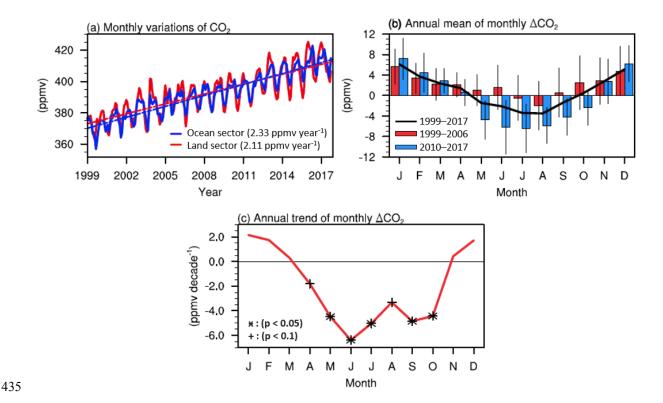


**Figure 3** Monthly variations of atmospheric CO<sub>2</sub> at Ulleungdo station (ULD; black line) and CO<sub>2</sub> concentrations for wind from the ocean sector (CO<sub>2</sub><sup>ocean</sup>; blue line) and land sector (CO<sub>2</sub><sup>land</sup>; red line) at Anmyeondo station (AMY) from January 2014 to October 2017.

## 411 Regional influence on atmospheric $CO_2$ concentration variations

Both time series of  $CO_2^{land}$  and  $CO_2^{ocean}$  at the AMY Station show gradually increasing trends and clear seasonal variations, with the maximum in winter and minimum in summer from January 1999 to October 2017 (Figure 4a). However, the seasonal variability of  $CO_2^{land}$  is greater than that of  $CO_2^{ocean}$ . In comparison with  $CO_2^{ocean}$ , annually averaged  $CO_2^{land}$  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 4b; black thick line). The maximum value of the difference between  $CO_2^{land}$  and  $CO_2^{ocean}$  (i.e.  $\Delta CO_2$ ) is found in January at 6.1 ppmv and the minimum is exhibited in August at -3.5 ppmv.

Notable decreases in annually averaged  $\Delta CO_2$  during the growing season appear in recent years from  $0.5 \pm 2.1$  ppmv in 1999–2006 to  $-5.0 \pm 2.2$  ppmv in 2010–2017 (Figure 4b; bar). Moreover, the annually averaged monthly  $\Delta CO_2$  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  $\Delta CO_2$  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. A statistically significant decreasing trend of the  $\Delta CO_2$  is also observed for the growing season, on average, at 4.75 ppmv decade<sup>-1</sup> (p < 0.05; Figure 4c). The maximum decreasing trend is exhibited in May at 6.39 ppmv decade<sup>-1</sup> (p < 0.05) and the minimum is found in April at 1.81 ppmv decade<sup>-1</sup>. An increase in the  $\Delta CO_2$  is observed from November to March at 1.27 ppmv decade<sup>-1</sup> but this is not significant. The results suggest that the increase in atmospheric  $CO_2$  concentration over South Korea is relatively lower than that in the surrounding ocean regions, especially during the growing season for the last two decades.



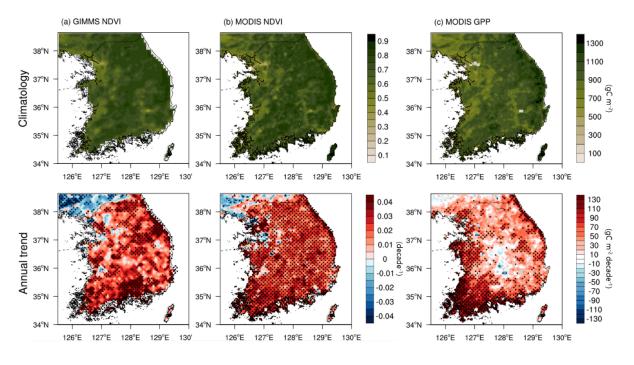
**Figure 4** Variations of (a) monthly mean CO<sub>2</sub> concentrations for wind from the ocean sector  $(CO_2^{\text{ocean}}; \text{ blue line})$  and land sector  $(CO_2^{\text{land}}; \text{ red line})$  for the period 1999–2017. (b) Annually averaged monthly  $\Delta CO_2$  for the entire study period (black thick line), the first eight years (1999–2006; red bar), and the last eight years (2008–2017; blue bar). Error bar shows the monthly standard derivation of the  $\Delta CO_2$ . (c) Trends of monthly mean  $\Delta CO_2$  for the same period. The asterisk and cross indicate statistical significance at the 95% and 90% confidence level, respectively.

Changes in vegetation and terrestrial carbon fluxes

To understand the reasons for the observed decreases in  $\triangle CO_2$  during the growing season, we investigated the changes in vegetation and terrestrial carbon fluxes over South Korea for the last two decades. South Korea, where two thirds of the country are forests (Korea Forest

Service, 2018), has a climatological NDVI of 0.69 (GIMMS: 0.69, MODIS: 0.69; Figure 5a, b) and an accumulated GPP of 919 gC m<sup>-2</sup> during the growing season (Figure 5c). Both linear trends of NDVI from the two different satellite measurements show nationwide positive values (Figure 5a, b). For the growing season, the NDVI from GIMMS increased by 0.030 decade<sup>-1</sup> over 90.2% of South Korea from 1999 to 2015; this accounts for a 4.3% increase. Similarly, the NDVI from MODIS increased 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% increase. The greening trends of NDVI from GIMMS and MODIS are significant at the 95% confidence level in 34.0% and 75.1% of the study area, respectively. In line with the observed nationwide vegetation greening trend, accumulated GPP from MODIS increased by 65.1 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 7.6% increase (Figure 5c). Some parts of central South Korea show decreases or relatively small increases in the GPP, but a significant increase in the GPP is observed in 45.4% of the analysis domain, especially over the southwestern part of South Korea.





**Figure 5** Spatial distributions of climatology and trends 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.

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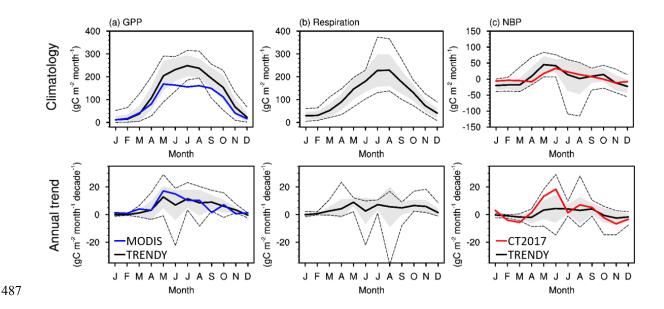
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The TRENDY multi-model mean overestimated the climatology of the annual GPP average of the study area as 350 gC m<sup>-2</sup>, but it estimated an increase in GPP for the growing season by  $54.8 \pm 27.5$  gC m<sup>-2</sup> decade<sup>-1</sup>, similar to the GPP from MODIS (61.4 gC m<sup>-2</sup> decade<sup>-1</sup> <sup>1</sup>) for 2000–2016 (Figure 6a). 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) increased by  $18.5 \pm 24.0$  gC m<sup>-2</sup> decade<sup>-1</sup> during the growing season; this accounts for  $14.2 \pm 18.5\%$  of its climatological value ( $125 \pm 105$  gC m<sup>-2</sup>) (Figure 6b, c). Even though large variations between the TRENDY models exist from -48.7 to 101.3 gC m<sup>-2</sup> decade<sup>-1</sup> (this accounts for 39.0% of decrease and 81.0% of 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; this accounts for 45.5% of its climatological value (94.2 gC m<sup>-2</sup>). Both TRENDY and CT2017 models estimate a relatively lower increasing trend of the carbon flux in July than adjacent months. This is due to the stagnant rain front created during summer rainy seasons which blocks solar radiation and inhibits vegetation growth over South Korea (Lee et al., 2017). These results suggest that the terrestrial carbon uptake in South Korea has increased during the growing season due to the nationwide vegetation greening trend for the last two decades.



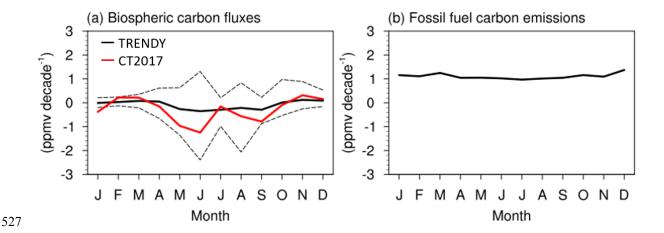
**Figure 6** Area-averaged climatology and trends in monthly (a) gross primary production (GPP), (b) total respiration (i.e. autotrophic respiration + heterotrophic respiration), and (c) net biome production (NBP) in South Korea according to simulations from TRENDY models (black line) over the period 2000–2016. The shading denotes one inter-model standard deviation range of each month. The dotted lines represent the minimum and maximum values of monthly trends in the carbon fluxes estimated from TRENDY models. Blue and red lines indicate GPP from MODIS and biosphere carbon flux from CT2017, respectively.

*Influences of changes in regional land-surface carbon fluxes on CO2 concentrations* 

We assessed the influence of model-estimated changes in regional terrestrial carbon fluxes on CO<sub>2</sub> concentrations in South Korea (35–38°N, 126.5–129.5°E) from the CTM simulations for the period 2000–2016. The increasing terrestrial carbon fluxes estimated in both CT2017 and TRENDY multi-model mean consistently induced decreases in CO<sub>2</sub> concentrations in South Korea during the growing season, distinct from the non-growing season (Figure 7a). The flux change estimated by the CT2017 decreased CO<sub>2</sub> concentration by

0.64 ppmv decade<sup>-1</sup> during the growing season and caused little change in the  $CO_2$  concentration by 0.06 ppmv decade<sup>-1</sup> during the non-growing season (Figure 7a; red line). The maximum decreasing trend appeared in June at 1.38 ppmv decade<sup>-1</sup>, likewise with the  $\Delta CO_2$  trend. Even though the flux change estimated by the TRENDY multi-model mean induces a relatively small decrease in the  $CO_2$  concentration during the growing season (0.24 ppmv decade<sup>-1</sup>) than that from CT2017, the models estimated that the  $CO_2$  concentration would decrease at most by 1.38 ppmv decade<sup>-1</sup> (Figure 7a; black line). A possible maximum decreasing trend of the  $CO_2$  concentration is also exhibited in June at 2.39 ppmv decade<sup>-1</sup>. The results suggest that the observed decreasing trend of  $\Delta CO_2$  is associated with the enhancement of terrestrial carbon uptake in South Korea 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 South 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>) is greater than that 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; this accounts for around 12% of the annual mean trends. The possible influence of the increasing carbon emissions was also evaluated. Our model estimates that the rise of domestic carbon emissions increases annual mean CO<sub>2</sub> concentrations in South Korea by 1.11 ppmv decade<sup>-1</sup> (Figure 7b). 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.



**Figure 7** Area-averaged changes in trends in monthly mean CO<sub>2</sub> concentrations due to (a) regional terrestrial carbon fluxes (red solid line: CT2017; E1 simulation minus E3 simulation, black solid and dotted lines: average, maximum, and minimum of TRENDY models' estimates; T2, T3, and T4 simulations minus T1 simulation, respectively) and (b) fossil fuel carbon emissions (i.e., E1 simulation minus E2 simulation) at the South Korea for the period 2000–2016.

Influences of atmospheric transport changes on CO<sub>2</sub> concentrations over South Korea

South 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 (i.e. Siberia). Considering the geographical location, the influences of transport changes on CO<sub>2</sub> concentrations over South Korea are also evaluated through model simulations. Even without considering the land-surface carbon flux changes in the region, annual mean CO<sub>2</sub> concentrations increase 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 8a). The spatial distributions of the increasing trends are 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 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 South Korea by more than 6 ppmv decade<sup>-1</sup> (Figure 8b). We compared the influences of transport change on the trends of monthly mean CO<sub>2</sub> concentrations in the surrounding area (i.e. West Sea of Korea) 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).

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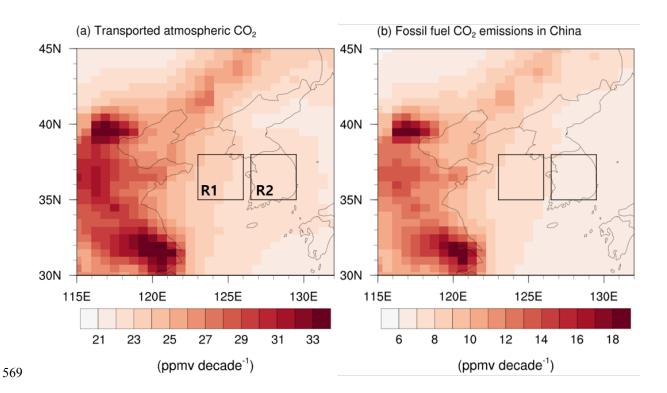
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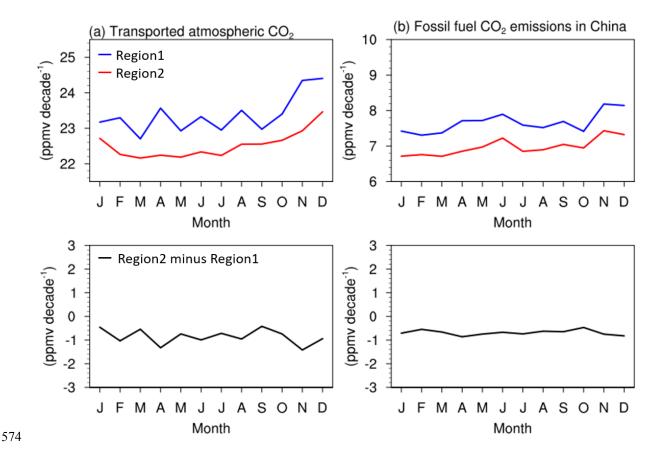
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The changes in atmospheric CO<sub>2</sub> transport raise the annual mean CO<sub>2</sub> concentration in Region1 more than that of in Region2 by 0.86 ppmv decade<sup>-1</sup> (Figure 9a). More than 1.3 ppmv decade<sup>-1</sup> of the regional differences in CO<sub>2</sub> concentration trends (i.e. Region2 minus Region1) are exhibited in April and November, and less than 0.5 ppmv decade-1 of the regional differences are found in August. The annual mean trends of the regional differences are mostly resulted from the increasing carbon emissions in eastern China (0.69 ppmv decade<sup>-1</sup>; Figure 9b). 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 South 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 that of during the non-growing season (0.95 ppmv decade<sup>-1</sup>). These results suggest that the observed decreasing trend in  $\triangle CO_2$  during the growing season, distinct during the nongrowing season, is not attributed to the changes in transported atmospheric CO<sub>2</sub> for the study period.



**Figure 8** Spatial distributions of changes in simulated annual mean CO<sub>2</sub> concentration trends due to (a) transported atmospheric CO<sub>2</sub> (i.e. E5 simulation) and (b) China's carbon emissions (i.e., E1 simulation minus E4 simulation) for the period 2000–2016.



**Figure 9** Area-averaged changes in simulated monthly mean CO<sub>2</sub> concentration trends due to (a) transported atmospheric CO<sub>2</sub> (i.e. E5 simulation) and (b) China's carbon emissions (i.e., E1 simulation minus E4 simulation) at the Region1 (35–38°N, 124.3–126.3°E), the Region2 (35–38°N, 126.3–128.3°E), and the difference between them.

Discussion

The best way to understand the terrestrial carbon cycle would be to use as much terrestrial carbon flux measurements as possible. However, the number of terrestrial carbon flux measurements are still limited in many parts of the globe (Schimel et al., 2015). Recent studies suggest the applicability of long-term measurements of atmospheric CO2 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 South Korea. However, unlike the clean-air environment in Alaska where previous studies have been carried out (i.e., Commane et al., 2016; Jeong et al., 2018), South 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>ocean</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 of CO<sub>2</sub> concentrations cannot be negligible over the region as highlighted in previous modeling (e.g. Ballav et al., 2012; Ballav et al., 2016) and observation studies (e.g. Umezawa et al., 2018).

In this study, the changes in regional land-surface  $CO_2$  fluxes were estimated from the difference between  $CO_2^{\text{ocean}}$  and  $CO_2^{\text{land}}$ . It is worth noting that Ballav et al. (2016) showed that the relatively high  $CO_2$  concentration over land the night before was sometimes observed over the open ocean region the next day mostly when wind blows from land to sea; thus, higher  $CO_2^{\text{land}}$  could be observed in those cases. However, our results show that  $CO_2^{\text{land}}$  has greater seasonal variability than that of both  $CO_2^{\text{ocean}}$  (Figure 4a) and the ULD  $CO_2$  concentrations (Figure 3) and clear seasonal variations in  $\Delta CO_2$  appeared (Figure 4b). This indicates that  $\Delta CO_2$  can capture land-surface  $CO_2$  flux signals that are distinct from the surrounding area (i.e. the East Sea and West Sea of Korea).

Based on the observed  $\triangle CO_2$ , we found that regional land carbon uptake significantly increases relative to the surrounding 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 increase in terrestrial carbon fluxes for the period and checked the changes in vegetation and terrestrial carbon fluxes using available satellite datasets and model simulation results. Our analysis of the satellite dataset showed that significant nationwide increasing trends in vegetation and its photosynthesis appeared 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 South Korea increased by 142% from 1999 to 2015 is another good evidence for the increase in vegetation production (Korea Forest Service, 2018). Generally, major reasons for the increase in vegetation growth is 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 trends in the NDVI between North and South Korea are observed in Figure 5. 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, Teplyakov, & Adams, 2014). The different forest management history caused a vegetation browning trend in the southern area of North Korea despite the favorable environmental conditions for vegetation growth.

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In accordance with the observed increasing trend of vegetation growth and 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 S3 simulation results considering all changes to climate, CO<sub>2</sub>, and land use, but found that there were little differences in the S2 simulation results, 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 CO2 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 (i.e. TRENDY) seem to estimate the increasing trends less than that of the inverse model constrained CO<sub>2</sub> measurements data (i.e. CT2017). Although our CTM model simulations have relatively weaker responses to the estimated terrestrial carbon flux changes, they show that the observed ΔCO<sub>2</sub> decrease is associated with the enhancement of terrestrial carbon uptake in South 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 an 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 are not treated as minimal components.

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This study suggests that increase in terrestrial carbon uptake is a key process causing changes in temporal variations of  $\triangle CO_2$ . However,  $\triangle CO_2$  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 increase in domestic fossil fuel carbon emissions do not make notable seasonal differences in the  $CO_2$  concentration trends in South Korea. That is because seasonal variability of the carbon emissions trends is relatively too small than that of terrestrial carbon fluxes, which

change its 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 is more than 40 times of that in South Korea, has potential to make around 1 ppmv decade<sup>-1</sup> annual amplitude of increasing rates of CO<sub>2</sub> concentrations over South Korea. However, because the seasonal patterns of CO<sub>2</sub> concentration trends are similar around and in the Korean Peninsula, China's emissions also seem to be a minor factor on the long-term trends in the seasonal cycle of  $\triangle CO_2$ . One thing to keep in mind is that the greater increasing rate of CO<sub>2</sub> concentrations in the West Sea of Korea than the Korean Peninsula could alleviate the ΔCO<sub>2</sub> increase caused by the increasing domestic carbon emissions. Readers need to be cautious when interpreting our results (i.e. decrease in annual ΔCO<sub>2</sub> trends does not mean that South 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 South Korea. Our model suggests, however, that there are no notable changes in atmospheric circulation patterns that lead to the observed  $\triangle CO_2$  trend for the study period. This study treated the anthropogenic emissions and atmospheric circulation as main drivers of transport changes, but 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 biospheric CO<sub>2</sub> flux appeared in boreal regions at 0.35% year-1 and in arctic regions at 0.85 % year-1 for the period 1982–2012 from inversion results. We evaluated the possible effect through a set of ideal model experiments with and without a 1% year-1 increase in seasonal amplitude of monthly terrestrial CO<sub>2</sub> fluxes in high-latitude regions (>45°N) for the study period. The model estimated that the increasing biospheric carbon uptake increases the seasonal amplitude of CO<sub>2</sub> concentration

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around 1.6 ppmv decade<sup>-1</sup> over South Korea, but it also did not make remarkable regional differences in the CO<sub>2</sub> concentration trends around the region (not shown here).

This study takes the advantages of the long-term continuous  $CO_2$  measurement at the AMY Station. In addition to the AMY Station, numerous surface stations are continuously measuring atmospheric  $CO_2$  concentration over the globe and provides hourly data. Because many of these observation sites are located on the coastal side, 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 (3397m) in the center of the Hawaiian Islands, observes  $CO_2$  concentrations above the planetary boundary layer, but  $CO_2$  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 of observed  $CO_2$  concentration between the two periods could play the role of  $\Delta CO_2$  of this study. In addition, local variations in land carbon flux within the continent could be detected from a comparison of atmospheric  $CO_2$  data from adjacent observation sites (e.g. Sargent et al., 2018).

In summary, this study showed that the terrestrial carbon uptake has been enhanced resulted from the nationwide vegetation greening over South 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 (e.g. 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  $CO_2$  concentrations. Inverse modeling using the added in situ, airborne, and satellite data would provide more reliable quantitative information on the increase in terrestrial carbon uptake by using the relationship between our results (i.e.  $\Delta CO_2$ ) and the estimated biosphere flux in future studies (e.g. Commane et al., 2016). Atmospheric  $CO_2$  measurements could open up 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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