Analysis of Four Years of Global XCO$_2$ Anomalies as Seen by Orbiting Carbon Observatory-2

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Abstract: NASA’s carbon dioxide mission, Orbiting Carbon Observatory-2, began operating in September 2014. In this paper, we analyze four years (2015–2018) of global (60°S–60°N) XCO$_2$ anomalies and their annual variations and seasonal patterns. We show that the anomaly patterns in the column-averaged CO$_2$ dry air mole fraction, XCO$_2$, are robust and consistent from year-to-year. We evaluate the method by comparing the anomalies to fluxes from anthropogenic, biospheric, and biomass burning and to model-simulated local concentration enhancements. We find that, despite the simplicity of the method, the anomalies describe the spatio-temporal variability of XCO$_2$ (including anthropogenic emissions and seasonal variability related to vegetation and biomass burning) consistently with more complex model-based approaches. We see, for example, that positive anomalies correspond to fossil fuel combustion over the major industrial areas (e.g., China, eastern USA, central Europe, India, and the Highveld region in South Africa), shown as large positive XCO$_2$ enhancements in the model simulations. We also find corresponding positive anomalies and fluxes over biomass burning areas during different fire seasons. On the other hand, the largest negative anomalies correspond to the growing season in the northern middle latitudes, characterized by negative XCO$_2$ enhancements from simulations and high solar-induced chlorophyll fluorescence (SIF) values (indicating the occurrence of photosynthesis). The largest discrepancies between the anomaly patterns and the model-based results are observed in the tropical regions, where OCO-2 shows persistent positive anomalies over every season of every year included in this study. Finally, we demonstrate how XCO$_2$ anomalies enable the detection of anthropogenic signatures for several local scale case studies, both in the Northern and Southern Hemisphere. In particular, we analyze the XCO$_2$ anomalies collocated with the recent TROPOspheric Monitoring Instrument NO$_2$ observations (used as indicator of anthropogenic fossil fuel combustion) over the Highveld region in South Africa. The results highlight the capability of satellite-based observations to monitor natural and man-made CO$_2$ signatures on global scale.

Keywords: carbon dioxide; satellite; remote sensing; anthropogenic; biogenic; global analysis

1. Introduction

The first routine measurements of atmospheric carbon dioxide (CO$_2$) were started at Mauna Loa Observatory in 1958 by Charles David Keeling. These measurements now form the iconic Keeling curve that shows, in addition to seasonal variations, rapidly increasing levels of CO$_2$ in the atmosphere due to the burning of fossil fuels. Now, 60 years later, atmospheric CO$_2$ concentrations are measured
all over the world and show levels that are about 100 ppm higher than they were in 1958, when they were about 315 ppm. However, the measurement networks are still denser in the Western world.

One of the most important applications of CO$_2$ measurements is to infer the surface CO$_2$ fluxes, i.e., sources and sinks (or emissions into and removal from the atmosphere). A variety of methods are commonly used to derive CO$_2$ fluxes from spatially resolved estimates of column-averaged CO$_2$ dry air mole fraction, XCO$_2$. These methods can be broadly classified as analytic mass balance, plume dispersion, or atmospheric inversion methods. All three approaches predict or optimize surface fluxes needed to reproduce the observed XCO$_2$ distribution in the presence of the time-varying wind field. Ground-based in situ measurements provide the best available constraints on CO$_2$ concentrations, but the existing ground based network is unevenly distributed and includes few stations in the tropics, high latitudes, or over the ocean. One way to extend this network is to use space-based measurements of the XCO$_2$ [1,2]. The main advantage of space-based observations is that they can provide high spatial and temporal resolution on global scale with the same measurement device. The principal challenge is the need for unprecedented levels of precision and accuracy to resolve and quantify CO$_2$ fluxes. High precision measurements are needed to clearly resolve CO$_2$ concentration anomalies in the context of their large background concentrations. For example, source pixel mass balance methods [3] indicate that a single-sounding precision of ~0.25% (~1 part per million (ppm) out of the 400 ppm background) is needed within a 4 square km satellite footprint to detect a ~10 kiloton/day CO$_2$ source in the presence of a ~5 km/h wind. Spatially and temporally coherent biases in the measurements must also be minimized because, regional-scale biases in XCO$_2$ no larger than a few tenths of a part per million can introduce carbon flux errors exceeding 0.7 Gigatons of carbon per year [4,5]. Ideally, the inverse modeling systems that estimate CO$_2$ fluxes from atmospheric measurements, can assimilate information from all available measurement types.

The first space-based instrument designed to retrieve estimates of XCO$_2$ throughout the atmospheric column from high resolution spectra of reflected sunlight was SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Chartography), launched by the European Space Agency onboard the Envisat spacecraft in 2002 [6]. The first mission that provided XCO$_2$ measurements at a precision better than 0.6% is the Japanese GOSAT (Greenhouse Gases Observing Satellite), which has produced CO$_2$ and methane (CH$_4$) measurements since April 2009 [7]. The follow-on mission GOSAT-2 was launched in 2018. In 2014, NASA launched the Orbiting Carbon Observatory-2 (OCO-2), which has been providing CO$_2$ measurements since September 2014 [1,2,8]. An increasing number of CO$_2$ measuring satellites, including the Chinese TanSat [9,10], Fen-Yun-3D, and Gaofen 5 have been launched since 2016 and others are being planned (see, e.g., https://en.wikipedia.org/wiki/Space-based_measurements_of_carbon_dioxide). Recently, TROPOMI (TROPOspheric Monitoring Instrument), on board the European Space Agency (ESA) Sentinel-5 Precursor (S5P) satellite, began retrieving estimates of the methane dry air mole fraction from observations of reflected sunlight, but not CO$_2$.

Although the current space-based CO$_2$ missions have focused primarily on the need to better understand the biospheric carbon fluxes [11], satellite-based CO$_2$ observations have also been used in anthropogenic CO$_2$ studies. For example, Reuter et al. [12] analyzed the CO$_2$-to-NO$_2$ emission ratios with collocated SCIAMACHY data, Janardanan et al. [13] studied localized CO$_2$ enhancements using GOSAT data, Kort et al. [14] studied the CO$_2$ signal from Los Angeles and Mumbai megacities from GOSAT data, Nassar et al. [11] estimated the CO$_2$ emissions from individual power plants using OCO-2 data, Schwandner et al. [15] analyzed localized carbon dioxide sources (e.g., Los Angeles) using OCO-2 data, and Ye et al. [16] evaluated the possibility of constraining the fossil fuel CO$_2$ emissions from urban areas using OCO-2 observations. Recently, Wang et al. [17] used OCO-2 XCO$_2$ anomalies for distinguishing anthropogenic CO$_2$ emissions from different energy intensive industrial sources in northern China. Since the Paris Agreement was signed in 2015, the focus has shifted more and more to anthropogenic CO$_2$ emissions. For example, the European Commission is currently planning a dedicated anthropogenic CO$_2$ monitoring mission within the Copernicus Programme.
One large obstacle limiting the quantification of anthropogenic CO$_2$ flux from space, is the limited mapping capability of current satellite missions. GOSAT provides single soundings with 10.5 km diameter surface footprints that are separated by ~250 km. OCO-2 provides measurements with eight 2.25 km long footprints along a narrow (less than 10 km) swath, but the swaths are separated by ~1.5 degrees of longitude. In both cases, the data coverage is quite sparse, and the sampling density is further reduced by optically thick clouds and aerosols, which preclude full-column XCO$_2$ measurements. Figure 1 illustrates an example of the OCO-2 data coverage on 1° × 1° latitude-longitude grid during different periods of time: one day, one week, one month and one year. The OCO-2 XCO$_2$ anomalies illustrated in Figure 1 are further discussed in Section 3. In addition to the nominal measurements, special target mode measurements are performed in selected measurement locations for calibration and/or validation purposes [18].

Figure 1. Mean Orbiting Carbon Observatory-2 (OCO-2) XCO$_2$ anomalies on 1° × 1° latitude-longitude grid during one day, one week, one month and one year. For the background we calculate the daily medians for each 10-degree latitude bands and linearly interpolate the resulting values to each OCO-2 data point.
The added value of satellite data in carbon cycle research has been demonstrated in several model-based studies (e.g., [19,20]). Our previous study [21] introduced the concept of XCO₂ anomaly and analyzed the first 18 months of OCO-2 measurements for three selected areas in the Northern Hemisphere. The objective of this paper is to describe the spatio-temporal variability of the XCO₂ anomalies (including anthropogenic emission patterns and seasonal variability related to vegetation and biomass burning) solely using satellite observations. The aim is to exploit the intrinsic value of satellite-based XCO₂ observations beyond their application as model input. We analyze four years (2015–2018) of global (60°S–60°N) XCO₂ anomalies as seen by OCO-2. We present the annual and seasonal anomaly patterns and compare the observed anomalies to modeled enhancements, estimated fluxes and solar-induced chlorophyll fluorescence (SIF, indicating the occurrence of photosynthesis). We also demonstrate the capability of the method to analyze small scale emission sources. In particular, we study the polluting emissions over the Highveld industrial region in South Africa by combining the XCO₂ anomalies to the new TROPOMI nitrogen dioxide (NO₂) observations (used as indicator of atmospheric pollution).

Section 2 introduces the datasets and Section 3 presents the method to derive the global XCO₂ anomalies. In Section 4, we study the annual and seasonal patterns of XCO₂ anomalies, and discuss them in terms of anthropogenic, biospheric and biomass burning contributions, including the analysis of local emission sources in South Africa. In Sections 5 and 6, we discuss the results and present the main conclusions.

2. Datasets

2.1. OCO-2 CO₂ Data

We use data from NASA’s OCO-2 satellite [8]. OCO-2 was launched on 2 July 2014, and now leads the 705 km Afternoon Constellation (also known as the A-Train). The spacecraft carries and points a three-channel imaging grating spectrometer designed to collect high resolution, bore-sighted spectra of reflected sunlight. Spectra are collected in the molecular oxygen A-band at 0.765 microns and the CO₂ bands at 1.61 and 2.06 microns. The retrieved quantity is column-averaged dry air mole fraction of CO₂, XCO₂. The instrument collects science data while viewing the local nadir (nadir mode), near the “glint spot,” where solar radiation is reflected specularly from the surface (glint mode) or stationary surface targets (target mode). The study by Crisp et al. [8] provides more details on the OCO-2 measurement principle, instrument design, calibration approach and on-orbit performance.

Validation and bias correction have been important elements of the data processing strategy for OCO-2 as the OCO-2 XCO₂ retrieval algorithm continues to evolve [22]. The study by Wunch et al. [18] provides a comprehensive validation of the Version 7 retrospective (V7r) XCO₂ product against the Total Carbon Column Observing Network (TCCON), and found that the absolute median difference is less than 0.4 ppm and the RMS difference less than 1.5 ppm. The bias appears to depend on latitude, surface properties, and scattering by aerosols [18]. Similar validation experiments on the V8r and V9r products were performed by O’Dell et al. [22] and Kiel et al. [23].

In this paper, we use the OCO-2 data version V9r, available from the MIRADOR platform at http://mirador.gsfc.nasa.gov. We use the lite files that include bias correction and data screening. We use data points from all three science viewing modes, where the quality flags are set to zero, indicating highest quality data.

2.2. CarbonTracker CO₂ Fluxes

In order to better understand the spatio-temporal patterns in the XCO₂ anomaly distribution, we compare our results to NOAA’s CarbonTracker flux maps (see also https://www.esrl.noaa.gov/gmd/ccgg/carbontracker/fluxes.php), version CT2017 ([24] with updates documented at http://carbontracker.noaa.gov). In particular, we consider the biospheric fluxes as well as those associated with biomass burning emissions. As input information, CT2017 uses Global Fire Emissions Database
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(GFED) 4.1, which is based on MODIS (Moderate Resolution Imaging Spectroradiometer) observations of fire counts. The a priori biospheric fluxes are based on the Carnegie–Ames Stanford Approach (CASA) biogeochemical model, and their scaling factors are estimated using CT2017.

2.3. Solar-Induced Chlorophyll Fluorescence (SIF)

We analyze the solar-induced chlorophyll fluorescence, which is also measured by the OCO-2 instrument [25] at 757 nm and 772 nm. We use the cloud-filtered 757 nm data from the OCO-2 lite files V8r that include quality filtering and bias correction. SIF can serve as a proxy for the vegetation gross primary production (GPP) [26], defined as the synthesis of organic compounds from atmospheric CO$_2$ and principally occurring through the process of photosynthesis. Therefore, SIF data are expected to provide information on the contribution of the CO$_2$ uptake by the vegetation on the XCO$_2$ anomalies.

2.4. XCO$_2$ Enhancements from FLEXPART Model

We use the Lagrangian particle dispersion model, FLEXPART (FLEXible PARTicle dispersion model) [27], to simulate anthropogenic and biospheric XCO$_2$ enhancements at OCO-2 measurement locations. We follow the approach used by Janardanan et al. [13], where the localized GOSAT CO$_2$ enhancements are compared to the inventory-based CO$_2$ estimates using the FLEXPART model outputs. We use the JRA-55 meteorology provided at 60 vertical levels and interpolated to a horizontal resolution of 1.25 degrees. As there are about 85 000 OCO-2 data points per day, for computational reasons, we aggregate OCO-2 data points into one-second averages, taken separately for OCO-2 footprints 1–4 and 5–8 (≤12 soundings in one set, 2 sets per second). The sampling corresponds to the OCO-2 data version V7r.

As input information for anthropogenic emissions, we use the high-resolution ODIAC (Open-Data Inventory for Anthropogenic Carbon dioxide) inventory [28]. In the ODIAC inventory, the anthropogenic CO$_2$ emissions are estimated at 1 km $\times$ 1 km resolution given the power plant emissions (intensity and geographical location) and satellite-observed nightlights. In the FLEXPART model simulations, from each aggregated OCO-2 data point, ten thousand virtual particles are released and transported three days backward in time with the three-dimensional wind field using parameterizations for turbulence and convection.

2.5. TROPOMI/S5P NO$_2$ Tropospheric Columns

In order to study local scale emission sources in South Africa, we use the TROPOMI/S5P NO$_2$ tropospheric columns [29,30] collocated with the XCO$_2$ anomalies, as indicators of anthropogenic fossil fuel emissions. Launched in October 2017, TROPOMI on board S5P flies on a sun-synchronous polar orbit satellite with overpass time 13:30 LT. TROPOMI NO$_2$ data are publicly available since July 2018 with a spatial resolution of 3.5 km $\times$ 7 km at nadir and daily global coverage.

3. Global XCO$_2$ Anomalies

CO$_2$ has a long atmospheric lifetime and relatively large background concentrations (~410 ppm). Because of this, XCO$_2$ varies by only about 2% from pole-to-pole and over the seasonal cycle. Meanwhile, XCO$_2$ variations corresponding to anthropogenic sources are even smaller on the scale of the satellite sounding (2–4 km$^2$). High precision is therefore essential to quantify XCO$_2$ anomalies associated with these sources. Methods like those used to map short-lived air pollutants, like NO$_2$, based on averaging out the outflow downwind from the emission sources, cannot be directly applied to space-based CO$_2$ measurements. The large CO$_2$ background and seasonal variability must be removed before being able to highlight the emission areas.

In order to extract information about the anthropogenic and biogenic signatures from OCO-2 retrievals, we use the concept of XCO$_2$ anomaly [21], defined as the difference between the individual XCO$_2$ value measured by OCO-2 and the daily background:
\[ XCO_2(\text{anomaly}) = XCO_2(\text{individual}) - XCO_2(\text{daily background}). \] (1)

This equation provides an anomaly value for each OCO-2 data point. Using the daily background allows us to remove the seasonal variability and the increasing trend of CO\(_2\) concentrations. Once we obtain the anomalies for each OCO-2 measurement point, we define a spatial grid (e.g., 1° × 1°, latitude-longitude) and calculate the mean at each grid point over a defined period of time. Figure 1 shows examples of mean XCO\(_2\) anomalies for one day, one week, one month and one year. The resulting anomaly maps illustrate the areas where CO\(_2\) is emitted (positive anomalies) into the atmosphere and those acting as sinks, where CO\(_2\) is absorbed at the surface (negative anomalies). The strength of this approach is that it only uses satellite-based measurements and is not dependent on patterns in a priori fields, external data or other assumptions in atmospheric chemistry-transport models.

Defining the area over which the daily background is calculated, is a critical step in the analysis. In [21], we focused on three selected anthropogenic emission areas: (1) North America, (2) Europe, northern Africa, and Middle East, and (3) East Asia. In this paper, our aim is to obtain comparable anomalies for different regions. In order to obtain the background, we calculate the daily medians for each 10-degree latitude band and linearly interpolate the resulting values to each OCO-2 data point. The median was chosen because it better represents the typical value in each latitude band, and it is not skewed towards extreme values.

Figure 2 shows the daily XCO\(_2\) background as a function of latitude interpolated at 1-degree resolution. The interpolation step removes the sharp latitudinal boundaries that would result if the daily medians at different latitude bands were used directly. In particular, we would obtain a sharp boundary between the northern high mid-latitudes (40°N–60°N) and lower latitudes during summer months due to the stronger (and shifted) CO\(_2\) uptake. The median XCO\(_2\) values for each 10-degree latitude band as well as their latitudinal distribution for selected days between September 2015 and September 2016 are included in Figure S1 (top and bottom panels, respectively). The background used in the calculation of the anomalies changes at different times of the year and over different regions. The peak of the seasonal cycle shifts during the year from the Northern to the Southern hemisphere. Also the amplitude of the seasonal cycle is much larger in the Northern Hemisphere due to the strong seasonal biospheric flux differences. Figure 2 also shows the XCO\(_2\) values increasing by about 10 ppm from September 2014 to September 2018. We note that no data are available at high middle-latitudes in autumn-winter when the Sun is too low for a successful XCO\(_2\) retrieval. Furthermore, a smaller amount of data is expected over areas frequently covered by clouds or with a large aerosol load.

Figure 2. OCO-2 XCO\(_2\) daily background, 2014–2018. Daily medians are calculated for each 10-degree latitude bands and interpolated at 1-degree resolution.
4. Results

4.1. Annual XCO₂ Anomalies

Figure 3 illustrates the mean annual OCO-2 XCO₂ anomaly for 2015–2018. The largest anthropogenic anomalies correspond to the areas in China, North-East India, Middle East, central Europe, and eastern USA, as also noted in [21]. In the Southern Hemisphere, the largest anthropogenic emission area, the Highveld region in South Africa, is clearly visible.

![Global XCO₂ anomalies as seen by OCO-2. The first panel corresponds to the four-year average 2015–2018, while the second, third, fourth, and fifth panels include the annual mean anomalies for the years 2015, 2016, 2017, and 2018, respectively.](image-url)
The largest positive XCO$_2$ anomalies are observed in the tropical regions in northern and southern Africa, Indonesia, Indochina, and South America. These correspond to large-scale biomass burning and to positive biospheric fluxes (respiration). These anomalies are most intense during the 2015–2016 El Niño, but persist through the four-year period considered here.

The largest negative anomalies are observed in the northern middle-latitudes ($40^\circ$N–$60^\circ$N) over Asia and North America. These are clearly associated with the strong biospheric sink during the growing season. This area is mainly sampled by OCO-2 during summer months and not during winter due to persistent cloudiness and low signal levels at high solar zenith angles (see Figure 2). This temporal sampling bias may introduce biases in the annual-average anomalies (examined in greater detail below). We also observe a large area with negative anomalies in the Southern Cone in South America.

The large areas with positive and negative anomalies are quite consistent from year to year. Also, smaller areas with large anomalies seem to be consistently visible in all years (e.g., the Highveld region). Some differences in the XCO$_2$ anomaly patterns between different years are, at least partly, related to the sampling of the instrument and the number of data points available. These are most visible for example at northern and southern mid-latitudes, where we can still identify individual “satellite tracks” from the map. In Figure S2 of the supplementary material, we present the maps of the number of data points that were used to calculate the mean in Figure 3. In addition to northern latitudes, the number of data points is lower also in places with high cloud density and/or large aerosol load (Himalaya, Amazonia, central Africa and China). Thus, the anomalies calculated over these areas are less robust. This is further illustrated in Figure S3, which shows the standard error of the mean, i.e., sample standard deviation divided by the square root of the sample size.

We find that the ocean XCO$_2$ anomalies are ~0.5 ppm lower than those over land. The XCO$_2$ concentrations are generally continuous across coastlines, clearly revealing outflow regions west of tropical Africa, and east of the east coasts of China and the U.S. The low bias over the ocean is generally consistent with the hypothesis that the ocean is a net sink for CO$_2$, absorbing about 8.9 GtCO$_2$/year [31]. However, it is important to note that the approach for defining the background and anomalies adopted here was optimized for identifying point emission sources. It might not be ideal for quantifying weak, spatially extensive sinks, like those expected over ocean and seen in our anomaly plots. For example, Gurney et al. [32] found that the simulations of oceanic CO$_2$ surface concentrations show about 1 ppm variation in the annual zonal mean. Also, the low XCO$_2$ values over the tropical and high latitude ocean are among the most difficult to validate given the lack of TCCON stations at those latitudes and very limited in situ measurements. This is therefore active area of research within the scientific community.

4.2. Seasonal XCO$_2$ Anomalies

Figure 4 illustrates the mean seasonal XCO$_2$ anomalies calculated from the period September 2014 to August 2018. The different seasons are defined as September-October-November (SON), December-January-February (DJF), March-April-May (MAM), and June-July-August (JJA). The large-scale patterns on SON, DJF, MAM look very similar to each other and to the annual mean, while in JJA the anomaly distribution looks quite different, with strong negative anomalies over the Northern Hemispheric mid-latitudes. We note that no OCO-2 observations are available during winter months (DJF) over the northern high- and mid-latitudes, due to persistent cloudiness and low signal levels. Figure S4 in the supplementary material shows the number of data points available for each season from September 2014 to August 2018. The standard error of the mean is illustrated in Figure S5.
Figure 4. Seasonal XCO$_2$ anomalies 2014–2018 for September-October-November, December-January-February, March-April-May, and June-July-August (top to bottom panels, respectively).

In order to better understand this seasonal variability, we analyze the seasonal distribution of the SIF (Figure 5), also measured by OCO-2, as well as the fluxes from NOAA’s CarbonTracker model (Figure 6). Both SIF and CarbonTracker flux spatial distributions show how the negative anomalies observed during JJA are related to the biospheric sink (i.e., high SIF and negative fluxes). This is particularly visible in the eastern part of North America, where the highest SIF values are observed. At the same time, low SIF values and high XCO$_2$ anomalies are observed in the western part of North America. The high XCO$_2$ anomalies observed in part of the northern hemisphere during JJA are partially related to the seasonal minimum of the background (Figures 2 and S1). These patterns,
however, do not contradict the patterns in the CarbonTracker flux map and are similar to the seasonal FLEXPART modeling results described in Section 4.3.

![Figure 5.](image)

Figure 5. Seasonal OCO-2 solar-induced chlorophyll fluorescence (SIF) at 757 nm for different seasons (from September 2014 to August 2015).

The XCO$_2$ anomaly patterns in Africa show positive values in the northern biomass burning area during winter months (DJF) and relatively smaller anomalies during SON and MAM. During summer months, we find mainly negative XCO$_2$ anomalies over the same area. These features correspond directly to those observed in the CarbonTracker flux maps, i.e., strong emissions from biomass burning during winter and uptake during summer (Figure 6, right column). SIF is also higher during summer months, when the XCO$_2$ anomalies are negative. In the southern biomass burning area in Africa, we observe the largest XCO$_2$ anomalies during SON and JJA, when the biomass burning emissions are the strongest (Figure 6, right column) and SIF values relatively small. The largest negative XCO$_2$ anomalies are seen in DJF and MAM, when there are reduced emissions from biomass burning. From Figure 4, we find that the XCO$_2$ anomalies over the Highveld area in South Africa are the largest during JJA and SON, when the CO$_2$ uptake by photosynthesis is minimum.

The limited number of data points available over Amazonia makes the quantification of the XCO$_2$ anomalies in South America more challenging (Figure S4). This is also illustrated in Figure S5 with high values of the standard error of the mean. The XCO$_2$ anomaly values are positive throughout the year, although, smaller anomalies are observed in JJA when the biospheric fluxes are negative. Signatures from biomass burning and positive biospheric fluxes are visible as larger positive anomalies during SON. In the Southern Cone, the XCO$_2$ anomalies are most negative during SON and DJF, which is in line with the SIF and biospheric flux maps, indicating that these anomalies are primarily biogenic.

In Indochina, negative XCO$_2$ anomalies correspond to the largest SIF values observed during JJA, although from the biospheric flux maps, we would expect positive fluxes during JJA. During DJF and MAM, we see the signature of strong emission from fires in the anomalies and flux maps (Figure 6, right column). We would expect however to see negative anomalies during SON due to the strong biospheric sink, but we find only slightly smaller positive anomalies. In the Indian peninsula we find positive XCO$_2$ anomalies as expected from anthropogenic emissions when the biospheric uptake is low.

Seasonal patterns are also evident in the largest anthropogenic emission areas, i.e., China, central Europe and eastern USA. For example, China and central Europe show positive anomalies during all seasons, while in eastern USA these are offset by a very strong biospheric sink in JJA. Also, the positive XCO$_2$ anomalies in China and central Europe are lower during JJA and MAM, respectively. Arid and semi-arid regions, including the Sahara, Tibetan Plateau and Australia, are out of sync with the rest of their hemispheres, with negative anomalies in the winter and small positive anomalies in the summer.
Finally, we note signatures of outflow over the Atlantic ocean with positive XCO$_2$ anomalies from the biomass burning emissions in Africa. Furthermore, the spring time outflow along the dominant westerlies from China to North America is also visible from Figure 4, as also observed, e.g., for tropospheric ozone in several studies (e.g., [33,34]).

![Figure 6. Seasonal biospheric (BIO) and biomass burning (FIRE) fluxes from NOAA’s CarbonTracker CT2017 from September 2014 to August 2015.](image)

**4.3. Modeling Results**

We analyze the XCO$_2$ enhancements related to fossil fuel combustion and biospheric fluxes corresponding to the OCO-2 sounding footprints using the Lagrangian FLEXPART model. This allows us to account for the effect of the OCO-2 sampling as well as the transport by the wind.

Figure 7 illustrates the modeled XCO$_2$ enhancements for the year 2015. We illustrate the contribution from the ODIAC fossil fuel fluxes alone (Figure 7, upper panel) and together with the biospheric contribution (Figure 7, lower panel). The anthropogenic component shows spatial patterns similar to those observed from the XCO$_2$ anomalies in Figure 3, with high positive enhancements in eastern USA, Europe, India, Middle East and China. In the Southern Hemisphere, the Highveld region in South Africa shows the strongest anthropogenic signal, together with the area around Sidney in Australia. When adding the biospheric component, we also find negative values in the northern mid-latitudes and in the Southern Cone in South America, as observed from the anomalies in Figure 3. In Europe, the large anthropogenic XCO$_2$ enhancements are drawn down by the biospheric sink (Figure 7, lower panel). The largest differences between the patterns in the FLEXPART simulations and the OCO-2 anomalies are in the tropical region, where the OCO-2 XCO$_2$ anomalies show large positive values while the FLEXPART enhancements are close to zero. Part of the reason is that the FLEXPART simulations do not include the biomass burning contribution, but also the biospheric contribution is lower.
Figure 7. FLEXPART simulation results for the year 2015 with contribution from fossil fuel combustion (FF) only and also with biospheric component (FF + BIO).

Figure 8 illustrates the results of the FLEXPART seasonal simulations. We find that the Highveld region is clearly visible during SON and JJA, while it is not detectable during DJF due to effect of the biospheric sink. The same feature is also visible in the seasonal anomalies in Figure 4. In addition, while OCO-2 shows negative XCO$_2$ anomalies over central North America and South Central Asia during the fall (SON) and winter (DJF), the FLEXPART enhancements over both regions are positive.

Four local “case studies” for both FLEXPART enhancements and OCO-2 anomalies are illustrated in Figure 9. The first one is the Iberian peninsula, where OCO-2 XCO$_2$ retrievals are available consistently throughout the year. Here, OCO-2 measurements indicate larger XCO$_2$ anomalies over coastal cities than the model results. The second area is the Highveld industrial region in South Africa. In this case, the anthropogenic signatures seem more localized in the FLEXPART simulations than in the OCO-2 anomaly maps, although positive anomalies are clearly visible over the area as well. The third case study is over India, where we can see a clear positive signal in both OCO-2 and FLEXPART data, but the OCO-2 results indicate more discrete and localized anthropogenic emissions than the model results. The last case is Mexico City, where we also see strong anthropogenic signatures (i.e., positive enhancements and anomalies), related to the emissions from the city as well as the power plants in the area, with some differences in the locations of the positive enhancements. These differences may reflect uncertainties in the emission source intensities or locations or significant local wind data errors at low wind conditions while using wind data from global reanalysis.
Figure 8. FLEXPART seasonal XCO$_2$ enhancement simulations from September 2014 to August 2015, including fossil fuel and biospheric contribution (FF + BIO).
4.4. Case Study: Collocated OCO-2 and TROPOMI Observations in the Highveld Region

We analyse the daily XCO$_2$ anomalies over the Highveld industrial region in South Africa together with NO$_2$ tropospheric columns. The area includes several coal-fired power plants as well as pollution from urban sites. We concentrate the analysis on the month of July 2018, in order to reduce the effect of the CO$_2$ drawdown. We find six direct overlaps with the OCO-2 swath (including a sufficient number of XCO$_2$ retrievals) passing through NO$_2$ local enhancements (plumes). In Figure 10, we illustrate the TROPOMI NO$_2$ tropospheric columns overlaid with local OCO-2 XCO$_2$ anomalies (with background calculated from the values inside a latitude-longitude box $[40^\circ S, 20^\circ S] \times [15^\circ E, 45^\circ E]$). In all six cases, we note clear XCO$_2$ gradients between the area inside the NO$_2$ plume and the surrounding background. On 11 and 20 July 2018, we identify smaller plumes originating from Matimba coal-fired power plant, but also on 13 July 2018 we see fine structures with high XCO$_2$ anomalies over the NO$_2$ plumes.

In order to analyze the cases further, we collocate the TROPOMI NO$_2$ observations to each OCO-2 footprint. In Figure S6, we illustrate the normalized histograms of OCO-2 XCO$_2$ anomalies labeled as “anthropogenic” and “surrounding,” discriminated when tropospheric NO$_2$ values are larger or smaller than $5 \times 10^{15}$ molec./cm$^2$, respectively. In all cases, we note that the anthropogenic histograms peak at higher positive XCO$_2$ anomalies than the histogram for the surrounding. We also note that on 4 July 2018, only a few data points are classified as “anthropogenic”. In Figure 11, we illustrate the correlation between the median NO$_2$ and XCO$_2$ anomaly values inside and outside the NO$_2$ plume as well as the difference between the values inside and outside the plume for each of the six days. We note that the points outside the NO$_2$ plumes (red dots in Figure 11) fall into the bottom-left part of the scatter plot, corresponding to negative XCO$_2$ anomalies and relatively low NO$_2$ levels. Meanwhile, the points falling inside the plume with enhanced NO$_2$ levels (blue dots in Figure 11) show a positive linear correlation between the median NO$_2$ tropospheric columns and XCO$_2$ anomalies. A similar correlation is observed for the differences between the values inside and outside the plume (yellow dots in Figure 11). In both cases, the slope, or the CO$_2$-to-NO$_2$ ratio, is about $-1.5 \times 10^{-16}$ ppm/(molec./cm$^2$). On 11 and 20 July 2018, the OCO-2 flight track passes right through the NO$_2$ plume generated from the Matimba power plant. Figure 12 shows the OCO-2 XCO$_2$ values (black dots) as a function of latitude during both days. Figure 12 (red dots) also includes the XCO$_2$ values obtained by empirically converting TROPOMI NO$_2$ tropospheric columns (collocated to the OCO-2 measurements) into XCO$_2$ using the results of the linear regression between the two data sets. The CO$_2$-to-NO$_2$ ratio in both
days was found to be $\sim 1.1 \times 10^{-16}$ ppm/(molec./cm$^2$). Both XCO$_2$ cross-sections show a similar enhancement when passing through the emission plumes (latitude 24.1°S).

Figure 10. TROPOMI NO$_2$ tropospheric columns and local OCO-2 XCO$_2$ anomalies for six days in July 2018. Positive OCO-2 XCO$_2$ anomalies (yellow-red colors) are visible over the plumes with the largest NO$_2$ enhancements (dark gray areas). Data are overlapped to the Google map background.

Figure 11. Scatter plot between the median XCO$_2$ anomalies and tropospheric NO$_2$ columns inside (blue dots) and outside (red dots) the NO$_2$ plumes. The difference between the values inside and outside the plume are also shown (yellow dots). The corresponding linear fits are also shown (blue and yellow line). The slope (CO$_2$-to-NO$_2$ ratio) in both cases is $\sim 1.5 \times 10^{-16}$ ppm/(molec./cm$^2$).
Figure 12. OCO-2 XCO₂ values (black dots) as a function of latitude on 11 July 2018 and 20 July 2018. The enhancement corresponds to plumes originating from Medupi and Matimba coal-fired power plants. The red dots indicate the XCO₂ values obtained by converting TROPOMI NO₂ tropospheric columns (collocated to the OCO-2 measurements) into XCO₂ using the results of the linear regression between the two data sets. The slope (CO₂-to-NO₂ ratio) is in both cases $\sim 1.1 \times 10^{-16}$ ppm/(molec./cm²).

5. Discussion

Current CO₂ missions have been primarily designed to extend the spatial coverage of the ground-based atmospheric observation networks and to improve the model estimates of biospheric fluxes on regional scale. The COP 21 Paris agreement emphasizes the need to monitor anthropogenic CO₂ emissions over a range of scales [35]. The anomaly approach presented here exploits the intrinsic value and capabilities of satellite-based CO₂ observations for mapping anthropogenic and natural emission patterns, beyond their application as model input information. The observed consistency between satellite-based XCO₂ anomalies and model outputs (distinguishing fossil fuel and biospheric contributions) demonstrates the capability of satellite observations for describing the CO₂ spatio-temporal variability and, in particular, for detecting anthropogenic CO₂ emission patterns.

A feature of the anomaly approach is that the XCO₂ anomalies (given in ppms) are not directly convertible to fluxes (given in ktons/day). The approach often taken with short-lived air pollutants is to use a statistical model that describes their spatial distribution near the emission sources as a function of wind speed and direction (e.g., [36,37]). In the future, a similar approach might also be possible with XCO₂ anomalies, for example using CO₂ measuring satellites with wider swaths, such as the anthropogenic CO₂ monitoring satellite constellation planned by the European Copernicus Programme. In this context, the results presented here highlight the need for satellite observations with high precision, good accuracy and dense sampling covering a wider swath (e.g., few hundred km) than OCO-2, in order to separate individual emission sources (e.g., cities or large power plants) from the background signal. Here we showed how overlapping OCO-2 XCO₂ data and TROPOMI NO₂ observations (covering a large swath) can be used to identify small scale anthropogenic CO₂ signatures.

The anomaly approach is also a useful tool for identifying CO₂ fluxes associated with the natural carbon cycle. For example, the XCO₂ anomaly maps show persistent positive XCO₂ anomalies (1–2 ppm) over the Amazon, Congo, and tropical Oceania. The intensity and spatial extent of the positive XCO₂ anomaly is largest during the 2015–2016 El Niño, but it spanned every season of every year included in this study. This appears to support the conclusions of Baccini et al. [38] who assert that the tropical deforestation and degradation have compromised these forests to the extent that they are now net sources instead of sinks of carbon, even if this conclusion is still under debate [39]. A recent study by Pugh et al. [40] also indicates that contrary to previous findings, the sink is predominantly in mid-high latitude, rather than tropical, forests.
The method presented here can also be applied to future planned XCO₂ missions, such as OCO-3, GOSAT-2, MicroCarb, GeoCarb, and the Chinese and European wide-swath constellations. The global approach can be easily applied to instruments that fly in near-polar, sun-synchronous orbits, however, adjustments to background definition are needed for instruments operating on the International Space Station (e.g., OCO-3) or on geostationary orbit (e.g., GeoCarb) due to the possible influence of diurnal variations and spatial coverage. Further applications of this method could combine anomalies from similar platforms, e.g. belonging to the same constellation.

6. Conclusions

In this paper, we analyzed four years (2015–2018) of Orbiting Carbon Observatory-2 (OCO-2) data to derive global (60°S–60°N) XCO₂ anomalies. We calculated regional-scale XCO₂ anomalies and identified the main anthropogenic emission areas, biomass burning regions, and biospheric sinks. We find that the patterns observed in OCO-2 annual XCO₂ anomalies are robust and consistent from year-to-year. From the seasonal XCO₂ anomalies, we identified the patterns in the Northern Hemispheric growing season and also the different fire seasons in Africa. In addition to large-scale features, we also visualized different local “case studies” with high anomalies associated with anthropogenic emissions in both the Northern and Southern Hemispheres. Finally, we combined daily XCO₂ anomalies with TROPOSpheric Monitoring Instrument (TROPOMI) NO₂ tropospheric columns, used as an indicator of anthropogenic pollution plumes, over the Highveld industrial region in South Africa. We find corresponding gradients and positive correlation between XCO₂ anomalies and NO₂ enhancements.

From a methodological point of view, instrument sampling, background choice, and bias correction do have an impact on the OCO-2 XCO₂ anomaly patterns. Thus, the results are most significant over areas where more data are available throughout the year. The consistency of the spatial features from year to year (or for the same season during different years) is an indicator of the robustness of the results. The FLEXible PARTicle dispersion model (FLEXPART) simulations also confirm the observed spatial patterns. On the other hand, the error associated with the anomaly is not purely statistical and tends to be more systematic due to errors in assumed source location, intensity, persistent wind patterns, and uneven geographic distribution of OCO-2 observations. For example, OCO-2 data in the northern high- and middle-latitudes are available predominantly during summer months so the annual maps are likely to be biased towards the anomalies derived during this period.

We showed that using a piecewise linear background for the calculation of the anomalies provides a robust tool for global analysis. We have tested different background choices, as well as different OCO-2 data versions (V7r, V8r). We conclude that the structure and amplitude of the anomalies is relatively insensitive to these choices, although, for example, using narrow latitude bands without interpolation would yield sharp latitudinal boundaries in the maps. When analyzing local case studies on regional scales, a different background region can be selected, as shown for example in the South Africa’s case study (Section 4.4) or by Wang et al. [17] in China.

Supplementary Materials: The following are available online at http://www.mdpi.com/2072-4292/11/7/850/s1, Figure S1: OCO-2 seasonal cycle at 10 degree latitude bands and latitudinal gradient at selected days between September 2015 and September 2016, Figure S2: Number of data points on each 1° × 1° grid cell, Figure S3: Standard error of the mean on each 1° × 1° grid cell, Figure S4: Number of data points on each 1° × 1° grid cell at each season from September 2014 to August 2018, Figure S5: Standard error of the mean for the seasonal XCO₂ anomalies 2014–2018, Figure S6: Histograms of XCO₂ anomalies inside and outside the NO₂ plumes.

Author Contributions: J.H. and I.I. analyzed the data and wrote the first draft of the manuscript. S.H. and D.C. provided data and expertise on FLEXPART and OCO-2, respectively, and helped in finalizing the paper.

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

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