



## Article

# Detecting the Responses of CO<sub>2</sub> Column Abundances to Anthropogenic Emissions from Satellite Observations of GOSAT and OCO-2

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**Abstract:** The continuing increase in atmospheric CO<sub>2</sub> concentration caused by anthropogenic CO<sub>2</sub> emissions significantly contributes to climate change driven by global warming. Satellite measurements of long-term CO<sub>2</sub> data with global coverage improve our understanding of global carbon cycles. However, the sensitivity of the space-borne measurements to anthropogenic emissions on a regional scale is less explored because of data sparsity in space and time caused by impacts from geophysical factors such as aerosols and clouds. Here, we used global land mapping column averaged dry-air mole fractions of CO<sub>2</sub> (XCO<sub>2</sub>) data (Mapping-XCO<sub>2</sub>), generated from a spatio-temporal geostatistical method using GOSAT and OCO-2 observations from April 2009 to December 2020, to investigate the responses of XCO<sub>2</sub> to anthropogenic emissions at both global and regional scales. Our results show that the long-term trend of global XCO<sub>2</sub> growth rate from Mapping-XCO<sub>2</sub>, which is consistent with that from ground observations, shows interannual variations caused by the El Niño Southern Oscillation (ENSO). The spatial distributions of XCO<sub>2</sub> anomalies, derived from removing background from the Mapping-XCO<sub>2</sub> data, reveal XCO<sub>2</sub> enhancements of about M due to anthropogenic emissions and seasonal biomass burning in the wintertime. Furthermore, a clustering analysis applied to seasonal XCO<sub>2</sub> clearly reveals the spatial patterns of atmospheric transport and terrestrial biosphere CO<sub>2</sub> fluxes, which help better understand and analyze regional XCO<sub>2</sub> changes that are associated with atmospheric transport. To quantify regional anomalies of CO<sub>2</sub> emissions, we selected three representative urban agglomerations as our study areas, including the Beijing-Tian-Hebei region (BTH), the Yangtze River Delta urban agglomerations (YRD), and the high-density urban areas in the eastern USA (EUSA). The results show that the XCO<sub>2</sub> anomalies in winter well capture the several-ppm enhancement due to anthropogenic CO<sub>2</sub> emissions. For BTH, YRD, and EUSA, regional positive anomalies of  $2.47 \pm 0.37$  ppm,  $2.20 \pm 0.36$  ppm, and  $1.38 \pm 0.33$  ppm, respectively, can be detected during winter months from 2009 to 2020. These anomalies are slightly higher than model simulations from CarbonTracker-CO<sub>2</sub>. In addition, we compared the variations in regional XCO<sub>2</sub> anomalies and NO<sub>2</sub> columns during the lockdown of the COVID-19 pandemic from January to March 2020. Interestingly, the results demonstrate that the variations of XCO<sub>2</sub> anomalies have a positive correlation with the decline of NO<sub>2</sub> columns during this period. These correlations, moreover, are associated with the features of emitting sources. These results suggest that we can use simultaneously observed NO<sub>2</sub>, because of its high detectivity and co-emission with CO<sub>2</sub>, to assist the analysis and verification of CO<sub>2</sub> emissions in future studies.

**Keywords:** Mapping XCO<sub>2</sub>; anthropogenic emission; GOSAT; OCO-2; NO<sub>2</sub> columns



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## 1. Introduction

Global atmospheric CO<sub>2</sub> concentration continues to increase by 2–3 ppm per year, which contributes significantly to global warming [1,2]. Changes in atmospheric CO<sub>2</sub> concentrations are primarily driven by emissions from human activities, photosynthesis of natural terrestrial ecosystems, and biogeochemical processes in the ocean. To achieve the goal of curbing global warming proposed by the Paris Agreement in 2015, many countries put forward the strategy of carbon neutrality. They are committed to limit global average temperature rise to be below 1.5° above pre-industrial levels through different effective ways of reducing greenhouse gas emissions [3,4]. To achieve these goals, it is critical to investigate the spatio-temporal changes of atmospheric CO<sub>2</sub> concentration and detect the influence mechanism of human activities in various regions on atmospheric CO<sub>2</sub> variations, so as to provide a basis for governments to evaluate the effects of CO<sub>2</sub> emission reduction measures.

Satellite measurements from GOSAT and OCO-2 have delivered the column-averaged dry air mole fractions of CO<sub>2</sub> (XCO<sub>2</sub>) data for more than 12 years, which provide data for studying long-time variations of global and regional carbon emissions [5–11]. It has become an effective data source for understanding the contributions of natural ecosystems and human activities to the increase of atmospheric CO<sub>2</sub> concentration. For example, using satellite XCO<sub>2</sub> observations from GOSAT and OCO-2, many studies have found that extreme climate related to the El Niño Southern Oscillation (ENSO) disturbs the interannual increase of atmospheric CO<sub>2</sub> concentration at global and regional scales [12–16]. Abnormal increase in CO<sub>2</sub> concentration mostly occurs in natural vegetation areas. The detection and attribution analysis of extreme CO<sub>2</sub> changes show that CO<sub>2</sub> anomalies are related to the abnormal carbon emissions from terrestrial ecosystems caused by extreme climate [17]. Previous studies using CO<sub>2</sub> data from model and ground observations also showed consistent results with that from satellite observations [18–22].

XCO<sub>2</sub> enhancements could be detected by satellite observations in large cities, power plants, volcanoes, and fire emissions. By differencing the observations over a megacity with those in the nearby background, XCO<sub>2</sub> enhancements can be derived. The enhancement is found to be more than 3 ppm in large cities, such as Beijing-Tianjin-Hebei areas and the Yangtze River Delta in China, the Los Angeles megacity in the USA, the Seoul Metropolitan area in South Korea, and Mumbai in India [23–29]. XCO<sub>2</sub> observations from OCO-2 have also been used to identify enhanced plume signals and estimate anthropogenic emissions from individual point sources such as power plants and volcanoes [30–32]. For Australian mega bushfires, fire-induced XCO<sub>2</sub> enhancement detected by three orbits of observations from OCO-2 during November–December in eastern Australia is approximately 1.5 ppm [33]. Global XCO<sub>2</sub> anomalies derived from satellite observations agree well with the spatial patterns of emission inventories and model simulations [34–36]. Furthermore, an assessment combining satellite XCO<sub>2</sub> observations and other relatively short-lived pollutants (e.g., CO and NO<sub>2</sub>) in cities found that urban CO<sub>2</sub> enhancements have a good correlation with air pollutants, which can be used to evaluate emission characteristics, such as combustion efficiency [36–38]. These results indicate that satellite XCO<sub>2</sub> observations have the detectability of natural and anthropogenic CO<sub>2</sub> emissions. Combined with ground-based measurements, they provide reliable data sources for constraining anthropogenic emission estimates and verifying bottom-up inventories.

However, previous studies on the detectivity of using satellite XCO<sub>2</sub> observations for anthropogenic emissions still have some limitations. Due to the impact of data sparsity in space and time caused by impacts from geophysical factors such as aerosols and clouds, most existing studies focus on individual areas or specific events, but lack sufficient analysis at global and regional scales. In response to this problem, we generated a dataset of global land mapping XCO<sub>2</sub> data (Mapping-XCO<sub>2</sub>) using GOSAT and OCO-2 observations. With these global spatio-temporal continuous XCO<sub>2</sub> data, this study is able to fully explore the changes of XCO<sub>2</sub> enhancements caused by anthropogenic emission at both global and regional scales.

In this paper, we investigate global XCO<sub>2</sub> variations in space and time, and analyze spatial patterns of seasonal XCO<sub>2</sub> changes affected by atmospheric transport and terrestrial biosphere. We further focus on urban agglomerations with high anthropogenic emissions and quantify the responses of regional XCO<sub>2</sub> to anthropogenic emissions. Our study aims to provide global spatial and temporal analysis of XCO<sub>2</sub> changes and quantify the responses of regional XCO<sub>2</sub> enhancements to anthropogenic emission using long-term mapping data generated from satellite XCO<sub>2</sub> observations.

## 2. Materials and Methods

### 2.1. Materials

#### 2.1.1. CO<sub>2</sub> Datasets

We use the global land mapping XCO<sub>2</sub> dataset (Mapping-XCO<sub>2</sub>) from April 2009 to December 2020, which has a spatial grid resolution of 1° latitude by 1° longitude and temporal resolution of 3 days. The dataset is produced by applying a spatio-temporal geostatistical method to satellite XCO<sub>2</sub> retrievals from GOSAT observations (from April 2009 to August 2014) and OCO-2 observations (from September 2014 to December 2020). The XCO<sub>2</sub> retrievals are the latest ACOS level 2 Lite data product (v9r) and the latest level 2 lite data product (v10r) for OCO-2 [7,10,11]. These products are both obtained from the Goddard Earth Science Data Information and Services Center (GES DISC) [39]. The workflow chart of mapping gridded XCO<sub>2</sub> data are illustrated in Figure A1, including the following key steps: (1) We adjusted the differences in XCO<sub>2</sub> retrievals induced by the *a priori* CO<sub>2</sub> profile and different overpass time using CO<sub>2</sub> profiles from CarbonTracker as reference data. Spatial and temporal scales of satellite observations are integrated to a uniform unit by averaging XCO<sub>2</sub> within 10.5 km and 3 days. (2) The global land is divided into different continental regions and processed separately. In each mapping region, the spatio-temporal correlation structures of the integrated XCO<sub>2</sub> data are assumed to be homogeneous and locally stationary. The spatio-temporal empirical variogram in each region was calculated after removing the spatial and temporal trend from the integrated XCO<sub>2</sub>. (3) Based on these variogram models, space-time kriging with moving cylinder kriging neighborhood was implemented to estimate the XCO<sub>2</sub> value at the center of 1° grids. A detailed description of the gap-filling method is referred to Zeng et al. [40–43] and He et al. [43]. We calculated estimation uncertainty for each grid according to the method described in Zeng et al. [42], which shows that the estimation uncertainty of Mapping-XCO<sub>2</sub> is less than 1.5 ppm on average. Compared to TCCON data, the overall bias of Mapping-XCO<sub>2</sub> obtained by ±0.5° box centered at the TCCON sites is  $0.16 \pm 1.19$  ppm.

Table 1 gives a summary of Mapping-XCO<sub>2</sub> and the comparisons with model simulations by CarbonTracker and ground-based observations from the World Data Centre for Greenhouse Gases (WDCGG). CarbonTracker simulates global atmospheric CO<sub>2</sub> mole fractions from a combination of CO<sub>2</sub> surface exchange models and an atmospheric transport model driven by meteorological fields [44]. We collected CO<sub>2</sub> data at the local time of 13:30 from CT2019B for comparison analysis with spatio-temporal variations of Mapping-XCO<sub>2</sub>. The dataset has a resolution of 3° × 2° grid in space and 1 day in time. In order to analyze long-term trends derived from Mapping-XCO<sub>2</sub>, we collected global analysis data of atmospheric CO<sub>2</sub> concentrations and rates of change from WDCGG. The data are produced based on ground observations from the WMO Global Atmosphere Watch (GAW) in situ observational network. Globally averaged CO<sub>2</sub> mole fractions and CO<sub>2</sub> trends cover the period of 1984–2019, with the growth rates range from 1985 to 2018. These data are also reported by the annual WMO Greenhouse Gas Bulletin [2].

**Table 1.** The products of CO<sub>2</sub> data from satellite observation, model, and ground observation.

Dataset	Description	Reference/Data Source
Mapping-XCO <sub>2</sub>	Global land mapping XCO <sub>2</sub> dataset produced by applying spatio-temporal geostatistics on GOSAT and OCO-2 observations from April 2009 to September 2020. The dataset is regularly distributed with a temporal interval of 3 days and spatial interval of 1° grid.	GES DISC [39] Zeng et al. [40–43]
CT-XCO <sub>2</sub>	The model XCO <sub>2</sub> data at the local time 13:30 (LST) from CT 2019B from 2009 to March 2019 in 3° × 2° grids with a temporal interval of 1 day.	NOAA [45]
WDCGG-CO <sub>2</sub>	Global CO <sub>2</sub> analysis based on ground-based observations, covering from 1984 to 2019 for global monthly mean concentrations and trends and from 1985 to 2018 for growth rates.	WDCGG [46]

### 2.1.2. Auxiliary Datasets

To analyze the mechanism of XCO<sub>2</sub> changes and its response to anthropogenic emissions, we collected various auxiliary datasets to compare with XCO<sub>2</sub> variations at global and regional scales.

The Open-source Data Inventory for Anthropogenic CO<sub>2</sub> (ODIAC) is used to evaluate high emission areas, which can potentially be detected by satellite observed XCO<sub>2</sub> data. ODIAC is a global gridded emission product based on spatial and temporal disaggregation of country scale emissions [47,48]. The latest version of ODIAC emission data product (ODIAC 2020B) provides monthly CO<sub>2</sub> emissions from 2000 to 2019, including two different spatial resolutions of 1° × 1° and 1 km × 1 km. CO<sub>2</sub> emission estimates of the product are based on the latest country fossil fuel CO<sub>2</sub> emission estimates made by the new Carbon Dioxide Information Analysis Center (CDIAC) team from 2000 to 2017 and its projection using fuel consumption data reported by the BP Statistical Review of World Energy in 2018 and 2019 [49]. We downloaded ODIAC data from 2009 to 2019 from the Center for Global Environmental Research, National Institute for Environmental Studies (CGER-NIES) [49].

We used two ENSO indices, including the Southern Oscillation Index (SOI) and the Oceanic Niño Index (ONI), to analyze the fluctuating response of the global CO<sub>2</sub> growth rate to ENSO events. The indices are both provided by the Physical Sciences Laboratory at the National Oceanic and Atmospheric Administration (NOAA). The SOI is defined as the normalized pressure difference between Tahiti and Darwin based on the method developed by Ropelewski and Jones [50]. The data are obtained from the Climate Research Unit [51]. The ONI is a three-month running mean of sea surface temperature (SST) anomalies in the El Niño region (5°N–5°S, 120°W–170°W). The data are obtained from the NOAA Climate Prediction Center [52].

In order to evaluate the latitudinal zonal pattern of seasonal XCO<sub>2</sub> changes revealed by the satellite XCO<sub>2</sub> data, we compared it with the spatial patterns of potential temperature, which acts as a dynamical tracer of transport of the air masses [53]. Potential temperature is most frequently used in atmospheric sciences because it is not affected by the physical lifting or sinking associated with flow over obstacles or large-scale atmospheric turbulence [26,27,54]. Lines of constant potential temperature are natural flow pathways that are largely horizontal near the surface, and it is tightly correlated with CO<sub>2</sub> in simulations with zonally uniform surface fluxes [53]. In this paper, we used the potential temperature at 1000 hPa and calculated the averaged contours during the period from 2009 to 2020. The potential temperature data are monthly means produced by the NCEP/NCAR reanalysis. The online website is <http://www.esrl.noaa.gov/psd/cgi-bin/data/composites/printpage.pl> (accessed on 15 June 2021).

To analyze the influence of the terrestrial ecosystem on the global carbon cycle, we collected the Normalized Difference Vegetation Index (NDVI) data and the land cover type derived from the Moderate Resolution Imaging Spectroradiometer (MODIS) observation. These datasets are downloaded from the website <https://ladsweb.modaps.eosdis.nasa.gov/> (accessed on 8 March 2021). NDVI data from the MOD13C2 product have temporal

and spatial resolutions of  $0.05^\circ$  and 2 days, respectively [55]. We calculated global monthly mean data with  $1^\circ$  resolution from 2009 to 2020, which are used for correlation analysis with seasonal XCO<sub>2</sub> changes from Mapping-XCO<sub>2</sub>. The land cover type is from the MCD12C1 product. We used the land cover type of the International Geosphere Biosphere Programme (IGBP) scheme, which includes 11 natural vegetation classes, 3 developed and mosaicked land classes, and 3 non-vegetated land classes. For regional studies, the land cover type is classified into urban, croplands, vegetation, and other.

NO<sub>2</sub> is a short-lived gas mostly co-emitted from fossil fuel combustion by industries and vehicles. It has been shown to be a good tracer for anthropogenic CO<sub>2</sub> emissions [36–38]. NO<sub>2</sub> data used in our studies is the level 3 offline NO<sub>2</sub> data product derived from TROPOMI/Sentinel-5 Precursor observations [56,57]. The data product provides the total vertical column of NO<sub>2</sub> concentrations with temporal and spatial resolutions of 2 days and  $0.01^\circ$  grid, respectively. The dataset is delivered by the European Space Agency (ESA) and publicly available on Google Earth Engine [57,58]. We obtained regional NO<sub>2</sub> columns in the study areas from July 2018 to December 2020 to assist the analysis of regional emission characteristics.

## 2.2. Methods

### 2.2.1. Calculation of Global Temporal XCO<sub>2</sub> Variations Using Mapping-XCO<sub>2</sub>

The time series of atmospheric XCO<sub>2</sub> is basically a combination of three signals: a long-term trend, a seasonal cycle, and short-term variations [59]. To extract the temporal characteristics of XCO<sub>2</sub> variations, the most common method is to assume that the long-term trend and seasonal cycles can be represented by a polynomial function and a sum of seasonal harmonics, respectively [17,42,60–62]. As shown in Equation (1), we applied curve fitting to global gridded XCO<sub>2</sub> from Mapping-XCO<sub>2</sub> using a linear least squares regression method:

$$f(t) = a_0 + a_1t + a_2t^2 + \sum_{i=1}^4 (\beta_i \sin(i\omega t) + \gamma_i \cos(i\omega t)) \quad (1)$$

$$XCO_2 = f(t) + \delta, \quad (2)$$

where  $f(t)$  is the fitting result,  $t$  is the time in a unit of 3 days (122 cycles per year),  $\omega$  is a parameter of the temporal period in yearly harmonics calculated by  $2\pi/122$ . The parameters of  $a_0, a_1, a_2, \beta_i, \gamma_i$  are obtained by least squares fitting. Note that the residuals ( $\delta$ ) between global mapping XCO<sub>2</sub> data and  $f(t)$  in Equation (2) include a part of information on interannual and short-term variations that are not represented by the function. We use a low-pass filter to filter the residuals and obtain the signals of interannual and short-term variations [59,60]. Global monthly XCO<sub>2</sub> and its long-term trend are calculated by combining the fitting part of the function and the filtered part. The growth rate of global XCO<sub>2</sub> is computed by taking the derivative of the long-term trend of XCO<sub>2</sub>.

### 2.2.2. Clustering Spatial Pattern of the Seasonal XCO<sub>2</sub> Cycle

The changes of XCO<sub>2</sub> show a seasonal cycle especially in the Northern Hemisphere, which is affected by CO<sub>2</sub> flux from atmospheric transport and the terrestrial biosphere. The seasonal XCO<sub>2</sub> cycle for each grid is obtained by fitting XCO<sub>2</sub> timeseries of the grid using Equation (1), which also characterizes the long-term trend and a seasonal cycle for each grid. We utilized an unsupervised K-means method to cluster the XCO<sub>2</sub> based on the similarities in its seasonal cycles in order to obtain the spatial pattern of seasonal XCO<sub>2</sub> changes. K-means is an iterative algorithm used to classify the given dataset based on the similarity of temporally changing features where those grids with similar seasonal XCO<sub>2</sub> changes are classified into the same cluster [63]. The temporal variation of XCO<sub>2</sub>, after removing long-term trends for each grid, reflects the biospheric fluxes from vegetation seasonal activities coupled with the atmospheric transport. This clustering method groups those grids with similar temporal variations to the same class. The clustering results are



able to reveal the spatial patterns of atmospheric transport and terrestrial ecosystems' CO<sub>2</sub> uptake.

### 2.2.3. Detecting CO<sub>2</sub> Anomalies at Global and Regional Scales

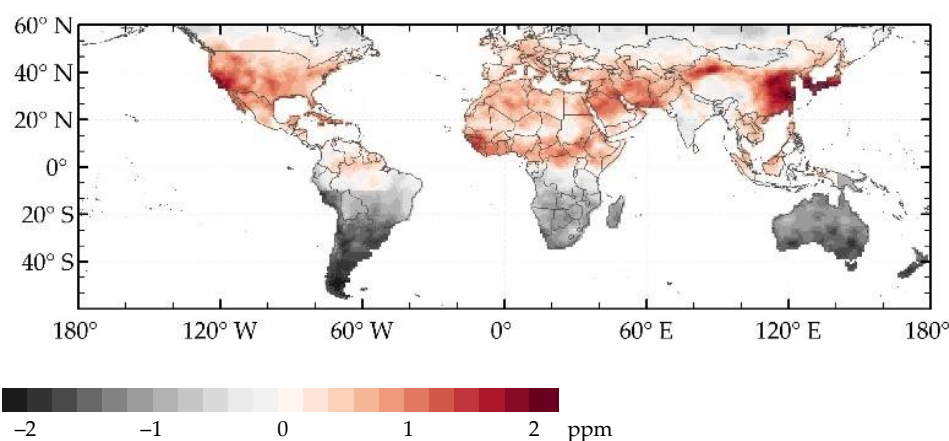
The global atmospheric CO<sub>2</sub> concentration represents a balance of all natural and anthropogenic CO<sub>2</sub> fluxes into and out of the atmosphere. Atmospheric CO<sub>2</sub> is well mixed by turbulent mixing and atmospheric transport [2]. We use global monthly averaged XCO<sub>2</sub> as the global background. Gridded XCO<sub>2</sub> anomalies are calculated as the differences between gridded XCO<sub>2</sub> data and the background, which is hereafter referred to as dXCO<sub>2</sub>. The dXCO<sub>2</sub> is associated with net CO<sub>2</sub> fluxes in the grid. A negative dXCO<sub>2</sub> implies a net sink of CO<sub>2</sub>, while positive dXCO<sub>2</sub> implies a net source relative to global background. The spatial distribution of global gridded dXCO<sub>2</sub> from Mapping-XCO<sub>2</sub> is described in Section 3.1 and is further compared with dXCO<sub>2</sub> from CT-XCO<sub>2</sub> data.

Lastly, we focus on urban agglomerations in China and the USA to demonstrate regional detectivity of CO<sub>2</sub> anomalies induced by anthropogenic emission. The urban agglomerations with high emissions are selected as study areas, which are basically located in the same latitude zone of 25°–45°. In order to remove large-scale CO<sub>2</sub> variations, median XCO<sub>2</sub> in the latitude zone is utilized as a background value. We computed regional XCO<sub>2</sub> anomalies ( $\Delta$ XCO<sub>2</sub>) by subtracting the “background” from regional averaged XCO<sub>2</sub> in the study areas.

## 3. Results

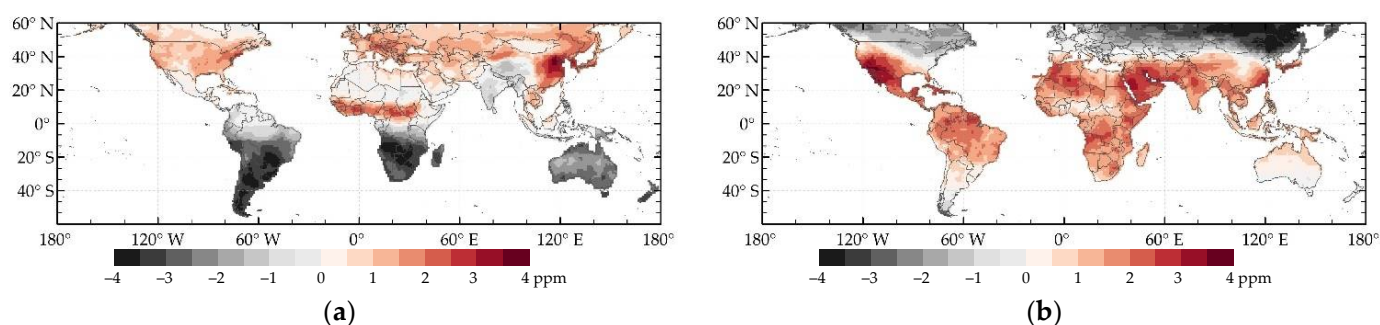
### 3.1. Spatio-Temporal Characteristic of Global XCO<sub>2</sub> Variations and Anthropogenic Emissions

We calculated global gridded anomalies (dXCO<sub>2</sub>) from Mapping-XCO<sub>2</sub> and CT-XCO<sub>2</sub> to analyze global XCO<sub>2</sub> variations in space and time. Figure 1 shows spatial distributions of multi-year averaged dXCO<sub>2</sub> of Mapping-XCO<sub>2</sub> during 2010–2018, which have a similar spatial pattern with that calculated from CT-dXCO<sub>2</sub> in Figure A2. Higher positive dXCO<sub>2</sub> is observed in the region of East Asia, Southeast Asia, Middle East, North America, and North Africa. The dXCO<sub>2</sub> shows a negative value in the Southern Hemisphere. The result from Mapping-XCO<sub>2</sub> is about 0.4 ppm lower than CT-XCO<sub>2</sub> in eastern Asia. However, it shows obvious higher dXCO<sub>2</sub> over the regions of Xinjiang in China and lower dXCO<sub>2</sub> in India. The overall difference of global monthly mean XCO<sub>2</sub> between Mapping-XCO<sub>2</sub> and CT-XCO<sub>2</sub> is  $-0.24 \pm 0.39$  ppm, which is less than the difference of dXCO<sub>2</sub>. Therefore, the differences of global XCO<sub>2</sub> anomalies between Mapping-XCO<sub>2</sub> and CT-XCO<sub>2</sub> are mostly induced by their gridded XCO<sub>2</sub> data. As can be seen in Figure A3, the large difference is mainly distributed in southern Eurasia. This large difference is very likely caused by sparse satellite observations that lead to higher mapping uncertainty, especially between 2010 and 2014.



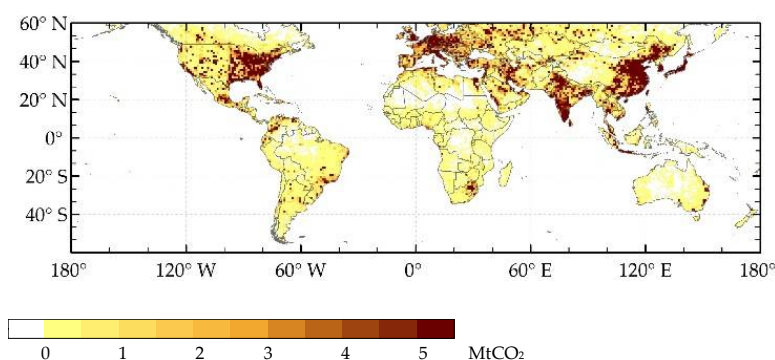
**Figure 1.** Spatial distributions of the averaged dXCO<sub>2</sub> from 2009 to 2018 calculated from Mapping-XCO<sub>2</sub>.

Seasonal  $\Delta\text{XCO}_2$  in winter and summer are computed by averaging  $\Delta\text{XCO}_2$  values during December-January-February (DJF) and June-July-August (JJA), respectively. Figure 2 maps spatial patterns of seasonal  $\Delta\text{XCO}_2$  from Mapping- $\text{XCO}_2$  from 2009 to 2020. During wintertime, ecosystem  $\text{CO}_2$  uptake tends to be minimal over the Northern Hemisphere so that the  $\Delta\text{XCO}_2$  is mostly caused by  $\text{CO}_2$  emissions from fossil fuel combustions. Positive  $\Delta\text{XCO}_2$  of 1–2 ppm could be clearly observed in eastern China, eastern USA, and Europe in the Northern Hemisphere. The regions from the equator to 15° N have positive  $\Delta\text{XCO}_2$  values greater than 1 ppm in winter and lower  $\Delta\text{XCO}_2$  about 0.31 ppm in summer, which may be attributed to seasonal biomass burning [23,35,64]. In summer, the regions over the northern high latitudes show the largest negative  $\Delta\text{XCO}_2$  because terrestrial ecosystems in the Northern Hemisphere take up  $\text{CO}_2$  emitted by human activities.  $\text{CO}_2$  anomalies in the Southern Hemisphere are negative in winter and positive in summer, excluding the regions in tropical Africa. These spatial characteristics are generally similar to  $\Delta\text{XCO}_2$  calculated by CT- $\text{XCO}_2$  in Figure A4. Positive  $\Delta\text{XCO}_2$  in summer from CT- $\text{XCO}_2$  is slightly higher than the result of satellite  $\text{XCO}_2$  data. The main difference is that there are no consistent changes of  $\Delta\text{XCO}_2$  in tropical Africa between Mapping- $\text{XCO}_2$  and CT- $\text{XCO}_2$ , which may be due to the underestimation of fire emissions in CT simulation.



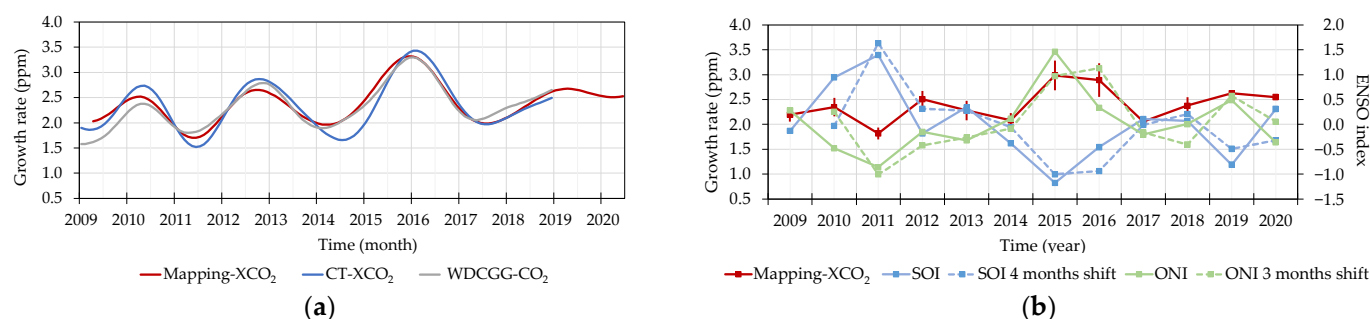
**Figure 2.** Spatial distributions of seasonal  $\Delta\text{XCO}_2$  in winter (a) and in summer (b) calculated using Mapping- $\text{XCO}_2$  from 2009 to 2020.

Comparing the spatial distribution of anthropogenic emissions from the ODIAC emission inventory in Figure 3, we can see that these regions with positive  $\Delta\text{XCO}_2$  of 1–2 ppm are very consistent with high anthropogenic emissions. As shown in Figures 2a and A4a, the pattern of  $\Delta\text{XCO}_2$  in the United States during wintertime shows larger  $\Delta\text{XCO}_2$  in the east than that in the west, which is similar to the pattern of  $\text{CO}_2$  emissions from ODIAC. Additionally, the high  $\text{CO}_2$  absorption by natural terrestrial biosphere in the western region during summertime, because of the high emissions as indicated by ODIAC, is not found in the multi-year mean  $\Delta\text{XCO}_2$ . These results indicate that global  $\text{CO}_2$  anomalies in winter can effectively represent the increase in atmospheric  $\text{CO}_2$  concentration caused by anthropogenic emissions and biomass burning.



**Figure 3.** Long-term average of global  $\text{CO}_2$  emissions in 1° grid from ODIAC during the period of 2009 to 2019.

Figure 4a shows the global CO<sub>2</sub> growth rates derived from Mapping-XCO<sub>2</sub>, CT-XCO<sub>2</sub>, and the ground-based CO<sub>2</sub> measurements from WDCGG. Compared with CT-XCO<sub>2</sub>, the global CO<sub>2</sub> growth rates calculated by Mapping-XCO<sub>2</sub> are more consistent with observational data. Annual mean CO<sub>2</sub> growth rates of 1.82 to 2.98 ppm are reflected on the continuous increases in atmospheric CO<sub>2</sub> concentration, which is mainly caused by anthropogenic CO<sub>2</sub> emissions. High growth rates appeared in 2010, 2012/2013, and 2015/2016. Among them, the growth rate in 2015/2016 was the highest. Many studies have pointed out that significant inter-annual fluctuations are induced by abnormal natural CO<sub>2</sub> emissions associated with ENSO events [2,16]. For that, we also compared the annual CO<sub>2</sub> growth rate from Mapping-XCO<sub>2</sub> with two ENSO indices, which are shown in Figure 4b. The result shows the satellite-derived growth rates agree well with ENSO indices. The correlation of the annual CO<sub>2</sub> growth rate with SOI and ONI are  $-0.52$  and  $0.68$ , respectively. The growth rate response as quantified by the correlation coefficient ( $R$ ) is largest after 4 months for SOI ( $R^2 = 40.24\%$ ) and after 3 months for ONI ( $R^2 = 58.46\%$ ). These results are consistent with previous reported findings [16,18,20,22].



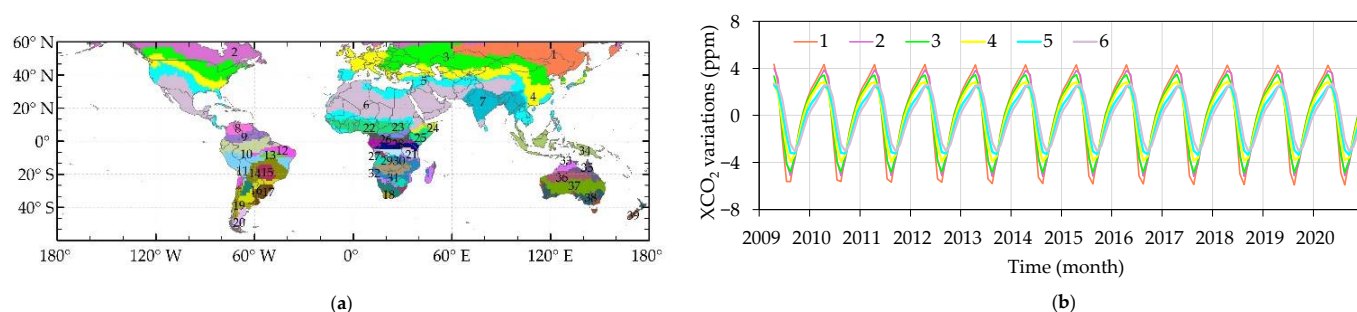
**Figure 4.** Time series of global CO<sub>2</sub> growth rate from 2009 to 2020 and comparison with ENSO indices. (a) Global growth rates of the long-term CO<sub>2</sub> trend from Mapping-XCO<sub>2</sub>, CT-XCO<sub>2</sub>, and ground-based observations of CO<sub>2</sub> data; (b) comparison of satellite-derived growth rate (red line) and ENSO indices. The 1 $\sigma$  uncertainty range of the growth rates are shown as vertical lines. The original ENSO indices are shown as solid lines and time-shifted data are shown as dotted lines.

### 3.2. Spatial Pattern of the Seasonal XCO<sub>2</sub> Cycle

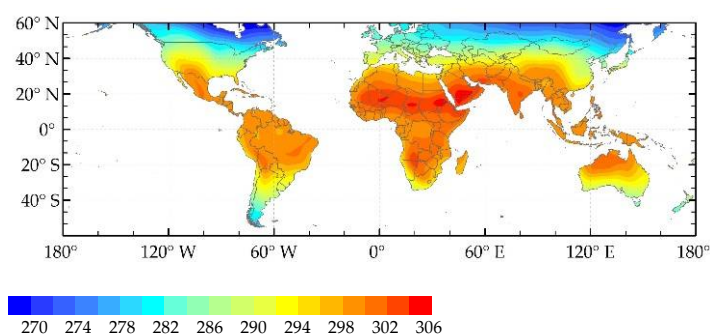
Global seasonal XCO<sub>2</sub> changes from 2009 to 2020 are grouped into 40 clusters based on the K-means method as described in Section 2.2.2. Figure 5 presents spatial distribution of the clustering results. We noted that seasonal XCO<sub>2</sub> changes show latitudinal zonal distribution but significantly offset to the southwest in the Northern Hemisphere. These interesting results are highly consistent with the pattern of clusters derived from CT-XCO<sub>2</sub> using the same approach in Figure A5. Compared to the distribution of potential temperature in Figure 6, the spatial pattern of seasonal XCO<sub>2</sub> changes is in good agreement with potential temperature contours, especially in the Northern Hemisphere. The result indicates that clustered XCO<sub>2</sub> variation is relatively homogeneous, which allows us to detect any perturbations due to the external CO<sub>2</sub> fluxes within each cluster region. Moreover, seasonal amplitudes of XCO<sub>2</sub> gradually reduce from north to south as shown in Figure 5b. The maximum is up to 10 ppm in cluster 1, and the minimum is 5 ppm in cluster 5, which is primarily caused by the strength of vegetation uptake at different latitudes.

We further investigated the relationship between seasonal XCO<sub>2</sub> changes and seasonal vegetation activities characterized by NDVI. Figure 7 shows the spatial distribution of correlation coefficients ( $R$ ) between their seasonal changes globally. The seasonal XCO<sub>2</sub> presents strong negative correlation with NDVI in most areas due to seasonal activities of vegetation CO<sub>2</sub> uptake in the northern high latitude area and regions of grassland and savannas. The regions with less or no vegetation present weak correlation between seasonal XCO<sub>2</sub> and NDVI. These regions with strong correlations indicate that the biosphere has large impacts on the variation of CO<sub>2</sub> concentration, which can also be seen in Figure A6.

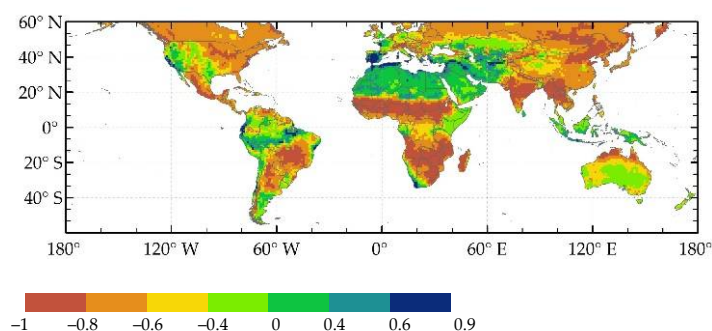




**Figure 5.** The clustering results of seasonal XCO<sub>2</sub> changes based on Mapping-XCO<sub>2</sub> from 2009 to 2020 (a) and the temporal variations of clusters in the Northern Hemisphere (b). The line colors correspond to the clusters in (a).



**Figure 6.** Spatial distribution of potential temperature contours at 1000 hPa from 2009 to 2020.



**Figure 7.** Spatial distribution of correlation coefficients between seasonal XCO<sub>2</sub> changes based on Mapping-XCO<sub>2</sub> and NDVI from 2009 to 2020.

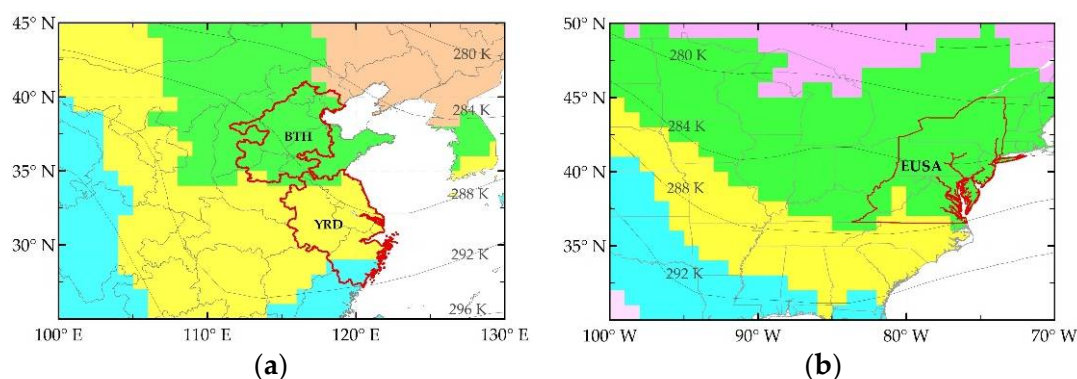
An accurate assessment of the contribution of the biosphere and atmospheric transport helps better disentangle the contribution of anthropogenic emissions to XCO<sub>2</sub> variations. This clustering result can help us understand globally spatial distribution characteristics of XCO<sub>2</sub> variation affected by the biosphere and atmospheric transport. Comparing Figure 5a with Figures 6 and 7, we can find that clustering results of XCO<sub>2</sub> after removing long-term changes effectively reveal the effects of fluxes from the biosphere and atmospheric transport. The ranges of clustering classes can be used to select and analyze interesting areas with similar biospheric fluxes and atmospheric transport.

### 3.3. Regional XCO<sub>2</sub> Anomalies and Anthropogenic Emissions

#### 3.3.1. Regional XCO<sub>2</sub> Anomalies in Urban Agglomeration Areas

We focus on the investigation of regional XCO<sub>2</sub> anomalies caused by anthropogenic emissions in the urban agglomeration areas in China and the United States. Based on the density of cities, we selected three source areas of anthropogenic emissions (AE), including the Beijing-Tian-Hebei region and nearby areas (BTH), the Yangtze River Delta urban agglomerations (YRD), and the urban agglomerations in the eastern United States

of America (EUSA), which are shown in Figure 8. Total CO<sub>2</sub> emissions from these areas account for about 13% of global CO<sub>2</sub> emissions according to anthropogenic emissions from ODIAC. These three regions are located in the same clustering areas that have similar seasonal XCO<sub>2</sub> cycles. They are clusters 3 and 4, as shown in Figure 5.



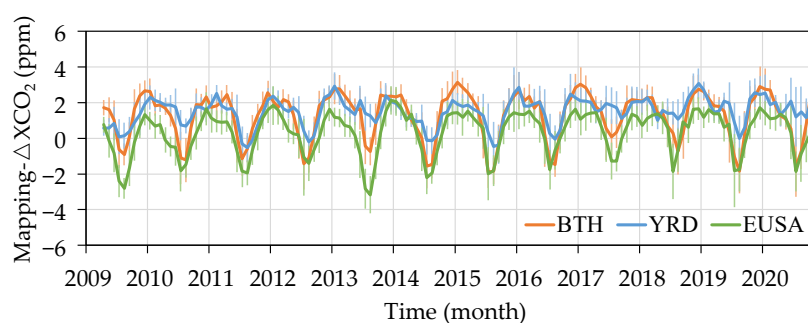
**Figure 8.** Location of source areas in China and the USA. (a) The areas for BTH and YRD; (b) the area for EUSA. The red boundary represents source areas. The clustering results from Figure 5 and the lines of potential temperature are also indicated.

Regional XCO<sub>2</sub> anomalies ( $\Delta$ XCO<sub>2</sub>) are calculated by removing the “background” trend of latitude zone from regional CO<sub>2</sub> concentrations as described in Section 3.1. We calculated the multi-year averaged  $\Delta$ XCO<sub>2</sub> for these three regions using Mapping-XCO<sub>2</sub> according to two stages during 2009–2014 and during 2015–2020, respectively. From Table 2,  $\Delta$ XCO<sub>2</sub> are generally 1–3 ppm and the values during the wintertime are up to 1 ppm larger than the multi-year mean, especially for BTH and EUSA. Both BTH and EUSA are basically located in cluster 3 with a seasonal amplitude of 8 ppm, which is larger than the amplitude of 6 ppm for YRD in cluster 4. From the first 5 years of 2009–2014 to the second 5 years of 2015–2020,  $\Delta$ XCO<sub>2</sub> increased in the three areas. Comparing the differences of  $\Delta$ XCO<sub>2</sub> among AE areas,  $\Delta$ XCO<sub>2</sub> in both BTH and YRD is greater than that in EUSA, while BTH is slightly larger than YRD.

**Table 2.** Regional characteristics in the emission source areas.

Source Areas	BTH	2009–2014 YRD	EUSA	BTH	2015–2020 YRD	EUSA
XCO <sub>2</sub> (ppm)	393.96 ± 3.55	394.14 ± 3.44	392.91 ± 3.56	407.56 ± 4.73	407.86 ± 4.87	406.77 ± 4.71
$\Delta$ XCO <sub>2</sub> (ppm)	1.24 ± 0.24	1.42 ± 0.31	0.19 ± 0.19	1.36 ± 0.16	1.66 ± 0.22	0.57 ± 0.08
XCO <sub>2</sub> in winter (ppm)	395.29 ± 3.49	395.12 ± 3.33	394.41 ± 3.55	409.40 ± 4.43	409.06 ± 4.56	408.14 ± 4.51
$\Delta$ XCO <sub>2</sub> in winter (ppm)	2.32 ± 0.38	2.16 ± 0.34	1.44 ± 0.41	2.59 ± 0.33	2.25 ± 0.37	1.33 ± 0.25
Total CO <sub>2</sub> emission (GtCO <sub>2</sub> /year)	1.54 ± 0.14	1.66 ± 0.04	0.72 ± 0.01	1.71 ± 0.19	1.86 ± 0.05	0.70 ± 0.01
Land cover (%)	7.6	7.8	9.7	8.4	8.3	9.7
(Urban; Croplands;	34.9	56.4	13.5	34.5	55.4	14.1
Vegetation; Other)	51.2	34.4	74.7	50.6	35.0	73.9
	6.3	1.4	2.2	6.6	1.5	2.3

Time series of XCO<sub>2</sub> anomalies in source areas from Mapping-XCO<sub>2</sub> and CT-XCO<sub>2</sub> are shown in Figures 9 and A7, respectively.  $\Delta$ XCO<sub>2</sub> shows seasonal variations in which BTH and EUSA present greater negative  $\Delta$ XCO<sub>2</sub> than YRD. This is likely induced by the vegetation CO<sub>2</sub> uptake as the vegetation coverage is larger in BTH and EUSA. As can be seen from Figure 7, the correlation coefficients between seasonal CO<sub>2</sub> cycles and NDVI are  $-0.80 \pm 0.11$  and  $-0.77 \pm 0.05$  for BTH and EUSA, respectively, which are greater than the obtained coefficients of  $-0.65 \pm 0.15$  for YRD.



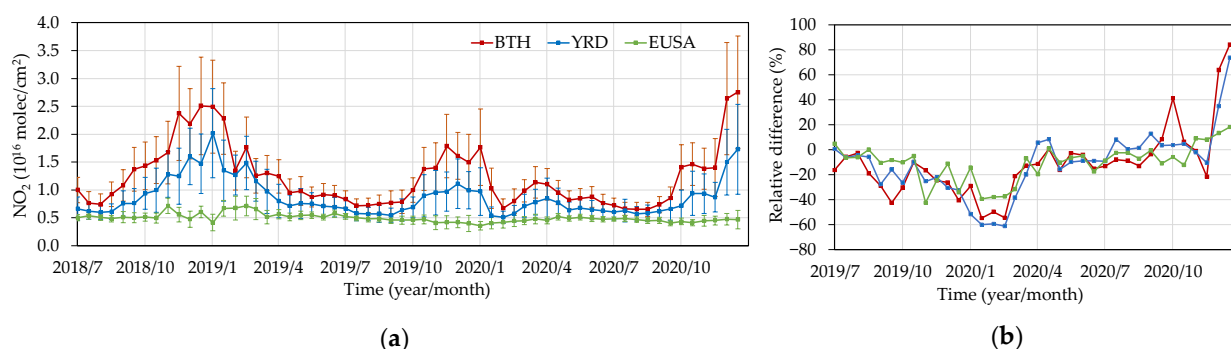
**Figure 9.** Time series of regional XCO<sub>2</sub> anomalies ( $\Delta XCO_2$ ) in the source areas derived from Mapping-XCO<sub>2</sub>. The  $1\sigma$  uncertainty estimate of regional XCO<sub>2</sub> anomalies is represented by the error bar, which is computed by the averaging mapping uncertainty and the standard deviation of regional statistics.

### 3.3.2. Response of Regional XCO<sub>2</sub> Anomalies during the COVID-19 Pandemic

Beginning from December 2019, Coronavirus disease 2019 (COVID-19) has occurred in numerous countries. The decline of economic activities caused by the pandemic lockdown measures has led to sharp reductions in anthropogenic CO<sub>2</sub> emissions in many countries. Regional-scale COVID-19-related CO<sub>2</sub> emission reductions are expected to be the largest in the first months of the pandemic outbreak. According to Le Quéré et al. [65], China's CO<sub>2</sub> emissions decreased by 242 MtCO<sub>2</sub> (uncertainty range 108–394 MtCO<sub>2</sub>) during January–April 2020. Buchwitz et al. [66] estimated the relative change of East China monthly emissions in 2020 relative to previous months. Their results showed significant differences across the ensemble of GOSAT and OCO-2 data products analysis. The ensemble mean indicates emission reductions by approximately  $8\% \pm 10\%$  in March 2020 and  $10\% \pm 10\%$  in April 2020 (uncertainties are 1-sigma), while somewhat lower reductions for the other months in 2020. These reduction months, however, should be investigated further, since the lockdown was mainly implemented during January–March; hence, the emission reduction should be in the same period.

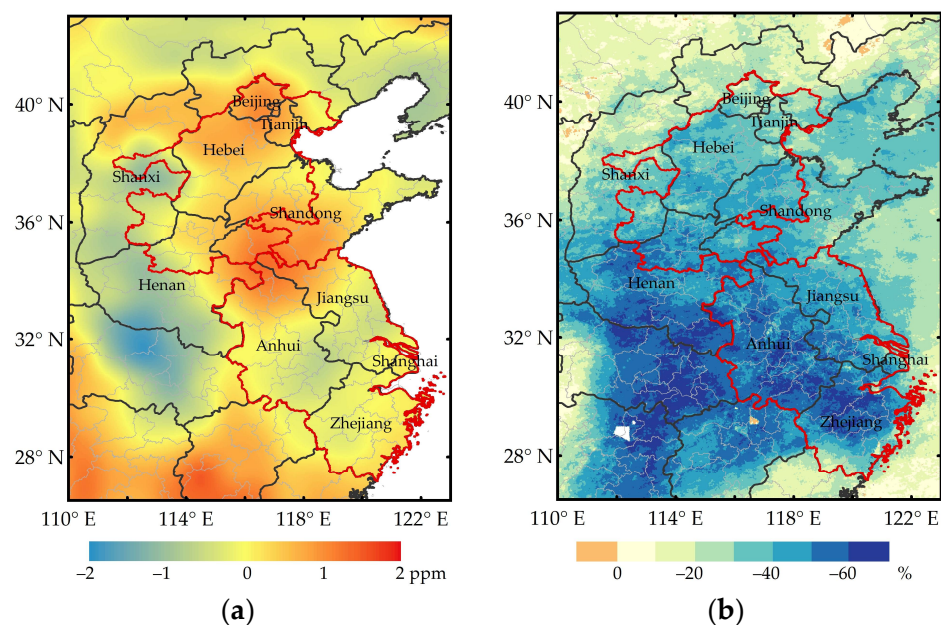
We compared the relative differences of regional XCO<sub>2</sub> anomalies during January–April between 2020 and 2019. CO<sub>2</sub> anomalies in YRD have a slight decrease of 0.17 ppm during January to February in 2020 relative to the same month of 2019, while there has been no decline in CO<sub>2</sub> anomalies for BTH and EUSA. This is because that CO<sub>2</sub> is a long-lived gas, and therefore, it has a high background concentration in the atmosphere. The increase of CO<sub>2</sub> concentration caused by anthropogenic emissions and the decline induced by emission reduction are small variables. The precision of satellite observations and mapping uncertainty makes it difficult to detect weak signals due to the emission reduction.

NO<sub>2</sub> concentration in the atmosphere has been used to infer CO<sub>2</sub> reductions and estimate China's CO<sub>2</sub> emissions during the COVID-19 pandemic [67]. Figure 10 illustrates the time series of regional NO<sub>2</sub> columns from July 2018 to December 2020 and the difference of 2020 relative to the previous year of 2019 for three areas. From Figure 10b, we can find that the sharp declines of NO<sub>2</sub> columns started in January and basically ended in April; NO<sub>2</sub> columns were reduced by 45–51%, 59%–61%, and 30% during January–March for BTH, YRD, and EUSA, respectively. The obvious reduction during the lockdown indicates that NO<sub>2</sub> columns are more sensitive to the reduction of anthropogenic emissions. The reduction, moreover, is lower in BTH than in YRD. This likely implies that the effects of reduced emissions from the decreased traffic volume were due to lockdown measures in YRD. However, there was increased demand for winter heating in BTH, as more people in 2020 had to stay in Beijing during the lockdown compared to former years. Additionally, the BTH area suffered a heavy pollution process from 11–13 February during the lockdown period [68].



**Figure 10.** Time series of NO<sub>2</sub> columns and the differences of NO<sub>2</sub> relative to the previous year. (a) Regional NO<sub>2</sub> columns every 16 days and 1 $\sigma$  uncertainty estimate is represented by error bar; (b) contemporaneous differences of NO<sub>2</sub> between 2019 and 2020.

In order to further analyze the response of XCO<sub>2</sub> to emission reduction in BTH and YRD, we focused on the period from January to March and compared the differences between 2019 and 2020 for  $\Delta$ XCO<sub>2</sub> and NO<sub>2</sub> columns.  $\Delta$ XCO<sub>2</sub> was resampled to a 0.01° grid by cubic convolution, which improves spatial resolution without changing the characteristics of the original data. As shown in Figure 11, the differences of  $\Delta$ XCO<sub>2</sub> between 2020 and 2019 tend to be negative in YRD, which means that emissions reduced, while they tended to increase by approximately 0.5 to 1 ppm in BTH. The spatial pattern of differences for  $\Delta$ XCO<sub>2</sub> is generally similar to NO<sub>2</sub> columns. The decrease of NO<sub>2</sub> columns in BTH is less than that in YRD. The NO<sub>2</sub> concentration decreased by approximately 35  $\pm$  5% in BTH, while it decreased by approximately 45  $\pm$  8% in YRD.

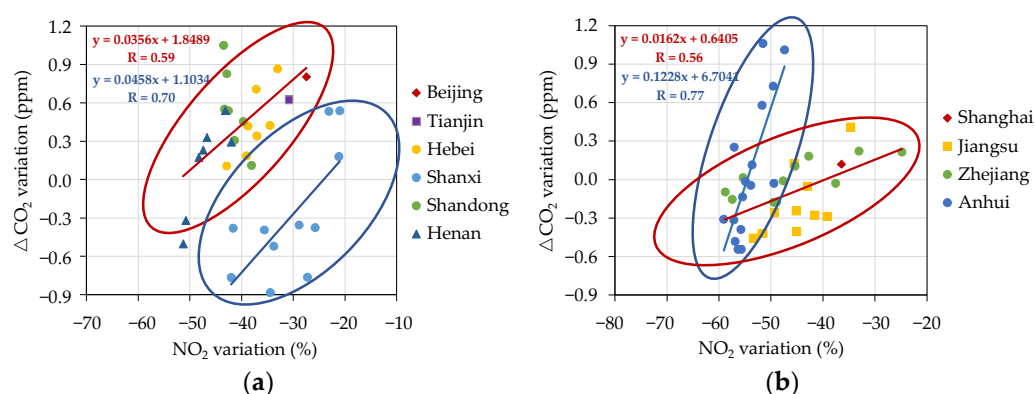


**Figure 11.** Spatial distribution of changes in XCO<sub>2</sub> anomalies and NO<sub>2</sub> columns from January to March in 2020 and 2019. (a) The variations of XCO<sub>2</sub> anomalies and (b) the variations of NO<sub>2</sub> columns. The bold gray lines represent the boundary of the provinces, while thin gray lines represent the boundary of cities.

In addition, we computed the variations of  $\Delta$ XCO<sub>2</sub> and NO<sub>2</sub> columns using the city district as a spatial unit. Figure 12 shows the result where the cities in AE areas are grouped according to provinces. The relationship between  $\Delta$ XCO<sub>2</sub> and NO<sub>2</sub> shows two distinct features in both BTH and YRD, Shanxi and other provinces in BTH, and Anhui and the



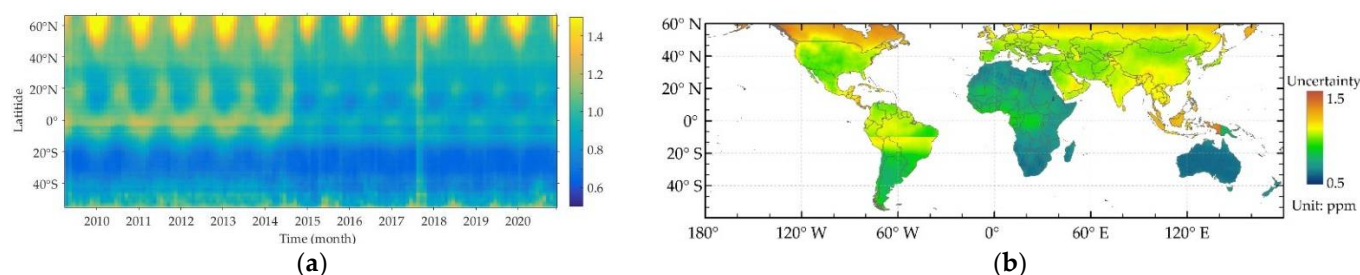
other provinces in YRD. These features are likely related to the types of emitting sources in these areas. The emitting sources in Shanxi and Anhui are mostly coal power plants and chemical plants, while the emitting sources of other provinces are mostly gas power plants and vehicles in the megacities of Beijing and Tianjin in BTH and Shanghai, Nanjing, Hangzhou, etc. in YRD. In Shanxi, the reduction of  $\Delta\text{XCO}_2$  is from  $-0.3$  to  $-0.9$  ppm and the decline of  $\text{NO}_2$  is 35% to 42%. In BTH, the reduction of  $\Delta\text{XCO}_2$  is approximately 0.3 to 0.9 ppm and the decline of  $\text{NO}_2$  is 30% to 45%. In comparison, in YRD, there is a larger range of  $\Delta\text{XCO}_2$  changes, from approximately  $-0.6$  to 1 ppm with declines of  $\text{NO}_2$  by 50% to 66% in Anhui. However, for other provinces in YRD other than Anhui, there are smaller changes of  $\Delta\text{XCO}_2$  from approximately  $-0.5$  to 0.3 ppm, with a decline of  $\text{NO}_2$  by 30% to 45%. These results indicate that the relationship between  $\text{XCO}_2$  and  $\text{NO}_2$  is available for the estimation of  $\text{CO}_2$  emissions. However, we should also consider the regional features of emitting sources, since their relationship highly depends on the types of emitting sources.



**Figure 12.** Comparison of  $\text{NO}_2$  variations and the changes of  $\text{XCO}_2$  anomalies for cities in (a) BTH and (b) YRD. The variations are relative differences in  $\text{CO}_2$  anomalies and  $\text{NO}_2$  columns from January to March in 2020 and 2019.

#### 4. Discussion

The accuracy of used Mapping- $\text{XCO}_2$  data will result in uncertainty around the findings of the spatio-temporal feature analysis. As described in Section 2.1.1, Mapping  $\text{XCO}_2$  data are obtained by processing different satellite observations using the spatio-temporal geostatistical method. The mapping uncertainty depends not only on the retrieval bias of original  $\text{XCO}_2$  retrievals, but also to a large extent on the number of available satellite observations. Mapping uncertainties are calculated by the method of Zeng et al. [43–46]. Figure 13 shows the spatio-temporal distribution of mapping uncertainties. The mapping uncertainties of global grids are generally less than 1.5 ppm. The areas with larger uncertainties are mainly in the high latitude of the Northern Hemisphere, which is due to sparse satellite observations. Mapping uncertainties during the period of GOSAT observations is higher than that corresponds to OCO-2 observations. This is because that the number of GOSAT observations is much less than OCO-2 observations. In the global analysis,  $\text{CO}_2$  growth rates derived from Mapping- $\text{XCO}_2$  during 2009 to 2020 are consistent with that from ground-based measures, which does not show the deviation, such as the uncertainty between GOSAT and OCO-2 data. The spatial patterns of mapping gridded  $\text{XCO}_2$ , in contrast to the global background, are consistent from year to year. These results indicate that the mapping  $\text{XCO}_2$  dataset using different satellite observations has consistent distribution characteristics in space and time. Moreover, the relative difference between regional  $\text{XCO}_2$  in source areas and the global background is in the range of 1.13 to 3.17 ppm during winter months, which is greater than mapping uncertainty in these areas.



**Figure 13.** Spatio-temporal distribution of mapping uncertainties from Mapping-XCO<sub>2</sub>. (a) Averaged mapping uncertainty in 1° latitudinal band and 1 month; (b) long-term averaged uncertainty from 2010 to 2020.

The land cover types in AE areas are dominated by croplands and vegetation, as shown in Table 2. Affected by the CO<sub>2</sub> uptake of terrestrial ecosystems and the accumulation of CO<sub>2</sub> from anthropogenic CO<sub>2</sub> emissions in the atmosphere, the regional CO<sub>2</sub> concentration reaches the highest value in the spring. The method of CO<sub>2</sub> anomalies can remove large-scale background information from regional CO<sub>2</sub> concentrations and enhance the signal of CO<sub>2</sub> changes. Many studies have pointed out that the calculation method of background concentration does not have a great impact on the results of regional CO<sub>2</sub> anomalies [35]. The temporal characteristic of regional CO<sub>2</sub> anomalies is consistent with that of the regional NO<sub>2</sub> concentration, as shown in Figure 10. Both of them have a maximum during the winter period of each year.

Regional CO<sub>2</sub> anomalies are mainly caused by anthropogenic CO<sub>2</sub> emissions and local ecological CO<sub>2</sub> fluxes. Regional ecological CO<sub>2</sub> flux has little impact on CO<sub>2</sub> changes in winter; CO<sub>2</sub> enhancement is in the range of 1.00 to 3.14 ppm in source areas during winter months, whereas the mapping uncertainty is 0.75 to 1.42 ppm in the same period. During the winter period, ΔXCO<sub>2</sub> of BTH is higher than that of YRD, which agrees with the emission characteristics of NO<sub>2</sub> concentrations. The ΔXCO<sub>2</sub> in BTH and EUSA show negative values in summer, which is because local ecological CO<sub>2</sub> fluxes have a greater impact on CO<sub>2</sub> anomalies in summer. On the other hand, the mapping uncertainty and standard deviation are relatively larger during the summer months. Therefore, it is challenging to detect the enhancement of regional CO<sub>2</sub> concentration caused by anthropogenic emissions in the growing season of vegetation.

## 5. Conclusions

We presented a global analysis of spatio-temporal XCO<sub>2</sub> variations and detected regional XCO<sub>2</sub> anomalies using satellite mapping XCO<sub>2</sub> data (Mapping-XCO<sub>2</sub>) from April 2009 to December 2020. The dataset has resolutions of 3 days in time and 1° grid in space, respectively. Mapping-XCO<sub>2</sub> is produced by a gap-filling technique using XCO<sub>2</sub> retrievals obtained by GOSAT and OCO-2.

The growth rates of global XCO<sub>2</sub> derived from Mapping-XCO<sub>2</sub> data show large fluctuations in inter-annual variabilities, which is in agreement with the long-term CO<sub>2</sub> trends calculated by WDCGG ground-based observations. Elevated XCO<sub>2</sub> of 1.5 to 3.5 ppm, which is mostly induced by anthropogenic emissions and seasonal biomass burning, can be observed using Mapping-XCO<sub>2</sub> data with background removed. Furthermore, the clustering analysis of gridded seasonal XCO<sub>2</sub> variations, after removing the long-term trend and background, reveal spatial pattern of atmospheric transport and terrestrial ecological CO<sub>2</sub> flux.

At the regional scale, XCO<sub>2</sub> enhancements during winter months are detected to be  $2.47 \pm 0.37$  ppm,  $2.20 \pm 0.36$  ppm, and  $1.38 \pm 0.33$  ppm for the Beijing-Tianjin-Hebei area, the Yangtze River Delta area, and the high-density urban areas in the eastern USA, respectively. The regional emission characteristic of XCO<sub>2</sub> enhancement is consistent with regional NO<sub>2</sub> columns. However, it is difficult to accurately detect enhanced CO<sub>2</sub> signals in the vegetation growing season due to impacts of local ecological CO<sub>2</sub> uptakes and relatively large uncertainty of the mapping data during summertime. The regional XCO<sub>2</sub> anomalies

did not clearly show the declines of anthropogenic CO<sub>2</sub> emissions during the lockdown of the COVID-19 pandemic from January to March 2020 compared with the same time in the previous year of 2019. However, the significant correlation between relative differences of XCO<sub>2</sub> and NO<sub>2</sub> columns calculated at urban scales indicates that different types of emitting sources show a significantly positive correlation. This result suggests that we could use space-observed NO<sub>2</sub> data to identify the anthropogenic emitting sources and rectify CO<sub>2</sub> emissions estimated from satellite observations since both gases are mostly co-emitted in cities.

Our studies provide new cases for investigating the responses of XCO<sub>2</sub> observed by satellites to anthropogenic emissions at global and regional scales. These results demonstrate the potential of the global land mapping XCO<sub>2</sub> dataset in monitoring the long-term XCO<sub>2</sub> variations and detecting regional XCO<sub>2</sub> enhancements caused by anthropogenic in non-growing seasons.

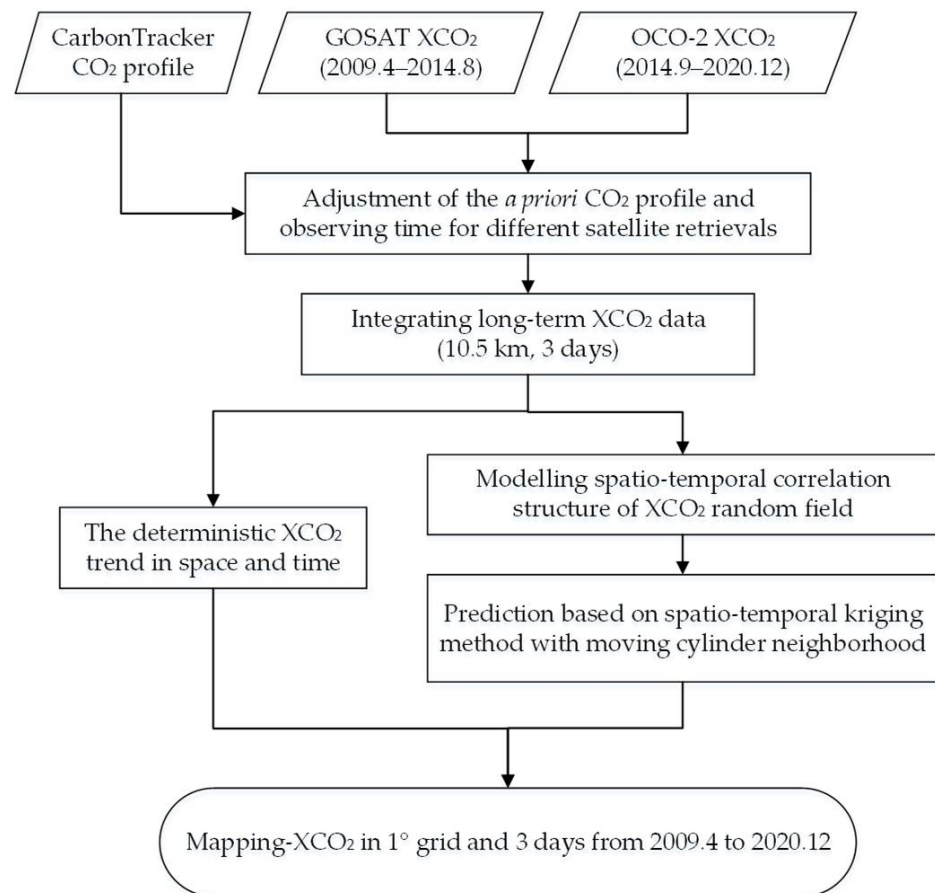
**Author Contributions:** Conceptualization, M.S., Z.-C.Z. and L.L.; Data curation, M.S. and S.Z.; Formal analysis, M.S., Z.-C.Z. and L.L.; Methodology, M.S. and L.L.; Software, M.S. and W.R. All authors have read and agreed to the published version of the manuscript.

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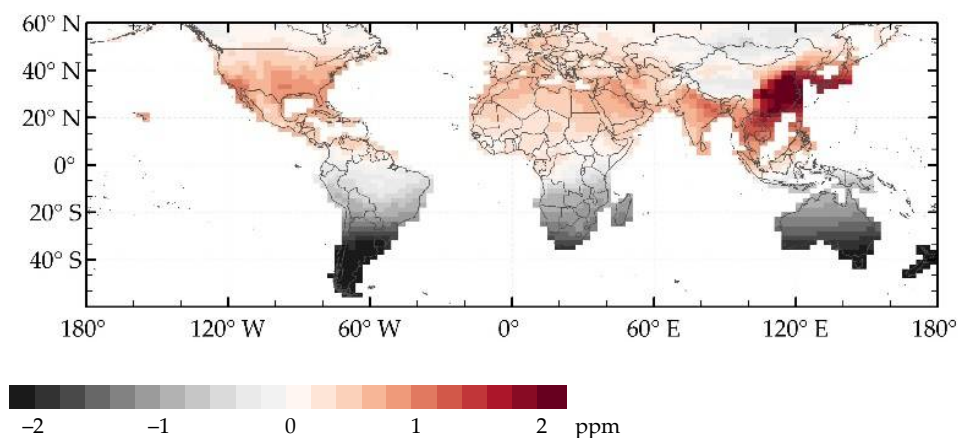
**Acknowledgments:** We are grateful for the ACOS-GOSAT v9r data and OCO-2 v10r data, which were provided by the ACOS/OCO-2 project at the Jet Propulsion Laboratory, California Institute of Technology and obtained from the ACOS/OCO-2 data archive maintained at the NASA Goddard Earth Science Data and Information Services Center. CarbonTracker CT2019B results were provided by NOAA ESRL, Boulder, Colorado, USA from the website at <http://carbontracker.noaa.gov> (assessed on 13 May 2021). We thank NOAA ESRL and PSL for providing potential temperature data and ENSO indices. We thank the European Space Agency (ESA) and Google Earth Engine for providing Sentinel-S5P NO<sub>2</sub> products, CGER-NIES for providing ODIAC data at <https://db.cger.nies.go.jp/dataset/ODIAC/> assessed on 29 November 2020 (Data DOI: doi:10.17595/20170411.001), and the World Data Centre for Greenhouse Gases (WDCGG) for providing global atmospheric CO<sub>2</sub> data. We also acknowledge the Land Processes Distributed Active Archive Center (LP DAAC) at the National Aeronautics and Space Administration (NASA) for sharing land cover type and NDVI data derived from MODIS.

**Conflicts of Interest:** The authors declare no conflict of interest.

## Appendix A

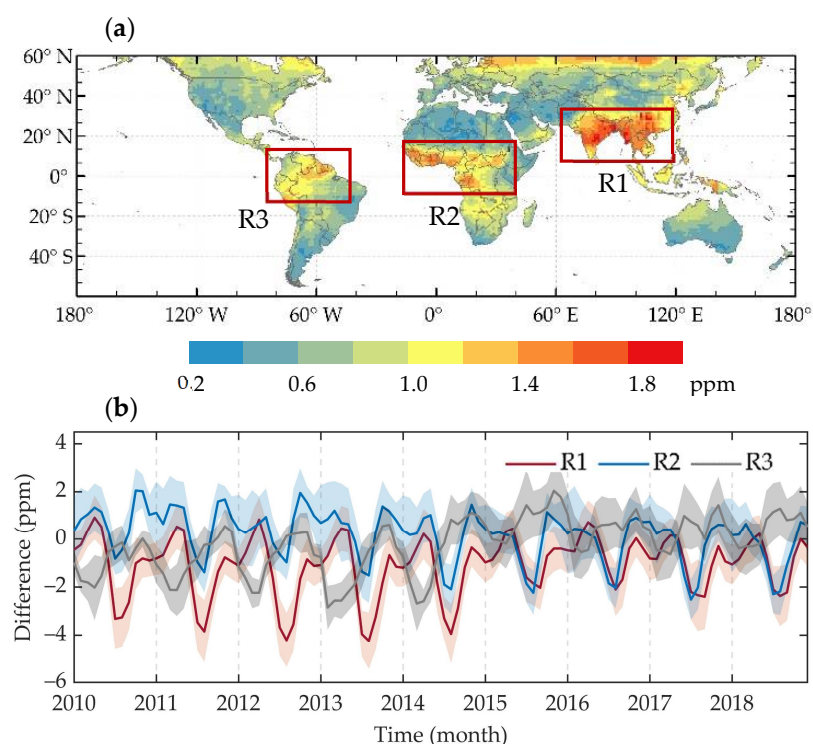


**Figure A1.** The workflow chart for generating Mapping-XCO<sub>2</sub> using satellite XCO<sub>2</sub> retrievals.

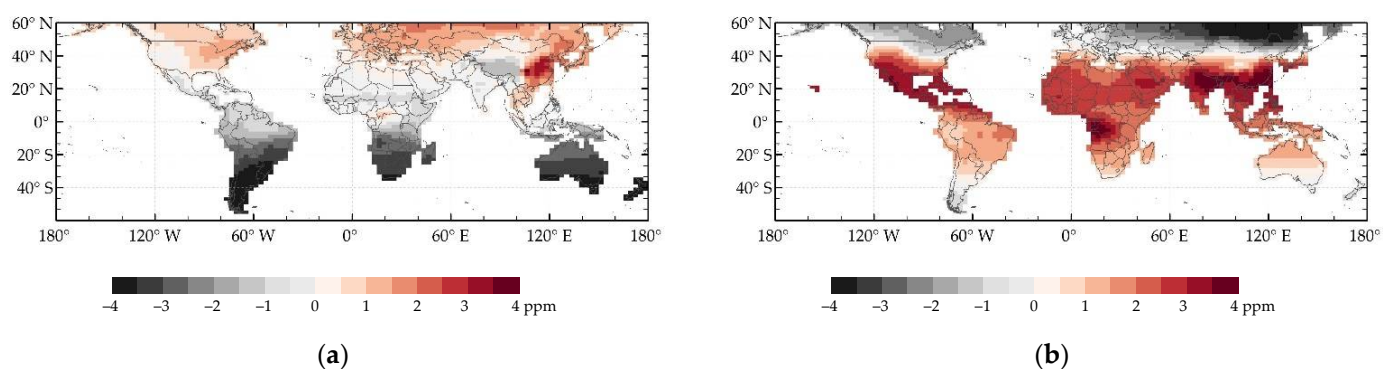


**Figure A2.** Spatial distributions of averaged dXCO<sub>2</sub> from 2009 to 2018 calculated from CT-XCO<sub>2</sub> following the same approach adopted by the Mapping-XCO<sub>2</sub> dataset.

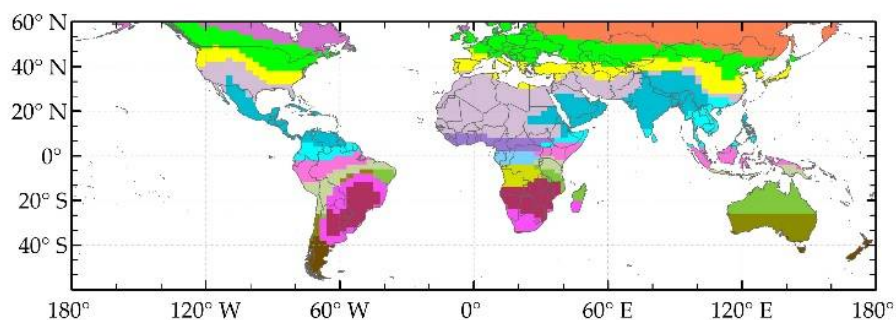




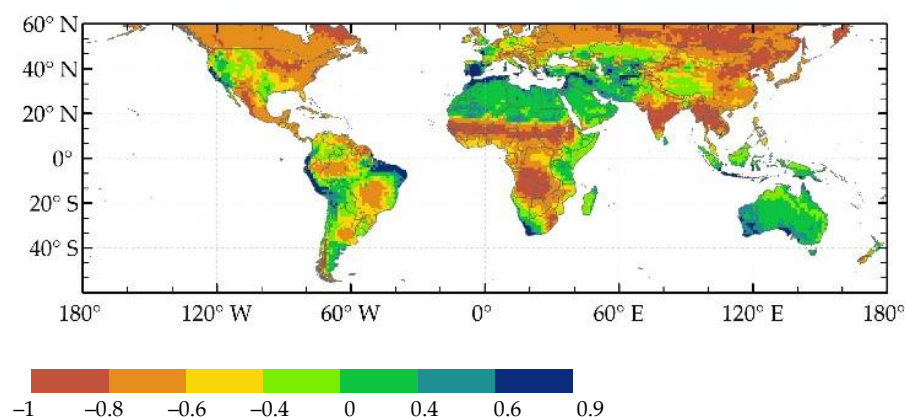
**Figure A3.** Comparison of Mapping-XCO<sub>2</sub> and CT-XCO<sub>2</sub> from 2010 to 2018. (a) The absolute mean difference of monthly gridded XCO<sub>2</sub> between Mapping-XCO<sub>2</sub> and CT-XCO<sub>2</sub> from 2010 to 2018; (b) time series of the mean difference in the regions of the red boxes shown in (a), in which the shaded colors represent one standard deviation.



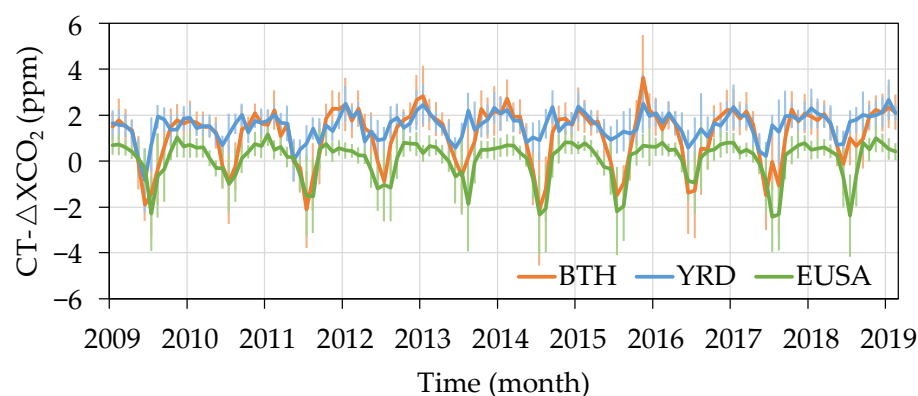
**Figure A4.** Spatial distributions of long-term averaged seasonal dXCO<sub>2</sub> in winter (a) and in summer (b) calculated from CT-XCO<sub>2</sub> from 2009 to 2018.



**Figure A5.** The clustering results of seasonal XCO<sub>2</sub> changes using CT-XCO<sub>2</sub> data from 2009 to 2019.



**Figure A6.** Spatial distribution of correlation coefficients in seasonal XCO<sub>2</sub> changes between CT-XCO<sub>2</sub> and NDVI from 2009 to 2019.



**Figure A7.** Time series of regional CO<sub>2</sub> anomalies ( $\Delta XCO_2$ ) in the source areas derived from CT-XCO<sub>2</sub>. The 1 $\sigma$  uncertainty estimate of regional XCO<sub>2</sub> anomalies is represented by error bar, which is one standard deviation of the regional statistics.

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