Article

Estimating CO₂ Emissions from Large Scale Coal-Fired Power Plants Using OCO-2 Observations and Emission Inventories

Yaqin Hu ^{1,2} and Yusheng Shi ^{1,*}

¹ Aerospace Information Research Institute, Chinese Academy of Sciences, Beijing 100101, China; huyaqin@mailsucas.ac.cn

² University of Chinese Academy of Sciences, Beijing 101408, China

* Correspondence: shiys@aircas.ac.cn; Tel.: +86-138-1042-0401

Abstract: The concentration of atmospheric carbon dioxide (CO₂) has increased rapidly worldwide, aggravating the global greenhouse effect, and coal-fired power plants are one of the biggest contributors of greenhouse gas emissions in China. However, efficient methods that can quantify CO₂ emissions from individual coal-fired power plants with high accuracy are needed. In this study, we estimated the CO₂ emissions of large-scale coal-fired power plants using Orbiting Carbon Observatory-2 (OCO-2) satellite data based on remote sensing inversions and bottom-up methods. First, we mapped the distribution of coal-fired power plants, displaying the total installed capacity, and identified two appropriate targets, the Waigaoqiao and Qinbei power plants in Shanghai and Henan, respectively. Then, an improved Gaussian plume model method was applied for CO₂ emission estimations, with input parameters including the geographic coordinates of point sources, wind vectors from the atmospheric reanalysis of the global climate, and OCO-2 observations. The application of the Gaussian model was improved by using wind data with higher temporal and spatial resolutions, employing the physically based unit conversion method, and interpolating OCO-2 observations into different resolutions. Consequently, CO₂ emissions were estimated to be 23.06 ± 2.82 (95% CI) Mt/yr using the Gaussian model and 16.28 Mt/yr using the bottom-up method for the Waigaoqiao Power Plant, and 14.58 ± 3.37 (95% CI) and 14.08 Mt/yr for the Qinbei Power Plant, respectively. These estimates were compared with three standard databases for validation: the Carbon Monitoring for Action database, the China coal-fired Power Plant Emissions Database, and the Carbon Brief database. The comparison found that previous emission inventories spanning different time frames might have overestimated the CO₂ emissions of one of two Chinese power plants on the two days that the measurements were made. Our study contributes to quantifying CO₂ emissions from point sources and helps in advancing satellite-based monitoring techniques of emission sources in the future; this helps in reducing errors due to human intervention in bottom-up statistical methods.

Keywords: carbon dioxide; Gaussian model; point source; Orbiting Carbon Observatory-2

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1. Introduction

Anthropogenic carbon dioxide (CO₂) emissions from industries (including fossil fuel burning) are the dominant contributors to the observed enhancement in global atmospheric CO₂ concentration [1], and the increase of CO₂ emissions account for approximately 78% of the total increase in greenhouse gas (GHG) emissions. As the most important GHG, CO₂ is the main driver of global climate change [2,3]. To date, 189 member states of the United Nations Framework Convention on Climate Change (UNFCCC) have achieved consensus on the Paris Agreement, which aims to reduce the incremental increase of the average temperature of earth's surface to no more than 2 °C higher than when global industrialization began [4]. China has been the largest emitter of CO₂ worldwide since 2006, and its emissions have been more than twice that of the next highest country, the United States, as of 2018 [5].

Based on the Emissions Database for Global Atmospheric Research (EDGAR) [6], the Science for Policy report by the Joint Research Center (JRC) indicates that of the main emission-producing sectors, the power industry has been the largest contributor to CO₂ emissions since 1990 [5]. In China, electrical equipment lags in efficiency; thus, CO₂ emissions from coal-fired power stations account for 80% of the nation's electricity sector emissions (the world average is 40%). Solving the environmental problems caused by the coal electric power industry has great significance for sustainable development and the mitigation of global climate change [7]. Consequently, the methods for estimating coal-burning power plants CO₂ emissions at the unit level must be improved to provide a reasonable scientific reference for the formulation of CO₂ emission reduction countermeasures for countries without well-established emission reporting systems [8].

There is increasing interest in estimating the CO₂ emissions from power plants in China. However, no global observation system for accurately monitoring point-source CO₂ emissions was previously available [9]. National CO₂ emission inventories of power plants are primarily obtained by statistical methods on account of factors such as the consumption of fossil fuel, the type of consumption, the purity and mix of fuel, and efficiency [10,11]. Therefore, the estimation of anthropogenic emissions at fine scales generally has a large uncertainty regarding the collection of accurate consumption data. With the launch of high-precision carbon satellite missions, such as Japan's Greenhouse Gases Observing Satellite (GOSAT) [12], the Orbiting Carbon Observatory-2 (OCO-2) [13] of the National Aeronautics and Space Administration (NASA), and other satellites, the remote sensing top-down retrieval method for quantifying CO₂ emissions has proven to be an effective method. OCO-2 incorporates three high-resolution spectrometers that take synchronous measurements of reflected sunlight; for CO₂, measurements are taken at the 1.61 and 2.06 μm bands in the shortwave infrared (SWIR) region, while measurements for oxygen (O₂) are taken at the 0.76 μm band in the near-infrared (NIR) region. These measurements are used to retrieve the average dry air mole fraction of the CO₂ column (herein, XCO₂). The precisions of GOSAT and OCO-2 are approximately 1–2 ppm and 1 ppm (or 0.25%), respectively [12,14].

Multiple studies have focused on estimating anthropogenic CO₂ flux on relatively larger spatial scales using GOSAT and OCO-2 data [15–19]. However, there is currently insufficient research on the estimation of emissions at point scales using remote sensing inversion methods, especially for power plants with high carbon emission intensities in China. Initially, a promising concept proposed by Bovensmann et al. [20] suggested that remote sensing satellites could detect strong localized CO₂ point sources and quantify their emissions. However, this achievement demands imaging with a spatial resolution of $2 \times 2 \text{ km}^2$ for satellites to map the concentration distribution of the atmospheric CO₂ column with an accuracy of 0.5% (2 ppm) or better. The CO₂ emission problem was solved on the scale of a single facility (power plant) for the first time by Nassar et al. [21] using a Gaussian plume model based on OCO-2 data with an assumed constant wind field. The gap between the estimated emissions of American power plants and the daily reported emissions was 1–17%. Following [21], Krings et al. [22] inferred the CO₂ emission estimation of a group of coal-fired power plants in a complex study area by combining the mass balance and inverse Gaussian plume model method. The data were collected through aerial remote sensing observation and airborne in-situ measurements of CO₂ with the MAMAP instrument, which is an airborne double-channel NIR–SWIR grating spectrometer that is used to accurately measure column-averaged gradients of CO₂ and methane concentrations.

Further, a high-resolution chemical transport model has been applied to evaluate the CO₂ emissions of power plants. Zheng et al. [23] used the Weather Research and Forecasting-Chem (WRF-Chem) simulation method, which provided a framework for addressing different wind fields, a method which could be extended to urban-level emission estimation. In addition, the WRF-Chem modeling system of complex components was designed to simulate the increase of CO₂ [23], yet its estimates of CO₂ emissions for the

target power plant did not show improved consistency with the reported value compared to Nassar's research [21], which used a relatively simple plume model. As for Krings' research [22], although more reliable observation data was obtained from the MAMAP instrument, which had higher precision (approximately 0.4 ppm) compared to OCO-2 data, air-based observations were not available for large-scale measurement; thus, national range point emissions estimates could not be achieved.

Therefore, this study adopted an improved Gaussian plume model method to estimate the CO₂ emissions at the unit level. By applying wind data with higher time resolution, the uncertainty of wind could be effectively reduced, which was the largest source of uncertainty in the estimation. Moreover, a physically based unit conversion method was used to enhance the applicability of the model for removing the limitations of terrain. In addition, the correlation of fitting was significantly improved by interpolating the observed data of OCO-2 at different resolutions. Overall, using this remote sensing satellite inversion method, we achieved the higher-accuracy conversion of OCO-2 satellite observation parameters of the CO₂ column to point-source emissions for large coal-fired power plants. We then compared these emission estimates with selected CO₂ emission inventories of previous statistics, as well as annual emissions calculated using the bottom-up statistical method. Finally, we briefly discuss the potential of future CO₂ satellites for CO₂ emission data retrievals.

2. Materials and Methods

2.1. Data

2.1.1. Power Plant Data

We obtained a list of thermal power plants in China by combining the Global Power Plant database (<https://datasets.wri.org/dataset/globalpowerplantdatabase> accessed on 23 June 2021) with Global Energy Observatory databases (GEO) (www.globalenergyobservatory.org, accessed on 23 June 2021). The list included geographic coordinates, total installed capacity, and estimated annual power generation. Further, the Carbon Brief database was used (<https://www.carbonbrief.org/mapped-worlds-coal-power-plants>, accessed on 23 June 2021) to obtain the distribution of large- and medium-sized power plants in China and their reported CO₂ emission information, in order to select the research objects of interest.

2.1.2. Orbiting Carbon Observatory-2 (OCO-2) Data

NASA's first mission for CO₂ monitoring, which was successfully launched on 2 July 2014, OCO-2, collects nearly one million observations over the hemisphere lit by sunlight routinely each day. The primary product delivered by OCO-2 is XCO₂, which is estimated by calculating the ratio of the CO₂ and O₂ column integral densities along the light path between the sun, surface detection, and instrument and then multiplying the values by the column's average concentration of O₂ (0.2095) [13,14,24]. In the spatial domain, the OCO-2 instrument has a narrow swath (0.8° width). It collects eight soundings over its swath every 0.333 s, detecting the earth surface at nadir mode with along-track dimensions < 2.25 km and cross-track dimensions ranging from 0.1 to 1.3 km. The data are available from 6 September 2014 to the present.

In this study, the data used, from OCO-2 L2 Lite FP V10r, was stored in NetCDF4 format (<https://disc.gsfc.nasa.gov/OCO-2>, accessed on 23 June 2021) and features bias-corrected data of quantified values, such as geolocation information, XCO₂, and atmospheric precipitable water vapor. By retrieving the OCO-2 standard data (inclusive of September 2014 to September 2020), we screened OCO-2 flybys near the power plants of interest, which contained sufficient high-quality soundings to read the XCO₂ data.

2.1.3. Wind Data

Wind speed and direction play a vital role in CO₂ diffusion simulation, which determines the speed and direction of CO₂ spread. The speed and direction of the wind are derived from the U and V wind vectors of the fifth generation European Center for

Medium-Range Weather Forecasts (ECMWF) atmospheric reanalysis of the global climate (ERA5) [25] as well as the Modern-Era [since 1980] Retrospective analysis for Research and Applications, Version 2 (MERRA-2) dataset (Global Modeling and Assimilation Office [26,27] ($0.5^\circ \times 0.625^\circ$, 72 levels, every 3 h) for validation. ERA5 provides data on multiple atmospheric, land-surface, and sea-state parameters, along with estimates of uncertainty per hour ($0.25^\circ \times 0.25^\circ$ resolution). Based on the assimilated 4D-Var data from the Integrated Forecast System (IFS) of ECMWF, ERA5 atmospheric parameters are available at 137 mixed sigma/pressure (model) levels in the vertical direction and then interpolated to 37 pressure levels [28]. Compared to the fourth-generation reanalysis data known as ERA-Interim, the horizontal and vertical resolutions of ERA5 data are significantly improved; furthermore, the time interval of reporting has transformed from 6 h to 1 h, and the overall data accuracy is also improved. These remarkable improvements make ERA5 data the best global atmospheric reanalysis data for climate research worldwide.

2.2. Gaussian Plume Model

Gaussian mathematical models are considered reliable for simulating pollutant movement and spread and are widely used at point scales or local levels. The Gaussian method is based on analytical formulas of plume distribution [29]. In this study, the Gaussian plume model was used to estimate the CO₂ emissions of an individual power plant by simulating the CO₂ dispersion process, as shown in Equations (1) and (2):

$$V(x, y) = \frac{F}{\sqrt{2\pi}\sigma_y(x)u} e^{-\frac{1}{2}\left(\frac{y}{\sigma_y(x)}\right)^2}, \quad (1)$$

$$\sigma_y(x) = a \cdot x^{0.894} \quad (2)$$

where $V(x, y)$ represents the vertical column (g/m^2) at coordinates (x, y) , and the variables x , y , and u refer to the along-wind distance (m), the across-wind distance (m), and wind speed (m/s), respectively. F is the release rate to be estimated (g/s), and $\sigma_y(x)$ conveys the standard deviation along the direction across the wind, reflecting the CO₂ diffusivity perpendicular to the wind. The standard deviation $\sigma_y(x)$ is a function of x and is related to atmospheric stability (here x is specified in kilometers to give $\sigma_y(x)$ in meters). a is determined by ranking the atmospheric stability near the power plant based on the Pasquill–Gifford stability [30], which relies on cloud cover, wind speed near the surface, and solar elevation angle, and the system consists of five classes with values 213, 184.5, 156, 130, and 104 [31]. Because the OCO-2 does not take measurements in dark or cloudy skies, the condition of cloud cover was not considered. Surface wind speed was collected from ERA5 data, and the solar elevation angle for each sounding was obtained from OCO-2 files.

CO₂ emissions are estimated within the plume region by linear least square fitting between the simulated enhancement of the Gaussian plume and the observed XCO₂ enhancement due to the power plant (the regressor is the simulated enhancement, and the response is the observed XCO₂ enhancement). The model plume is defined as the area from the x -axis (wind vector) down to a threshold of 1% intensity in the positive/negative y directions (the area where CO₂ concentration increases sharply). The simulated value is calculated using Equation (1), wherein the wind speed and direction are transformed from the UV vector of the ERA5 wind data and then linearly interpolated temporally and spatially to the sounding time and the stack height (240 m by default if there is no relevant information), respectively. The observed enhancement is achieved by subtracting the background value from the observed XCO₂. To determine the background, we selected an area of the OCO-2 swath not affected by the plume as the background (generally in the upwind vicinity of emission sources) with reference to [32] and obtained the background value by averaging the XCO₂ of every sounding in this area.

According to Bovensmann et al. [20], the higher the resolution, the more detailed the demonstrated plume; thus, more accurate fitting results can be obtained. For every sounding, each dimension of OCO-2 data was interpolated to a different resolution by the

Kriging interpolation method, which is most suitable for simulated smooth plumes [33]. After interpolation, not only can more plume points for regression be obtained but retrievals of CO₂ columns can also be better revealed by the corresponding interpolated values of the imaging area, thus reducing the edge effect of adjacent soundings. It should be noted that more fitting plume points and more practical observations for simulation would make the estimated emissions increasingly convincing.

Note that the units of observed and simulated enhancement are different; thus, we applied Equation (3) to convert $V(x, y)$ (g/m²) to XCO₂ (ppm):

$$XCO_2 = V(x, y) \cdot \frac{M_{air}}{M_{CO_2}} \cdot \frac{g}{P_{surf} - w \cdot g} \cdot 1000 \quad (3)$$

where M_{air} , M_{CO_2} , and g are three constants, representing the molecular weights (kg/mol) of air, CO₂, and the gravitational acceleration (m/s²), respectively; P_{surf} is the surface pressure (Pa), and w means the total water vapor of atmosphere column (kg/m²). P_{surf} and w are both derived from OCO-2 data.

2.3. Bottom-Up Estimates

The former power plant emission inventory of CO₂ in China at the unit level is the Carbon Monitoring for Action (CARMA) database [34,35] which was operated by a prominent Washington, D.C.-based think-tank, the Center for Global Development (CGD). The existing inventories include the China coal-fired Power Plant Emissions Database (CPED) [36], which is supported by the European Union Seventh Research Framework Programme (FP-7) MarcoPolo, and the Carbon Brief database. From CARMA, information was only gathered for the years 2004 and 2009, along with estimates of future production (from the latter date onwards), while CPED contains 7657 coal-fired power plants in mainland China, which comprised ~5700 units operating and ~1900 units closing. Therefore, these two databases were only partially suitable for verification in this study; this was attributed to the successful launch of OCO-2 in 2014. However, the Carbon Brief database (2000–2020) lacked specificity for each power plant, that is, the estimation details were not adjusted according to the different years.

To verify our estimated CO₂ emissions, we applied the approach used by the Global Energy Monitor [37] to roughly estimate the annual CO₂ emissions of an individual power plant, as shown in Equation (4):

$$E_{CO_2s,k} = C \cdot CF \cdot HR_{s,k} \cdot EF_{CO_2k} \cdot 9.2427 \times 10^{12}, \quad (4)$$

where s represents the combustion technology type, and k represents different coal types. The E_{CO_2} is the annual CO₂ emission to be estimated in Mt from a proposed coal plant, which depends on the following parameters: C , which is the capacity of a power plant in MW; CF , which is the capacity factor, measuring the ratio of the actual power generation and the generating power when the coal power plant operates uninterrupted at its rated capacity as determined by its capacity and practicality; estimates of the generated energy was mainly obtained from the information given in listed company announcements; $HR_{s,k}$, which is the heat rate in Btu/kWh, measuring the efficiency of a coal-fired power plant to transform the energy of coal into electricity, determined by the combustion technology type, the coal type, and the capacity of the generation, and the values used in this study are from Sargent and Lundy [38] and Beér [39]; EF_{CO_2k} , which is the CO₂ emission factor varying with the coal type in kg/TJ from the IPCC [40], and the constant term, which is a coefficient that allows the emissions to be described in Mt.

3. Results

3.1. Screening

The Chinese government has promulgated policies to promote extra-large-sized and large-sized power plants and to eliminate small ones. The share of large units (generating

power ≥ 300 MW) expanded rapidly from 18% to 74% between 1990 and 2010, whereas the proportion of small-sized units (generating power < 100 MW) decreased to 9% [36]. Hence, our study focuses on large and medium-sized coal-fired power plants. The data used for retrieval are based on the standard data from the OCO-2 official website, and a refined range of $0.5^\circ \times 0.5^\circ$ was considered around the power plant. Eight medium- and large-sized power plants were selected to verify the feasibility of the method and the reliability of the estimation. In the process of targeted power plant screening, several conditions needed to be satisfied. First, at least one or more overpasses were relatively close to the proposed power plant in the historical data of OCO-2. In addition, at the overpass hour, the enhancement of XCO_2 caused by CO_2 emission plumes, under the influence of the dominant wind direction, was able to be observed from the OCO-2 swath.

3.2. Configuration

For the large-sized power plants, two representative plants were screened: the Waigaoqiao Power Plant (hereinafter, Waigaoqiao) in Shanghai Province and the Qinbei Power Plant (hereinafter, Qinbei) in Henan Province (Figure 1). Waigaoqiao generates 5000 MW of power. The distribution of all power plants in China is shown in Figure 1, while information about the OCO-2 flyby is shown in Table 1. For Waigaoqiao, the flight path of a satisfactory flyby of OCO-2 on 12 March 2015, and the relevant wind direction at that overpass hour, is shown in Figure 2a. The nearest distance between the power plant and the OCO-2 swath was approximately 2.3 km. From the OCO-2 swath, an increase in the retrieved XCO_2 within the CO_2 emission plume region can be clearly observed (1–6 ppm). However, some disadvantages exist for this overpass. After meeting the above two requirements, the wind direction was not approximately parallel to the OCO-2 swath but instead intersected with the swath direction at a certain angle ($\sim 45^\circ$), as shown in Figure 2a. Under the observation conditions, only 29 soundings were within the plume area, indicating that the regression could only be performed for 29 sets of corresponding values. In applying these 29 points for estimation, the correlation of later regression was greatly reduced, making the estimation of the CO_2 emissions less reliable. In addition, we noticed that the enhancement compared to the regional upwind was not only detected in the plume area corresponding to the wind direction but also shown in the OCO-2 swath, which was consistent with the wind direction (Figure 3) (in an ideal state, XCO_2 distribution of the region resemble that of the upwind area). We have checked Google Earth to confirm that there is no other industrial emission source near Waigaoqiao. Based on the analysis of the wind-direction data before and after the overpass hour, it was found that the above-mentioned phenomenon is caused by the residual effect of wind before the soundings of OCO-2. Therefore, we included the residual effect of wind in the calculation of the background value by combining the downwind area with the upwind area. Certain parameters needed for the estimation process are shown in Figure 2.

Table 1. Elementary information for two coal-fired power plants.

Coal Power Plant	Province	Capacity (MW)	Date	OCO-2 Mode	Overpass Hour in UTC	Configuration	Number of OCO-2 Points in Plume
Waigaoqiao	Shanghai	5000	12 March 2015	Glint	5:01	Flyby (~2.3 km)	29
Qinbei	Henan	4400	24 November 2017	Glint	5:27	Overpass	6

Figure 1. Distribution of coal-fired power plants in China (left image); the size of the circles indicates the scale of the power generation. The capacities of different-sized power plants are as follows: extra-large-sized, >5000 MW; large-sized, 3000–5000 MW; medium-sized, 1000–3000 MW, and small-sized, <1000 MW. Aerial images of two power plants are on the right.

Figure 2. (a) XCO₂ from the OCO-2 overpass near the Waigaoqiao Power Plant was displayed with ERA5 wind vectors (red arrow) with simulated XCO₂ in the left corner. Wd: wind direction, Ws: wind speed, SEA: solar elevation angle, a: atmospheric stability parameters, and bg: background. (b–e) XCO₂ observation data was interpolated to 1.00 × 1.00, 0.75 × 0.75, 0.50 × 0.50, and 0.25 × 0.25 km², respectively, showing plume range and wind direction (red lines). (f) Plume points (red), background points (black), other observations in the swath (blue), and the background mean (orange line) of XCO₂ at the resolution of 0.75 × 0.75 km².

Figure 3. Wind direction before and after the overpass hour (from 23:00 on 11 March to 12:00 on 12 March). The arrows display the wind direction.

As for the Waigaoqiao power plant, an insufficient number of plume points could be detected for the Qinbei Power Plant, which generates 4400 MW of power. The OCO-2 screened flyby for Qinbei on 24 November 2017 is shown in Figure 4a, and the wind direction at the overpass hour was nearly perpendicular to the OCO-2 swath. For both plants, we noticed a large deviation between the footprint center given by OCO-2 data and the actual observation range in the OCO-2 swath (Figure 5). In fact, the deviation is related to misalignment between the OCO-2 spectrometers for each band resulting in pointing errors [41] and is largest in the presence of steep topography of the type found to the north of the Qinbei power plant (according to Google Earth elevation). However, the observations corresponding to each sounding were allocated by the given geographic coordinates at the center of the sounding field-of-view. Such deviations lead to the misallocation of observations. According to the formula of the polygon centroid, we adjusted each sounding center; thus, the adjusted coordinates were located at the geometric center of the parallelogram defining each field-of-view. However, in adopting the revised center coordinates for each sounding, only six soundings could be divided into plumes. By adopting only six plume points for regression, the estimated emissions would have an order of magnitude gap with the reported value. This situation was attributed to not only the low number of corresponding points but also to the relatively large resolution of the OCO-2 field-of-view. According to the wind speed and atmospheric stability at the overpass hour, the range of the CO₂ emission plume was relatively narrow, indicating that the simulated value would be expected to increase or decrease rapidly as the cross-wind distance changed. In contrast, the XCO₂ of OCO-2 did not vary in a sounding parallelogram because of the OCO-2 field-of-view. This contradiction leads to a weaker correspondence and correlation between the simulated value and the observed value, indicating that the simulation result would not be sufficiently accurate for practical use.

Figure 4. (a) XCO₂ from OCO-2 overpass near the Qinbei Power Plant displayed with ERA5 wind vectors (blue arrow) and simulated XCO₂ displayed in the left corner. (b–d) XCO₂ observation data interpolated to 0.75×0.75 , 0.50×0.50 , and 0.25×0.25 km², respectively, showing plume range and wind direction (blue lines). (e) Plume points (red), background points (black), other observations in the swath (blue), and the background mean (orange line) of XCO₂ at the resolution of 0.50×0.50 km².

3.3. Estimated Emissions

As described in Section 2.2, the Kriging interpolation method was applied to OCO-2 data to increase the number of plume points and improve the correlation coefficient of the linear regression. In the CO₂ plume of Waigaoqiao, when OCO-2 soundings were interpolated to resolutions of 1.0×1.0 , 0.75×0.75 , 0.5×0.5 , and 0.25×0.25 km² with a maximum searching distance of 0.5 km (Figure 2b–e), after simple filtering, 31, 75, 102, and 438 observed values could be applied to the fitting with simulated values. The improvement for fitting can be clearly observed by comparing Figure 2a–e. The former shows a sharp increase of CO₂ column concentration within the plume region, while the latter further demonstrates that the decrease of CO₂ concentration corresponds with an increase of the distance between the point source and each sounding; this is consistent with CO₂ diffusion simulated by the Gaussian model.

For extrapolating instantaneous flux data to annual mean emissions, the temporal variability of emissions must be considered. As for the diurnal cycle of CO₂ emissions, there are no CO₂ on-site monitoring systems for power plants in China, so we collected emission information on NO_x from power plants considering that NO_x is co-emitted with CO₂ during fossil-fuel combustion. China's continuous emissions monitoring systems (CEMS) network (<http://www.envsc.cn/>, accessed on 23 June 2021) provide the direct, actual, real-time measurements of emission concentrations for a variety of air pollutants at power plant stacks nationwide. The gathered NO_x emissions through field measurements indicated that the power plant's operating power within a day is relatively constant especially for large coal power plants. The tiny variation in concentration doesn't affect our ability to extrapolate instantaneous estimation to daily emissions. According to the quarterly power generation of power plants, we extended daily emissions to annual emissions.

The quantified emissions using OCO-2 V10 data from Waigaoqiao, based on different interpolation resolutions, were 22.57, 23.00, 23.06, and 21.99 Mt/yr (Table 2), respectively. Different estimations of our study and the correlation coefficient (R) of our Gaussian model method are shown in Table 2. The estimations for Qinbei are also shown in Table 2. The CO₂ emissions for Qinbei could not be estimated with the column concentration of OCO-2 at the resolution of 1.0×1.0 , 0.75×0.75 km² due to the lack of plume points.

3.4. Bottom-Up Estimations

To verify our estimations, the bottom-up estimation method mentioned in Section 2.3 was conducted to quantify the emissions of the Waigaoqiao and Qinbei power plants. Phase I of Waigaoqiao was established in 1993, including four domestic subcritical units (300 MW); phase II was built in 2004, including two imported supercritical units (900 MW); and phase III was built in 2008, including two ultra-supercritical units (1000 MW). As for Qinbei, phase I was constructed in 2004 and comprised two supercritical units (600 MW); phase II was built in 2007, with two supercritical units (600 MW), and phase III was added in 2013, with two ultra-supercritical units (1000 MW). The parameters of the bottom-up method for the estimation for the two power plants are shown in Table 3.

The estimation for Waigaoqiao is 16.28 Mt/yr and is 14.08 Mt/yr for Qinbei. In addition, other studies have performed bottom-up estimations of the power plants investigated in this study. The estimations of CARMA for the future, CPED for 2010, and Carbon Brief for 2015 were 21.2, 25.4, and 24.7 Mt/yr for Waigaoqiao, respectively. However, 5–6 units at Qinbei were constructed in 2013, whereas the CO₂ emissions collected from CPED were from no later than 2010. Thus, the estimate for CPED was obtained in proportion to its capacity ratio in 2010 and 2017. Figure 6 shows the CO₂ emissions estimated via different methods and based on data from different sources for these two power plants.

Table 2. Correlation coefficients and estimates at different resolutions for two power plants.

Power Plant	Resolution	Number of OCO-2 Points in Plume	R	Annual CO ₂ Emissions (Mt/yr)
Waigaoqiao	1.00×1.00 km ²	31	R ₁ = 0.75	22.57
	0.75×0.75 km ²	75	R ₂ = 0.83	23.09
	0.50×0.50 km ²	102	R ₃ = 0.90	23.06
	0.25×0.25 km ²	438	R ₄ = 0.86	21.99
Qinbei	0.75×0.75 km ²	19	–	–
	0.50×0.50 km ²	41	R ₂ = 0.71	14.86
	0.25×0.25 km ²	80	R ₃ = 0.79	14.58

Table 3. Estimation details of the bottom-up method for two power plants.

Power Plant	Units	Capacity (MW)	Power Generation (TWh)	Capacity Factor (%)	Heat Rate (Btu/kWh)	Emission Factor (kg/TJ)	CO ₂ Emissions (Mt/yr)
Waigaoqiao	1–4	300	3.12	29.70	8863	94,600	16.28
	5–6	900	7.82	49.61	8540		
	7–8	1000	9.59	54.72	7896		
Qinbei	1–2	600	16.60	43.07	8564	94,600	14.08
	3–4	600			8564		
	5–6	1000			8418

Figure 5. Left: coordinate deviation, right: coordinates after bias-correction (the lines form parallelograms of the soundings and the red dots represent the center coordinates).

Figure 6. Comparison of CO₂ emissions with different methods for Waigaoqiao and Qinbei power plants.

4. Discussion

4.1. Power Plant Screening

In the screening process for large-size power plants, in addition to Waigaoqiao and Qinbei, we considered six other power plants: Tuoketuo ($111^{\circ}21' \text{ E}$, $40^{\circ}12' \text{ N}$), Guohua Taishan ($112^{\circ}55' \text{ E}$, $21^{\circ}52' \text{ N}$), Huaneng Yueyang ($113^{\circ}10' \text{ E}$, $29^{\circ}27' \text{ N}$), Shanxi Zouxian ($116^{\circ}56' \text{ E}$, $35^{\circ}20' \text{ N}$), Yangcheng ($112^{\circ}35' \text{ E}$, $35^{\circ}28' \text{ N}$), and Yaomeng ($113^{\circ}15' \text{ E}$, $33^{\circ}44' \text{ N}$). By analyzing close flybys or direct overpasses of these power plants, we concluded that there were no OCO-2 data available for these power plants to retrieve CO_2 emissions, a problem which was also noted by Zheng et al. [42]. Considering the Tuoketuo Power Plant in Inner Mongolia as an example, within the region centered at the power plant set up for filtering, 23 OCO-2 overpasses were screened out between September 2014 and September 2020. Among the 23 overflights, 13 had no retrievals around the power plant, six swaths were excessively far from the plant ($\sim 10\text{--}25 \text{ km}$), and two occurred over the power plant, but were missing a large fraction of soundings close to the point of power generation, which is imperative in emissions estimation. The remaining two swaths were extremely close to the plant (~ 1 and $\sim 2.5 \text{ km}$, respectively) with an adequate amount of data, yet the plume areas did not intersect with the corresponding OCO-2 swaths. The lack of available OCO-2 data for these plants is primarily attributed to the sounding limitation, such that OCO-2 yields 4–8 cross-track footprints along the spectrometer slit with dimensions $\leq 1.29 \times 2.25 \text{ km}^2$. Such a narrow swath leads to fewer close flybys or direct overpasses; thus, XCO_2 enhancement caused by power plants is less likely to be observed. In addition, the lack of available data could be partly because of the ABO2 algorithm ($\text{O}_2 \text{ A}$ band cloud screening algorithm) removing data from the OCO-2 data processing to avoid wrong retrieval results of corresponding footprints with excessive interference of clouds and aerosols; this is common since approximately 70% of Earth is covered by clouds at any given moment [43].

4.2. Estimation Details and Validation of Emissions

Concerning the correlation coefficient, fitting coefficients in our top-down estimation were much higher than those from Nassar et al. [21]. In terms of CO_2 emissions estimation, the pixel size of the OCO-2 soundings after interpolation had little effect on the estimated values; however, the resolution influenced the correlation of regression. As previously described in Section 3 for Waigaoqiao, the correlation coefficient was 0.69 in the absence of interpolation for OCO-2 data; however, the correlation coefficients using interpolated OCO-2 retrievals were 0.75, 0.83, 0.90, and 0.86, at resolutions of 1.0×1.0 , 0.75×0.75 , 0.5×0.5 , and $0.25 \times 0.25 \text{ km}^2$, respectively. This improvement demonstrates that the interpolation method can enable some cancellation of random noise. After interpolating, the retrievals of OCO-2 could better match the in-situ values simulated by the Gaussian model, and furthermore, could reduce the influence of excessive deviation between the observed values of boundary points and the practical values resulting from the abrupt change of the retrievals for adjacent pixels, which significantly promotes the correlation. Nevertheless, the resolution after interpolation has restrictions. When the OCO-2 retrievals are interpolated to each pixel whose area is less than $0.25 \times 0.25 \text{ km}^2$, the correlation can no longer be improved; this stems from the restriction of the OCO-2 resolution. Thus, the observations with higher resolution cannot be clearly expressed, and the interpolated XCO_2 cannot produce any significant changes. As shown in Figure 7 (considering Waigaoqiao for an example), observations for a simulation with different geographic coordinates share the same parameters, such as column concentration and surface pressure, especially in the 0.5×0.5 and $0.25 \times 0.25 \text{ km}^2$ figures. Therefore, a higher resolution and increased number of plume points do not necessarily indicate better correspondence to the simulation points.

Figure 7. Distribution of observations after interpolation at different resolutions for Waigaoqiao. Plume points (red), background points (black), and other observations in the swath (blue).

In the case of the ultimate estimation of point-scale CO₂ emissions, all emissions estimated by the various methods showed obvious consistency in terms of the order of magnitude. However, on comparing our estimation with the previous studies mentioned in Section 2.3, several emission inventories estimated using the bottom-up method spanning different time frames had overestimated the CO₂ emissions of one of two Chinese power plants on the two days of measurements, which is also reflected in Nassar’s research [21]. As described in previous research [36], the heating values of coal used for bottom-up estimation have declined significantly since 2007, indicating that the estimated CO₂ emissions with previous parameter values are likely to be larger than the actual value. Generally, the lack of independent “truth” data does not allow a full validation of our estimation. A due diligence report on CARMA’s data and methodology [44] concluded that CARMA’s data are unfit for policy and business decisions, and that statistical shortcuts are not an acceptable substitute for real data. Compared with the statistical methods, our estimation is based on the reliable and robust real-time observation of satellites.

As further proof of the improvement in our study, we estimated CO₂ emissions from three coal power plants with reported CO₂ emissions through in-site measurement in Nassar et al. [21]. The estimated emissions and corresponding correlation coefficients of our study are shown in Table 4 (observations interpolated to $1.00 \times 1.00 \text{ km}^2$). The increased correlation coefficients indicated that the liner-fitting effects of the three power plants were improved, and the estimated daily emissions in our study are closer to the reported emissions. There are sufficient plume points in the three power plants for which we simulated the emissions, before and after data interpolation. We noticed that, for the Westar Jeffrey Energy Center, which has more plume points, only a slight improvement is obtained due to observations interpolation. Compared with Nassar et al. [21], the optimization of the estimation results for the power plant mainly benefits from unit conversion method and more accurate direction and speed of wind from reanalysis data with higher spatial and temporal resolution. In the case of Gavin and Kyger Creek Power Plants and the Ghent Generating Station with fewer plume points, observations interpolation improved the fitting effect and obtains emissions closer to the reported amount. Furthermore, some of the improvement in correlation (R) can be attributed to the improvement in OCO-2 data

going from version 7 to version 10, which have improved precision (lower noise) for all of the above power plants.

Table 4. Estimations of three coal power plants in the United States. The rows labeled with ‘N’ are the numbers of points in plumes; the rows labeled ‘Report’ are the reported emission; the rows labeled with ‘N17’ and ‘Estimation’ are the estimations, and the rows labeled with ‘R_N’ and ‘R’ are correlation coefficients in Nassar et al. [21] and this paper, respectively.

Power Plant	Date	Background	N	Report (kt/d)	N17 (kt/d)	R _N	Estimation (kt/d)	R
Westar	4 December 2015	400.96 ppm	130	26.67	31.21	0.468	24.95	0.751
Ghent	13 August 2015	392.67 ppm	33	29.17	29.46	0.707	28.94	0.824
Gavin/Kyger	30 July 2015	396.62 ppm	17	50.54	48.66	0.688	51.03	0.812

4.3. Limitation and Prospects

Our simulation with the Gaussian plume model is generally applied to instantaneous emissions, which simulate the practical emissions at the time of OCO-2 overpasses. The main limitation of our study is that the Gaussian plume model method assumes constant wind velocity and turbulent diffusivities values. Therefore, our top-down estimation assumed a constant emission rate and constant wind speed and direction, which were expected to fail over longer distances and times. As shown for Waigaoqiao, the OCO-2 observation values were affected by the changing wind direction. Furthermore, the Gaussian plume model cannot reproduce plumes overlapping or atmospheric circulation [45], and therefore, there is a mismatch between real emission plumes and Gaussian plumes due to atmospheric turbulence. In addition, the bias and precision of OCO-2 also contributed to the simulation uncertainty, yet the accuracy and precision of OCO-2 proved to be qualified for estimating CO₂ emissions from large-scaled coal-fired power plants [21].

The uncertainty of this study results from the variations in wind, background, observed enhancement, secondary emission sources, and interpolation. As another potential source of error, the uncertainty of estimating an annual emission from a single OCO-2 overpass was not quantified for the reason that more detailed data on the actual operation of power plants were unavailable. Regarding the wind, a linear relationship exists between the estimated emission and wind speed; thus, the wind speed error is linearly propagated with the error of the emission estimate [20]. Therefore, a wind vector with a high temporal and spatial resolution is critical. The uncertainties of the remaining two factors are related to the OCO-2 observations [46]. We defined the background by setting different distances from the point source and selecting the assembly with the lowest standard deviation as the background area for reducing uncertainty. The uncertainty of observed enhancement refers to the uncertainty of OCO-2 XCO₂ retrievals provided in the OCO-2 data. We calculated the emission uncertainties of Waigaoqiao and Qinbei according to Nassar et al. [21] (Table 5). The uncertainty from secondary emission sources was not considered because there were no other industrial emission sources near the two power plants. As for the uncertainty from Kriging, although no additional OCO-2 observations are added in the interpolation process, the interpolation brought new plume points associated with the observations for fitting and caused XCO₂ of the same geographical coordinate to vary. The uncertainty from the interpolation method is determined by calculating the standard deviation of estimated emissions at different resolutions. The reduction of uncertainty in our study was mainly due to the application of high-resolution wind vectors from ERA5.

Table 5. Uncertainty from five terms for two power plants.

Power Plant	Total Uncertainty (Mt/year)	Wind Speed Uncertainty (Mt/year)	Background Uncertainty (Mt/year)	Enhance Uncertainty (Mt/year)	Secondary Uncertainty (Mt/year)	Interpolation Uncertainty (Mt/year)
Waigaoqiao	2.86	2.59	0.22	1.10	–	0.45
Qinbei	3.38	3.12	0.30	1.25	–	0.14

To quantify CO₂ emissions from a coal-fired power plant from space accurately, CO₂ detection with effective spatial and temporal resolution and sufficient coverage is imperative. The first Chinese satellite dedicated to the detection and monitoring of CO₂, TanSat [47], was launched in December 2016, yet gaps still remain in the TanSat measurements between the footprints of each orbit. The unobserved areas can be filled effectively by combining data from the OCO-2 satellite with that of the TanSat satellite because the footprint tracks are almost parallel and interlaced. However, only level 1 primary products of TanSat are available, and CO₂ flux data are difficult to access. For future data collection, the Orbiting Carbon Observatory-3 (OCO-3) [48], launched in 2019, continues the collection of NASA's CO₂ measurements from space, including XCO₂, and emphasizes the targeting of cities and power plants. OCO-3 released an "Early" version [49], which is a temporary edition for comprehending the spectral and calibration characteristics of the satellite, but it has not been widely used. We attempted to use OCO-3 lite data in our estimation; however, the XCO₂ data around power plants showed no obvious distinction between the plume area and background area on the imaging coverage. However, improved versions of TanSat, OCO-2, and OCO-3 observations are promising for future research efforts. In addition, the Copernicus Anthropogenic CO₂ Monitoring (CO2M) mission can detect CO₂ plumes effectively, and there will be a huge benefit of adding an NO₂ instrument to a constellation of CO2M satellites for detecting city plumes and weaker point sources [50].

Another aspect related to obtaining a more accurate estimation for individual power plant CO₂ emissions is the revisit rate. This parameter can be used to achieve the perspectives of real-time monitoring of the emissions of power plants at the unit level with CO₂ satellites in China, where currently, real-time measured CO₂ data obtained from proper on-site monitoring and verification are lacking. A formula for the revisit rate or the number of overpasses required to achieve ideal accuracy in annual emission estimates has been derived to provide observations with the necessary characteristics to monitor anthropogenic CO₂ emissions in a manner that would support international climate agreements [51].

5. Conclusions

This study estimated CO₂ emissions from two individual coal-fired power plants in China, using XCO₂ values from OCO-2 by employing an improved Gaussian plume model simulation and bottom-up method. A spatial distribution map of power plants in China was first produced for the screening of eligible medium- and large-sized power plants. As a result, the Waigaoqiao and the Qinbei power plants were selected for estimation. Then, we improved the application of the Gaussian model using the method and data. The observation data for CO₂ column concentration of the latest version (with bias correction) and wind vector data with the highest temporal and spatial resolution were applied as the input parameters. The physically based unit conversion method and OCO-2 observation interpolations were performed to quantify CO₂ emissions more accurately. In particular, when the power plant was situated within the OCO-2 flight strip and the wind direction was not parallel to the strip direction, few plume points were suitable for fitting. We found that the interpolated OCO-2 XCO₂ data for simulation in small scales significantly improved the correlation of the simulation, thus enhancing the credibility of CO₂ estimation.

The estimated emissions values of Waigaoqiao with the Gaussian and bottom-up methods were 23.06 ± 2.82 (95% CI) and 16.28 Mt/yr, respectively, while those for Qinbei were 14.58 ± 3.37 (95% CI) and 14.08 Mt/yr, respectively. We verified our estimates by comparing them with the emissions of the CARMA, CPED, and Carbon Brief databases. The results indicated that previous emission inventories may have overestimated the emissions of Qinbei on the day of measurement.

This study demonstrated that the proposed method could estimate coal-fired power plants' CO₂ emissions at the unit level from space more accurately, which is independent of human intervention. This achievement is expected to provide a data reference for relevant policies promoting the commitment of the Chinese government to reach the target CO₂ emissions by 2030. However, the screening process of available power plants illustrated that

satellite measurements such as OCO-2 are simply inadequate for small-scale monitoring of the type described in this study, and that other techniques that can see through cloud cover are needed to provide the necessary observations. Furthermore, the improved method provides a foundation for real-time monitoring and quantification of CO₂ emissions from point sources in the future.

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