Theoretical assessment of the ability of the MicroCarb satellite city-scan observing mode to estimate urban CO$_2$ emissions

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Abstract.

We assess the theoretical capability of the upcoming French-UK MicroCarb satellite, which has a city-scan observing mode, to determine integrated urban emissions of carbon dioxide (CO$_2$). To achieve this we report results from a series of closed-loop numerical experiments that use an atmospheric transport model with anthropogenic and biogenic fluxes to determine the corresponding changes in atmospheric CO$_2$ column, accounting for changes in measurement coverage due to clouds loading. We use a Maximum A Posteriori inverse method to infer the CO$_2$ fluxes based on the measurements and the a priori information. Using an urban CO$_2$ inversion system, we explore the relative performance of alternative two-sweep and three-sweep city observing strategies to quantify CO$_2$ emissions over the cities of Paris and London in different months when biospheric fluxes vary in magnitude. We find that both the two-sweep and three-sweep observing modes are able to reduce a priori flux errors by 20–40% over Paris and London. The three-sweep observing strategy, which generally outperforms the two-sweep mode by virtue of its wider scan area that typically yields more cloud-free observations, can retrieve the total emissions of the truth within 7% over Paris and 21% over London. by virtue of its wider scan area that typically yields more cloud-free scenes. The performance of the limited-domain city-mode observing strategies is sensitive to cloud coverage and particularly sensitive to the prevailing wind direction. We also find that seasonal photosynthetic uptake of CO$_2$ by the urban biosphere weakens atmospheric CO$_2$ gradients across both cities thereby reducing the sensitivity of urban CO$_2$ enhancements and subsequently compromising the ability of MicroCarb to estimate urban CO$_2$ emissions. This suggests that additional trace gases co-emitted with anthropogenic CO$_2$ emissions, but unaffected by the land biosphere, are needed to quantify sub-city scale CO$_2$ emissions during months when the urban biosphere is particularly active.

1 Introduction

Over Urban areas account for about 70% of global energy-related anthropogenic CO$_2$ emissions are from urban areas (Bulkeley, 2013) (including Scope 1 and 2 emissions), while comprise only 3% of Earth's surface area (Turnbull et al., 2018). Recent studies
have empirically related city-scale CO$_2$ emission estimates with urban population density (Wu et al., 2020; Yang et al., 2020). With urbanization rates projected to increase to 68% by 2050 (Chaouad and Verzeroli, 2018), urban CO$_2$ emissions have become a focus of emission mitigation efforts to limit global warming to 1.5°C above pre-industrial levels and to achieve carbon neutrality by 2050 (Masson-Delmotte et al., 2018, 2021). Satellite-based measurements of atmospheric CO$_2$ are dedicated to support the monitoring of surface CO$_2$ fluxes (Crisp et al., 2017). Here, we explore the theoretical potential of the city-observing mode of the upcoming French-UK MicroCarb satellite to quantify urban emissions of CO$_2$.

Quantifying urban-scale greenhouse gas emissions is currently limited to individual and networks of ground-based (in situ) instruments (Lopez et al., 2013; Bréon et al., 2015; Turner et al., 2016; Helfter et al., 2016; Lauvaux et al., 2016; Davis et al., 2017; Verhulst et al., 2017; Sargent et al., 2018; Kunik et al., 2019) and to sensors on mobile platforms (Mallia et al., 2020; Ionov et al., 2021; Makarova et al., 2021). Emissions of CO$_2$ from the largest emission hotspots, including cities, can already be observed along individual orbits using existing low Earth orbiting satellites of GOSAT (Yokota et al., 2009; Morino et al., 2011; Kort et al., 2012; Janardanan et al., 2016) and OCO-2 (Hakkarainen et al., 2016; Schwandner et al., 2017; Crisp et al., 2017; Eldering et al., 2017) (Hakkarainen et al., 2016; Schwandner et al., 2017; Crisp et al., 2017; Eldering et al., 2017) that will increase the probability of clear-sky data being collected over cities.

MicroCarb, due for launch in late 2023/early 2024, will be the first European satellite dedicated to measuring atmospheric CO$_2$ with sufficient precision to detect the changes associated with surface fluxes across the world (https://microcarb.cnes.fr/en). Data from MicroCarb will ensure continuity with other satellites collecting atmospheric CO$_2$ from low Earth orbit, e.g., GOSAT, OCO-2, TanSat, and GOSAT-2. The instrument is a passive spectrometer that uses an echelle grating to disperse re-
flected sunlight (Pascal et al., 2017) into four short-wave infrared bands sensitive to changes in atmospheric CO$_2$ (1.61 µm and 2.06 µm) and O$_2$ (0.76 µm and 1.27 µm) used as a proxy for the atmospheric path length accounting for changes in, for example, orography, aerosols, and clouds (Bertaux et al., 2020). MicroCarb will employ three observing modes: (1) nadir measurements over land, which have a ground pixel size of 4.5 km (across track) × 8.9 km (along track); (2) sunglint measurements over oceans and lakes with a variable footprint size but with the same order of magnitude as the nadir configuration; and (3) city-mode observations that collect a series of fore- and aft- measurements of atmospheric CO$_2$ at a spatial resolution of 2.25 km × 2.25 km as the satellite passes over a target region, e.g., a city or a calibration reference point collocated with a ground-based remote sensing instrument, e.g., Total Carbon Column Observing Network (TCCON) (Wunch et al., 2011, 2017).

We use a series of closed-loop numerical experiments (i.e., observing system simulation experiments) to evaluate the theoretical ability of MicroCarb city-mode measurements to estimate integrated city-wide CO$_2$ emissions over Paris and London, considering a realistic instrument noise model. We quantify the impacts of different observing modes, random measurement errors, cloud cover, and biogenic fluxes on inferring urban fossil fuel CO$_2$ emissions using synthetic MicroCarb observations. We report results in three different months, accounting for differences in cloud cover, prevailing wind patterns, and biospheric CO$_2$ fluxes. We also compare the performance of two proposed sampling strategies being considered by MicroCarb involving two and three measurement sweeps across a city. In the next section, we describe our modelling approach and provide details about how we simulate MicroCarb measurements using synthetic MicroCarb observations. In section 3, we report our results, including a sensitivity analysis. We conclude the study in section 4, including a discussion of our results and how we anticipate they will be used when MicroCarb is launched.

2 Data and Methods

Here we describe the MicroCarb instrument and its city-mode sampling strategy, and the individual components of the closed-loop numerical experiments (Figure 1) focused on Paris and London during April, July, and December 2018.

2.1 MicroCarb city-mode configuration

The two-sweep and three-sweep city-scan sampling configurations (Figure 2) are being considered by the MicroCarb science team. Both city-scan configurations cover a city the size of Paris or London. For the nominal nadir acquisition mode, each satellite sounding the spatial across-track (ACT) swath is acquired by a detector matrix using that has 90 pixels in a dimension corresponding to the spatial across-track (ACT) swath the spatial dimension and 1024 pixels in the other dimension corresponding to the spectrum, which is used that collects spectral information for the four infrared bands described above. Each individual ACT sounding results from the binning of 30 pixels. The spatial along-track (ALT) dimension is determined by the satellite velocity during the 1.3 s measurement integration time, which leads to the nominal pattern of three soundings of 4.5 km ACT × 8.9 km ALT, leading to a 13.5 km ACT swath and a continuous ALT sampling. The city-mode differs from the nominal nadir pattern in the following three ways: 1) the satellite makes a permanent pitch rotation to slow the projection of the line of sight on the ground, leading
to an ALT footprint size of 2.25 km (for the same 1.3 m s\(^{-1}\) measurement integration time); 2) the ACT footprint binning includes each individual ACT footprint results from the binning of 15 ACT pixels, leading to 6 ACT footprints—six ACT footprints each with an ACT size of 2.25 km; and 3) the ACT swath is increased by making several of the complete city-mode measurement configuration is constructed by the juxtaposition of two or three ALT sweeps, each shifted by 13.5 km. For that purpose, the satellite makes additional enabled by satellite pitch manoeuvres before and after flying over the target — to get a that provides forward and backward viewing of the target. Each successive sweep is shifted by 13.5 km ACT using the a roll-axis pointing mirror to get contiguous spatial observations. Two configurations are implemented. The roll-axis pointing mirror also provides the opportunity to centre the city-mode sampling pattern over cities that lie aside of the satellite track. Further instrument details that enable the city-scale mode can be found in Jouglet et al. (2021).

We study two configurations in the MicroCarb satellite city-scan observing mode (Figure 2): 1) the two-sweep configuration has only forward and backward observations, leading to a \(\sim 40\) km ACT swath (2 x 13.5 km including the off-nadir pointing footprint deformation); and 2) the three-sweep configuration has forward, nadir, and backward observations, leading to a \(\sim 60\) km ACT swath. These are typical values including off-nadir centred city modes. For a city mode centred at nadir, the ACT swath is 34 km for the two-sweep mode, 52 km for the three-sweep mode. The allowed duration of each sweep mode leads to an ALT swath of \(\sim 40\) km.

### 2.2 Cloud screening

The MicroCarb science team will use ECMWF cloud forecasts to program city-mode measurements, in order to maximize the success of cloud-free observations. However, the cloud forecast has a limited precision and small-scale clouds still exist:\(\text{may not be predicted.}\) Here, to realistically describe (and remove) scene observations contaminated by clouds on the spatial scale of the observations, we use ERA5 total cloud cover reanalysis data at 0.25\(^\circ\)\(\times\)0.25\(^\circ\) resolution (Hersbach et al., 2018) for the expected satellite equatorial overpass time (12:00 UTC) of MicroCarb. We use a downscaling method that assumes the dependence of the probability \((f)\) of a cloud-free scene observation to its pixel size \((s)\) and large-scale cloud cover \((P)\) (Palmer et al., 2011):

\[
f(P, s) = F_p(s)g(1 - P),
\]

where \(g\) is defined as a constant factor for alignment between the instrument pixel and the cloud-free subgrid region, which we choose to be 1. The pixel size \((s)\) of MicroCarb city-scan observing mode is approximately 2 km \(\times\) 2 km. Using a constant alignment factor \((g)\) will cause an overestimation of the probability for cloud-free observations at high cloud cover and an underestimation at low cloud cover. \(F_p(s)\) is the penalty function accounting for the adverse effects on cloud-free probability when the effective instrument pixel size is larger than the nominal 1 km\(^2\) (Boesch et al., 2011):

\[
F_p(s) = \frac{26.098s^{-0.45} + 10.18}{26.098 + 10.18}.
\]

For an individual sampling, we generate a pseudo-random number between 0 to each individual measurement pixel, we calculate its cloud-free probability \((f\text{ in Equation 1 and compare it -})\) based on ERA5 total cloud cover reanalysis data at 0.25\(^\circ\)\(\times\)0.25\(^\circ\) resolution, then we generate a random number for each measurement to compare with its cloud-free probability.
to determine its quality flag (QF). If the pseudo-random number is smaller than its estimated probability \( f \), we define it as a \( f \) and determine whether a measurement is cloud-free observation (QF=0). Otherwise, the sampling is defined as a cloud-contaminated observation (QF=1). The first step is related to the organization of cloud distribution based on the ERA5 data, and the second step is related to the randomness of the impact of cloud on satellite measurements.

We report our results over Paris and London for three arbitrary days (5th, 15th and 25th) from the months of April, July, and December 2018 (Figure A1). For some of these days (five for Paris and three for London), we find that the cloud cover is 100%. In these cases, we arbitrarily halve the cloud cover to 50% to generate cloud-free scenes observations (Table A1). Out of the total 228 (two-sweep mode) and 342 (three-sweep mode) individual scenes observations over Paris and London, our method assigns 72 (two-sweep mode) and 124 (three-sweep mode) samplings to be cloud-free in Paris and 69 (two-sweep mode) and 119 (three-sweep mode) cloud-free samplings in London at 12:00 UTC on 15 April 2018, respectively (Figure 2).

### 2.3 Anthropogenic CO\(_2\) emissions

We use the Open-source Data Inventory for Anthropogenic CO\(_2\) (ODIAC, 2019 version) for monthly-mean fossil fuel emissions of CO\(_2\) in Paris and London at a spatial resolution of 1 km \( \times \) 1 km (Oda and Maksyutov, 2011; Oda et al., 2018). The data product uses satellite observations of nighttime light and power plant profiles (including emissions intensity and geographical location) to distribute country-level CO\(_2\) emission estimates from fossil fuel combustion. We use ODIAC to guide our broader study domains for Paris and London, ensuring we include anthropogenic source regions that lie outside the spatial extent of MicroCarb city-mode configurations (Figures 3a and 3b). For our calculations, we exclude large point sources (such as, we exclude a small number of extremely large emissions sources (e.g., power plants and some industrial emission points that typically lie outside the urban core), whose emissions are typically more than ten emissions) that are more than 100 times larger than other diffused (area) emissions in the study domains and will likely (areal and point sources) emission pixels but have lower uncertainties than other urban sectors. In addition, in total, we exclude nine pixels over Paris (about 0.1% of 10080 pixels) and 22 pixels are excluded in London (about 0.2% of 10368 pixels). We exclude these sources because: 1) the atmospheric transport model is not sufficiently accurate and precise to infer the location and emissions strength of these sources and errors in the locations of these point sources diminish the ability to infer sub-city scale emissions (Oda et al., 2017; Roten et al., 2022)—; and 2) these anomalously large emissions complicate the matrix-based inverse method calculation. We acknowledge that real measurements will likely detect these large emission sources, subject to changes in cloud cover and wind direction and speed. For meteorological circumstances where the resulting plume takes the form of thin, anomalously large CO\(_2\) enhancement, only very high-resolution models will be able to describe the plume and estimate its parent emissions with fidelity. Paris emissions of CO\(_2\) (1.71 tCO\(_2\) s\(^{-1}\) in April 2018) are concentrated in the city centre, home to about 2.17 million people (The National Institute of Statistics and Economic Studies). They are 1.4 times the size of those from London (1.25 tCO\(_2\) s\(^{-1}\)), home to about 8.9 million people (United Nations - World Population Prospects), where emissions are aligned in the East-West direction along the Thames River.
2.4 Urban biosphere CO$_2$ fluxes

We use the SMUrF (Solar-Induced Fluorescence for Modeling Urban biogenic Fluxes) model (Wu et al., 2021) to estimate the net ecosystem exchange (NEE) across Paris and London. SMUrF estimates gross primary productivity (GPP) using OCO-2-derived solar-induced chlorophyll fluorescence (SIF) (Zhang et al., 2018) and neural networks to determine the relationship between ecosystem respiration and air and soil temperatures and GPP. SMUrF determines biome-specific model parameters by aggregating multiple flux tower sites from similar biomes across the globe to describe urban biogenic fluxes. We interpolate NEE values to the same grid used by ODIAC.

We evaluate SMUrF NEE estimates in France and UK by using eddy-covariance flux measurements from the European Fluxes Database Cluster: five sites in France (FR-Aur, FR-Hes, FR-Lam, FR-LGt, FR-Pue) and two sites in UK (UK-AMo and UK-EBu). Details of each site can be found at http://www.europe-fluxdata.eu/home/sites-list. These flux sites correspond to a variety of land types, including croplands (CRO), deciduous broadleaf forests (DBF), permanent wetlands (WET), evergreen broadleaf forests (EBF), and grasslands (GRA). We acknowledge these sites are not located in urban areas due to the lack of sufficient urban biosphere flux measurements (only one urban flux site in London and the data are not public), thus this evaluation provides us with limited confidence to understand the gradients in atmospheric CO$_2$ across these cities. All flux data in France are from 2018 (same year as our model meteorology). In the absence of UK flux data from 2018 we use data from 2015 (UK-AMo) and 2011 (UK-EBu).

The seasonal-mean NEEs simulated by SMUrF in July 2018 agree reasonably with the eddy-covariance flux data ($r=0.98$ and slope=1.07 for the ensemble mean of all sites) indicating that the simulated biogenic CO$_2$ fluxes are in a realistic range, although there are differences between site-specific model and observed photosynthetic uptake due to random errors, interpolation errors, and different spatial and temporal scales (Figure A2). Figures 3c and 3d show the spatial distribution of urban biosphere CO$_2$ fluxes on the 15 April 2018. We find substantial biospheric uptake over Paris and London during this month, comparable to the magnitude of fossil fuel emissions but with peak values located in contrasting regions. As we discuss below, these different emission distributions have implications for inferring fossil fuel combustion fluxes from atmospheric data.

2.5 Atmospheric transport model

We use the column version of the Stochastic Time-Inverted Lagrangian Transport model (X-STILT) (Wu et al., 2018a; Fasoli et al., 2018; Lin et al., 2003) to link surface CO$_2$ fluxes to variations of atmospheric CO$_2$ mole fractions at the locations of satellite soundings (Figures A3 and A4). X-STILT traces the movement of air parcels backwards in time for 24 hours. For the purposes of this study, we use a typical averaging kernel profile of OCO-2 that describes the vertical sensitivity of the column observations to changes in CO$_2$ in the column. We apply this averaging kernel and pressure weighting functions to the model fields to describe the footprints of satellite-based atmospheric CO$_2$ observations. The footprints describe the sensitivity of CO$_2$ columns at the receptors (locations where the satellite observes the atmosphere) to upwind surface fluxes.
To drive air parcels in X-STILT we use Global Forecast System meteorological data with a horizontal resolution of 0.25 degree (GFS0.25) (National Centers for Environmental Prediction, National Weather Service, NOAA, U.S. Department of Commerce, 2015). At the receptor, 3000 air parcels evenly distributed from the surface to 3 km height are released from the atmospheric column of each observed scene. We simulate footprints for the cloud-free scenes sampled by MicroCarb. The sum of the convolution of footprints and the ODIAC and SMUrF fluxes represents the urban CO\textsubscript{2} enhancements from upwind CO\textsubscript{2} fluxes, as sampled by air parcels arriving at the locations of each observed scene.

### 2.6 Maximum A Posteriori inverse method

We use the Maximum A Posteriori (MAP) inverse method (Enting, 2002; Tarantola, 2004; Feng et al., 2009) in which we solve for a posteriori CO\textsubscript{2} emissions by minimizing a cost function that describes the mismatch between the model-calculated enhancements and the measurements, accounting for a priori and measurement uncertainties. Minimizing the cost function results in the following expressions:

\[
\hat{x} = x_0 + (HB)^T( HBH^T + R)^{-1}(y - Hx_0),
\]

\[
\hat{S} = B - (HB)^T( HBH^T + R)^{-1}(HB),
\]

where \(\hat{x}\) and \(\hat{S}\) denote the a posteriori state of grid-based CO\textsubscript{2} emission estimates across a city and the associated error covariance matrix, respectively. \(x_0\) and \(B\) denote the a priori emissions and the associated error covariance matrix. The measurement vector \(y\), includes the atmospheric CO\textsubscript{2} column enhancements, with the associated errors described by the observation error covariance matrix \(R\) (including measurement errors and atmospheric transport errors). \(H\) denotes the Jacobian matrix that describes the sensitivity of atmospheric CO\textsubscript{2} column enhancements to changes in CO\textsubscript{2} emissions.

To evaluate the theoretical performance of the MicroCarb city-mode data on improving a priori knowledge of urban CO\textsubscript{2} emissions over Paris and London, we use an error reduction metric (\(\eta\)) that takes into account differences between the a priori and a posteriori error covariance matrices (Palmer et al., 2000):

\[
\eta = \left[ 1 - \left( \frac{S_{i,i}}{B_{i,i}} \right)^{1/2} \right] \times 100\%,
\]

where the subscripts denote the diagonal elements of the error covariance matrices. The larger value of \(\eta\) the more reduction in uncertainty of CO\textsubscript{2} emissions due to using the atmospheric CO\textsubscript{2} measurements.

### 2.7 Closed-loop numerical experiments

Figure 1 describes the experimental approach we take to assess the theoretical ability of MicroCarb city-scan mode measurements to quantify urban emissions of CO\textsubscript{2} from Paris and London. Here, we discuss the individual steps involved, following Wu et al. (2018b). We use the ODIAC emissions and the SMUrF fluxes, described above, as the true state. We optimize individual flux components as separate state vector elements. The spatial resolution of fluxes is 1 km × 1 km and the temporal resolution is monthly. In Paris, the size of the state vector is 10080 × 1 for only fossil component and 20160 × 1 for including...
fossil and biogenic fluxes. The introduction of biogenic fluxes doubles the size of the state vector because we optimize them separately. The corresponding CO₂ column enhancements are generated from the true fluxes by the column-mean X-STILT transport model, as described above.

We then add realistic observation noise to each cloud-free scene (determined by the cloud coverage) based on estimates from detailed simulations of the MicroCarb instrument. The random measurement error budget. These simulations transfer the instrumental performances (signal to noise ratio, spectral sampling, and spectral band positions) to random measurement error using the gain matrix formalism from Rodgers (2000). Only the radiometric noise has been included here. The measurement random error is expressed as the standard deviation of a Gaussian probability distribution law, which is computed for each footprint given its Sentinel-2 L1C reflectance, solar zenith angles, and atmospheric profiles for temperature, H₂O, and CO₂. The random measurement errors range between 0.4–1.6 ppm with a mean value of 0.93 ppm for both the two-sweep and three-sweep configurations over Paris and mean values of 0.88 ppm (two-sweep) and 0.91 ppm (three-sweep) over London (Figure A5). We add a realistic additional 20% random error to the urban CO₂ column enhancement to account for atmospheric transport errors (Deng et al., 2017; Lauvaux et al., 2019) assuming the transport model is unbiased. A more formal estimation of atmospheric transport errors would suggest more confidence than we have in our estimate of the uncertainty in the X-STILT model. We will examine the impacts of different transport errors (described by the observation error covariance matrix) on the error reduction (η) of flux inversion. Based on these error estimates in the observation error covariance matrix (R), we generate synthetic observed CO₂ enhancements, which are assumed to be unbiased.

Evaluating our ability to reduce a priori flux errors (including systematic and random errors) is the primary objective of this study. We respectively assume a mean systematic and random flux error of 2 μmol m⁻² s⁻¹ (about 34%) for fossil fuel emissions (Oda et al., 2019; Wu et al., 2018b). The mean random error of the biological fluxes is estimated as 25% (which is an approximately mean value of the standard deviations of the seasonally-averaged biogenic CO₂ fluxes) based on the eddy-covariance flux measurements at the ecosystem sites described above. The total systematic and random flux errors are constrained by the Chi-square test that should be close to one. We use an eigenvalue decomposition method to generate a priori flux noise from the flux error covariance matrix (B). The flux error vector of flux noise is calculated by multiplying the eigenvector of the flux error covariance matrix (after considering the spatial correlation of errors) with a normal distribution vector characterizing the systematic and random flux errors. The flux errors are spatially correlated with an exponentially decaying function of the distance between emission grids and the spatial correlation length is estimated to be 10 km (Saide et al., 2011; Wu et al., 2011). We then sample the resulting a priori distribution of CO₂ column enhancements where there are cloud-free measurements and use the MAP inverse method to infer the a posteriori flux estimates and associated uncertainties over Paris and London. We can assess the ability of using the synthetic measurements to retrieve the “true” urban emissions from Paris and London at different times throughout the year.
3 Results

We report our results over Paris and London for three arbitrary days (5th, 15th and 25th) from the months of April, July, and December 2018. This allows us to investigate the impacts of different cloud cover and wind fields on the flux inversion of the proposed MicroCarb sampling strategies, and to understand how seasonal changes in the urban biosphere will influence our ability to determine urban anthropogenic emissions of CO$_2$. In total, we report the results of nine scenarios (three days per month for three months). In terms of structuring our results, we first use the results on 15th April 2018 to demonstrate the effectiveness of the inversion system, and then generalize these results for all nine scenarios. We firstly only include the fossil component and focus on estimating fossil fuel CO$_2$ emissions (Sections 3.1 and 3.2), and then extend the inversion system to include the biogenic component (Sections 3.3 and 3.4).

3.1 Flux estimates on 15th April 2018

Figure 4 shows the spatial distribution of the true and a priori fossil fuel CO$_2$ fluxes over Paris and London. Application of our random and systematic errors, as part of the closed-loop experimental configuration, introduces a significant difference between the truth and a priori emissions (Figures A6a and A7a). We use cloud-free satellite measurements to sample urban CO$_2$ enhancements that correspond to these emissions, as observed by MicroCarb using its proposed two-sweep and three-sweep city mode configurations (Figure 2). The number of cloud-free data over Paris at 12:00 (UTC) on 15th April 2018 using the two-sweep and three-sweep observing modes is 72 and 124, respectively. The corresponding number of cloud-free data over London is 69 and 119, respectively. The a posteriori CO$_2$ fluxes over Paris (Figures 4c and 4d) and London (Figures 4g and 4h) illustrate that cloud-free MicroCarb observations can broadly retrieve the spatial structure and magnitude of the true emissions. The a posteriori emission optimized by the two-sweep configuration can retrieve the true integrated emission for Paris within 18% from an a priori state of 52% larger than the truth. Similarly, this configuration results in an a posteriori emission estimate for London that is within 28% of the true value from an initial a priori difference of 74%. The three-sweep mode performs better to retrieve the total emissions of the truth within 7% (Paris) and 21% (London). For both cities, the a posteriori uncertainty is approximately half that of its a priori value. The three-sweep configuration outperforms the two-sweep configuration due to a higher number of cloud-free observations. For this day, the X-STILT footprints, describing the sensitivity of column-averaged CO$_2$ measurements to urban emissions, come from south of both Paris (Figure A3b) and London (Figure A4b). Consequently, sizeable flux correction and error reduction are shown in the south of Paris (Figure A6) and London (Figure A7). The error reduction is more significant for the three-sweep configuration due to more cloud-free observations.

Figure 5 shows the urban CO$_2$ column enhancements that correspond to the true, a priori, and a posteriori emissions. The a posteriori CO$_2$ enhancements are closer to the “perfect” observations (OBS), which are generated from the true emissions, demonstrating the effectiveness of using the MicroCarb city-mode samplings to correct flux errors and improve the estimation of urban CO$_2$ enhancements. The prior and posterior column CO$_2$ enhancements for Paris and London on 15th April 2018 range 0–4 ppm with mean values of the “perfect” observations 1.28±0.71 ppm over Paris and 0.67±0.46 ppm over London.
for the two-sweep mode, consistent with previous studies on other cities (Kort et al., 2012; Hedelius et al., 2017, 2018; Kiel et al., 2021). Variation in enhancements reflects the spatial distribution of emissions across the cities and the footprints related to local meteorological conditions.

3.2 Sensitivity to cloud cover and measurement uncertainty

We estimate the sensitivity of flux error reduction to observation uncertainties (including atmospheric transport errors) and the data availability to a different number of cloud-free observations within the three-sweep configuration (Figure 6). The peak spatially averaged error reduction is approximately 18% with the observation uncertainty of 0.5 ppm and the largest number of measurements. The lowest error reduction is for the minimal data availability with 80% cloud cover. Moreover, the scenario of all data (342 OBS) with 1.25 ppm observation uncertainty shows similar error reduction to the scenario of clear sky with 1 ppm observation uncertainty for both Paris and London, indicating better measurement precision can partially compensate for fewer measurements. For both cities, the sensitivity of error reduction to changes in the observation uncertainty are similar for different cloud coverage, with increasing cloud coverage resulting in a near-uniform decrease in the flux error reduction.

3.3 Influence of biogenic CO$_2$ fluxes

Figure 7 shows how the introduction of biospheric CO$_2$ fluxes can influence the ability to infer anthropogenic CO$_2$ emissions. For this calculation we use the same true and a priori fields for the anthropogenic emissions of CO$_2$ over Paris on 15th April 2018 (Figures 4a and 4b). The effect of adding biospheric CO$_2$ fluxes is to reduce the urban CO$_2$ enhancements. These smaller column enhancements, within the context of the measurement uncertainties, limit our ability to reduce the random and systematic flux errors we impose as part of our closed-loop experiment, and consequently to quantify correctly anthropogenic CO$_2$ emissions. The total a posteriori anthropogenic emission estimate over Paris inferred using the three-sweep mode is 2.19 tCO$_2$ s$^{-1}$ (Figure 7a), which is further from the true state than the emission estimate for the same scenario but in the absence of biospheric fluxes (Figure 4d). The a posteriori uncertainty increases by 51% from 0.45 to 0.68 tCO$_2$ s$^{-1}$ relative to the scenario without biospheric fluxes. The a posteriori flux noise is larger (Figure 7b) with a degraded error reduction (Figure 7c) relative to the scenario without biospheric fluxes (Figures A6f and A6g). We find that biospheric uptake reduces the signal to noise ratio in satellite measurements (Figure 7d), which limits the ability to retrieve the true anthropogenic CO$_2$ emissions. We acknowledge that the degree of the influence from biosphere is subject to the uncertainty of a priori biogenic fluxes, the separation of anthropogenic signals from net CO$_2$ measurements, and the scale of inverted fluxes.
3.4 Ensemble analysis of Paris and London scenes

We generalize the results for nine individual scenarios over Paris and London, distributed evenly between April, July, and December 2018 (Figures 8 and 9). The nine scenes differ because of different biospheric fluxes and meteorological conditions, including cloud cover and prevailing wind that impact the data availability and footprints. In general, all configurations improve the \emph{a priori} estimate, the extent of which is determined by the number of cloud-free scenes observations over each city, the sweep configuration used, and the introduction of seasonal biospheric CO$_2$ fluxes. This general discussion is consistent with our analysis from the 15th April 2018, described above. The three-sweep observing mode results in the most accurate reproduction of true urban emissions over Paris and London due to a higher number of cloud-free scenes observations (Figure 8). Ignoring the impact of biospheric CO$_2$ fluxes, the mean values of error reduction (including systematic and random error corrections) are 39% (two-sweep) and 46% (three-sweep) for Paris and those values are 44% (two-sweep) and 50% (three-sweep) for London (Figures 9a and 9c). In Paris, the scenario that results in the largest improvement to the \emph{a priori} estimate is on 25th December 2018. This is due to a greater number of cloud-free scenes observations (Table A1) and weakly dispersed footprints from the south of Paris (Figure A3i) that is correlated with the distribution of the \emph{a priori} flux noise (Figure A6a). A contrasting scenario, associated with weak improvement to the \emph{a priori} estimate, is over Paris on 5th July 2018. This is mainly due to the footprint originating from the north of the city (Figure A3d) and therefore insensitive to the \emph{a priori} flux noise. Generally, scenarios associated with the weakest improvements are associated with the lowest number of cloud-free scenes observations, e.g., 25th April 2018 over Paris (Table A1). We find similar results over London. Our analysis suggests that cloud cover and prevailing wind (speed and direction) are both determinants of city-scan observations being able to infer accurately urban anthropogenic emissions of CO$_2$.

Including the impact of seasonal biospheric CO$_2$ fluxes (with the associated uncertainties) generally reduces the associated atmospheric urban CO$_2$ column enhancements and consequently limits the ability to infer fossil fuel CO$_2$ emissions reduce systematic and random errors in the fossil component (Figures 8b and 8d), particularly during April and July when the biosphere is more active and the flux error reductions are 20–40% over Paris and London (Figures 9b and 9d). We acknowledge our results are dependent on the uncertainties estimated for these biospheric fluxes. Decreasing the uncertainty of the \emph{a priori} biospheric CO$_2$ fluxes will strengthen the relationship between the data and simulated urban CO$_2$ enhancements that consequently contributes to accurately quantifying anthropogenic CO$_2$ emissions.

4 Conclusions and Discussion

We evaluate the ability of MicroCarb city-scan observing modes to infer sub-city scale anthropogenic CO$_2$ emissions with sufficient measurement accuracy and density. Both the two-sweep and three-sweep observing modes are able to reduce \emph{a priori} flux errors for 20–40% (including bias and random errors) over Paris and London, and retrieve the emission spatial structures of the truth. The three-sweep mode generally outperforms the two-sweep mode due to a wider scan area and a higher number of cloud-free observations. Cloud cover and prevailing wind are two determinants of city-scan observations for inferring accurately urban anthropogenic emissions of CO$_2$. The inversion system corrects more flux errors when there is less
cloud cover and the footprints cover the area correlated to the distribution of a priori flux noise. The column-averaged urban CO₂ enhancements are 0–4 ppm for the cities we examined, which is about 1% of atmospheric background concentration with mean values of 1.28±0.71 ppm over Paris and 0.67±0.46 ppm over London. Quantifying sub-city scale emissions requires improving the accuracy and precision of satellite-based CO₂ measurements. Moreover, the presence of biospheric CO₂ fluxes diminishes the ability to estimate anthropogenic CO₂ emissions. Combining additional trace gases that are emitted with CO₂ during combustion (e.g., CO or NO₂) can help to infer urban CO₂ emissions (Konovalov et al., 2016; Reuter et al., 2019; Hakkarainen et al., 2021; Park et al., 2021; Wu et al., 2022a), especially during the growing season when the land biosphere absorbs atmospheric CO₂.

Estimating the regional/local atmospheric background. We demonstrate the ability of using synthetic unbiased MicroCarb data to infer urban CO₂ emissions, but there are additional limitations and uncertainties introduced by using real data that will have systematic and random errors (Broquet et al., 2018). Reducing random errors can be achieved by increasing the number of independent measurements. Addressing regional systematic errors is more difficult. For example, we find that column-averaged urban CO₂ enhancements are typically less than 1% of the atmospheric background column concentration (about 415 ppm), so that a regionally varying systematic measurement error of the order of 1 ppm would significantly degrade emission estimates inferred from the data. We expect that larger scale systematic errors would be identified and corrected by ground-based remote sensing instruments (e.g., TCCON). Establishing city-scale ground-based atmospheric remote sensing networks would help correct regional systematic errors.

Estimating the regional background CO₂ column concentration is important for quantifying the magnitude of urban CO₂ enhancements due to net urban emissions, including anthropogenic and biospheric CO₂ emissions. A range of methods being employed to determine the background, such as calculating the mean value of column CO₂ surrounding the city area (Kort et al., 2012), estimating a dynamic background derived from a two-step linear regression method (Ye et al., 2020), or using the independent TCCON data of a remote site to derive background concentration (Kiel et al., 2021). In this study, we have been able to sidestep this issue but only because it is a have been employed in different studies to quantify regional background values (Kort et al., 2012; Ye et al., 2020; Kiel et al., 2021). Here, we have sidestepped this issue by taking advantage of the closed-loop simulation. For inversions with experiment configuration, To estimate city-scale CO₂ emissions from real MicroCarb data, we will need to consider an additional uncertainty associated with the calculation of the elevated CO₂ column. In addition, This study makes the simplifying assumption that the three-sweep observing mode outperforms the two-sweep mode because of the larger number of observations that help to reduce random errors. The three-sweep mode also has a wider scan outside the urban plume that will help to reduce bias in estimating regional background CO₂ values.

This study assumes unbiased and uncorrelated atmospheric transport errors. However, these errors are likely to be correlated at the sub-city scale. Our simplifying assumptions yield the maximum flux error reduction for the proposed MicroCarb samplings and represent a We acknowledge that X-STILT used in the study is not tuned for simulating elevated emissions given the use of vertical-integrated 2-D footprint. Future work will include the use of the vertical resolved footprint associated with each air parcel, and the vertical profiles of sector-specific emissions to improve the hyper-nearfield simulations.
of point sources following prior approaches (Maier et al., 2022; Mallia et al., 2018). Also, relatively low (high) wind speed will produce the strong (weak) urban enhancement signals but are associated with relatively large (small) transport errors. Better understanding atmospheric transport and a priori flux errors is essential to improve the accuracy and precision of high-resolution urban a posteriori CO₂ flux estimates. Our assumptions about atmospheric transport errors likely result in the best-case scenario for the level of error reduction that could be achieved by the MicroCarb city-scan observing modes.

As with other polar-orbiting satellites, MicroCarb provides a limited amount of data over urban areas due to seasonal cloud cover (especially for mid-to-high latitude cities) and elevated aerosol loading in urban areas, and because of their orbital configuration. As a result, the instrument will not see CO₂ immediately overhead of some parts of the city and some cloud-free pixels, particularly between cloud breaks, will likely have a bias due to 3-D cloud radiation effects (Massie et al., 2017). Table A1 lists the number and percentage of cloud-free data over our test cities. In Paris, four of the nine days collect cloud-free data (the other five days are covered by cloud at 12 UTC). For these four days, only 40-50% of the data are cloud-free. Lei et al. (2021) found only 17% of OCO-2 soundings over the 70 most populated cities are of high-quality, consistent with our calculations (18-22%). The number of available measurements is reduced further considering the effective footprints over urban areas. Using satellites to collect data throughout the day over a particular location only be achieved by with a constellation of satellites or with a satellite in a geostationary orbit, neither solution is currently planned.

Seasonal biospheric uptake of CO₂ within and around major cities will weaken observed CO₂ gradients across cities that would otherwise be determined by anthropogenic emissions. The use of 14C isotope and CO has been demonstrated to separate fossil and biogenic fluxes inferred by ground-based atmospheric CO₂ measurements (Turnbull et al., 2015; Basu et al., 2020; Miller et al., 2020; Wu et al., 2022b). There are number of candidate approaches to alleviate this challenge for satellite-based CO₂ inversions but none have been proven extensively. A coupled assimilation of CO₂ with other trace gases that are co-emitted with anthropogenic CO₂ emissions such as CO or NO₂ provides constraints of the activity rates of individual anthropogenic sectors but still require sector-based knowledge of emission factors. Likewise, being able to reliably relate net biospheric fluxes to data products observed from satellites could also help to isolate fossil fuel emissions of CO₂ from natural fluxes over cities.

**Code and data availability.** The ODIAC emission inventory is available at http://dx.doi.org/10.17595/20170411.001 hosted by the Center for Global Environmental Research, National Institute for Environmental Studies (https://db.cger.nies.go.jp/dataset/ODIAC/). Biogenic CO₂ flux estimates from SMUrF are available at https://doi.org/10.3334/ORNLDAAC/1899. The X-STILT model is available at https://github.com/uataq/X-STILT. Codes of this study are available upon request.

**Author contributions.** KW and PIP designed the experiments; KW conducted the calculations using the X-STILT model and input data provided by DW, DJ, and TO; KW and PIP analysed the results and wrote the paper, with comments from all co-authors.
Competing interests. The authors declare no competing interests.

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References


Lauvaux, T., Díaz-Isaac, L. I., Bocquet, M., and Bousserez, N.: Diagnosing spatial error structures in CO\textsubscript{2} mole fractions and X\textsubscript{CO\textsubscript{2}} column mole fractions from atmospheric transport, Atmospheric Chemistry and Physics, 19, 12 007–12 024, 2019.


Lopez, M., Schmidt, M., Delmotte, M., Colomb, A., Gros, V., Janssen, C., Lehman, S., Mondelain, D., Perrussel, O., Ramonet, M., et al.: CO, NO\textsubscript{x} and \textsuperscript{13}CO\textsubscript{2} as tracers for fossil fuel CO\textsubscript{2}: results from a pilot study in Paris during winter 2010, Atmospheric Chemistry and Physics, 13, 7343–7358, 2013.


Oda, T. and Maksyutov, S.: A very high-resolution (1 km × 1 km) global fossil fuel CO₂ emission inventory derived using a point source database and satellite observations of nighttime lights, Atmospheric Chemistry and Physics, 11, 543, 2011.


Figure 1. Flow chart of the Observing System Simulation Experiment (OSSE) for urban CO$_2$ inversion.
Figure 2. MicroCarb city-mode sampling patterns for the two-sweep and three-sweep modes in Paris (a and b) and London (c and d). Samplings are marked by quality flag (QF) using ERA5 total cloud cover at 12:00 UTC on 15 April 2018 (halve the cloud cover from one to 0.5). QF=0 means the samplings are cloud-free and QF=1 means the samplings are cloud-contaminated. Numbers in the parentheses are the number of observations and the percentage relative to the total number of observations for the two-sweep (228) and three-sweep (342) modes. The red star marks the city centre. The satellite moves from northeast to southwest and wind comes from southwest.
Figure 3. Monthly mean fossil fuel CO$_2$ emissions in April 2018 from ODIAC (without strong point sources) in Paris (a) and London (b). Biogenic CO$_2$ fluxes at 12:00 (UTC) on 15 April 2018 simulated by the SMUrF model in Paris (c) and London (d).
Figure 4. Truth, prior state, and posterior state based on the cloud-free samplings of the two-sweep and three-sweep modes in Paris (top panels, a to d) and London (bottom panels, e to h) at 12:00 (UTC) on 15 April 2018. The values in parentheses are the total CO$_2$ emissions within the domain (exclude strong point sources) and its uncertainty.
Figure 5. Urban CO$_2$ enhancements for the two-sweep and three-sweep modes in Paris (a and b) and London (c and d).
Figure 6. Spatially-averaged flux error reduction with the change of observation uncertainty under different sampling scenarios in Paris (a) and London (b). Numbers in the parentheses are the number of observations and the percentage relative to the total number of observations for the three-sweep mode (342).
Figure 7. Posterior fossil fuel CO$_2$ fluxes (a), posterior flux noise (b), flux error reduction (c), and urban CO$_2$ enhancements (d) after including biogenic CO$_2$ fluxes in Paris for the three-sweep mode. The values in parentheses are the total emissions within the domain (exclude strong point sources) and its uncertainty.
Figure 8. Total fossil fuel CO$_2$ emissions within the domain in Paris (a and b) and London (c and d) for different sampling scenarios. Left panels do not include the biogenic CO$_2$ flux component while right panels include the biogenic fluxes.
Figure 9. Similar as Figure 8, but for the error reduction of the total city-scale emissions.
Figure A1. ERA5 total cloud cover at 12:00 (UTC) on the 5\textsuperscript{th}, 15\textsuperscript{th}, and 25\textsuperscript{th} in April, July, and December 2018 over Paris and London. The black lines are the coastline and the red stars mark the city center of Paris and London.
<table>
<thead>
<tr>
<th>Two-sweep, Three-sweep</th>
<th>5</th>
<th>15</th>
<th>25</th>
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<td><strong>Paris</strong></td>
<td></td>
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<tr>
<td>APR</td>
<td>67 (29%), 114 (33%)</td>
<td>72 (32%), 124 (36%)</td>
<td>27 (12%), 33 (10%)</td>
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<td>114 (50%), 169 (49%)</td>
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<tr>
<td>DEC</td>
<td>69 (30%), 128 (37%)</td>
<td>76 (33%), 108 (32%)</td>
<td>101 (44%), 151 (44%)</td>
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<tr>
<td><strong>London</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>APR</td>
<td>85 (37%), 151 (44%)</td>
<td>69 (30%), 119 (35%)</td>
<td>81 (36%), 130 (38%)</td>
</tr>
<tr>
<td>JUL</td>
<td>121 (53%), 159 (46%)</td>
<td>148 (65%), 226 (66%)</td>
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<tr>
<td>DEC</td>
<td>78 (34%), 116 (34%)</td>
<td>70 (31%), 111 (32%)</td>
<td>57 (25%), 59 (17%)</td>
</tr>
</tbody>
</table>

Table A1. The number of cloud-free observations and its percentage relative to the total number of observations for the two-sweep (228) and three-sweep (342) modes in Paris and London. Bold numbers mark the dates that we artificially halve the cloud cover from one to 0.5, otherwise there are no cloud-free samplings at 12 UTC on those dates.
Figure A2. Diurnal variation of seasonally-averaged eddy-covariance flux measurements (a) and net ecosystem exchange (NEE) simulated by the SMUrF model (b). Error bars are the standard errors of the seasonal means. Flux data are downloaded from the European Fluxes Database Cluster, with five sites in France (FR-Aur, FR-Hes, FR-Lam, FR-LGt, FR-Pue) and two sites in UK (UK-AMo and UK-EBu). Site information is described on the website (http://www.europe-fluxdata.eu/home/sites-list). The land types are croplands (CRO), deciduous broadleaf forests (DBF), permanent wetlands (WET), evergreen broadleaf forests (EBF), and grasslands (GRA).
Figure A3. Monthly mean ODIAC emissions (without strong point sources) and the simulated footprint of a cloud-free sampling in the two-sweep mode at 12:00 (UTC) on the 5th, 15th, and 25th in April, July, and December 2018 over Paris. The values of footprint are displayed on a logarithmic color scale.
Figure A4. Same as Figure A3, but for London.
Figure A5. Probability density of the observation uncertainty (random measurement error) in Paris (a) and London (b).
Two-sweep mode

Three-sweep mode

Figure A6. Prior flux noise (prior state minus truth), flux correction (posterior minus prior state), posterior flux noise (posterior state minus truth), and flux error reduction using the cloud-free samplings of the two-sweep (top panels, b to d) and three-sweep (bottom panels, e to g) modes in Paris (considering only anthropogenic emissions.).
Two-sweep mode

Three-sweep mode

Figure A7. Same as Figure A6, but for London.