Detecting the Responses of CO₂ Column Abundances to Anthropogenic Emissions from Satellite Observations of GOSAT and OCO-2

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Abstract: The continuing increase in atmospheric CO₂ concentration caused by anthropogenic CO₂ emissions significantly contributes to climate change driven by global warming. Satellite measurements of long-term CO₂ 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 CO₂ (XCO₂) data (Mapping-XCO₂), generated from a spatio-temporal geostatistical method using GOSAT and OCO-2 observations from April 2009 to December 2020, to investigate the responses of XCO₂ to anthropogenic emissions at both global and regional scales. Our results show that the long-term trend of global XCO₂ growth rate from Mapping-XCO₂ is consistent with that from ground observations, shows interannual variations caused by the El Niño Southern Oscillation (ENSO). The spatial distributions of XCO₂ anomalies, derived from removing background from the Mapping-XCO₂ data, reveal XCO₂ enhancements of about 0.37 ppm, 2.20 ± 0.36 ppm, and 1.38 ± 0.33 ppm, respectively, can be detected during winter months from 2009 to 2020. These anomalies are slightly higher than model simulations from CarbonTracker-CO₂. In addition, we compared the variations in regional XCO₂ anomalies and NO₂ columns during the lockdown of the COVID-19 pandemic from January to March 2020. Interestingly, the results demonstrate that the variations of XCO₂ anomalies have a positive correlation with the decline of NO₂ during this period. These correlations, moreover, are associated with the features of emitting sources. These results suggest that we can use simultaneously observed NO₂ because of its high detectivity and co-emission with CO₂ to assist the analysis and verification of CO₂ emissions in future studies.

Keywords: Mapping XCO₂; anthropogenic emission; GOSAT; OCO-2; NO₂ columns
1. Introduction

Global atmospheric CO$_2$ concentration continues to increase by 2–3 ppm per year, which contributes significantly to global warming [1,2]. Changes in atmospheric CO$_2$ 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$^\circ$ 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$_2$ concentration and detect the influence mechanism of human activities in various regions on atmospheric CO$_2$ variations, so as to provide a basis for governments to evaluate the effects of CO$_2$ emission reduction measures.

Satellite measurements from GOSAT and OCO-2 have delivered the column-averaged dry air mole fractions of CO$_2$ (XCO$_2$) 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$_2$ concentration. For example, using satellite XCO$_2$ 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$_2$ concentration at global and regional scales [12–16]. Abnormal increase in CO$_2$ concentration mostly occurs in natural vegetation areas. The detection and attribution analysis of extreme CO$_2$ changes show that CO$_2$ anomalies are related to the abnormal carbon emissions from terrestrial ecosystems caused by extreme climate [17]. Previous studies using CO$_2$ data from model and ground observations also showed consistent results with that from satellite observations [18–22].

XCO$_2$ 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$_2$ 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$_2$ 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$_2$ enhancement detected by three orbits of observations from OCO-2 during November–December in eastern Australia is approximately 1.5 ppm [33]. Global XCO$_2$ 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$_2$ observations and other relatively short-lived pollutants (e.g., CO and NO$_2$) in cities found that urban CO$_2$ 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$_2$ observations have the detectability of natural and anthropogenic CO$_2$ 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$_2$ 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$_2$ data (Mapping-XCO$_2$) using GOSAT and OCO-2 observations. With these global spatio-temporal continuous XCO$_2$ data, this study is able to fully explore the changes of XCO$_2$ enhancements caused by anthropogenic emission at both global and regional scales.
In this paper, we investigate global XCO$_2$ variations in space and time, and analyze spatial patterns of seasonal XCO$_2$ 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$_2$ to anthropogenic emissions. Our study aims to provide global spatial and temporal analysis of XCO$_2$ changes and quantify the responses of regional XCO$_2$ enhancements to anthropogenic emission using long-term mapping data generated from satellite XCO$_2$ observations.

2. Materials and Methods

2.1. Materials

2.1.1. CO$_2$ Datasets

We use the global land mapping XCO$_2$ dataset (Mapping-XCO$_2$) from April 2009 to December 2020, which has a spatial grid resolution of $1^\circ$ latitude by $1^\circ$ longitude and temporal resolution of 3 days. The dataset is produced by applying a spatio-temporal geostatistical method to satellite XCO$_2$ retrievals from GOSAT observations (from April 2009 to August 2014) and OCO-2 observations (from September 2014 to December 2020). The XCO$_2$ 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$_2$ data are illustrated in Figure A1, including the following key steps: (1) We adjusted the differences in XCO$_2$ retrievals induced by the a priori CO$_2$ profile and different overpass time using CO$_2$ profiles from CarbonTracker as reference data. Spatial and temporal scales of satellite observations are integrated to a uniform unit by averaging XCO$_2$ 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$_2$ 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$_2$. (3) Based on these variogram models, space-time kriging with moving cylinder kriging neighborhood was implemented to estimate the XCO$_2$ value at the center of $1^\circ$ 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$_2$ is less than 1.5 ppm on average. Compared to TCCON data, the overall bias of Mapping-XCO$_2$ obtained by $\pm 0.5^\circ$ box centered at the TCCON sites is $0.16 \pm 1.19$ ppm.

Table 1 gives a summary of Mapping-XCO$_2$ 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$_2$ mole fractions from a combination of CO$_2$ surface exchange models and an atmospheric transport model driven by meteorological fields [44]. We collected CO$_2$ data at the local time of 13:30 from CT2019B for comparison analysis with spatio-temporal variations of Mapping-XCO$_2$. The dataset has a resolution of $3^\circ \times 2^\circ$ grid in space and 1 day in time. In order to analyze long-term trends derived from Mapping-XCO$_2$, we collected global analysis data of atmospheric CO$_2$ 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$_2$ mole fractions and CO$_2$ 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$_2$ data from satellite observation, model, and ground observation.

<table>
<thead>
<tr>
<th>Dataset</th>
<th>Description</th>
<th>Reference/Data Source</th>
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<tbody>
<tr>
<td>Mapping-XCO$_2$</td>
<td>Global land mapping XCO$_2$ dataset produced by applying spatio-temporal</td>
<td>GES DISC [39] Zeng et al. [40–43]</td>
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<td></td>
<td>geostatistics on GOSAT and OCO-2 observations from April 2009 to September</td>
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<td></td>
<td>2020. The dataset is regularly distributed with a temporal interval of 3 days</td>
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<td></td>
<td>and spatial interval of 1° grid.</td>
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<tr>
<td>CT-XCO$_2$</td>
<td>The model XCO$_2$ data at the local time 13:30 (LST) from CT 2019B from 2009</td>
<td>NOAA [45]</td>
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<tr>
<td></td>
<td>to March 2019 in 3° × 2° grids with a temporal interval of 1 day.</td>
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<tr>
<td>WDCGG-CO$_2$</td>
<td>Global CO$_2$ analysis based on ground-based observations, covering from 1984</td>
<td>WDCGG [46]</td>
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<tr>
<td></td>
<td>to 2019 for global monthly mean concentrations and trends and from 1985 to</td>
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<td>2018 for growth rates.</td>
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2.1.2. Auxiliary Datasets

To analyze the mechanism of XCO$_2$ changes and its response to anthropogenic emissions, we collected various auxiliary datasets to compare with XCO$_2$ variations at global and regional scales.

The Open-source Data Inventory for Anthropogenic CO$_2$ (ODIAC) is used to evaluate high emission areas, which can potentially be detected by satellite observed XCO$_2$ 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$_2$ emissions from 2000 to 2019, including two different spatial resolutions of 1° × 1° and 1 km × 1 km. CO$_2$ emission estimates of the product are based on the latest country fossil fuel CO$_2$ 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$_2$ 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$_2$ changes revealed by the satellite XCO$_2$ 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$_2$ 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° and 2 days, respectively [55]. We calculated global monthly mean data with 1° resolution from 2009 to 2020, which are used for correlation analysis with seasonal XCO₂ changes from Mapping-XCO₂. 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.

NO₂ 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 CO₂ emissions [36–38]. NO₂ data used in our studies is the level 3 offline NO₂ data product derived from TROPOMI/Sentinel-5 Precursor observations [56,57]. The data product provides the total vertical column of NO₂ concentrations with temporal and spatial resolutions of 2 days and 0.01° grid, respectively. The dataset is delivered by the European Space Agency (ESA) and publicly available on Google Earth Engine [57,58]. We obtained regional NO₂ 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 XCO₂ Variations Using Mapping-XCO₂

The time series of atmospheric XCO₂ 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 XCO₂ 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 XCO₂ from Mapping-XCO₂ using a linear least squares regression method:

\[ f(t) = a_0 + a_1 t + a_2 t^2 + \sum_{i=1}^{4} (\beta_i \sin(i\omega t) + \gamma_i \cos(i\omega t)) \]  

\[ XCO₂ = f(t) + \delta, \]  

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 XCO₂ 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 XCO₂ and its long-term trend are calculated by combining the fitting part of the function and the filtered part. The growth rate of global XCO₂ is computed by taking the derivative of the long-term trend of XCO₂.

2.2.2. Clustering Spatial Pattern of the Seasonal XCO₂ Cycle

The changes of XCO₂ show a seasonal cycle especially in the Northern Hemisphere, which is affected by CO₂ flux from atmospheric transport and the terrestrial biosphere. The seasonal XCO₂ cycle for each grid is obtained by fitting XCO₂ 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 XCO₂ based on the similarities in its seasonal cycles in order to obtain the spatial pattern of seasonal XCO₂ 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 XCO₂ changes are classified into the same cluster [63]. The temporal variation of XCO₂, 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’ CO₂ uptake.

2.2.3. Detecting CO₂ Anomalies at Global and Regional Scales

The global atmospheric CO₂ concentration represents a balance of all natural and anthropogenic CO₂ fluxes into and out of the atmosphere. Atmospheric CO₂ is well mixed by turbulent mixing and atmospheric transport [2]. We use global monthly averaged XCO₂ as the global background. Gridded XCO₂ anomalies are calculated as the differences between gridded XCO₂ data and the background, which is hereafter referred to as dXCO₂. The dXCO₂ is associated with net CO₂ fluxes in the grid. A negative dXCO₂ implies a net sink of CO₂, while positive dXCO₂ implies a net source relative to global background. The spatial distribution of global gridded dXCO₂ from Mapping-XCO₂ is described in Section 3.1 and is further compared with dXCO₂ from CT-XCO₂ data.

Lastly, we focus on urban agglomerations in China and the USA to demonstrate regional detectivity of CO₂ 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 CO₂ variations, median XCO₂ in the latitude zone is utilized as a background value. We computed regional XCO₂ anomalies(ΔXCO₂) by subtracting the “background” from regional averaged XCO₂ in the study areas.

3. Results

3.1. Spatio-Temporal Characteristic of Global XCO₂ Variations and Anthropogenic Emissions

We calculated global gridded anomalies (dXCO₂) from Mapping-XCO₂ and CT-XCO₂ to analyze global XCO₂ variations in space and time. Figure 1 shows spatial distributions of multi-year averaged dXCO₂ of Mapping-XCO₂ during 2010–2018, which have a similar spatial pattern with that calculated from CT-dXCO₂ in Figure A2. Higher positive dXCO₂ is observed in the region of East Asia, Southeast Asia, Middle East, North America, and North Africa. The dXCO₂ shows a negative value in the Southern Hemisphere. The result from Mapping-XCO₂ is about 0.4 ppm lower than CT-XCO₂ in eastern Asia. However, it shows obvious higher dXCO₂ over the regions of Xinjiang in China and lower dXCO₂ in India. The overall difference of global monthly mean XCO₂ between Mapping-XCO₂ and CT-XCO₂ is −0.24 ± 0.39 ppm, which is less than the difference of dXCO₂. Therefore, the differences of global XCO₂ anomalies between Mapping-XCO₂ and CT-XCO₂ are mostly induced by their gridded XCO₂ 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.
Seasonal dXCO$_2$ in winter and summer are computed by averaging dXCO$_2$ values during December-January-February (DJF) and June-July-August (JJA), respectively. Figure 2 maps spatial patterns of seasonal dXCO$_2$ from Mapping-XCO$_2$ from 2009 to 2020. During wintertime, ecosystem CO$_2$ uptake tends to be minimal over the Northern Hemisphere so that the dXCO$_2$ is mostly caused by CO$_2$ emissions from fossil fuel combustions. Positive dXCO$_2$ 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 dXCO$_2$ values greater than 1 ppm in winter and lower dXCO$_2$ 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 dXCO$_2$ because terrestrial ecosystems in the Northern Hemisphere take up CO$_2$ emitted by human activities. 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 dXCO$_2$ calculated by CT-XCO$_2$ in Figure A4. Positive dXCO$_2$ in summer from CT-XCO$_2$ is slightly higher than the result of satellite XCO$_2$ data. The main difference is that there are no consistent changes of dXCO$_2$ in tropical Africa between Mapping-XCO$_2$ and CT-XCO$_2$, which may be due to the underestimation of fire emissions in CT simulation.

Comparing the spatial distribution of anthropogenic emissions from the ODIAC emission inventory in Figure 3, we can see that these regions with positive dXCO$_2$ of 1–2 ppm are very consistent with high anthropogenic emissions. As shown in Figures 2a and A4a, the pattern of dXCO$_2$ in the United States during wintertime shows larger dXCO$_2$ in the east than that in the west, which is similar to the pattern of CO$_2$ emissions from ODIAC. Additionally, the high 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 dXCO$_2$. These results indicate that global CO$_2$ anomalies in winter can effectively represent the increase in atmospheric CO$_2$ concentration caused by anthropogenic emissions and biomass burning.

Figure 2. Spatial distributions of seasonal dXCO$_2$ in winter (a) and in summer (b) calculated using Mapping-XCO$_2$ from 2009 to 2020.

Figure 3. Long-term average of global CO$_2$ emissions in 1° grid from ODIAC during the period of 2009 to 2019.
Figure 4a shows the global CO$_2$ growth rates derived from Mapping-XCO$_2$, CT-XCO$_2$, and the ground-based CO$_2$ measurements from WDCGG. Compared with CT-XCO$_2$, the global CO$_2$ growth rates calculated by Mapping-XCO$_2$ are more consistent with observational data. Annual mean CO$_2$ growth rates of 1.82 to 2.98 ppm are reflected on the continuous increases in atmospheric CO$_2$ concentration, which is mainly caused by anthropogenic CO$_2$ 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$_2$ emissions associated with ENSO events [2,16]. For that, we also compared the annual CO$_2$ growth rate from Mapping-XCO$_2$ 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$_2$ 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 4a](image1.png)  ![Figure 4b](image2.png)

**Figure 4.** Time series of global CO$_2$ growth rate from 2009 to 2020 and comparison with ENSO indices. (a) Global growth rates of the long-term CO$_2$ trend from Mapping-XCO$_2$, CT-XCO$_2$, and ground-based observations of CO$_2$ data; (b) comparison of satellite-derived growth rate (red line) and ENSO indices. The 1σ 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$_2$ Cycle

Global seasonal XCO$_2$ 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$_2$ 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$_2$ using the same approach in Figure A5. Compared to the distribution of potential temperature in Figure 6, the spatial pattern of seasonal XCO$_2$ changes is in good agreement with potential temperature contours, especially in the Northern Hemisphere. The result indicates that clustered XCO$_2$ variation is relatively homogeneous, which allows us to detect any perturbations due to the external CO$_2$ fluxes within each cluster region. Moreover, seasonal amplitudes of XCO$_2$ 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$_2$ 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$_2$ presents strong negative correlation with NDVI in most areas due to seasonal activities of vegetation CO$_2$ 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$_2$ and NDVI. These regions with strong correlations indicate that the biosphere has large impacts on the variation of CO$_2$ concentration, which can also be seen in Figure A6.
Figure 5. The clustering results of seasonal XCO$_2$ changes based on Mapping-XCO$_2$ 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$_2$ changes based on Mapping-XCO$_2$ 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$_2$ variations. This clustering result can help us understand globally spatial distribution characteristics of XCO$_2$ variation affected by the biosphere and atmospheric transport. Comparing Figure 5a with Figures 6 and 7, we can find that clustering results of XCO$_2$ 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$_2$ Anomalies and Anthropogenic Emissions

3.3.1. Regional XCO$_2$ Anomalies in Urban Agglomeration Areas

We focus on the investigation of regional XCO$_2$ 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 CO₂ emissions from these areas account for about 13% of global CO₂ emissions according to anthropogenic emissions from ODIAC. These three regions are located in the same clustering areas that have similar seasonal XCO₂ cycles. They are clusters 3 and 4, as shown in Figure 5.

![Figure 8](image_url)

**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 XCO₂ anomalies (ΔXCO₂) are calculated by removing the “background” trend of latitude zone from regional CO₂ concentrations as described in Section 3.1. We calculated the multi-year averaged ΔXCO₂ for these three regions using Mapping-XCO₂ according to two stages during 2009–2014 and during 2015–2020, respectively. From Table 2, ΔXCO₂ 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, ΔXCO₂ increased in the three areas. Comparing the differences of ΔXCO₂ among AE areas, ΔXCO₂ in both BTH and YRD is greater than that in EUSA, while BTH is slightly larger than YRD.

<table>
<thead>
<tr>
<th>Source Areas</th>
<th>BTH</th>
<th>YRD</th>
<th>EUSA</th>
<th>BTH</th>
<th>YRD</th>
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<tr>
<td>XCO₂ (ppm)</td>
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<td>394.14 ± 3.44</td>
<td>392.91 ± 3.56</td>
<td>407.56 ± 4.73</td>
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<td>406.77 ± 4.71</td>
</tr>
<tr>
<td>ΔXCO₂ (ppm)</td>
<td>1.24 ± 0.24</td>
<td>1.42 ± 0.31</td>
<td>0.19 ± 0.19</td>
<td>1.36 ± 0.16</td>
<td>1.66 ± 0.22</td>
<td>0.57 ± 0.08</td>
</tr>
<tr>
<td>XCO₂ in winter (ppm)</td>
<td>395.29 ± 3.49</td>
<td>395.12 ± 3.33</td>
<td>394.41 ± 3.55</td>
<td>409.40 ± 4.43</td>
<td>409.06 ± 4.56</td>
<td>408.14 ± 4.51</td>
</tr>
<tr>
<td>ΔXCO₂ in winter (ppm)</td>
<td>2.32 ± 0.38</td>
<td>2.16 ± 0.34</td>
<td>1.44 ± 0.41</td>
<td>2.59 ± 0.33</td>
<td>2.25 ± 0.37</td>
<td>1.33 ± 0.25</td>
</tr>
<tr>
<td>Total CO₂ emission (GtCO₂/year)</td>
<td>1.54 ± 0.14</td>
<td>1.66 ± 0.04</td>
<td>0.72 ± 0.01</td>
<td>1.71 ± 0.19</td>
<td>1.86 ± 0.05</td>
<td>0.70 ± 0.01</td>
</tr>
<tr>
<td>Land cover (%)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(Urban; Croplands; Vegetation; Other)</td>
<td>7.6</td>
<td>7.8</td>
<td>9.7</td>
<td>8.4</td>
<td>8.3</td>
<td>9.7</td>
</tr>
<tr>
<td>(Urban; Croplands; Vegetation; Other)</td>
<td>34.9</td>
<td>56.4</td>
<td>13.5</td>
<td>34.5</td>
<td>55.4</td>
<td>14.1</td>
</tr>
<tr>
<td>(Urban; Croplands; Vegetation; Other)</td>
<td>51.2</td>
<td>34.4</td>
<td>74.7</td>
<td>50.6</td>
<td>35.0</td>
<td>73.9</td>
</tr>
<tr>
<td>(Urban; Croplands; Vegetation; Other)</td>
<td>6.3</td>
<td>1.4</td>
<td>2.2</td>
<td>6.6</td>
<td>1.5</td>
<td>2.3</td>
</tr>
</tbody>
</table>

Time series of XCO₂ anomalies in source areas from Mapping-XCO₂ and CT-XCO₂ are shown in Figures 9 and A7, respectively. ΔXCO₂ shows seasonal variations in which BTH and EUSA present greater negative ΔXCO₂ than YRD. This is likely induced by the vegetation CO₂ uptake as the vegetation coverage is larger in BTH and EUSA. As can be seen from Figure 7, the correlation coefficients between seasonal CO₂ cycles and NDVI are −0.80 ± 0.11 and −0.77 ± 0.05 for BTH and EUSA, respectively, which are greater than the obtained coefficients of −0.65 ± 0.15 for YRD.
3.3.2. Response of Regional XCO₂ 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 CO₂ emissions in many countries. Regional-scale COVID-19-related CO₂ 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 CO₂ emissions decreased by 242 MtCO₂ (uncertainty range 108–394 MtCO₂) 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% ± 10% in March 2020 and 10% ± 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 XCO₂ anomalies during January–April between 2020 and 2019. CO₂ 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 CO₂ anomalies for BTH and EUSA. This is because that CO₂ is a long-lived gas, and therefore, it has a high background concentration in the atmosphere. The increase of CO₂ 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.

NO₂ concentration in the atmosphere has been used to infer CO₂ reductions and estimate China’s CO₂ emissions during the COVID-19 pandemic [67]. Figure 10 illustrates the time series of regional NO₂ 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 NO₂ columns started in January and basically ended in April; NO₂ 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 NO₂ 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 NO2 columns and the differences of NO2 relative to the previous year. (a) Regional NO2 columns every 16 days and 1σ uncertainty estimate is represented by error bar; (b) contemporaneous differences of NO2 between 2019 and 2020.

In order to further analyze the response of XCO2 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 ΔXCO2 and NO2 columns. ΔXCO2 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 ΔXCO2 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 ΔXCO2 is generally similar to NO2 columns. The decrease of NO2 columns in BTH is less than that in YRD. The NO2 concentration decreased by approximately 35 ± 5% in BTH, while it decreased by approximately 45 ± 8% in YRD.

Figure 11. Spatial distribution of changes in XCO2 anomalies and NO2 columns from January to March in 2020 and 2019. (a) The variations of XCO2 anomalies and (b) the variations of NO2 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 ΔXCO2 and NO2 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 ΔXCO2 and NO2 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 ΔXCO$_2$ is from −0.3 to −0.9 ppm and the decline of NO$_2$ is 35% to 42%. In BTH, the reduction of ΔXCO$_2$ is approximately 0.3 to 0.9 ppm and the decline of NO$_2$ is 30% to 45%. In comparison, in YRD, there is a larger range of ΔXCO$_2$ changes, from approximately −0.6 to 1 ppm with declines of NO$_2$ by 50% to 66% in Anhui. However, for other provinces in YRD other than Anhui, there are smaller changes of ΔXCO$_2$ from approximately −0.5 to 0.3 ppm, with a decline of NO$_2$ by 30% to 45%. These results indicate that the relationship between XCO$_2$ and NO$_2$ is available for the estimation of 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.

![Comparison of NO$_2$ variations and the changes of XCO$_2$ anomalies for cities in (a) BTH and (b) YRD. The variations are relative differences in CO$_2$ anomalies and NO$_2$ columns from January to March in 2020 and 2019.](image)

### 4. Discussion

The accuracy of used Mapping-XCO$_2$ data will result in uncertainty around the findings of the spatio-temporal feature analysis. As described in Section 2.1.1, Mapping 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 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, CO$_2$ growth rates derived from Mapping-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 XCO$_2$, in contrast to the global background, are consistent from year to year. These results indicate that the mapping XCO$_2$ dataset using different satellite observations has consistent distribution characteristics in space and time. Moreover, the relative difference between regional 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.
The land cover types in AE areas are dominated by croplands and vegetation, as shown in Table 2. Affected by the CO₂ uptake of terrestrial ecosystems and the accumulation of CO₂ from anthropogenic CO₂ emissions in the atmosphere, the regional CO₂ concentration reaches the highest value in the spring. The method of CO₂ anomalies can remove large-scale background information from regional CO₂ concentrations and enhance the signal of CO₂ changes. Many studies have pointed out that the calculation method of background concentration does not have a great impact on the results of regional CO₂ anomalies [35]. The temporal characteristic of regional CO₂ anomalies is consistent with that of the regional NO₂ concentration, as shown in Figure 10. Both of them have a maximum during the winter period of each year.

Regional CO₂ anomalies are mainly caused by anthropogenic CO₂ emissions and local ecological CO₂ fluxes. Regional ecological CO₂ flux has little impact on CO₂ changes in winter; CO₂ 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, ΔXCO₂ of BTH is higher than that of YRD, which agrees with the emission characteristics of NO₂ concentrations. The ΔXCO₂ in BTH and EUSA show negative values in summer, which is because local ecological CO₂ fluxes have a greater impact on CO₂ 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 CO₂ concentration caused by anthropogenic emissions in the growing season of vegetation.

5. Conclusions

We presented a global analysis of spatio-temporal XCO₂ variations and detected regional XCO₂ anomalies using satellite mapping XCO₂ data (Mapping-XCO₂) from April 2009 to December 2020. The dataset has resolutions of 3 days in time and 1° grid in space, respectively. Mapping-XCO₂ is produced by a gap-filling technique using XCO₂ retrievals obtained by GOSAT and OCO-2.

The growth rates of global XCO₂ derived from Mapping-XCO₂ data show large fluctuations in inter-annual variabilities, which is in agreement with the long-term CO₂ trends calculated by WDCGG ground-based observations. Elevated XCO₂ of 1.5 to 3.5 ppm, which is mostly induced by anthropogenic emissions and seasonal biomass burning, can be observed using Mapping-XCO₂ data with background removed. Furthermore, the clustering analysis of gridded seasonal XCO₂ variations, after removing the long-term trend and background, reveal spatial pattern of atmospheric transport and terrestrial ecological CO₂ flux.

At the regional scale, XCO₂ enhancements during winter months are detected to be 2.47 ± 0.37 ppm, 2.20 ± 0.36 ppm, and 1.38 ± 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 XCO₂ enhancement is consistent with regional NO₂ columns. However, it is difficult to accurately detect enhanced CO₂ signals in the vegetation growing season due to impacts of local ecological CO₂ uptakes and relatively large uncertainty of the mapping data during summertime. The regional XCO₂ anomalies
did not clearly show the declines of anthropogenic CO$_2$ 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$_2$ and NO$_2$ 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$_2$ data to identify the anthropogenic emitting sources and rectify CO$_2$ 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$_2$ observed by satellites to anthropogenic emissions at global and regional scales. These results demonstrate the potential of the global land mapping XCO$_2$ dataset in monitoring the long-term XCO$_2$ variations and detecting regional XCO$_2$ 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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**Conflicts of Interest:** The authors declare no conflict of interest.
Appendix A

Figure A1. The workflow chart for generating Mapping-XCO\textsubscript{2} using satellite XCO\textsubscript{2} retrievals.

Figure A2. Spatial distributions of averaged dXCO\textsubscript{2} from 2009 to 2018 calculated from CT-XCO\textsubscript{2} following the same approach adopted by the Mapping-XCO\textsubscript{2} dataset.
Figure A3. Comparison of Mapping-XCO2 and CT-XCO2 from 2010 to 2018. (a) The absolute mean difference of monthly gridded XCO2 between Mapping-XCO2 and CT-XCO2 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 dXCO2 in winter (a) and in summer (b) calculated from CT-XCO2 from 2009 to 2018.

Figure A5. The clustering results of seasonal XCO2 changes using CT-XCO2 data from 2009 to 2019.
Figure A6. Spatial distribution of correlation coefficients in seasonal XCO₂ changes between CT-XCO₂ and NDVI from 2009 to 2019.

Figure A7. Time series of regional CO₂ anomalies(ΔXCO₂) in the source areas derived from CT-XCO₂. The 1σ uncertainty estimate of regional XCO₂ anomalies is represented by error bar, which is one standard deviation of the regional statistics.

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