

## RESEARCH ARTICLE

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Constraining Fossil Fuel CO<sub>2</sub> Emissions From Urban Area Using OCO-2 Observations of Total Column CO<sub>2</sub>Xinxin Ye<sup>1,2</sup> , Thomas Lauvaux<sup>1,3</sup> , Eric A. Kort<sup>4</sup> , Tomohiro Oda<sup>5,6</sup> , Sha Feng<sup>1</sup> , John C. Lin<sup>7</sup> , Emily G. Yang<sup>4</sup> , and Dien Wu<sup>7</sup> 

## Key Points:

- Inversion method is utilized to constrain whole-city fossil fuel emissions with measurement and transport model errors considered
- Potential of incorporating multiple tracks to obtain regular emission estimates is evaluated by pseudo data experiments
- Significant contribution of the biospheric fluxes variability to local X<sub>CO<sub>2</sub></sub> variation is demonstrated

## Supporting Information:

- Supporting Information S1

## Correspondence to:

X. Ye,  
xuy112@psu.edu;  
xinxinye@ucla.edu

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<sup>1</sup>Department of Meteorology and Atmospheric Science, Pennsylvania State University, University Park, PA, USA,<sup>2</sup>Department of Atmospheric and Oceanic Sciences, University of California, Los Angeles, CA, USA, <sup>3</sup>Laboratoire des Sciences du Climat et de l'Environnement, CEA, CNRS, UVSQ/IPSL, Université Paris-Saclay, Gif-sur-Yvette, France,<sup>4</sup>Department of Climate and Space Sciences and Engineering, University of Michigan, Ann Arbor, MI, USA, <sup>5</sup>Global Modeling and Assimilation Office, NASA Goddard Space Flight Center, Greenbelt, MD, USA, <sup>6</sup>Goddard Earth Sciences Technology and Research, Universities Space Research Association, Columbia, MD, USA, <sup>7</sup>Department of Atmospheric Sciences, University of Utah, Salt Lake City, UT, USA

**Abstract** Satellite observations of the total column dry-air CO<sub>2</sub> (X<sub>CO<sub>2</sub></sub>) are expected to support the quantification and monitoring of fossil fuel CO<sub>2</sub> (ffCO<sub>2</sub>) emissions from urban areas. We evaluate the utility of the Orbiting Carbon Observatory 2 (OCO-2) X<sub>CO<sub>2</sub></sub> retrievals to optimize whole-city emissions, using a Bayesian inversion system and high-resolution transport modeling. The uncertainties of constrained emissions related to transport model, satellite measurements, and local biospheric fluxes are quantified. For the first two uncertainty sources, we examine cities of different landscapes: “plume city” located in relatively flat terrain, represented by Riyadh and Cairo; and “basin city” located in basin terrain, represented by Los Angeles (LA). The retrieved scaling factors of emissions and their uncertainties show prominent variabilities from track to track, due to the varying meteorological conditions and relative locations of the tracks transecting plumes. To explore the performance of multiple tracks in retrieving emissions, pseudo data experiments are carried out. The estimated least numbers of tracks required to constrain the total emissions for Riyadh (<10% uncertainty), Cairo (<10%), and LA (<5%) are 8, 5, and 7, respectively. Additionally, to evaluate the impact of biospheric fluxes on derivation of the ffX<sub>CO<sub>2</sub></sub> enhancements, we conduct simulations for Pearl River Delta metropolitan area. Significant fractions of local X<sub>CO<sub>2</sub></sub> enhancements associated with local biospheric X<sub>CO<sub>2</sub></sub> variations are shown, which potentially lead to biased estimates of ffCO<sub>2</sub> emissions. We demonstrate that satellite measurements can be used to improve urban ffCO<sub>2</sub> emissions with a sufficient amount of measurements and appropriate representations of the uncertainty components.

## 1. Introduction

The global atmospheric CO<sub>2</sub> concentration has increased by more than 40% since the preindustrial era to more than 400 ppm in recent years and remains the main driver to current and future climate changes (Le Quéré et al., 2018). The increase in CO<sub>2</sub> concentration predominantly originates from anthropogenic CO<sub>2</sub> emissions by combustion of fossil fuels such as coal, petroleum, and natural gas (Andres et al., 2012; Ciais et al., 2013; Rotty, 1983). In order to foster the mitigation and management of anthropogenic CO<sub>2</sub> emissions, the international community has pursued treaties and agreements in the recent decades, such as the Kyoto Protocol (United Nations, 1998) and the Paris Agreement (UNFCCC, 2015).

A large percentage of the anthropogenic CO<sub>2</sub> is emitted from urban areas, about 40% as estimated by production-based figures (i.e., adding up emissions from entities located within cities) and as high as 60–70% with a consumption-based method (i.e., adding up emissions resulting from the production of all goods consumed by urban residents) (International Energy Agency, 2008; Satterthwaite, 2008; UN-Habitat, 2011). Given their significant contributions to fossil fuel CO<sub>2</sub> (ffCO<sub>2</sub>), cities can perform as leading entities in implementing emission reduction plans. Comprehensive, accurate, and comparable emission quantifications are crucial for transparent monitoring of ffCO<sub>2</sub> emissions from urban areas and implementing effective mitigation schemes (Duren & Miller, 2012; Gurney et al., 2015; Pacala et al., 2010).

Emission inventory compilations with two methods referred to as “bottom-up” and “downscaling” are generally used to quantify  $\text{ffCO}_2$  emissions. However, urban emissions usually bear large uncertainties, due to the missing socioeconomic information and inaccurate emission conversion factors, which are critical elements affecting the quality of emission inventories at the urban scale (Gately & Hutyra, 2017). For example, urban energy consumption and industrial activity data needed for “bottom-up” method are reported on a voluntary basis or under climate action activities in only a few cities, for example, those participating the Global Covenant of Mayors (<http://www.globalcovenantofmayors.org/>) and are usually spatially inexplicit, incomplete, and unverified (Hutyra et al., 2014). Similarly, for inventories developed by disaggregating (“downscaling”) national or regional emissions at fine scales, the uncertainty is large at high spatial and temporal resolutions caused by the disaggregation methods (Janssens-Maenhout et al., 2012; Kurokawa et al., 2013; Oda & Maksyutov, 2011), resulting in significant discrepancies among different emission inventories (Ackerman & Sundquist, 2008; van der Gon et al., 2011; Gurney et al., 2012, 2019; Hogue et al., 2016; Oda & Maksyutov, 2011; Oda et al., 2018; Turnbull, Karion et al., 2011). The two-sigma uncertainties of national annual  $\text{ffCO}_2$  emissions are estimated to be 2–4% for countries with well-developed energy statistics and inventories (Rypdal & Winiwarter, 2001) and are at a possible order of 10% for countries with less well-developed energy data systems (IPCC, 2006). Nevertheless, urban-scale emission inventories can exhibit large differences (50–250%) compared to other downscaled data sets at local scales (Gately & Hutyra, 2017). The large uncertainties in  $\text{ffCO}_2$  emissions not only impose difficulties on evaluating the effects of emission reduction strategies but also would lead to significant biases in the regional carbon budget estimations (Corbin et al., 2010). It is reported by European Space Agency (2015) that accuracies of inferred emissions in the order of 10% of the total would be needed for providing constraints that allows emission inventories to be evaluated at the time of an overpass. For cities that do not have inventories, an accuracy of 20% is already an important gain in information.

Atmospheric observations-based methods are becoming important ways to objectively obtain  $\text{ffCO}_2$  emission estimates, allowing existing emission inventories to be improved. Some attempts have been made to derive local-scale emissions for the urban areas by utilizing the inverse modeling method (Bousquet, 2000; Ciais et al., 2010) with ground-based observations (Bréon et al., 2015; Lauvaux et al., 2016; McKain et al., 2012; Staufer et al., 2016; Wunch et al., 2009) or by the mass-balance approach with aircraft measurements (Cambaliza et al., 2014). However, one of the key limitations of these approaches is the unavailability of direct, continuous, and high-frequency atmospheric  $\text{CO}_2$  measurements representing  $\text{CO}_2$  enhancement in urban areas (Bréon et al., 2015), as only a handful of cities, mostly in Europe and North America, are instrumented with networks of  $\text{CO}_2$  sensors (Bréon et al., 2015; Davis et al., 2017; McKain et al., 2012; Miles et al., 2017; Verhulst et al., 2017).

In recent decades, space-based satellite measurements of column-averaged  $\text{CO}_2$  dry air mole fractions ( $X_{\text{CO}_2}$ ) have been highly recommended for quantification and monitoring of urban  $\text{ffCO}_2$  emissions, especially for cities where ground-based observations are sparse or unavailable (Duren & Miller, 2012; Kort et al., 2012; McKain et al., 2012; Schneising et al., 2013). Initial attempts have been made to relate satellite  $X_{\text{CO}_2}$  measurements to  $\text{ffCO}_2$ . Based on the retrievals from Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) on ENVISAT (2002–2012) (Bovensmann et al., 1999; Buchwitz et al., 2005), regional  $X_{\text{CO}_2}$  enhancements over the industrial areas in Germany were revealed to be correlated with yearly increase of anthropogenic  $\text{CO}_2$  emissions (Schneising et al., 2008, 2013). In addition, with the launch of Greenhouse gases Observing SATellite (GOSAT) in 2009 (Kuze et al., 2009; Morino et al., 2011), discernible  $X_{\text{CO}_2}$  contrasts between the emission and background regions (Janardanan et al., 2016; Keppel-Aleks et al., 2013) and local  $X_{\text{CO}_2}$  enhancements over megacities (Kort et al., 2012) have been reported. However, due to coarse spatial resolution ( $\sim 60 \text{ km} \times 30 \text{ km}$ ) and relatively low sensitivity (4–8 ppm) of the SCIAMACHY instrument, applications of its data are limited to large and intense emission regions. For the GOSAT instrument, the major limitation is its low sounding density, with a single 85-km<sup>2</sup> measurement per 250 km, resulting in fewer than 1,000 cloud-free soundings each day (Eldering, Wennberg, et al., 2017). Hence, these data sets are insufficient to enable accurate assessment of  $\text{ffCO}_2$  urban emissions at high spatial and temporal resolutions.

National Aeronautics and Space Administration's Orbiting Carbon Observatory 2 (OCO-2) satellite mission (Crisp, 2008; Crisp et al., 2004) has been providing continuous and global retrievals of  $X_{\text{CO}_2}$  since September

2014 (Crisp, 2015). Although the OCO-2 mission is primarily developed for assessing regional carbon sources and sinks, its unique characteristics allow for detection of  $X_{\text{CO}_2}$  enhancements over cities at fine scales. The OCO-2 measurements have a higher spatial resolution than GOSAT and SCIAMACHY and collect more data per day. In particular, the small nadir footprint ( $\sim 1.29 \text{ km} \times 2.25 \text{ km}$ ) helps to maximize the detectability of local emissions and increase the probability of cloud-free observations in the presence of patchy clouds. In addition, the high spectral resolution of OCO-2 enables high precision of  $X_{\text{CO}_2}$  with single sounding random errors of 0.5–1 ppm (Eldering, O'Dell, et al., 2017), which will greatly help to detect the small anthropogenic  $X_{\text{CO}_2}$  signals from the large background driven by biospheric sources and sinks and large-scale atmospheric transport (Keppel-Aleks et al., 2013; Turnbull et al., 2016). There have been some studies to examine  $X_{\text{CO}_2}$  imprints linked to anthropogenic emissions with the OCO-2 data. For example, spatial enhancements have been reported over the Northern Hemisphere regionally (Hakkarainen et al., 2016) and across the Los Angeles basin (Schwandner et al., 2017). For local-scale emissions and point sources, atmospheric transport modeling approach is applied to disentangle  $\text{ffCO}_2$  emissions from background. Wu et al. (2018) developed a Lagrangian model to interpret the  $X_{\text{CO}_2}$  retrievals and constrain emissions from some cities in the Middle East. Nassar et al. (2017) presented the capability of quantifying  $\text{ffCO}_2$  emissions from individual power plants by utilizing a Gaussian plume model.

Although the abovementioned studies have demonstrated the utility of OCO-2  $X_{\text{CO}_2}$  on revealing  $\text{ffCO}_2$  emissions, the emission estimation has not been carried out with high spatial resolution forward transport modeling so far, which has an advantage in capturing the fine-scale structure of  $\text{ffCO}_2$  plume. Meanwhile, it is essential to evaluate the uncertainty in emission estimate related to atmospheric transport model error with the high-resolution plume simulations, which has been identified as a major source of uncertainty in inverse modeling (Gerbig et al., 2003; Houweling et al., 2010; Lauvaux & Davis, 2014; Lauvaux et al., 2012; Lin & Gerbig, 2005; S. M. Miller et al., 2015; Pacala et al., 2010). Additionally, the uncertainty in biosphere  $X_{\text{CO}_2}$  at local scale increases the difficulty of unambiguously disentangling fossil fuel emission signals in  $X_{\text{CO}_2}$  from the observations for cities in vegetated areas, which has not been evaluated in respect of space-based total column observations.

The previous studies were focused on cases using individual OCO-2 tracks. However, from the perspective of tracking  $\text{ffCO}_2$  emissions for global cities on a regular basis, the potential of OCO-2 retrievals has yet to be investigated, which will be helpful to provide regular and policy-relevant references in support to improving the emission reduction strategies. Given current limitations in remote sensing that create a trade-off in sampling coverage and measurement precision, OCO-2 was designed as a sampling mission to provide measurements at high precision but only samples a small fraction of the globe each day (Eldering, Wennberg, et al., 2017), with a narrow swath ( $\sim 10.3 \text{ km}$ ) and a long revisit cycle ( $\sim 16$  days). These features enable a small percentage of local emissions to be detected in each cycle (Pacala et al., 2010). Moreover, because the OCO-2 sampling locations vary among different observation modes (nadir, glint, and target), and the atmospheric transport condition changes, overpasses that happen to detect fossil fuel  $X_{\text{CO}_2}$  enhancements across a certain city are limited. For the cities examined in this work, about 5–15% of nearby OCO-2 tracks show detectable urban plumes, based on 15 months of OCO-2 data (September 2014 to November 2015). Hence, it would be still difficult to regularly monitor emissions by using individual overpasses. Despite these limitations, there is a potential to constrain fossil fuel emissions regularly by utilizing data collected along multiple tracks over one or more revisit cycle(s), which could enable tracking emission variations, although at a lower temporal resolution.

In this paper, we present the utility of OCO-2  $X_{\text{CO}_2}$  data to constrain  $\text{ffCO}_2$  emission estimations for urban areas. Several sources of uncertainty in emission estimate are evaluated, including the transport model errors, measurement errors, and variations in local biospheric carbon fluxes. High-resolution forward simulations are performed using the Weather Research and Forecast (WRF) model, which is capable of capturing fine-scale variability in  $X_{\text{CO}_2}$  distributions caused by transport and emission processes at urban scales. We first evaluate the emission estimate uncertainty related to transport model errors and measurement errors for three selected cities with different topographic influences and negligible impact of variations in local biospheric fluxes, that is, Riyadh and Cairo, classified as “plume cities” located in relatively flat terrain, and Los Angeles as a “basin city.” Based on these simulations, we carried out Observing System Simulation Experiments (OSSEs) to evaluate the potential of tracking urban emissions regularly by utilizing  $X_{\text{CO}_2}$

data from multiple OCO-2 tracks. We also evaluated the uncertainty induced by local biospheric fluxes variability for the Pearl River Delta (PRD) metropolitan area. We discuss additional uncertainty sources in the inverse emission estimates, for example, the prior emission error correlations and the daytime-only sampling, and conclude with the implications for utilizing future satellite observations to monitor urban emissions.

## 2. Data and Method

### 2.1. OCO-2 $X_{CO_2}$ Observations

The OCO-2 Lite files (Version 7r) from September 2014 to November 2015 are used in this study (obtained online at <https://co2.jpl.nasa.gov>). The OCO-2 satellite operates in a sun-synchronous polar orbit at the altitude of about 705 km and crosses the equator nominally at 13:36 LT (local time). It provides high-resolution spectroscopic measurements at eight adjacent 2.25-km-long footprints within a narrow swath every 0.333 s, with a cross-track resolution of 0.1–1.3 km (Crisp, 2008; Eldering, O'Dell, et al., 2017). The  $X_{CO_2}$  data are retrieved from the spectroscopic observations of reflected sunlight in near-infrared  $CO_2$  and  $O_2$  bands near midday, using the Atmospheric  $CO_2$  Observations from Space algorithm (O'Dell et al., 2012). We used the bias-corrected  $X_{CO_2}$  data and filter them with “xco2\_quality\_flag” (QF) equals to 0, which is an indicator of data passing the internal quality check. The bias correction and data quality assessment have been detailed in the OCO-2 documentation (Mandrake et al., 2015). In addition, we analyzed each individual track over the selected cities for possible interferences by complex terrain, aerosols, and clouds. Some tracks were excluded due to contaminations by aerosols or clouds, as confirmed with the Cloud-Aerosol Lidar with Orthogonal Polarization data (Winker, 2016). The OCO-2  $X_{CO_2}$  data are averaged over time windows of 1 s, consisting of 24 consecutive soundings (at most) representing  $\sim 10.32$  km (cross track)  $\times$  6.75 km (along track) in area. Note that we only derive 1-s averaged  $X_{CO_2}$  when there are at least 5 soundings (at most 24) in that time window passing the data selection criteria.

The measurement error of each  $X_{CO_2}$  sounding consists of two parts: a random error related to noise and a systematic error that is in principle bounded by the calculated interference error owing primarily to aerosol optical depth, surface albedo, and surface pressure (Boxe et al., 2010). For the random error, the OCO-2 data products include an estimate of the uncertainty on  $X_{CO_2}$ , which is generally smaller over water than the land surface and larger at the extreme latitudes (Eldering, O'Dell, et al., 2017). However, this estimation is a lower bound (Connor et al., 2016). Worden et al. (2017) evaluated the OCO-2 uncertainty by examining the  $X_{CO_2}$  variability within small neighborhoods of  $\sim 100$  km  $\times$  10.5 km, in which natural  $CO_2$  variability is expected to be small. It is shown that the random error in the data product over land ( $\sim 0.36$  ppm) is smaller than the empirically derived  $X_{CO_2}$  random error ( $\sim 0.75$  ppm) by a factor of approximately 2. Conservatively, we consider a random error for each  $X_{CO_2}$  sounding with the standard deviation of 1 ppm, which leads to 0.20–0.45 ppm for the 1-s averaged data calculated with 5–24 soundings.

For the systematic error, as we used the bias-corrected data, some biases have been removed, for example, systematic footprint-to-footprint differences, mode-to-mode differences, and systematic differences that appear to be correlated to other retrieval variables (Mandrake et al., 2015). However, as validated using the Total Carbon Column Observing Network (TCCON)  $X_{CO_2}$  measurements by Wunch et al. (2017), residual biases remain in the OCO-2 retrievals after bias correction, with the absolute median differences  $< 0.4$  ppm and RMS differences  $< 1.5$  ppm. These biases appear to depend on latitude, surface properties, and scattering by aerosols. We note that for OCO-2 nadir and glint modes, these biases are evaluated using aggregated  $X_{CO_2}$  data within a box centered around the TCCON station that spans  $5^\circ$  in latitude ( $\sim 555$  km) and  $10^\circ$  in longitude ( $\sim 1,100$  km) on the same day as a TCCON measurement. As we examine the variability in  $X_{CO_2}$  at a smaller spatial scale ( $\sim 200$ – $300$  km, see Table 1 for the domain sizes), these biases in background can be removed when we extract local  $X_{CO_2}$  enhancements associated with fossil fuel emissions ( $ffX_{CO_2}$ ) by subtracting the background  $X_{CO_2}$ . Due to the lack of an accurate representation of aerosol contamination in the current retrieval algorithm, we assume that the effects of urban aerosols on  $X_{CO_2}$  retrievals are negligible. Hence, the local  $ffX_{CO_2}$  derived from OCO-2 observations is assumed to be unbiased in this paper. Details of the derivation of background  $X_{CO_2}$  can be found in section 2.2.

**Table 1**  
Summary of the Simulations Carried Out for the Selected Cities

| City type  | City/Metropolitan region                    | Land cover (% innermost domain) <sup>a</sup>   | Transport error propagation method | Innermost domain size and resolution | Simulation time  |
|------------|---|--|------------------------------------|--------------------------------------|--|
| Plume city | Riyadh                                      | Barren or sparse (92.2%)   | Perturbed plume                    | 201 × 201 (1 km)                     | 1–16 November 2014<br>17 December 2014 to 5 January 2015<br>27–30 January 2015                 |
|            | Cairo                                       | Barron or sparse (57.4%)<br>Croplands (32.8%)  | Perturbed plume                    | 201 × 201 (1 km)                     | 4–7 October 2014<br>16–19 March 2015<br>17–20 May 2015<br>13–16 July 2015<br>14–17 August 2015 |
| Basin city | Los Angeles (LA) metropolitan region        | Water (45.2%)<br>Open shrublands (34.9%)<br>Barren or sparse (6.1%)<br>Woody savannas (3.6%)<br>Croplands (2.9%) | Ensemble simulation                | 207 × 150 <sup>b</sup> (4 km)        | 3 July to 20 August 2015<br>6–19 October 2015  |
| Multi-city | Pearl River Delta (PRD) metropolitan region | Water (26.7%)<br>Croplands (26.2%)<br>Evergreen broadleaf (20.0%)<br>Woody savannas (10.1%)<br>Urban (8.6%)      | Perturbed plume                    | 240 × 240 (1.333 km)                 | 12–15 January 2015<br>1–4 August 2015  |

<sup>a</sup>The domain of interest for LA is set to 119.0°W to 116.3°W, 32.2–35.7°N, which is smaller than the innermost domain of simulations. <sup>b</sup>Land cover is based on MODIS IGBP 21-category data. The land cover types accounting for more than 90% of the innermost domain in aggregate are listed.

## 2.2. Background X<sub>CO2</sub>

We extract  $\text{ffX}_{\text{CO}_2}$ , that is,  $\text{X}_{\text{CO}_2}$  enhancements caused by  $\text{ffCO}_2$  emissions, by subtracting background  $\text{X}_{\text{CO}_2}$  from the OCO-2  $\text{X}_{\text{CO}_2}$  retrievals. A typical method to derive  $\text{ffCO}_2$  from in situ  $\text{CO}_2$  measurements is to calculate the difference between  $\text{CO}_2$  at an upwind site and a downwind site (e.g., Bréon et al., 2015; Lauvaux et al., 2016; Super et al., 2017). The best application condition of this background method is when the wind vector is aligned with the stations. Similarly, when the wind is aligned with the orbit, we could get background  $\text{X}_{\text{CO}_2}$  by averaging the measurements in the upwind region of a city. However, the alignment is very rare for OCO-2 (Nassar et al., 2017). In previous studies, a constant background is often used for  $\text{X}_{\text{CO}_2}$  retrievals collected over a time period, which is calculated as the median  $\text{X}_{\text{CO}_2}$  over a latitudinal band (Hakkarainen et al., 2016) or the average  $\text{X}_{\text{CO}_2}$  in a “background area,” for example, the desert located close to Los Angeles (Kort et al., 2012). However, for a single overpass, a constant background would not represent the spatial variability of the background concentrations. Thus, for each single overpass across a city of interest, we derived a “background line,” as shown with the black lines in Figures 4, 5, and 7. The “background line” is derived by a two-step linear regression. We firstly decompose the  $\text{X}_{\text{CO}_2}$  data into two parts, that is,  $\text{X}_{\text{CO}_2} = \text{X}_{\text{CO}_2\text{-trend}} + \text{X}_{\text{CO}_2\text{-local}}$ , where the  $\text{X}_{\text{CO}_2\text{-trend}}$  is the nonlocal trend represented using a linear function:  $\text{X}_{\text{CO}_2\text{-trend}} = a \cdot x + b$ . Here  $x$  is latitude, and  $a$  and  $b$  are the slope and interception derived by linear regression. With the standard deviation of  $\text{X}_{\text{CO}_2\text{-local}}$  ( $\sigma_{\text{local}}$ ) representing the local-scale variability, we filtered the  $\text{X}_{\text{CO}_2}$  samples with  $\text{X}_{\text{CO}_2} < \text{X}_{\text{CO}_2\text{-trend}} + 0.5\sigma_{\text{local}}$ . These filtered data are chosen as “background samples” (the black triangles in Figures 4, 5, and 7), as they have lower spatial variability at local scales compared to the samples affected by urban  $\text{ffCO}_2$  emissions. Then we recalculate the linear regression line, that is, the “background line,” based on the “background samples.” This “background line” method allows considering the spatial trend in the background.

## 2.3. Cities of Interest

Four cities/urban areas are selected in this work considering the availability of OCO-2 data, topography, and vegetation coverage to highlight different sources of uncertainty in the emission estimates. To explore the impact of transport model errors, three different cities are chosen according to the following criteria: (i)

distant isolation from other large anthropogenic emission sources nearby, (ii) large fossil fuel carbon emissions, (iii) low cloud cover and relatively preferable data availability, and (iv) weak contribution of biospheric signals. Cities are categorized into two different types based on the impact of local topography on dispersion: “plume cities” located in relatively flat terrain and “basin cities” located in complex terrain trapping the dispersion. In this paper, Riyadh, Saudi Arabia, with a population of 6.2 million, and Cairo, Egypt, with 18.4 million are chosen as typical “plume cities,” which are characterized by  $\text{ffX}_{\text{CO}_2}$  enhancements distributed as plumes. The Los Angeles metropolitan area (referred to as LA hereafter) with a population of more than 13 million is chosen as a characteristic “basin city.” The LA basin presents large elevation gradients from the sea surface to the top of Mount Wilson to the north. The strong enhancements in  $\text{X}_{\text{CO}_2}$  are mainly due to air masses trapped in the basin, which has been referred to as “urban dome” (Idso et al., 1998), albeit we do not adopt this terminology in this work due to the potential confusion with actual accumulation of  $\text{CO}_2$ .

To evaluate the impact of uncertainties in biospheric fluxes on the emission estimate, the PRD region of China is selected, where an agglomeration of several cities is located, including Guangzhou, Hong Kong, Shenzhen, Zhuhai, Dongguan, and Zhongshan. The PRD region is one of the largest metropolitan areas in the world with about 45 million people. The cities are less vegetated compared to their surrounding area, leading to a distinctive contrast in the net ecosystem exchange (NEE) in the urban area and surrounding rural area (see Figure S2 in the supporting information and section 2.4.3).

## 2.4. Model Setup

### 2.4.1. Atmospheric Transport Model

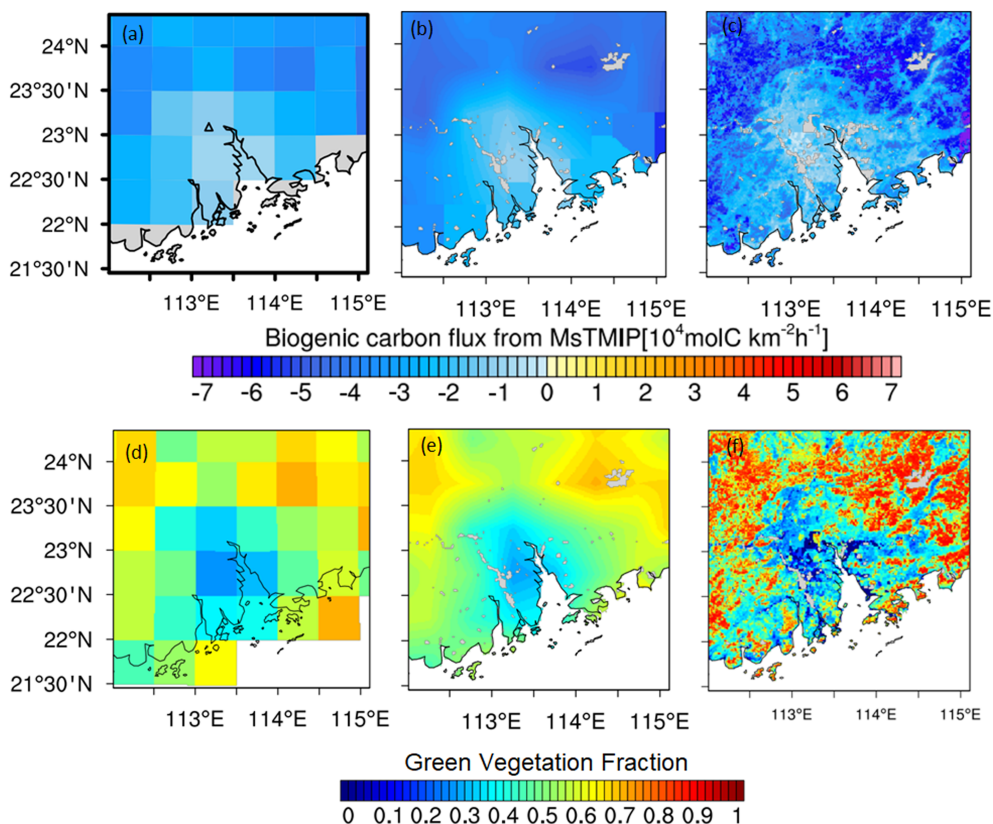
The spatial heterogeneity of emissions and intense point sources (e.g., power plants) lead to complex spatial structures of urban emissions, resulting in complex  $\text{ffCO}_2$  plume combined with local atmospheric dynamics (e.g., Feng et al., 2016). In order to capture the fine-scale variations, we simulate the  $\text{ffX}_{\text{CO}_2}$  using the Weather Research and Forecasting model (V3.6.1) with chemistry (WRF-Chem) (Grell et al., 2005; Skamarock et al., 2008), coupled to  $\text{CO}_2$  emissions and biospheric fluxes using the passive tracer mode (Lauvaux et al., 2012). Model grids are configured with 51 vertical (eta) levels. The 6-hourly NCEP FNL (Final) Operational Global Analysis data on  $0.5^\circ \times 0.5^\circ$  grids are used as the initial and boundary conditions of meteorological and land surface fields. The boundary condition of  $\text{CO}_2$  concentration for the outermost domain is 390.0 ppm. The simulations are initiated every 4 days at 12:00 UTC with an integration time of 108 hr, including a spin-up time of 12 hr and producing hourly outputs.

One-way nested domains with resolutions of 27, 9, 3, and 1 km are used for Riyadh and Cairo, 36, 12, and 4 km for LA, and 36, 12, 4, and 1.333 km for the PRD region. The innermost domains and distributions of  $\text{ffCO}_2$  emission for the selected cities are shown in Figure S3. For Riyadh, Cairo, and PRD, the innermost domains are used as the domains of interest to filter the OCO-2 observations. Note that for LA, the domain of interest is set to  $119.0\text{--}116.3^\circ\text{W}$ ,  $32.2\text{--}35.7^\circ\text{N}$ , which is smaller than the innermost domain, and the spatial resolution is coarser than others. As reported by Feng et al. (2016), the 1.3-km run does not show significant improvement compared to the 4-km run at the surface, even though it resolves the vertical gradient of horizontal winds and planetary boundary layer (PBL) better. Given that aggregated  $\text{ffX}_{\text{CO}_2}$  along track is used to compute the scaling factor of a priori emissions, we compared the aggregated  $\text{ffX}_{\text{CO}_2}$  using 4- and 1.3-km runs, and they also present similar results owing to  $\text{ffX}_{\text{CO}_2}$  mostly being trapped within the basin during daytime. Therefore, we used 4-km resolution for LA in this study.

A summary of the simulations performed in this study is shown in Table 1. The fossil fuel  $\text{CO}_2$  emission data and NEE data are detailed in sections 2.4.2 and 2.4.3. Note that the simulations for tracers imposed by NEE are only conducted for the PRD region. Moreover, an ensemble of modeling based on model physics parameterizations is deployed to represent the transport model errors in the simulated  $\text{ffX}_{\text{CO}_2}$  over LA (see section 2.5.1).

### 2.4.2. Fossil Fuel $\text{CO}_2$ Emissions

The Open-source Data Inventory for Atmospheric Carbon dioxide (ODIAC) Version 2015a (Oda et al., 2017; 2018; Oda & Maksyutov, 2011, 2015) is used in this paper for emissions from the cities of interest. The ODIAC emission product provides 1-km  $\times$  1-km gridded global and monthly  $\text{ffCO}_2$  emissions. It is developed based on country-level  $\text{ffCO}_2$  emission estimates, fuel consumption statistics, satellite-observed nightlight data, and point source information (geographical locations and emission intensities) from the CARbon

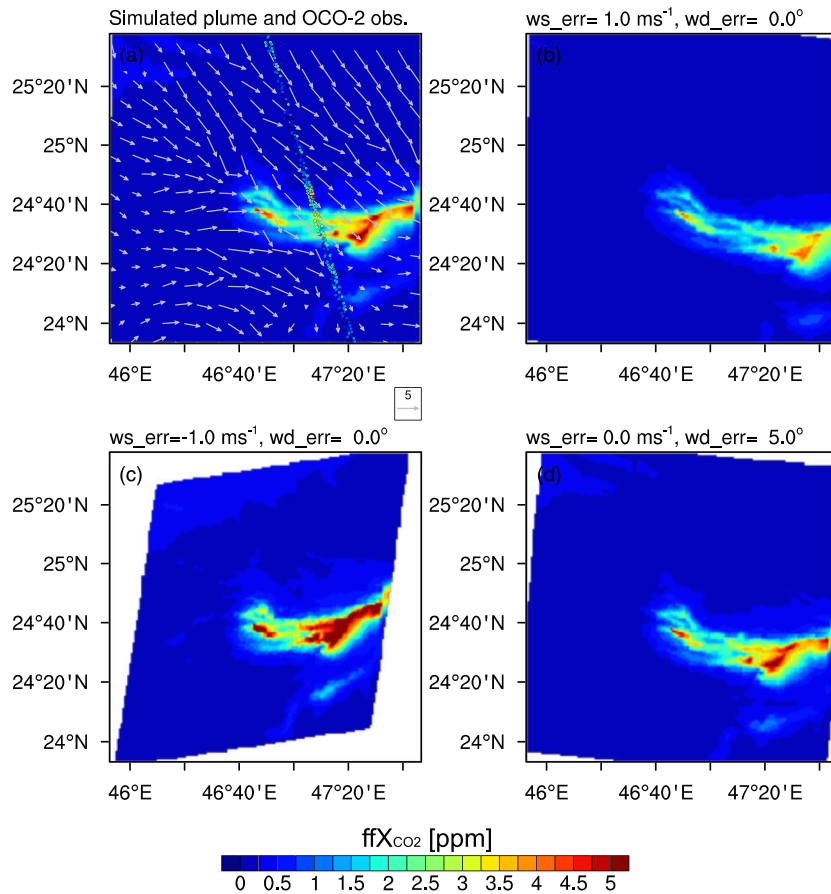


**Figure 1.** Example of biogenic carbon fluxes (net ecosystem exchange, NEE) downscaling in PRD region. The top panels show the NEE from 3-hourly MsTMIP data at 12:00 LT 12 January 2010 on (a) the original  $0.5^\circ \times 0.5^\circ$  grid, (b) WRF grid ( $1.333 \times 1.333$  km), derived by bilinear interpolation of original NEE, and (c) WRF grid ( $1.333 \times 1.333$  km), derived by scaling the interpolated NEE. The bottom panels show the green vegetation fraction (GVF) in January on (d) the  $0.5^\circ \times 0.5^\circ$  grid, (e) WRF grid ( $1 \times 1$  km) by bilinear interpolation of GVF in (d), and (f) WRF grid ( $1 \times 1$  km). See section 2.4.3 for further details.

Monitoring for Action power plant database (Oda et al., 2018). The global nightlight data were used as a georeferenced, spatial proxy to determine the spatial extent of anthropogenic emissions from the line and diffused (area) sources (e.g., road traffic, residential, or commercial fuel consumption). The ODIAC gridded emission fields defined on a global rectangular (latitude  $\times$  longitude) coordinate are remapped to meet the grid resolutions for each simulation domain. Note that temporal variability of emissions at diurnal and weekly scales is not included in the modeling and the pseudo data experiments. We remapped the monthly emission distributions for the time periods investigated in our simulation. The  $\text{ffCO}_2$  emission distributions are shown in Figure S3. All  $\text{ffCO}_2$  is released at the ground surface.

### 2.4.3. Biogenic CO<sub>2</sub> Fluxes

The NEE fluxes in the PRD region are provided by the 15 different global Terrestrial Biogeochemical Models ( $0.5^\circ \times 0.5^\circ$ ) in the Multi-scale Synthesis and Terrestrial Model Intercomparison Project (MsTMIP) (Huntzinger et al., 2013). In order to better characterize the diurnal variability and spatial distribution of biogenic fluxes, a 3-hourly data set for global biogenic fluxes (Fisher et al., 2016) is used, which is temporally downsampled from the monthly global models. Furthermore, we spatially downscale the 3-hourly NEE from the original MsTMIP grid ( $0.5^\circ \times 0.5^\circ$ ) (e.g., Figure 1a) to the WRF domains (36-, 12-, 4-, and 1.333-km resolutions) using the green vegetation fraction (GVF), with the assumption that vegetation productivity and respiration scales linearly with canopy coverage in each grid cell. We note that using this method, besides plant productivity and respiration, soil respiration is also downsampled by GVF, which could lead to some misrepresentation of soil respiration although the impact on the results would be small. The GVF is defined as the fraction of the grid cell for which midday downward solar insolation is intercepted by a photosynthetically active green canopy. A robust relationship between canopy cover and biomass was observed in Boston,



**Figure 2.** (a) Local  $ffX_{CO_2}$  derived from OCO-2 data (colored dots) at about 10:00 UTC 29 December 2014 by subtracting the background concentration and the simulated  $ffX_{CO_2}$  enhancement (color shading) using ODIAC emissions, which is the truth in the OSSEs. Panels (b) and (c) show the rescaled plumes of panel (a) with wind speed error of 1.0 and  $-1.0$  m/s. Panel (d) shows the rescaled plume with wind direction error of  $5.0^\circ$ .

which supports the use of GVF as a proxy for biomass, and hence as a scaling parameter for biogenic fluxes (Briber et al., 2013; Raciti et al., 2012). The NEE can be downscaled as follows:

$$E_{wrf,i,j} = (E_{blin,i,j}/GVF_{blin,i,j}) \times GVF_{wrf,i}, \quad (1)$$

where the subscripts  $i$  and  $j$  represent the coordinates of a WRF grid cell,  $E_{wrf}$  the NEE at WRF grid (e.g., Figure 1c), and  $E_{blin}$  the bilinear interpolated NEE from the original  $0.5^\circ \times 0.5^\circ$  grids to WRF grid (e.g., Figure 1b).  $GVF_{blin}$  is interpolated using Moderate Resolution Imaging Spectroradiometer (MODIS) climatological GVF (e.g., Figure 1d) in the same way of deriving  $E_{blin}$  (e.g., Figure 1c), ensuring the same spatial representativeness of  $GVF_{blin}$  and  $E_{blin}$ , and  $GVF_{wrf}$  (e.g., Figure 1e) is the GVF projected to the WRF grid. The GVF data used in this study are based on MODIS climatological observations from 2001 to 2010, which is available in the geographic data since WRF v3.6. The uncertainties in biogenic  $X_{CO_2}$  are represented by the spread of simulated biogenic  $X_{CO_2}$  using the NEE from the 15-member models in MsTMIP.

### 2.5. Representation of Transport Model Errors

The impact of transport model errors in wind speed and wind direction on the uncertainty of emission estimates is considered in the inversions. In this section, we introduce the methods to propagate the transport model errors to the modeled  $ffX_{CO_2}$  fields across different types of cities. The method applied for plume city and basin city are explained in sections 2.5.1 and 2.5.2, respectively.



**Table 2**  
WRF Model Configurations of the Ensemble of Simulations Conducted for Los Angeles

| Ensemble member | PBL scheme | Surface layer scheme        | Urban canopy model |
|-----------------|------------|-----------------------------|--------------------|
| MYJ             | MYJ        | Eta similarity (Janjić Eta) | None               |
| MYJ_UCM         | MYJ        | Eta similarity (Janjić Eta) | Noah UCM           |
| MYNN            | MYNN       | Nakanishi and Niino         | None               |
| MYNN_UCM        | MYNN       | Nakanishi and Niino         | Noah UCM           |
| BouLac_BEP      | BouLac     | Eta similarity (Janjić Eta) | BEP                |
| BouLac_UCM      | BouLac     | Eta similarity (Janjić Eta) | Noah UCM           |

### 2.5.1. Plume City: Transformation of Plumes

For a “plume city,” the transport model errors are propagated by transformation of the modeled  $\text{ffX}_{\text{CO}_2}$  plume. More detail of this method is included in the supporting information (Text S1.1). The errors for the “plume city” are assumed to be unbiased based on the previous study on Indianapolis (Deng et al., 2017). For a random error ( $\theta$ ) in wind direction, we rotate the simulated plume  $\bar{c}(x, y, t)$  at a given time ( $t$ ) by an angle  $\theta$  about the emission center  $(x_0, y_0)$  to get  $\bar{c}_r(x_r, y_r, t)$ . Note that  $x$  axis is aligned with the dominant wind direction in the domain. Then the rotated plume is transformed to incorporate random wind speed error ( $\varepsilon$ ) as follows:

$$\bar{c}_r(x_r, y_r, t) = \bar{c}(x_r, y_r, t), \quad (2)$$

where

$$x_r = \frac{\bar{u} + \varepsilon}{\bar{u}}(x_r - x_0) + x_0, \quad y_r = y_r, \quad z_r = z_r, \quad \bar{c}_r = \bar{c} \frac{\bar{u}}{\bar{u} + \varepsilon} \quad (3)$$

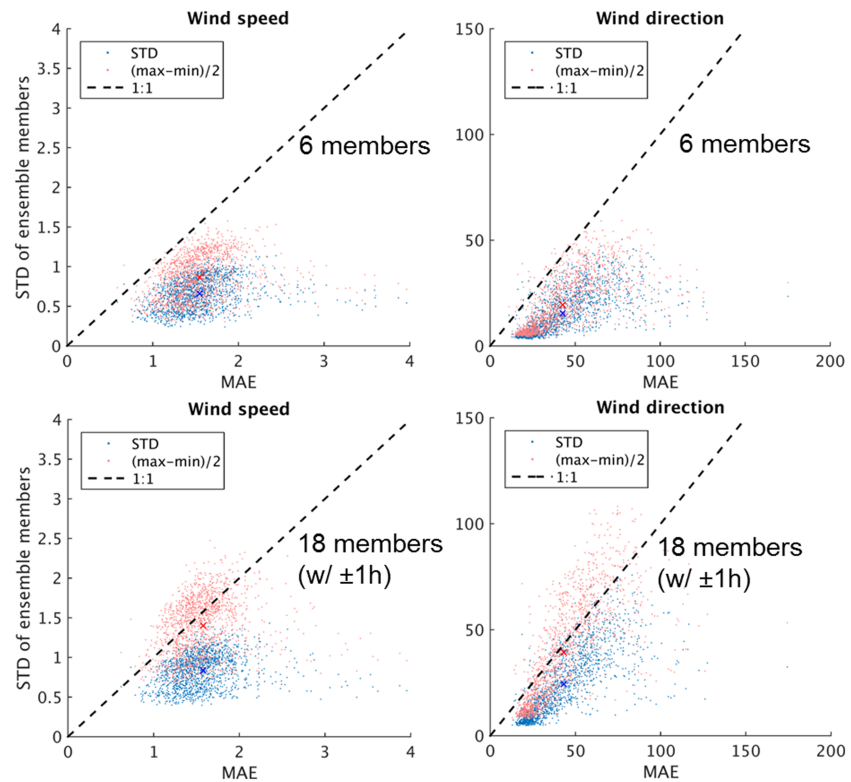
Since the  $\text{ffCO}_2$  is confined within the PBL and well mixed during the daytime, we used the domain average wind speed within the PBL and its typical error statistics for  $\bar{u}$  and  $\varepsilon$ . The errors are assumed to follow normal distributions of  $N(0, 1.0)$  (unit: m/s) for wind speed and  $N(0, 15)$  (unit: °) for wind direction, respectively. The selection of the error ranges is consistent with the model error statistics in the lower troposphere (<2 km) from model evaluation studies, for example, Indianapolis (Deng et al., 2017), Los Angeles (Feng et al., 2016), and more generally over the U.S. Upper Midwest (Diaz-Issac et al., 2018).

Examples of transformed plumes using this method are shown in Figure 2 based on a simulated  $\text{ffX}_{\text{CO}_2}$  plume over Riyadh at about 10:00 UTC 29 December 2014. The impacts of positive and negative wind speed errors are represented by transformation (Figures 2b and 2c), rescaling the plume along the domain average wind direction. The impact of a wind direction error is represented by a rotation of the plume (Figure 2d).

To obtain the transport model uncertainty in the modeled  $\text{ffX}_{\text{CO}_2}$ , we transform the modeled plume by multiple times (here  $10^4$ ) with random wind speed and wind direction errors, and extract the uncertainty spread of  $\text{ffX}_{\text{CO}_2}$  by using the 25th and 75th percentiles. The transformation method is a trade-off between running a simplified model (e.g., a Gaussian plume model) and running an ensemble of simulations. With this method, we can retain the complexity of  $\text{X}_{\text{CO}_2}$  plume structures while exploring the transport error impact at low computational costs. Note that this method is derived under the assumption of spatially uniform wind errors, which is generally valid within a few tens of kilometers from the emission center.

### 2.5.2. Basin City: Model Physics-Based Ensemble Simulation

For LA, a typical basin city, the transport model errors are represented by an ensemble of WRF simulations with different PBL and urban canopy physics parameterizations, following Feng et al. (2016). This method is suitable for transport conditions with the dispersion of  $\text{CO}_2$  trapped by local topography. Four different PBL parameterizations are used, that is, the Mellor-Yamada-Nakanishi-Niino 2.5 (Nakanishi & Niino, 2004) scheme, the Mellor-Yamada-Jancic scheme (Janjić, 1994), and the Bougeault and Lacarrère (BouLac) (Bougeault & Lacarrere, 1989) scheme. For the land surface processes in urban canopy, the single-layer urban canopy model (Kusaka & Kimura, 2004) and the multilayer building environment parameterization (Martilli et al., 2002) are used. The ensemble of simulations with different combinations of the model physics schemes (Table 2) can represent the model uncertainties in wind field, PBL structure, and PBL height.



**Figure 3.** Comparison of the modeling uncertainty and mean absolute error (MAE) of 10-m wind speeds and wind directions over 43 surface sites located in the 1.333-km resolution domain for LA. The modeling random uncertainty is calculated as the standard deviation (STD, blue scatters) and semifull range of the modeling results, that is, half of the difference between maximum and minimum values among the ensemble members (red scatters). The top two panels show results for the original six members, and the bottom two panels are for the 18 members with modeling results at  $\pm 1$  hr included. The red and blue crosses in each panel stand for average points of the scatters in the corresponding colors.

In order to evaluate the performance of the ensemble on representing the model-observation mismatches, the modeling results of wind speed and wind direction are compared with surface wind observations. Surface observations of wind speed and wind direction at 43 synoptic weather stations located within the 4-km domain covering LA were used, derived from the global hourly Integrated Surface Data and accessible at the National Centers for Environmental Information (<https://gis.ncdc.noaa.gov/geportal/catalog/search/resource/details.page?id=gov.noaa.ncdc:C00532>).

For each observation time, the mean absolute error (MAE) of modeling result is calculated to evaluate the magnitude of the model error. We compared the ensemble spread presented by two approaches: (1) the standard deviation of the ensemble results and (2) semifull range of the ensemble results (half of the difference between the maximum and minimum values). As shown in Figure 3, both of the approaches based on the simulation results of the six members exhibit a somewhat lower ensemble spread of wind speed compared to the observed MAE, suggesting an underestimation of the transport uncertainty (Figures 3a and 3b). When taking the model results at  $\pm 1$  hr relative to the observation time into account (Figures 3c and 3d with the ensemble size of 18 members), the ensemble spread is found to be enlarged, suggesting a better agreement with the observed MAE. The ensemble spreads for wind speed and wind direction both show better agreement with the MAE. It can also be notable that the semifull range yields a better representation of the model uncertainty compared to the standard deviation (Figures 3c and 3d). Hence, for LA we use the 18 ensemble members and the semifull range to estimate transport model uncertainty spread in the simulated  $\text{ffX}_{\text{CO}_2}$ .

For the systematic errors in the transport model, we compared the ensemble mean surface wind speed to the observations. The result shows a positive bias of 0.48 m/s. Similarly, Angevine et al. (2012) found a wind

speed bias of  $1.1 \pm 2.7$  m/s. Feng et al. (2016) reported a slightly smaller bias of  $\sim 1.0$  m/s for LA and showed larger biases near mountainous sites owing to complex topography. We note that the wind speed error is evaluated at surface in this work. The wind speed bias varies at different altitudes through the PBL, with usually larger value near the surface (Feng et al., 2016).

The wind speed bias can result in systematic error in emission estimates. In order to represent the impact of wind speed bias in the pseudo modeling data in the OSSEs, we apply a factor ( $k$ ) to the simulated  $\text{ffX}_{\text{CO}_2}$  for the pseudo truth, where

$$k = \frac{\bar{u}}{\bar{u} + \Delta}, \quad (4)$$

$\bar{u}$  is the average wind speed over the domain and  $\Delta$  is the corresponding bias (in m/s). This factor assumes a single ratio over the entire domain but allows to increase/decrease the  $\text{ffX}_{\text{CO}_2}$  to represent the impact of negative/positive wind speed bias.

## 2.6. Emission Optimization Method

A Bayesian inversion method similar to that used by Pillai et al. (2015) is implemented, which optimizes parameters of  $\text{ffCO}_2$  emissions with observational constraints to obtain the best consistency between modeled and observed  $\text{ffX}_{\text{CO}_2}$  enhancements. Specifically, we optimize the emissions by adjusting a scaling factor ( $\lambda$ ) upon the a priori emissions from the entire city, without modifying the spatial distribution. The integrated  $\text{ffX}_{\text{CO}_2}$  signals along a latitudinal range of interest of OCO-2 tracks are used as the observational constraint, which can be represented as follows:

$$y_o = \int_{\text{lat1}}^{\text{lat2}} \text{ffX}_{\text{CO}_2,o}, \quad y_m = \int_{\text{lat1}}^{\text{lat2}} \text{ffX}_{\text{CO}_2,m} \quad (5)$$

where  $y_o$  and  $y_m$  are the observed and modeled results, respectively. Here,  $\text{ffX}_{\text{CO}_2,o}$  is derived by subtracting the background  $\text{X}_{\text{CO}_2}$  from observations, and  $\text{ffX}_{\text{CO}_2,m}$  is derived by interpolating the modeling results of the tracer tagged with fossil fuel emissions at the coincident geolocations of the observations. Compared to deriving the scaling factor with the least square error method using all the soundings, the integral  $\text{ffX}_{\text{CO}_2}$  shows less sensitivity to wind direction error (Figure S2 and Text S1.2).

For  $n$  pairs of observations and modeling results obtained with  $n$  OCO-2 tracks, the integrated enhancements can be represented as:

$$\mathbf{y}_o = \mathbf{y}_m \cdot \lambda + \boldsymbol{\varepsilon}_o \quad (6)$$

Here the state vector has been simplified as a scalar, that is, the scaling factor  $\lambda$ , and the Jacobian matrix that represents the sensitivity of the observations to the state vector is given by a vector  $\mathbf{y}_m$ . The term  $\boldsymbol{\varepsilon}_o$  is an observational error vector, including errors in the OCO-2 measurements, forward model, and model parameters. It is assumed to follow the Gaussian distribution described with the error covariance matrix  $S_o$ . As the observation errors are assumed to be uncorrelated for different tracks,  $S_o$  is a diagonal matrix with the main diagonal entries representing the error variance of the observation ( $\sigma_o^2$ ) for each track. As the measurement and model uncertainty are unbiased and not correlated, we estimate  $\sigma_o^2$  by adding the error variances:

$$\sigma_o^2 = \sigma_{\text{measurement}}^2 + \sigma_{\text{model}}^2, \quad (7)$$

where  $\sigma_{\text{measurement}}^2$  is the measurement error variance and  $\sigma_{\text{model}}^2$  is the forward model error variance. The estimations of these two terms have been detailed in sections 2.1 and 2.5, respectively.

The posterior estimate of  $\lambda$  is derived by minimizing the cost function ( $J$ ):

$$J(\lambda) = (\mathbf{y}_o - \mathbf{y}_m \lambda)^T S_o^{-1} (\mathbf{y}_o - \mathbf{y}_m \lambda) + (\lambda - \lambda_a)^2 \sigma_a^{-2} \quad (8)$$

where  $\sigma_a^2$  is the error variance of the prior estimate,  $\lambda_a$ . The prior estimate  $\lambda_a$  is set to unity. The prior emission uncertainty  $\sigma_a$  is set up based on reported estimations in literature, as the ODIAC data product did not

provide uncertainty estimates. At annual scales, Gurney et al. (2019) investigated the difference between the ODIAC data and a high-resolution bottom-up estimate product (Hestia) in four U.S. urban areas, showing the differences of whole-city emissions ranging from  $-1.5\%$  to  $+20.8\%$ . Oda et al. (2019) found differences of about 40% by comparing a satellite-derived annual emissions product to a gridded national inventory at 25-km resolution. However, the uncertainty would become larger at a smaller time scale. Considering the variability in anthropogenic activities, for example, different power demand on weekdays and weekends, weather-related events, domestic heating, and air conditioning, the day-to-day variability of 20 to 50% can be found in emission inventories. As most of the temporal patterns in current emission products are prescribed, and based on recent publications at different timescales, we suggest uncertainties of 50% at the daily timescale as a lower bound. As this work focuses on quantifying the whole-city emissions, we conservatively set the prior flux uncertainty  $\sigma_a$  to 20% for LA, since the emissions from the U.S. megacities are relatively well characterized. For Riyadh and Cairo, the prior uncertainty is set to 40%, as the fuel consumptions are expected to have higher uncertainties than over the United States.

By solving the minimum of  $J$ , the optimal estimate of scaling factor,  $\hat{\lambda}$ , and the posterior error variance,  $\hat{\sigma}^2$  can be obtained as follows:

$$\hat{\lambda} = (\mathbf{y}_m^T \mathbf{S}_o^{-1} \mathbf{y}_m + \sigma_a^{-2})^{-1} (\mathbf{y}_m^T \mathbf{S}_o^{-1} \mathbf{y}_o + \sigma_a^{-2} \lambda_a) \quad (9)$$

$$\hat{\sigma}^2 = (\mathbf{y}_m^T \mathbf{S}_o^{-1} \mathbf{y}_m + \sigma_a^{-2})^{-1}. \quad (10)$$

### 2.7. OSSEs: Emission Optimization Using Multiple Tracks Under Different Meteorological Conditions

Given the limited amount of real OCO-2 overpasses and the observation geometry, OSSEs are implemented to examine the potential of using multiple OCO-2 tracks to constrain urban  $\text{ffX}_{\text{CO}_2}$  emissions in a statistical prospective, which allows examining the performances under different atmospheric transport conditions. Specifically, we examine the relation between the number of available tracks and the emission estimate uncertainty.

For each city, we use a specified and typical OCO-2 ground track locations in nadir observation mode to extract the pseudo observations and pseudo modeling data, based on the hourly modeled  $\text{ffX}_{\text{CO}_2}$ . We avoided random selection of track locations across the domain, as OCO-2 tracks repeat over time without major variations. As the OCO-2 overpasses are available only during daytime, we selected the modeling results during 09:00–15:00 LST with domain-averaged surface wind speed  $\geq 2$  m/s. Additionally, for Riyadh and Cairo, in order to ensure that plumes are transected by the satellite, the simulation results are also filtered with the angle from a plume axis to the typical track  $\geq 10^\circ$  and  $\leq 170^\circ$ .

The prior emissions, that is, the ODIAC data, are set as the true emissions in the OSSEs. Hence, pseudo observations of  $\text{ffX}_{\text{CO}_2}$  are obtained by sampling from the modeled  $\text{ffX}_{\text{CO}_2}$  at the locations of the 1-s averaged soundings along the typical track. The pseudo observations are assumed to be unbiased relative to the truth. A random error per sample has been added, following a Gaussian distribution with the standard deviation of 0.2 ppm representing the lower bound of the measurement errors (section 2.1).

The pseudo modeling data of  $\text{ffX}_{\text{CO}_2}$  are obtained in different ways for different cities. For Riyadh and Cairo, the pseudo modeling data are set directly to the modeled  $\text{ffX}_{\text{CO}_2}$  sampled along the typical track and perturbed by random transport errors. Thus, the posterior emission estimate would be unbiased, and the truth of scaling factor would be unity. For LA, as the positive wind speed bias has been seen with the comparison to surface observations, we apply a factor to the simulated  $\text{ffX}_{\text{CO}_2}$  to represent the impact of wind speed bias on the pseudo modeling  $\text{ffX}_{\text{CO}_2}$  (see equation 4), which lead to a biased posterior scaling factor as will be shown in the results. The transport model uncertainty is estimated with the same methods detailed in section 2.5. We note that the uncertainty in background  $\text{X}_{\text{CO}_2}$  is not included in the observation errors.

The impact of numbers of available tracks ( $N = 1, \dots, 20$ ) on emission estimate uncertainty is evaluated by a Monte Carlo approach. For each number of tracks ( $N$ ), we randomly select  $N$  pairs of pseudo observation and modeling data. The scaling factor is derived with the same inversion method (section 2.6) as used for the real

**Table 3**  
Inversion Results of Scaling Factors of the Whole-City Emissions for the Selected Cities Using OCO-2  $X_{CO_2}$  Data

| City        | Date of OCO-2 track | Prior total emission (Mt C/month) | Prior total emission uncertainty ( $\sigma_a$ ) | Measurement uncertainty ( $\sigma_{\text{measurement}}$ , units: ppm) | Transport model uncertainty ( $\sigma_{\text{model}}$ , units: ppm) | Scaling factor ( $\lambda$ ) $\pm$ posterior uncertainty ( $\hat{\sigma}$ ) |
|-------------|---------------------|-----------------------------------|---|---|---|---|
| Riyadh      | 27 December 2014    | 3.08                              | 40%   | 1.34  | <b>2.80</b>   | $0.92 \pm 0.24$   |
|             | 29 December 2014    | 3.08                              |   | 1.30  | <b>2.83</b>   | $0.83 \pm 0.17$   |
| Cairo       | 28 February 2015    | 2.52                              | 40%   | <b>1.07</b>   | 0.71  | $0.79 \pm 0.15$   |
|             | 18 March 2015       | 2.70                              |   | <b>0.55</b>   | 0.31  | $1.18 \pm 0.25$   |
|             | 19 May 2015         | 2.48                              |   | <b>1.18</b>   | 0.36  | $0.95 \pm 0.11$   |
|             | 15 July 2015        | 2.62                              |   | <b>1.08</b>   | 0.16  | $0.70 \pm 0.13$   |
|             | 16 August 2015      | 2.61                              |   | <b>0.94</b>   | 0.54  | $0.81 \pm 0.10$   |
| Los Angeles | 6 July 2015         | 4.68                              | 20%   | 1.48  | <b>2.99</b>   | $1.00 \pm 0.15$   |
|             | 15 July 2015        | 4.68                              |   | <b>2.11</b>   | 1.62  | $1.84 \pm 0.16$   |
|             | 7 August 2015       | 4.74                              |   | 1.11  | <b>1.35</b>   | $1.17 \pm 0.15$   |
|             | 16 August 2015      | 4.74                              |   | 1.93  | <b>3.03</b>   | $1.43 \pm 0.14$   |
|             | 10 October 2015     | 4.49                              |   | <b>1.20</b>   | 1.08  | $1.64 \pm 0.15$   |
|             | 12 October 2015     | 4.49                              |   | 1.40  | <b>2.70</b>   | $0.66 \pm 0.11$   |

*Note.* The scaling factors and their posterior uncertainties are shown for each individual track, as well as for integrating the information from all the selected tracks. Uncertainty components are listed for each track, including the prior uncertainty of scaling factor, and the measurement and transport uncertainties of the integral  $\text{ff}X_{CO_2}$  (the larger value between these two is shown in bold).

tracks analyses. Probability distributions of scaling factor and the associated posterior uncertainty are obtained by repeating the random selection procedure.

### 3. Results

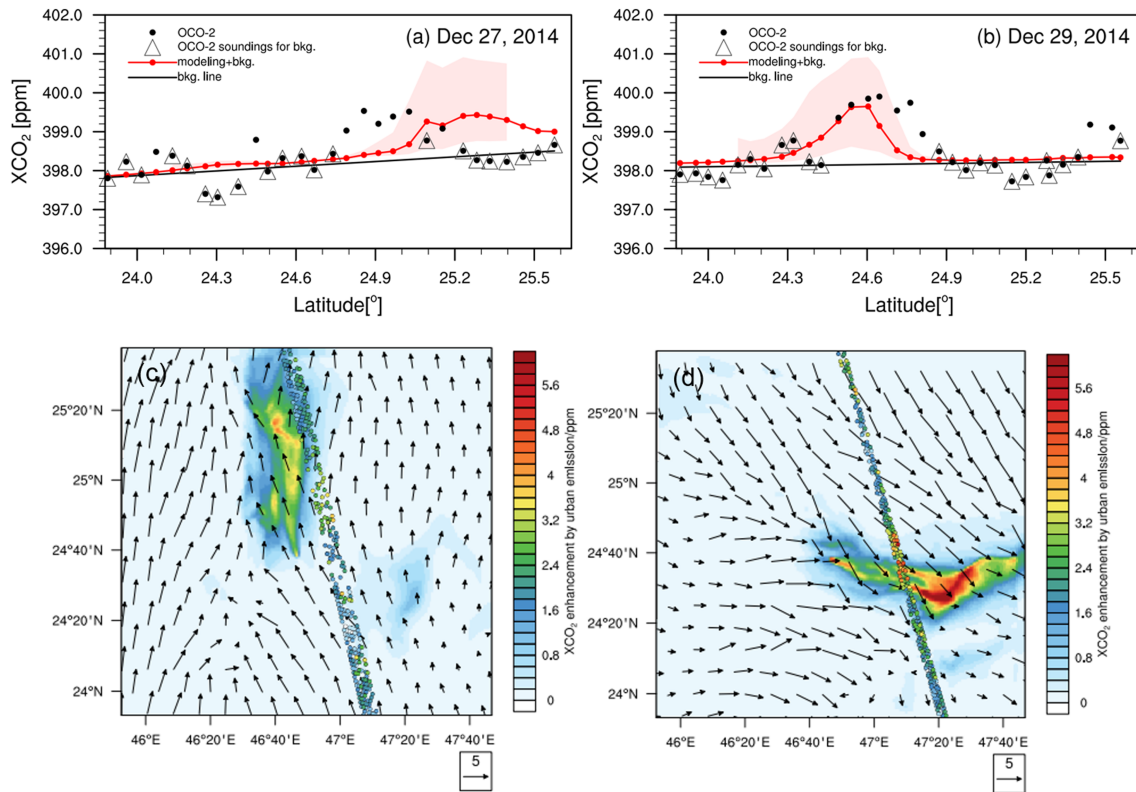
#### 3.1. Local Fossil Fuel $X_{CO_2}$ Enhancement ( $\text{ff}X_{CO_2}$ )

The  $\text{ff}X_{CO_2}$  enhancement, defined as the enhancement in  $X_{CO_2}$  associated with local fossil fuel emissions relative to the background concentration, is used to constrain emissions in this study. In this section, the  $\text{ff}X_{CO_2}$  enhancements are shown for Riyadh, Cairo, and LA, estimated by the simulations of  $\text{ff}CO_2$  tracers tagged with the ODIAC emissions. The results are compared to the observed enhancements derived from the OCO-2  $X_{CO_2}$  data, in order to evaluate the magnitude of  $\text{ff}X_{CO_2}$ , that is, to assess whether the signals of local emissions are robust and detectable. Note that the whole-city emissions from these three cities in the months of the selected tracks are about 3.1, 2.5–2.7, and 4.5–4.7 Mt C/month based on the ODIAC data (see Table 3). The OCO-2 tracks shown in this section are chosen by examining the observed  $X_{CO_2}$  and the simulated plumes, to ensure that the satellite overpasses are downwind of the city with  $X_{CO_2}$  signals attributable to local emissions.

##### 3.1.1. Comparison of Modeled and Observed $\text{ff}X_{CO_2}$

Figure 4 shows the  $\text{ff}X_{CO_2}$  enhancements derived over Riyadh on 27 and 29 December 2014. Overall, the modeled enhancements over this domain are pronounced and can be up to about 6 ppm (Figures 4c and 4d), indicating signals that can be unambiguously detected from space, given the precision and accuracy of the OCO-2 observations. The modeled  $\text{ff}X_{CO_2}$  distributions are characterized by elongated and non-Gaussian plume structures, mostly due to complex horizontal wind fields (Figures 4c and 4d). By examining the simulated  $\text{ff}X_{CO_2}$  at one hour earlier and 1 hr later, rapidly changing fine-scale structures can be seen (Figure S4), indicating notable variations in the distributions of  $\text{ff}X_{CO_2}$  over a few hours. It is also noteworthy that, to examine the impact of domain resolution on the simulated  $\text{ff}X_{CO_2}$ , we compared the results of 1-, 3-, and 9-km spatial resolutions. Lower and smoother peaks can be seen for the coarser resolutions (see Figure S5), which is expected given the larger aggregation errors of the emissions at the lower spatial resolution, particularly for intense point sources. This result suggests the necessity of using a high spatial resolution to reproduce the complex plume structures for the “plume cities”.

Figures 4a and 4b show the modeled and observed  $X_{CO_2}$  along the two tracks. Note that the simulated  $X_{CO_2}$  shown in these plots is derived as the sum of background line derived with the OCO-2 data and the simulated  $\text{ff}X_{CO_2}$ . The observed  $\text{ff}X_{CO_2}$  enhancements reach  $\sim 1.5$  and  $\sim 2.0$  ppm for these two tracks. The magnitudes of

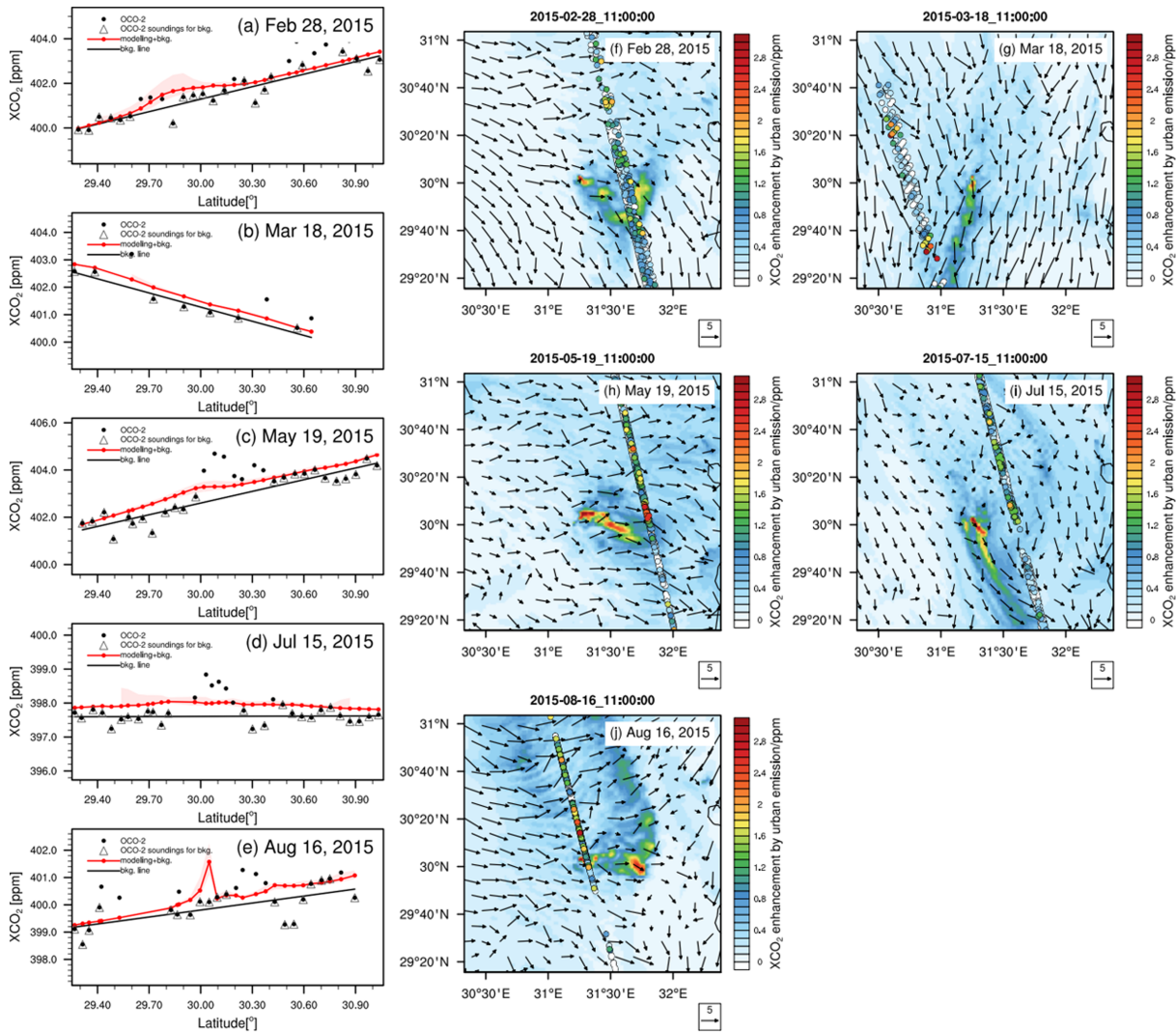


**Figure 4.** Comparisons of modeled and observed  $ffX_{CO_2}$  enhancements by the OCO-2 data on 27 and 29 December 2014 at about 10:00 UTC over Riyadh. Panels (a) and (b) show the OCO-2  $X_{CO_2}$  (black dots) and simulated  $X_{CO_2}$  (red dotted line, sum of  $ffX_{CO_2}$  and the background concentrations) along the two tracks. The OCO-2 retrievals are filtered with quality flag ( $QF = 0$ ) and are 1 s averaged. The blank triangles represent the data used for the derivation of background concentrations (black solid line) by linear regression with these data versus latitude. The uncertainty in the simulated  $ffX_{CO_2}$  related to transport model errors is shown by the light red shading. Panels (c) and (d) show the simulated  $ffX_{CO_2}$  and the observed  $ffX_{CO_2}$  obtained from the OCO-2 data, filtered with  $QF = 0$  only. The background  $X_{CO_2}$  concentrations have been subtracted. The vectors represent 10-m wind with the reference vector standing for the wind speed of 5 m/s.

modeled  $ffX_{CO_2}$  are generally in agreement with the observations, although there is a prominent spatial displacement of  $\sim 0.3^\circ$  in latitude ( $\sim 33.4$  km, Figure 4a) between the observed and simulated peaks on 27 December, and the modeled peak on 29 December is narrower than observed. The large spatial offset might be due to the satellite track transecting the edge of the plume in a nearly parallel way, so that the modeled  $ffX_{CO_2}$  values are very sensitive to errors in horizontal wind field.

For Cairo, the local  $ffX_{CO_2}$  enhancements are examined for five tracks on 28 February, 18 March, 19 May, 15 July, and 16 August 2015, respectively. As shown Figure 5, the modeled  $ffX_{CO_2}$  enhancements over Cairo are mostly  $< 3.0$  ppm over the simulation domain, with some hot spots located close to some intense sources (Figure S3). In comparison to Riyadh, the  $ffX_{CO_2}$  enhancements are overall smaller, as expected given the lower total  $ffCO_2$  emission. Compared to the simulated  $ffX_{CO_2}$ , the observed local enhancement peaks are mostly higher and narrower, especially for the tracks on 19 May and 15 July 2015, while the modeled enhancements are smoother and diffuser. Spatial displacements in the signals are also seen for the tracks on 28 February and 16 August 2015.

It is notable that the background  $X_{CO_2}$  values represented with the background lines exhibit higher latitudinal gradients for most of the selected tracks over Cairo (Figures 5a and 5b) compared to over Riyadh. The background line method used here provides likely reasonable estimations of the background  $X_{CO_2}$ , as general agreements are seen between the observed and modeled results of the integrated  $ffX_{CO_2}$  enhancements along the satellite track within the domain of interest, which can be seen in the inverse estimates of total emission scaling factors (see section 3.2). Hence, this result indicates the advantage of the background line method in deriving background  $X_{CO_2}$  for satellite observations analyzed at a spatial scale relevant to

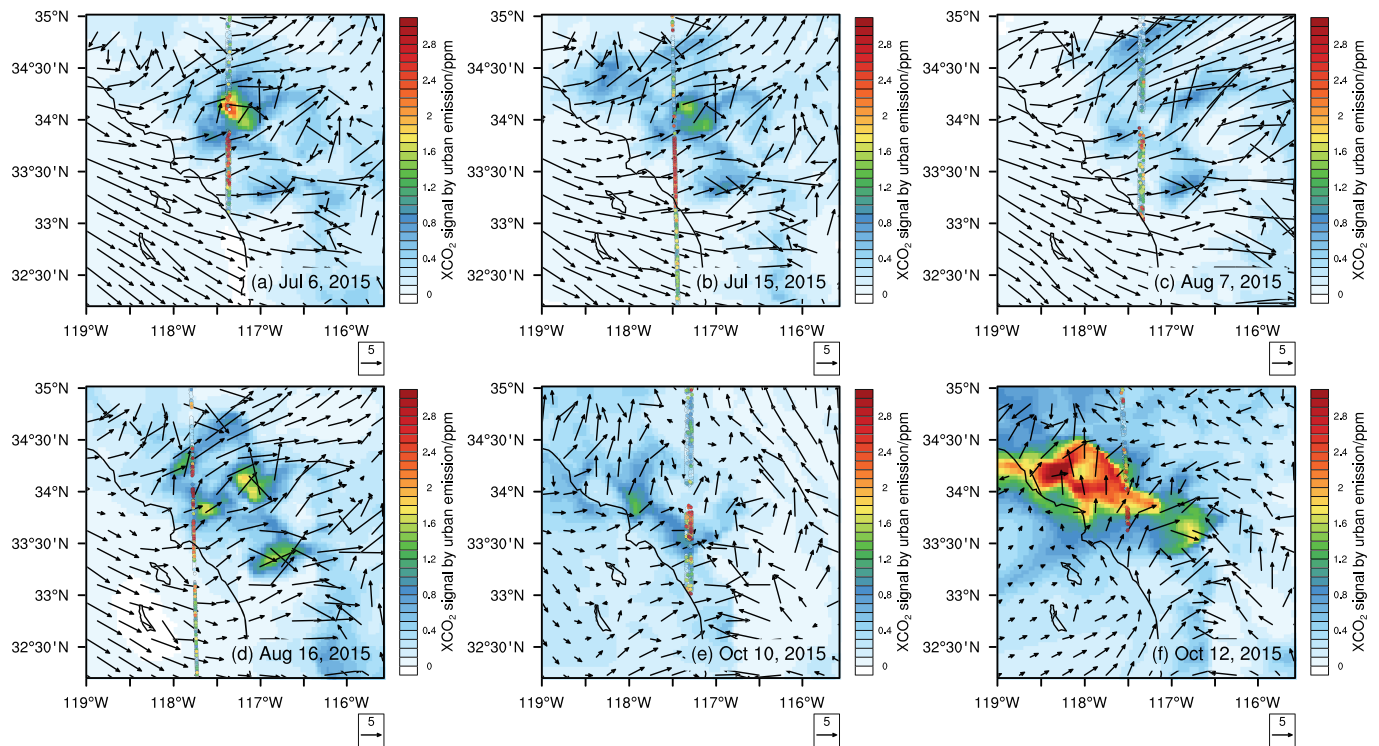


**Figure 5.** Similar to Figure 4 but for the OCO-2 tracks over Cairo on (a and f) 28 February, (b and g) 18 March, (c and h) 19 May, (d and i) 15 July, and (e and j) 16 August 2015 at about 11:00 UTC.

constraining local emission sources. Neglecting the latitudinal gradients in background  $X_{CO_2}$  could lead to biases in  $ffX_{CO_2}$ , as well as in emissions derived from observations.

In respect of LA, the  $ffX_{CO_2}$  enhancements on 6 and 15 July, 7 and 16 August, and 10 and 12 October 2015 are examined with the OCO-2 data and modeled  $ffX_{CO_2}$ . As shown in Figure 6, the  $ffX_{CO_2}$  over the domain can be up to  $\sim 3.0$  ppm and varies notably depending on meteorological conditions, as well as variations in the emissions. In comparison to the  $ffX_{CO_2}$  distributions over Riyadh and Cairo characterized by elongated plumes and rapid changing structures, the  $ffX_{CO_2}$  distributions over LA are more spread and composed of multiple plumes, mostly trapped in the basin during daytime owing to the onshore winds and local terrain. The study by Hedelius et al. (2017) demonstrated persistent differences of  $\sim 0.8$  ppm in  $X_{CO_2}$  between two locations only 9 km apart within the LA basin induced by the steep terrain near the basin. Hence, topography plays a critical role in the distributions of  $ffX_{CO_2}$ .

Figure 7 shows the comparison of the modeled and observed  $X_{CO_2}$  over LA. Note that the OCO-2  $X_{CO_2}$  retrievals north to the northern edge of the desert are excluded in the analysis, in order to avoid using the soundings with high warm levels owing to complex topography and high albedo of the desert area, as can be seen in Figure S6. The thresholds for latitude are  $35.21^\circ N$ ,  $35.05^\circ N$ ,  $35.20^\circ N$ ,  $34.90^\circ N$ ,  $35.25^\circ N$ , and  $35.01^\circ N$ , respectively, determined by examining the observations along with terrain height. Again, the background line has



**Figure 6.** Comparisons of the simulated  $\text{ffX}_{\text{CO}_2}$  (color shading) and observed  $\text{ffX}_{\text{CO}_2}$  enhancements (colored dots, background concentration subtracted) derived from the OCO-2 data collected at about 21:00 UTC over LA. The observation dates are labeled in each panel. The OCO-2 data are filtered by quality flag of zero (QF = 0). The vectors represent 10-m wind, with the reference vector standing for the wind speed of 5 m/s.

been added to the modeled  $\text{ffX}_{\text{CO}_2}$  to present the total values; that is, the  $\text{ffX}_{\text{CO}_2}$  can be seen by the increments above the background lines. Overall, the modeled  $\text{ffX}_{\text{CO}_2}$  is smaller than observed, which is likely owing to the positive biases in the modeled wind speeds, and the possible underestimation of the emissions. Similar to Cairo, prominent latitudinal gradients in the background are seen for some tracks, for example, on 15 July and 12 October 2015.

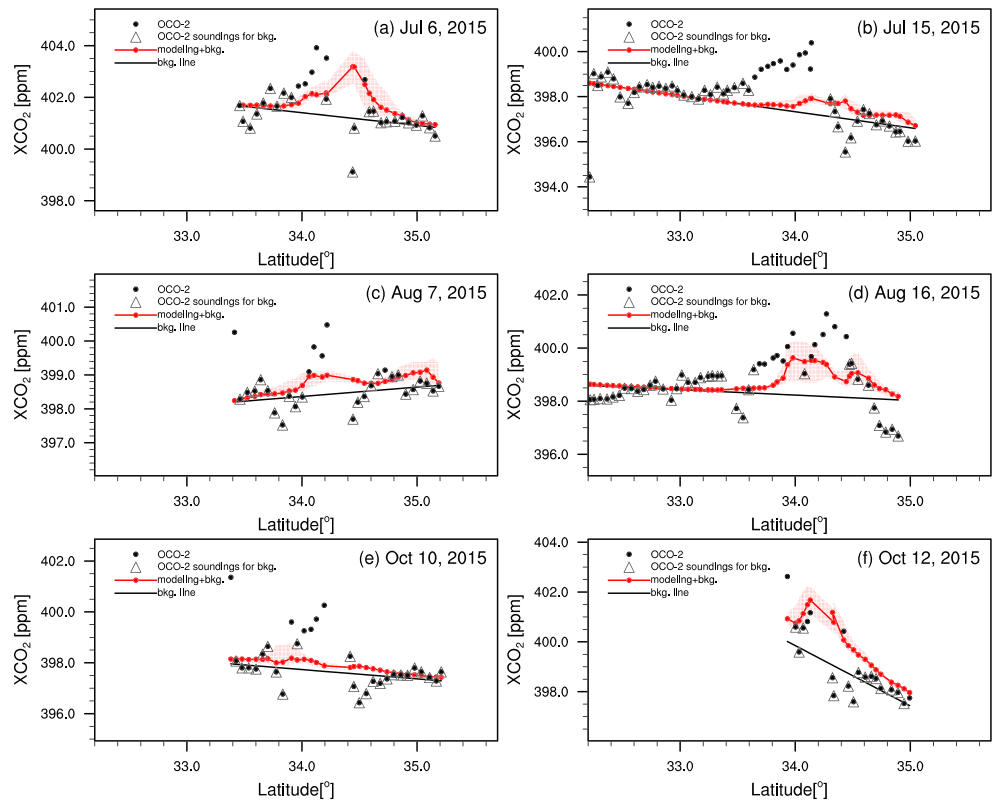
### 3.1.2. Transport Model Uncertainty of Modeled $\text{ffX}_{\text{CO}_2}$

The uncertainty in  $\text{ffX}_{\text{CO}_2}$  related to the transport model error is an important source of uncertainty in inverse emission estimate. Here we evaluate the impact of transport model error for the above-mentioned OCO-2 tracks. Note that different error estimation methods are used for the “plume cities” and the “basin city.”

For Riyadh and Cairo, the transport model uncertainty in  $\text{ffX}_{\text{CO}_2}$  has been shown with the light red shadings in Figures 4a, 4b, 5a, and 5b, estimated with the plume transformation method, as detailed in section 2.5.1. Specifically, for each track the simulated plume is perturbed  $10^4$  times with random errors in wind speeds and wind directions. Due to the non-Gaussian nature of the simulated plumes, as well as the nonlinearity of the sampling process, the perturbed plumes are also non-Gaussian. Hence, we used the interquartile range (difference between the 25th and the 75th percentiles) as the uncertainty spread. Note that the uncertainty spread near the domain border is not shown because of the large portion of missing data after transforming the plume, which can also be seen in Figure 2. The uncertainty in the modeled  $\text{ffX}_{\text{CO}_2}$  over Cairo is up to 0.5 ppm and up to 1.2 ppm over Riyadh. Given that the same probability distribution estimations of wind speed and wind direction errors are used for both cities, the smaller transport model uncertainty in  $\text{ffX}_{\text{CO}_2}$  over Cairo can be mostly attributed to its lower total emission leading to both the smaller  $\text{ffX}_{\text{CO}_2}$  signals and the lower uncertainty spread. Another reason could be the relative locations of the tracks against those plumes they transect.

To evaluate the transport model errors in the modeled  $\text{ffX}_{\text{CO}_2}$  over LA, an ensemble of simulations has been carried out as described in section 2.5.2. The result has been shown by the light red shadings in Figure 7.





**Figure 7.** Similar to Figures 4a and 4b, but for OCO-2  $X_{CO_2}$  measurements over LA and the corresponding simulated results at about 21:00 UTC on (a) 6 July, (b) 15 July, (c) 7 August, (d) 16 August, (e) 10 October, and (f) 12 October 2015.

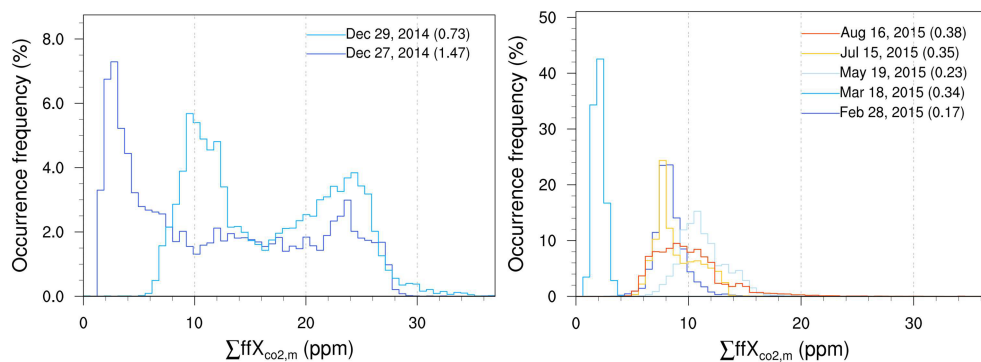
Note that this uncertainty spread is related to the random transport model error; therefore, it is independent of the systematic negative bias of the modeled  $ffX_{CO_2}$  mentioned above. As represented by the range between the minimum and maximum  $ffX_{CO_2}$  among the ensemble members, the uncertainty spread is larger where the  $ffX_{CO_2}$  is stronger, with the maximum uncertainty for each track ranging from 0.32 to 0.93 ppm. The uncertainty spread is found to be overall smaller compared to over Riyadh and comparable to over Cairo. Given that the whole-city emission is the largest for LA among these three cities, this result is likely related to the effect of local terrain trapping the  $ffCO_2$  within the LA basin and making the uncertainty decrease, as well as differences in the uncertainty spreads of wind fields.

### 3.1.3. Integral $ffX_{CO_2}$ Enhancement

In this work, we optimize the whole-city emissions using the latitudinal integral  $ffX_{CO_2}$  enhancements. Hence, in this section we present the modeled integral  $ffX_{CO_2}$  for each OCO-2 tracks shown in the above section and reveal how the integral  $ffX_{CO_2}$  relates to transport model errors.

Figure 8 shows the probability distributions of the simulated integral  $ffX_{CO_2}$  for the tracks over Riyadh and Cairo. The distribution for each track is derived by sampling the plumes perturbed with random transport errors  $10^4$  times. The distributions are mostly non-Gaussian, due to the nonlinearities in sampling transformed plumes at a specified track. To isolate the impact of transport model errors, we also show the normalized uncertainty spread, represented by the ratio between the interquartile range and the median. For Riyadh, the result shows bimodal distributions with normalized spread of 1.47 and 0.73 (Figure 8a). In comparison, the results for Cairo show narrower, less skewed, and unimodal distributions of the modeled integral  $ffX_{CO_2}$  (Figure 8b). The narrowest distribution is seen for the track on 18 March 2015 with the smallest integral  $ffX_{CO_2}$  among the five tracks. The normalized uncertainty spreads range from 0.17 to 0.38, smaller compared to Riyadh. This result is consistent with what has been shown along the tracks (Figures 4 and 5).

The large uncertainty spread over Riyadh can be attributed to the relative location of the track against the plume and structure of the plume. For example, on 27 December 2014, the large uncertainty spread is



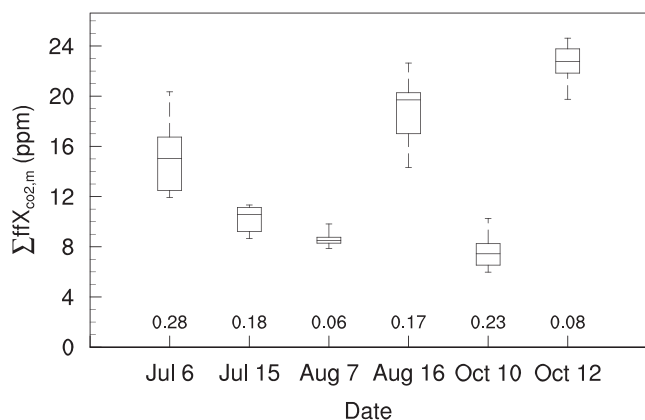
**Figure 8.** Probability distributions of the modeled integral  $\text{ffX}_{\text{CO}_2}$  enhancements ( $\Sigma\text{ffX}_{\text{CO}_2, m}$ ) for the OCO-2 tracks on 27 and 29 December 2014 over Riyadh (left) and 28 February, 28 March, 19 May, 15 July, and 16 August 2015 over Cairo (right). The total number of samples is  $10^4$  for each track. The distributions represent the uncertainty related to random transport model errors in wind speed and wind direction. Note that the y axis limits of the two plots are different. The numbers in the parentheses are the ratios of the interquartile range ( $q_3 - q_1$ ) and the median ( $q_2$ ).

likely due to high sensitivity of the simulated  $\text{ffX}_{\text{CO}_2}$  to transport errors, since the transaction is at the edge of the plume. For the track on 29 December, strong  $\text{ffX}_{\text{CO}_2}$  enhancements ( $>5.0$  ppm) are found downwind of the track due to local accumulation. Those enhancements can be sampled if the wind speed is lower, corresponding to the peak centered at  $\sim 24$  ppm in Figure 8a. Hence, the uncertainty spread also turned out to be large. Thus, it can be concluded that the transport model uncertainty in the integral  $\text{ffX}_{\text{CO}_2}$  is related to combined impacts of the magnitude of prior emissions, errors in winds, transection location relative to a plume, and the plume structure.

For LA, the modeled integral  $\text{ffX}_{\text{CO}_2}$  for each selected track is shown in Figure 9, based on the 18 ensemble members. The normalized uncertainty spread ranges between 0.06 and 0.28, smaller than the results over Riyadh and Cairo. This confirms the results shown earlier for the transport model uncertainty in  $\text{ffX}_{\text{CO}_2}$  along tracks in section 3.1.2, suggesting again that the trapped dispersion by local terrain can make the transport model uncertainty in  $\text{ffX}_{\text{CO}_2}$  smaller.

### 3.2. Emission Estimates and Uncertainty

In this section, we show the results of inverse emission estimates obtained by using the OCO-2 tracks shown in the above section for Riyadh, Cairo, and LA. Different sources of uncertainties are taken into consideration, including measurement errors and transport model errors. The inversions are performed to derive scaling factors of total emissions over each of the selected cities. Note that the fluxes that can be retrieved or viewed from the  $X_{\text{CO}_2}$  measurements vary from track to track, due to temporal variations in wind fields and emissions (Pillai et al., 2015). For example, if we consider an OCO-2  $X_{\text{CO}_2}$  sounding collected at 50 km downwind of the center of an urban emission area, it takes about 5 hr for air parcels to arrive traveling with a velocity of 3 m/s. In other words, the  $\text{ffX}_{\text{CO}_2}$  obtained over or near an urban emission area can be determined by the emissions during several hours ahead of the time of the overpass (Pillai et al., 2015). Given the overpass time of the OCO-2 in early afternoon (with the equatorial crossing time at 13:30 LT), the emission estimates constrained by the OCO-2  $X_{\text{CO}_2}$  measurements of each track could represent the emissions during morning to early afternoon on the date of the overpass. These should be taken into consideration when analyzing the results. The potential biases caused by the OCO-2 satellite observation strategy have been discussed in section 4.



**Figure 9.** Box plot of the modeled integral  $\text{ffX}_{\text{CO}_2}$  enhancements ( $\Sigma\text{ffX}_{\text{CO}_2, m}$ ) for the selected OCO-2 tracks over LA on the dates (in 2015) labeled at the x axis. For each box, the central line indicates the median ( $q_2$ ), and the bottom and top edges of the box indicate the 25th and 75th percentiles ( $q_1$  and  $q_3$ ), respectively. The whiskers extend to the maximum and the minimum. The numbers are the ratios of the interquartile range ( $q_3 - q_1$ ) versus the median ( $q_2$ ).

The inversions are first implemented separately for every track among the selected ones. The inverse estimates of the emission scaling factor have been shown in Table 3, as well as the measurement and transport model uncertainties of the integral  $\text{ffX}_{\text{CO}_2}$ , which are estimated with the methods described earlier in sections 2.1 and 2.5, respectively. Note that the prior scaling factor uncertainty,  $\sigma_a$  for LA has been set to a smaller value than those over Riyadh and Cairo, as in general there is a better knowledge of the emissions over the megacities in the United States (see also section 2.6).

The posterior scaling factors range between 0.92–0.83, 0.70–1.18, and 0.66–1.84 for the selected tracks over Riyadh, Cairo, and LA, respectively (Table 3), indicating notable temporal variations in the emission estimates from case to case. As has been explained, here the estimations represent emissions during a time period of several hours ahead of the time of overpass depending on the meteorological conditions. The posterior uncertainties,  $\hat{\sigma}$ , of the scaling factors for the three cities are found to range between 0.17–0.24, 0.10–0.25, and 0.11–0.16. The posterior scaling factor uncertainty has been shown overall larger for Riyadh and Cairo, compared to over LA.

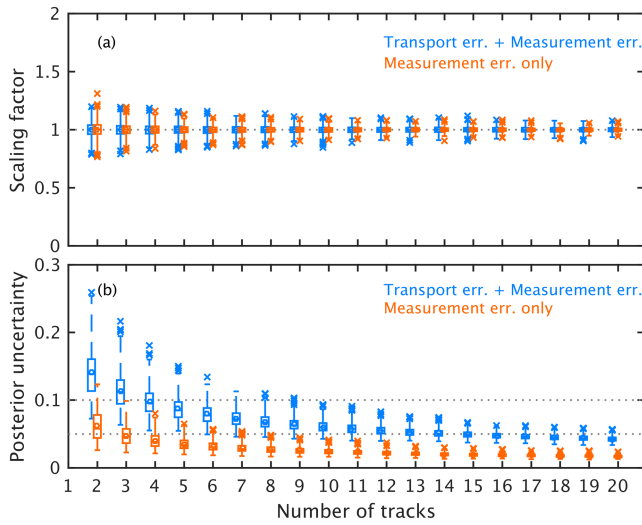
With specified prior emission uncertainty for each city, the posterior uncertainty is related to both the measurement and transport model errors. The relative contribution of transport model uncertainty,  $\sigma_{\text{model}}$ , and measurement uncertainty,  $\sigma_{\text{measurement}}$ , are found to be different over the three cities. The  $\sigma_{\text{model}}$  for Riyadh are larger than the  $\sigma_{\text{measurement}}$  by a factor of about 2 for the two tracks shown here (Table 3). By contrast, the  $\sigma_{\text{model}}$  for Cairo is generally smaller than the  $\sigma_{\text{measurement}}$ , consistent with what has been demonstrated in section 3.1.1. For LA, the relative magnitude of  $\sigma_{\text{model}}$  and  $\sigma_{\text{measurement}}$  varies. The results show a larger  $\sigma_{\text{model}}$  than  $\sigma_{\text{measurement}}$  for four among the six tracks analyzed here. The spread of transport model uncertainty in  $\text{ffX}_{\text{CO}_2}$  over LA is smaller compared to over Riyadh, owing to likely the trapped dispersion within the basin, which is consistent with what has been shown earlier. It can be seen that the transport model uncertainty is closely associated with the magnitude of urban emissions, relative location of plume and satellite track transection, transport model performance, and local topography. Variations of these factors lead to the temporal variability of the posterior emission uncertainty from track to track.

Although the inversion results of whole-city emissions have been demonstrated for each track separately, the sparseness of nearby OCO-2 tracks for many cities is still one of the main limitations to quantify emissions on a regular basis, as well as to track the temporal variations in emissions objectively from space. The numbers of useful tracks that have been analyzed in the above sections are 2, 5, and 6 over Riyadh, Cairo, and LA, respectively, which are chosen by examining the OCO-2 data during September 2014 to November 2015. Hence, we investigated the effect of using  $X_{\text{CO}_2}$  data from two or more tracks on the inverse emission estimates. Given that there is no correlation between the observation uncertainties for any two tracks on different days, the  $\text{ffX}_{\text{CO}_2}$  enhancement obtained from one track cannot affect the emission on another day. Therefore, in order to investigate the effect of incorporating observations on different days, we derive one single scaling factor for all the days of the useful overpasses, as shown in Table 3. Note that the resulting scaling factor for each city would represent the best estimate of emission during several hours before the general time of local overpasses.

The resulting scaling factors ( $\pm 1\sigma$  uncertainty) are  $0.85 \pm 0.16$  and  $0.83 \pm 0.074$  for Riyadh and Cairo (see Table 3), suggesting overestimation of the prior emissions over those time periods that the observations can represent, that is, the monthly ODIAC product. While for LA, the scaling factor ( $\pm 1\sigma$  uncertainty) is  $1.36 \pm 0.074$ , indicating underestimation of the prior emission. As expected, the posterior uncertainty of scaling factor decreases substantially by using all the selected tracks over each city compared to the results with each track separately. The posterior uncertainties of the constrained whole-city emissions are reduced to about 19%, 9%, and 5% for the three cities, with the OCO-2 tracks obtained over about 1, 7, and 4 months, respectively. This result indicates the potential to obtain the emission estimate with a lower uncertainty by utilizing OCO-2 tracks over a longer time period. As explained earlier, the inversion result is closely dependent on meteorological conditions and relative location of urban plume and satellite transection. Hence, we further investigate the performance of utilizing multiple OCO-2 tracks by the OSSEs (section 3.3).

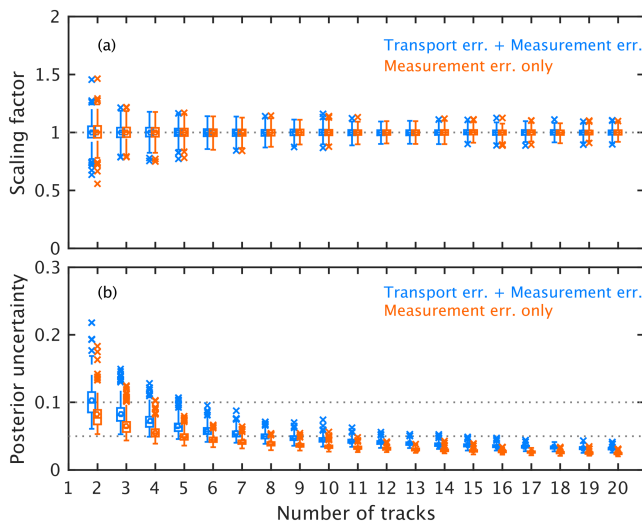
### 3.3. Potential of Constraining the Emissions With Multiple Tracks

In this section, we explore the effect of multiple tracks of  $X_{\text{CO}_2}$  measurements on constraining the  $\text{ffCO}_2$  emissions. OSSEs are carried out for Riyadh, Cairo, and LA to examine the performance of OCO-2



**Figure 10.** Box plot of the inverse estimates of (a) whole-city emission scaling factor and (b) the posterior uncertainty for Riyadh, derived by the OSSE for different number of OCO-2 tracks ( $N$ ). The number of repetition times of inversion with each specified value of  $N$  is  $10^3$ . For each box, the central line indicates the median, the circle represents the average, and the bottom and top edges of the box indicate the 25th and 75th percentiles ( $q_1$  and  $q_3$ ), respectively. The outliers are plotted with “x,” which are greater than  $q_3 + 2 \times (q_3 - q_1)$  or less than  $q_1 - 2 \times (q_3 - q_1)$ . The whiskers extend to the most extreme value that is not an outlier.

truth, that is, 1, for both T01 and T02, which can be expected given that the transport model and measurement errors are assumed to be unbiased. While for LA, the median and average values are around 1.1, suggesting an overestimation by about 10% relative to the truth. This can be attributed to the overall underestimation in  $\text{ffX}_{\text{CO}_2}$  due to the positive bias in the modeled wind speed. Variability of the retrieved scaling factors for different scenes and a specified  $N$  can be seen from the height of each box, that is, the interquartile range, and the range between whiskers extending to the extreme values that are not outliers. Consistently for the three cities, the spread is found to be not obviously affected by the model errors but mainly related to the random measurement errors. The spread becomes less and less with  $N$  increasing from 2 to 20, indicating that the measurement errors tend to have less impact of biasing the emission estimate when we have more data to constrain the emission.



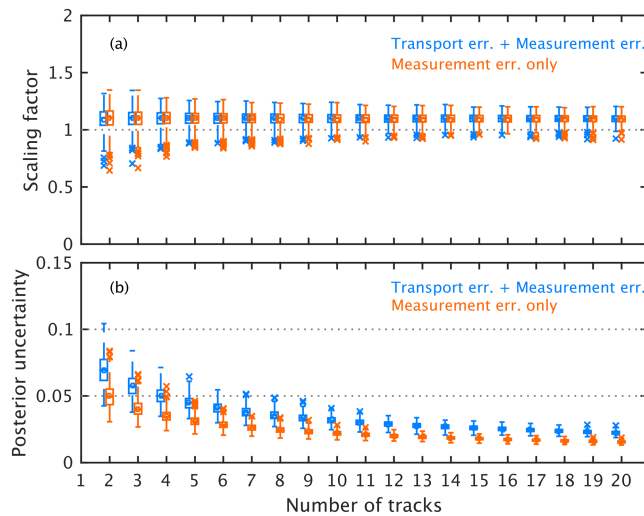
**Figure 11.** Similar to Figure 10 but for the OSSE results for Cairo.

observations from a number of tracks ( $N$ ) ranging from 2 to 20. In order to take scenes with meteorological conditions into consideration, we extract the pseudo observation and pseudo modeling data using all the hourly  $\text{ffX}_{\text{CO}_2}$  modeling results in daytime for each city, rather than using only the modeling results close to the actual overpassing time. In this way we derived 217, 140, and 420 hrs of  $\text{ffX}_{\text{CO}_2}$  data in total, from which the pseudo data are generated accordingly (see section 2.7 for details). Note that the random error term used to generate pseudo observation  $\text{ffX}_{\text{CO}_2}$  represents a lower bound of measurement uncertainty, and only the times with domain-averaged surface wind speed  $\geq 2$  m/s are included. For Riyadh and Cairo, the pseudo data are further filtered with a criterion of the relative location of satellite transection upon  $\text{ffX}_{\text{CO}_2}$  plume (see also section 2.7).

For a specific number of tracks ( $N$ ), we randomly chose  $N$  tracks from the filtered pseudo data and derive the scaling factor with these  $N$  tracks. It is assumed that these  $N$  satellite tracks were collected over a time period during which there was no bias in the prior emissions. We use a Monte Carlo method to evaluate the general performance of multiple tracks by repeating the selection and inversion process for  $10^3$  times. Two tests T01 and T02 are defined here. For T01, only measurement error is applied to the pseudo data, and for T02 both measurement errors and transport model errors are included.

The posterior scaling factors retrieved with different numbers of tracks ( $N$ ) are shown in Figures 10a, 11a, and 12a for the three cities respectively. For Riyadh and Cairo, the median and average values are very close to the truth, that is, 1, for both T01 and T02, which can be expected given that the transport model and measurement errors are assumed to be unbiased. While for LA, the median and average values are around 1.1, suggesting an overestimation by about 10% relative to the truth. This can be attributed to the overall underestimation in  $\text{ffX}_{\text{CO}_2}$  due to the positive bias in the modeled wind speed. Variability of the retrieved scaling factors for different scenes and a specified  $N$  can be seen from the height of each box, that is, the interquartile range, and the range between whiskers extending to the extreme values that are not outliers. Consistently for the three cities, the spread is found to be not obviously affected by the model errors but mainly related to the random measurement errors. The spread becomes less and less with  $N$  increasing from 2 to 20, indicating that the measurement errors tend to have less impact of biasing the emission estimate when we have more data to constrain the emission.

Additionally, the posterior uncertainties of scaling factors are found to become smaller when the number of tracks increases (see Figures 10b, 11b, and 12b). As expected, the posterior uncertainty is overall larger for T02 in comparison to T01, suggesting the prominent contribution of the transport model errors. For each box, 75% of data are lower than the top edge, that is, the 3/4 quartile. The top whisker corresponds to about  $3.275\sigma$  if the data are normally distributed, with about 99.85% coverage below it. Hence, for Riyadh, to retrieve the scaling factor with uncertainty  $\leq 0.1$  (i.e., 10% uncertainty of total emission) with both the measurement and transport model errors considered, 5 or 8 tracks are needed at a confidence level of 75% or 99.85% (Figure 10b). For Cairo, the numbers are found to be lower, that is, 3 or 5 at the two confidence levels (Figure 11b), owing to the smaller transport errors that have been also shown for the inversions based on real tracks. For LA, we consider a lower threshold of posterior uncertainty of 5%, given that the knowledge on prior emissions from the U.S. megacities is generally better, and the prior scaling factor uncertainty has already been set to 0.2. As shown in Figure 12b, 5 or 7 tracks are needed to obtain



**Figure 12.** Similar to Figure 10 but presents the OSSE results for LA.

the goal of 5% uncertainty at the two above-mentioned confidence levels. Note that in the OSSEs we have assumed no spatial correlation in the observation uncertainty. The number of tracks required to reduce the uncertainty would increase accordingly with the spatial correlation considered.

### 3.4. Impact of Local Biospheric CO<sub>2</sub> Variations on the Interpretation of Local X<sub>CO<sub>2</sub></sub> Enhancements

Forward simulations are carried out to investigate the influence of biospheric carbon fluxes on the interpretation of local X<sub>CO<sub>2</sub></sub> variation and the associated uncertainties. Two cases over the PRD region are analyzed using the simulation results and OCO-2 X<sub>CO<sub>2</sub></sub> obtained on 15 January and 4 August 2015, with the results shown in Figure 13. These two cases correspond to conditions of fast atmospheric transport (i.e., high wind speeds) and weak transport (i.e., stagnant winds). The coastal circulation contrasts with the continental wind regimes, with high wind speed over the sea on 15 January and low wind speed on 4 August, opposite to the inland circulation patterns. Because the ffCO<sub>2</sub> emissions come from several cities located in the PRD region, the modeled ffX<sub>CO<sub>2</sub></sub> is characterized by features of multiple overlapping plumes extending downwind from the major sources across the region.

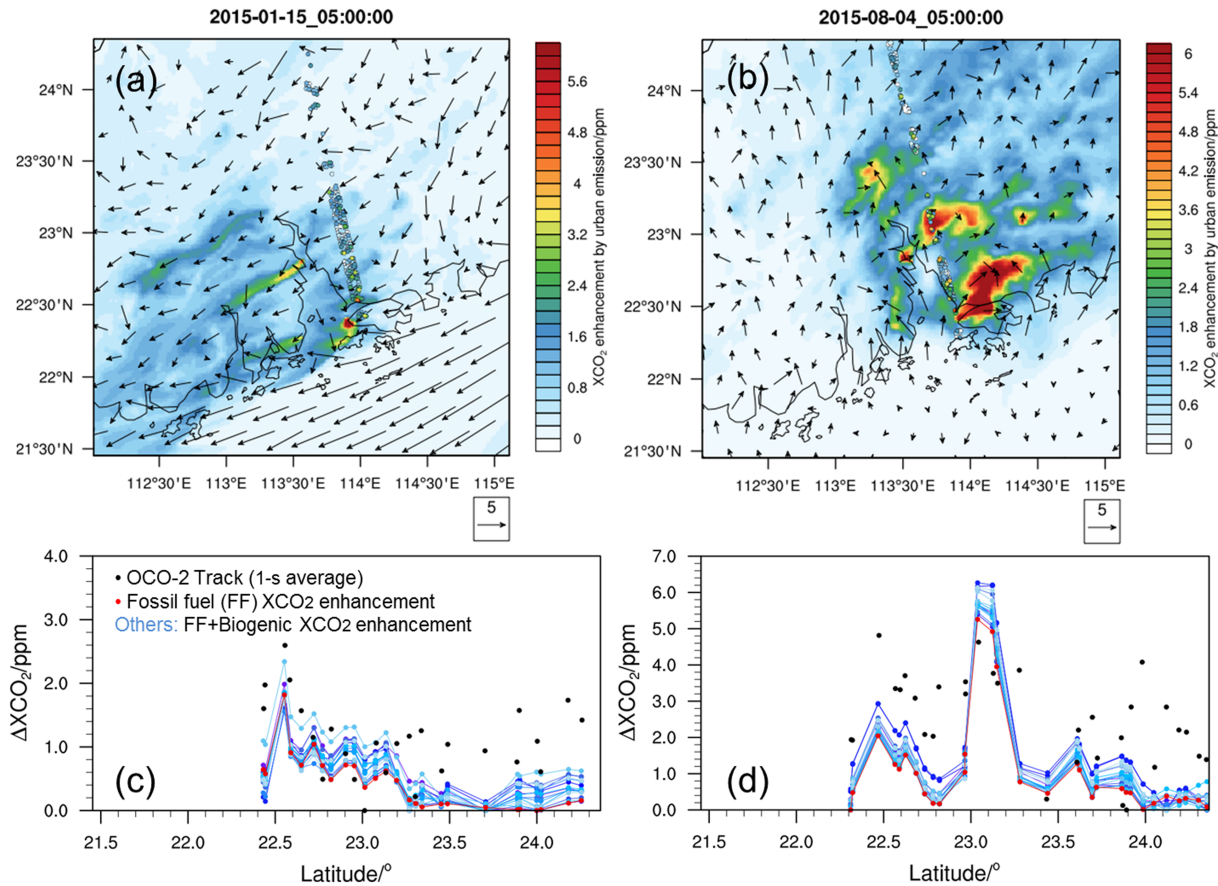
To demonstrate the impact of local biospheric CO<sub>2</sub> variations on local X<sub>CO<sub>2</sub></sub> enhancements, we derive the local X<sub>CO<sub>2</sub></sub> enhancement (referred to as ΔX<sub>CO<sub>2</sub></sub>) from the simulated total X<sub>CO<sub>2</sub></sub> by subtracting the minimum value of the simulated results along a track. The constant background is valid for these simulation results, as the spatial gradient in background concentration is negligible due to a constant boundary condition used in the simulations. The observed ΔX<sub>CO<sub>2</sub></sub> is extracted using the same method as the preceding sections. As can be seen in Figure 13, overall the simulated local enhancements including biospheric X<sub>CO<sub>2</sub></sub> signals are larger than the simulated ffX<sub>CO<sub>2</sub></sub>, and show a better agreement with the observed ΔX<sub>CO<sub>2</sub></sub>. Imposed by NEE from 15 MsTMIP biospheric models, the spread of simulated ΔX<sub>CO<sub>2</sub></sub> indicates the uncertainty in the local enhancement associated with the uncertainty in biospheric fluxes. Considering the tracks shown in Figure 13, the biogenic X<sub>CO<sub>2</sub></sub> variability at local scales account for  $\sim 32 \pm 27\%$  ( $1\sigma$ ) and  $\sim 24 \pm 18\%$  ( $1\sigma$ ) of the latitudinally integrated local enhancement, respectively. In other words, if the biospheric signal is not separated from the local ffX<sub>CO<sub>2</sub></sub> enhancements, the total emissions would be overestimated by about  $47 \pm 37\%$  and  $32 \pm 22\%$  for the two cases examined here.

For Riyadh and Cairo, the biospheric contribution is negligible compared to the local fossil fuel signals, since the local spatial gradient of NEE is much smaller than the ffCO<sub>2</sub> emission. Therefore, simulations for the two cities are not shown. But we note that biogenic fluxes for Cairo (Nile River delta) might be underestimated by the vegetation models. For LA, there are two reasons for not implementing a simulation like the PRD. First, the default MODIS-based GVF climatological maps in WRF show nonrealistic values compared with the real-time MODIS-based GVF maps derived by Vahmani and Ban-Weiss (2016) (Figure S7). Second, NEE is downscaled assuming a constant vegetation productivity within a NEE grid cell ( $0.5^\circ \times 0.5^\circ$ ). However, LA has a variety of climate zones because of its proximity to the Pacific Ocean and the nearby mountain ranges, where a variety of vegetation species exist with different growth patterns (McPherson et al., 2008). Thus, it could be inappropriate to assume a constant productivity. Also, a study based on in situ flask observations in LA of <sup>14</sup>CO<sub>2</sub> indicated about 25% biogenic contributions to the midday CO<sub>2</sub> enhancement (J. B. Miller et al., 2017), in agreement with Newman et al. (2016), but poorly simulated by vegetation model (Feng et al., 2016). More comprehensive data and method are needed to fulfill the estimation of biospheric contribution.

## 4. Discussion

### 4.1. Challenges for Other Cities

In this paper, typical cities with different local topography features are examined, which are selected following the criteria in section 2.3. The simulations over Riyadh and LA demonstrate ffX<sub>CO<sub>2</sub></sub> enhancements



**Figure 13.** Simulated  $fX_{CO_2}$  over the PRD region and the 10-m wind vectors in the 1.333-km resolution domain at (a) 05:00 UTC 15 January 2015 and (b) 05:00 UTC 4 August 2015. The reference vector stands for wind speed of 5 m/s. The colored dots represent the OCO-2 data at about 05:00 UTC over this domain, filtered with quality flag of 0 (QF = 0). The background has been subtracted from the OCO-2 data. The black dots in (c) and (d) show the 1-s averaged observations in the two tracks, with the modeled  $fX_{CO_2}$  and  $\Delta X_{CO_2}$  (owing to both fossil fuel and biogenic fluxes) represented by the red and blue dotted lines, respectively. The bunch of blue lines represents the results using NEE from 15 biospheric models.

overall larger than 1.0 ppm and up to about 6.0 ppm, greater than the uncertainty of retrievals over land (~1 ppm) (Eldering, O'Dell, et al., 2017). However, for Cairo the  $fX_{CO_2}$  values are mostly <3.0 ppm with some hot spots near the large emission sources. For some smaller cities, it would be even challenging to optimize their emissions from space due to limited detectability of fossil fuel imprints. The factors limiting the detectability would include (i) large cloud coverage obscuring the sensor, (ii) occasional high anthropogenic aerosol loading leading to larger measurement uncertainty, (iii) overlapping enhancements from other cities or point sources nearby, and (iv) low  $fX_{CO_2}$  emission. To obtain a bigger chance of unambiguously detecting plumes from cities, an imaging satellite with a wider swath and sufficient precision like the concept of CarbonSat (Buchwitz et al., 2013; European Space Agency, 2015) would be helpful.

Another challenge for many cities is to interpret and distinguish local  $X_{CO_2}$  variations introduced by the biospheric fluxes and fossil fuel emissions. In this work, the variations resulting from the inhomogeneity of local biospheric fluxes have been evaluated by the simulations in section 3.4. Similarly, Dayalu et al. (2017) showed equivalent magnitudes of the vegetation and the anthropogenic signal with the Vegetation, Photosynthesis, and Respiration Model (VPRM) simulations at a surface observation site in Northern China. We note that the NEE data used in this work still need further verification for regions and seasons. In addition, the downscaling of NEE data is based on the assumption of uniform local vegetation productivity, while some studies have reported impact of human interventions on urban vegetation (Hutyra et al., 2014). For example, fertilization is likely to increase both gross primary productivity and respiration of ecosystems in urban areas compared to their natural counterparts. Given the limitation in biosphere

models, many observational data have been used to extract  $\text{ffCO}_2$  signals against the large and varying background, for example, measurements of coemitted components such as CO and  $\text{NO}_x$  (Reuter et al., 2014; Silva et al., 2013; Turnbull, Tans, et al., 2011) with efforts to determine the emission ratios accurately. Additionally, the radiocarbon content of  $\text{CO}_2$  ( $^{14}\text{CO}_2$ ) (Turnbull et al. 2016, 2015) has been used, although existing technology limits  $^{14}\text{CO}_2$  measurement to laboratory-based analysis of individual samples at low sampling resolution. These data could provide more constraints of  $\text{ffCO}_2$  emissions when assimilated in the inversion system jointly.

#### 4.2. Insights From Results of the OSSEs

The performance of multiple tracks on retrieving the scaling factor of whole-city emission has been evaluated by the OSSEs, with transport model errors and measurement errors considered. The results suggest the potential of obtaining emission estimates at a lower uncertainty level and over a longer time window. At a confidence level of 99.85%, the estimated least number of tracks required to constrain the total emissions for Riyadh (<10% uncertainty), Cairo (<10%), and LA (<5%) are 8, 5, and 7, respectively. As the pseudo tracks represent different meteorological conditions, the OSSEs' results can indicate the potential of OCO-2 data in the long run. For example, we examined the number of available tracks over the three cities by counting the tracks located downwind of city and captured peaks in  $X_{\text{CO}_2}$  using OCO-2 data and surface horizontal wind data in NCEP FNL during September 2014 to November 2015, that is, 15 months in total. There are 8 (13 and 17) tracks over Riyadh (Cairo and LA) matching the criteria, corresponding to about one track per 1.75 (1.08 and 0.82) months. Therefore, the general time it takes to collect the above-mentioned number of tracks would be about 14, 5, and 6 months over the three cities. It takes the longest time for Riyadh, which can be likely owing to the larger emission than Cairo, and the less complex terrain than LA. With some other satellite missions being planned and carried out, a shorter time can be expected to retrieve emissions with uncertainty at policy-relevant level.

Positive biases in the emission scaling factors are found for LA, while the estimates are centered at the truth for Riyadh and Cairo. We suspect that the emission bias resulted from mostly the propagation of the positive wind speed bias in the atmospheric model over LA, as the wind speed bias has been considered when constructing the pseudo modeling data. In comparison, the simulations over plume cities located in flat terrain show better results of wind speed and direction; for example, wind speed errors of <1 m/s are reported for modeling results without data assimilation over Indianapolis (Deng et al., 2017) and < 0.8 m/s around Paris (Lac et al., 2013). Data assimilation systems are proven to be useful to significantly improve model performances with decreased systematic errors (<0.5 m/s) (Deng et al., 2017), which can be an effective way of reducing the wind biases for basin cities (Ware et al., 2016).

#### 4.3. Remaining Error Components in the Inverse Emission Estimates

In addition to the uncertainties in transport model, OCO-2 measurements, and biospheric fluxes, there are several sources of errors remaining to be considered. First, the measurement errors of OCO-2 data are assumed to be noncorrelated spatially, as the correlations are yet to be characterized at high resolution at present. Additionally, the nonlinearities in the retrievals and the random components of interference errors (Connor et al., 2016) and imperfection in cloud screening especially for low clouds (Taylor et al., 2016) could introduce large errors in the retrievals. In the OSSEs, the measurement uncertainty has been evaluated using a lower bound.

For the emission inversion system, the prior emissions have been assumed to be perfectly distributed and optimized with a whole-city scaling factor. As noted by Pillai (2015), the flexibility to capture the true spatial variation of fluxes is more limited in the inversion system with one scaling factor for entire city, compared to in pixel- or parameter-wise inversions. However, the pixel-level emission uncertainties would be significantly larger than those of the whole-city emissions. Assessment of prior emissions errors in gridded field is difficult (Andres et al., 2016), which is usually done by comparing emission inventories at an aggregated spatial resolution (Hutchins et al., 2017; Oda et al., 2015). The ODIAC data have been compared to the Hestia emission product (Gurney et al., 2019), which is one of the most accurate and complete emission inventories at the scale of buildings and street segments as evaluated against in situ tower measurements (Lauvaux et al., 2016). At 1-km  $\times$  1-km spatial scale, the result reveals a low-emission limit in ODIAC driven by saturation of the nighttime light spatial proxy, and the median difference ranging between 47% and 84%.

The largest discrepancies were found for large point sources and the on-road sector. More studies on emission comparisons would allow us to realistically constrain the emissions at sector and pixel levels.

Additionally, we note that the temporal representation of emission estimates by OCO-2 measurements can be limited given the sampling strategy. As the satellite measurements are only available in daytime clear-sky scenes,  $X_{\text{CO}_2}$  cannot be evenly sampled in time. This makes it difficult to quantify the diurnal variability in the emissions. It is suggested that  $X_{\text{CO}_2}$  retrievals must be taken into inversion modeling at the original temporal and spatial representativity (Corbin & Denning, 2006), since the clear-sky sampling biases the  $X_{\text{CO}_2}$  if they are used as daily values or averaged over a longer time period. To estimate the effect of sampling bias, we calculated the daily emission from Indianapolis by using data during daytime hours (09:00–14:00 LST) and clear-sky daytime hours, based on the hourly emission of the Hestia product (Gurney et al., 2012) for a full year and cloud cover data from a surface synoptic observation site nearby. The average daily emission for daytime hours has a +14% bias compared to the average including all hours. In addition, the bias increases to +28% when sampling only days with clear-sky conditions. Therefore, the retrieved emissions using OCO-2 measurements would be overall larger than the daily average. As another estimation, we examined the diurnal variability pattern of emissions estimated by Nassar et al. (2013). For Riyadh, Cairo, and LA, the ratios between emission at about the satellite overpassing time against the monthly average value are about 1.195, 1.127, and 1.288, corresponding to biases of about +19.5%, +12.7%, and +28.8% by only sampling in daytime compared to the monthly total emissions. More space observation missions including the Geostationary Carbon Cycle Observatory (GeoCarb) (Moore et al., 2018), OCO-3 (Eldering, Wennberg, et al., 2017), and MicroCARB ([https://microcarb.cnes.fr/en/MICROCARB/GP\\_mission.htm](https://microcarb.cnes.fr/en/MICROCARB/GP_mission.htm)) will further enhance the uniform sampling over urban areas.

## 5. Conclusions

In this paper, we presented the potential of using  $X_{\text{CO}_2}$  observations from OCO-2 to optimize the  $\text{ffCO}_2$  emission from urban areas. High-resolution forward modeling of the atmospheric transport has been implemented to reproduce fine-scale structures of  $\text{ffX}_{\text{CO}_2}$  plumes, as well as to link emissions with observed  $X_{\text{CO}_2}$ . The contributions of transport model errors, measurement errors, and local variability of biospheric fluxes on the inverse estimates of whole-city emissions have been evaluated.

We used a Bayesian inversion approach to optimize the  $\text{ffCO}_2$  emissions from three cities (Riyadh, Cairo, and LA), using the OCO-2 tracks with detectable enhancements collected between September 2014 and November 2015, namely, 15 months in total. The retrieved scaling factors ranged between 0.92–0.83, 0.70–1.18, and 0.66–1.84 for the three cities, indicating notable temporal variations in the inverse emissions from day to day. The posterior uncertainties were largest for Riyadh, mostly due to the transport model uncertainty. Prominent variability of posterior scaling factor uncertainties for the individual tracks was also due to varying meteorological conditions and locations of satellite tracks relative to city plumes. By incorporating all the selected tracks for each city, the posterior uncertainty of scaling factor was found to decrease substantially, corresponding to about 19%, 9%, and 5% uncertainty of the posterior emissions. This indicates a potential to improve current emissions estimates by utilizing OCO-2 tracks over a time period, since the frequency of nearby OCO-2 measurements is limited for each city.

We evaluated the potential of using multiple OCO-2 tracks by performing pseudo data experiments based on the high-resolution forward simulation results for the real cases analyzed above, taking the impacts of both measurement errors and transport model errors into account. For a certain number of tracks, it is assumed here that those satellite tracks were collected over a time period, during which there was no bias in the prior emissions. As revealed by the experiments, to obtain posterior uncertainty  $\leq 0.1$  (i.e., 10% uncertainty of total emission), five or eight (three or five) tracks are needed for Riyadh (Cairo), at a confidence level of 75% or 99.85%. For LA, we consider a lower threshold of the posterior uncertainty of 5%, and five or seven tracks are needed to achieve this goal at 75% or 99.85% confidence level.

The impact of local variability in biospheric fluxes on spatial  $X_{\text{CO}_2}$  variations is evaluated, with the uncertainty of the biospheric fluxes represented by using downscaled fluxes from the 15 biosphere models adopted in the MsTMIP intercomparison. Despite the large  $\text{ffCO}_2$  emissions from the PRD, significant fractions, that is,  $32 \pm 27\%$  ( $1\sigma$ ) and  $24 \pm 18\%$  ( $1\sigma$ ) for the two cases shown, of the local  $X_{\text{CO}_2}$  enhancements are driven by



the local biogenic fluxes. This would lead to an overestimation of total emissions by about  $47 \pm 37\%$  and  $32 \pm 22\%$ . For cities with biospheric fluxes of comparable magnitude but smaller fossil fuel emissions, the contribution is expected to be larger than the values shown above. Therefore, for the cities in midlatitudes and the equatorial areas with prominent local and regional biospheric fluxes, the biospheric contribution is essential for appropriate interpretation of the  $X_{\text{CO}_2}$  retrievals.

For future improvements of the quantification and monitoring of urban  $\text{ffCO}_2$  emissions with OCO-2 data or other polar-orbit measurements, temporal and spatial correlations in prior emissions errors will likely be critical terms to be considered, which are not included in the inversions here. Given the limited satellite overpasses owing to cloud cover, retrieval issues, sampling geometry, and satellite revisiting cycle, etc., the information on prior emissions error correlations will allow us to retrieve the temporal variations and spatial structures of emissions in a more effective way, compared to using one scaling factor for the entire city. In that case, the appropriate number of tracks to constrain urban emissions will depend on the granularity, that is, the spatiotemporal resolutions of emissions in a target city, as well as the precision level required to inform policy decisions.

In addition, compared to the long-term trends in emissions that are more easily detectable, biases in retrieved emissions due to daytime-only sampling are somewhat difficult to be recovered with observations similar to OCO-2 data. Fortunately, with the continuing OCO-2 observations during its extended mission and space-based  $\text{CO}_2$  measurement missions being deployed and planned with geostationary observations or targeting strategy over cities, the  $\text{CO}_2$  records will be extended to potentially allow us to achieve emissions with a better temporal representativity. Our results in this work indicate a promising potential of measurements from OCO-2 or similar missions to constrain urban emissions for cities with robust  $\text{ffX}_{\text{CO}_2}$  enhancements, by using high-resolution transport modeling and the inversion approach. It can be expected that, the OCO-2 data would be more effectively used to improve the accuracy and precision of urban fossil fuel carbon fluxes, in combination with observations from other platforms to support emission reduction strategies.

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