Urban greenhouse gas emissions from the Berlin area: A case study using airborne CO$_2$ and CH$_4$ in situ observations in summer 2018

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Urban areas are recognised as a significant source of greenhouse gas emissions (GHG), such as carbon dioxide (CO$_2$) and methane (CH$_4$). The total amount of urban GHG emissions, especially for CH$_4$, however, is not well quantified. Here we report on airborne in situ measurements using a Picarro G1301-m analyser aboard the DLR Cessna Grand Caravan to study GHG emissions downwind of the German capital Berlin. In total, five aircraft-based mass balance experiments were conducted in July 2018 within the Urban Climate Under Change [UC]$^2$ project. The detection and isolation of the Berlin plume was often challenging because of comparatively small GHG signals above variable atmospheric background concentrations. However, on July 20th enhancements of up to 4 ppm CO$_2$ and 21 ppb CH$_4$ were observed over a horizontal extent of roughly 45 to 65 km downwind of Berlin. These enhanced mixing ratios are clearly distinguishable from the background and can partly be assigned to city emissions. The estimated CO$_2$ emission flux of $1.39 \pm 0.76$ t s$^{-1}$ is in agreement with current inventories, while the CH$_4$ emission flux of $5.20 \pm 1.70$ kg s$^{-1}$ is almost two times larger than the highest reported value in the inventories. We localized the source area with HYSPLIT trajectory calculations and the global/regional nested chemistry climate model MECO(n) (down to ~1 km), and investigated the contribution from sewage-treatment plants and waste deposition to CH$_4$, which are treated differently by the emission inventories. Our work highlights the importance of strong CH$_4$ sources in the vicinity of Berlin and shows, that there is limited understanding of CH$_4$ emissions from urban regions, even for major cities in highly developed countries like Germany. Furthermore, we show that a detailed knowledge of GHG inflow mixing ratios is necessary to suitably estimate emission rates for Berlin.

Keywords: Urban; Greenhouse gas; Carbon dioxide; Methane; Airborne mass balance approach; In-situ

1 Introduction

Atmospheric mixing ratios of well-known greenhouse gases (GHGs), such as carbon dioxide (CO$_2$) and methane (CH$_4$), have increased by a factor of 1.4 and 2.5, respectively, compared to pre-industrial times (Hartmann et al., 2013). These gases contribute substantially to global warming due to the increased absorption of outgoing infrared radiation. The Paris Agreement (UNFCCC, 2015) aims to keep the global temperature rise below 2°C compared to pre-industrial levels. To achieve this ambitious goal, GHG emissions have to be reduced. However, for the development of efficient mitigation strategies and for the prediction of future climate impacts, an improved understanding and quantification of the GHG budget is needed. Typically two different approaches are used to estimate emissions.

The bottom-up approach has the advantage that emissions can be calculated globally and sector-wise. Furthermore, it provides detailed information about the different emission processes, but relies on statistical activity data and emission factors. It is well known that such input data are subject to large spatial and temporal heterogeneity, as well as to simplified assumptions with large uncertainties (Nisbet and Weiss, 2010; Brandt et al., 2014). The so-called top-down approach is based on atmospheric observations (on different scales), which are used to derive emission rates either with a mass-balance approach, as done in this study, or with inverse modelling techniques. These emission rates are commonly used to independently validate total bottom-up emissions, while the source...
attrition to different emission processes usually is more challenging. Estimating emissions with bottom-up and top-down approaches can either provide insight into large discrepancies (as shown e.g. by an ensemble of seven different inverse models estimating ~30% higher European \( \text{CH}_4 \) emissions than bottom-up reported values (Bergamaschi et al., 2018)), or can confirm reported emissions (as shown e.g. for the city of Indianapolis by Turnbull et al. (2019)).

Currently urban areas cover less than 3% of the terrestrial earth’s surface (Liu et al., 2014), but accommodate more than half of the world’s population, with a persistently increasing trend (UN, 2018). This spatial concentration of human activities and their high energy consumption make cities a hotspot for GHG emissions (Kennedy et al., 2012; Marcotullio et al., 2013). With respect to \( \text{CH}_4 \), global anthropogenic emissions are relatively well-known, but the contributions of single source sectors on the national, regional and city scale are often not well quantified (Kirschke et al., 2013; Saunois et al., 2017). Total \( \text{CO}_2 \) emissions of developed countries can be estimated with high accuracy (due to the precise knowledge of national fossil fuel consumption amounts), however, the spatial and temporal distribution of emissions is less well known because of lack of appropriate data (Bréon et al., 2015). Therefore, several observational studies have focused on estimating \( \text{CH}_4 \) and \( \text{CO}_2 \) emissions from urban areas. Based on airborne measurements a major source of elevated \( \text{CH}_4 \) from cities in the United States was attributed to emissions related to natural gas and landfills (e.g. Cambaliza et al., 2015; Ren et al., 2018; Plant et al., 2019). Airborne studies on European cities are relatively sparse and to the best of our knowledge, they have been conducted in London (O’Shea et al., 2014; Pitt et al., 2019) and Rome (Gioli et al., 2014). While airborne derived \( \text{CO}_2 \) emissions from Rome were found to agree with inventorial data (Gioli et al., 2014), \( \text{CO}_2 \) and \( \text{CH}_4 \) emissions from London need to be scaled by a factor of 1.57 and 0.7, respectively, in order to agree with the airborne derived fluxes from Pitt et al. (2019). In this estimate peri-urban emissions from the greater London area were taken into account, which downscales their previous estimate based on the conventional mass balance approach (factors of 3.08 for \( \text{CO}_2 \) and 1.22 for \( \text{CH}_4 \), given in the same study). Ground-based observations in London (Helfter et al., 2016), Paris (Bréon et al., 2015), Cracow (Kuc et al., 2003; Zimnoch et al., 2019) and Florence (Gioli et al., 2012) provide additional information on urban GHG emissions, e.g. long-term \( \text{CO}_2 \) observations in Florence indicate 19% higher emissions than reported by inventories (Gioli et al., 2012). Atmospheric monitoring sites in Paris, together with an atmospheric inversion approach, point towards an overestimation of urban emissions by the local inventory (Bréon et al., 2015).

Here we report on aircraft-based in situ measurements of atmospheric \( \text{CH}_4 \) and \( \text{CO}_2 \) mixing ratios in order to estimate GHG emissions of Berlin. Five research flights up- and downwind of Berlin were conducted in July 2018 with the DLR Cessna aircraft in the framework of the Urban Climate Under Change project [UC]² (Scherer et al., 2019b). Berlin itself is an interesting target for studying urban GHG emissions, as it is the largest German city in terms of area and second largest city in terms of population density (Statistisches Bundesamt, 2018). \( \text{CO}_2 \) emissions for the greater Berlin area (~33 Mt a⁻¹) are expected to be roughly similar to emissions from the well-studied London area (~32 Mt a⁻¹), while \( \text{CH}_4 \) emissions are ~50% lower (Berlin ~28 kt a⁻¹, London ~53 kt a⁻¹) when comparing estimates from Pitt et al. (2019) with the CAMS GHG inventory for 2015 (Kuenen et al., 2014). The relatively isolated location of Berlin and its flat topography makes the city an ideal target for the aircraft-based mass balance approach. To the best of our knowledge, the only published study on experimentally determined GHG fluxes from Berlin is by Hase et al. (2015) who estimated the \( \text{CO}_2 \) source strength of Berlin with ground-based Fourier-Transform infrared spectrometers. The city of Berlin has also been in the focus of several studies on the potential use of satellite measurements for estimating \( \text{CO}_2 \) emissions. They show, that even for future and highly sophisticated satellite systems, the identification and quantification of the Berlin city plume is extremely challenging (Pillai et al., 2016; Kuhlmann et al., 2019b), so that dedicated measurement campaigns will be an invaluable tool for validating both top-down and bottom-up inventories.

This paper is structured as follows: Section 2 introduces the aircraft instrumentation, the mass balance approach, the model setups, and compares three different emission inventories for the Berlin area. A mission overview is presented in Sect. 3.1. In Sect. 3.2 we assess the \( \text{CO}_2 \) and \( \text{CH}_4 \) emission rate for a selected flight on July 20th using the mass balance approach. We consider the history of the encountered air masses and discuss our estimate with respect to current GHG inventories. In Sect. 3.3 we compare our observations with simulated GHG mixing ratios from the global/regional nested chemistry climate model MECO(n) (MESSy-field ECHAM and COSMO models nested n times) to localize the source area and to investigate the contribution from different source sectors. Section 4 finally summarises the findings of the present study.

2 Material and methods

2.1 Airborne instrumentation

Aircraft measurements were performed with the DLR Cessna 208B Grand Caravan within the scope of the Urban Climate Under Change program [UC]² (Scherer et al., 2019b). While the project’s main objective is to develop, apply and validate an urban climate model at high spatial resolution (<10 m) for three German cities, our airborne observational wind and temperature data of Berlin were used for the validation subproject (Scherer et al., 2019a). One of our other objectives was to analyse the urban GHG emissions measured from the airborne platform. The aircraft was equipped with a series of instruments to measure meteorological parameters and trace gas species. The meteorological sensor package (METPOD) was mounted at the left underwing pod and included a boom with a five-hole probe at its tip for basic data like temperature (standard deviation \( \sigma = 0.15 \) K), pressure (\( \sigma = 0.25 \) hPa), humidity, wind direction (\( \sigma = 2^\circ \)) and wind speed (\( \sigma = 0.3 \) m s⁻¹); for details see Mallaun et al. (2015). Wind measurements during strong turns (i.e. more than 10° roll
angle) were discarded for further analysis as deviations of up to 1 m s\(^{-1}\) are possible (personal communication with C. Mallaun, 2019). The AEROControl IGI system, which is a combination of differential GPS (Global Positioning System) and IRS (Inertial Reference System), provided aircraft position data. The following trace gas analysers were used with their backward facing inlets at the right underwing pod: (1) a Picarro cavity ring-down spectroscopy analyser (G1301-m) for CO\(_2\), CH\(_4\) and water vapour (H\(_2\)O); (2) a modified Aerodyne Quantum and inter-band Cascade Laser Spectrometer (QCLS) for CO\(_2\), CH\(_4\), ethane (C\(_2\)H\(_6\)), carbon monoxide (CO) and nitrous oxide (N\(_2\)O); for details see Kostinek et al. (2019); (3) a Cavity Attenuated Phase Shift (CAPS) Sensor for nitrogen dioxide (NO\(_2\)) and a 2B ozone (O\(_3\)) monitor.

For this study, we used CH\(_4\) and CO\(_2\) data at 0.5 Hz from the Picarro analyser (for a general description see Chen et al., 2010). During the campaign, the analyser was frequently calibrated on ground with four multi gas cylinders from Air Liquide. Before and after the deployment these secondary standards were calibrated against two NOAA (National Oceanic and Atmospheric Administration) multi gas primary standards (cylinder #CB11542 and #CB11361), which are traceable to the WMO-X2007 and WMO-X2004A scale (Dlugokencky, 2005; Zhao and Tans, 2006). The overall measurement uncertainty is determined to be better than 0.2 µmol mol\(^{-1}\) for CO\(_2\) (hereafter referred to as ppm) and 1.1 nmol mol\(^{-1}\) for CH\(_4\) (hereafter referred to as ppb) based on the summation of the known sources of uncertainty in quadrature, which are listed in Table 1. Details on the error estimate can be found in Sect. S1 in the supplement.

### 2.2 Aircraft-based mass balance approach

To infer CH\(_4\) and CO\(_2\) emission rates, the aircraft-based mass balance approach is a well-established method (e.g. Mays et al., 2009; Karion et al., 2013; Heimbürger et al., 2017; Ren et al., 2018). A mass balance flight pattern typically includes one or multiple transects, that are in the ideal case aligned perpendicular to the wind direction and at an appropriate downwind distance from the source. Dependent on the study, they may also include an upwind transect to determine the background mixing ratios. The upwind transect usually is centred at the middle of the planetary boundary layer (PBL) and is used both to identify possible emission inflows (already being transported into the target region from a distant source) and to identify the natural atmospheric variability. The downwind transects, stacked at several altitudes within the PBL, capture the urban outflow downstream of the source. Vertical profiles, extending into the free troposphere, are used to determine the PBL depth. Flights are usually conducted in the afternoon, when the PBL is fully developed and its height is less variable during the time of the research experiment. Further assumptions are constant emission fluxes, steady wind speed and wind direction several hours before and during the flight experiment itself. The flux or mass flow rate [g s\(^{-1}\)] through a normal plane downwind of a source is then derived using Eq. (1):

\[
f = \int_{0}^{PBL} \int_{z}^{z_{top}} \left[ (c_{ij} - \bar{c}_{ij}) \right] \frac{P_{ij}}{T_{ij}} - R M \ u_{ij} \ dx \ dz,
\]

The background mixing ratio \(c_{ij}\) is subtracted from the observed mixing ratio \(c\) at location \(x_{ij}\) and \(z_{ij}\) and is converted to molar density using the ideal gas law with observed pressure \(p\) and temperature \(T\), and the ideal gas constant \(R\). Combined with the molar mass \(M\) and the perpendicular component \(u\) of the horizontal wind, a mass flow per unit area is obtained. By integrating along the horizontal boundaries of the plume (\(a\) to \(a\)) and in the vertical from ground to the top of the PBL, the mass flux through the plane is calculated. Uncertainties in the mass flux arise mainly from varying wind speed, the choice of background determination (e.g. using the mean or the lowest detected mixing ratios from an upwind transect or from edges of the downwind transects (Cambaliza et al., 2014; Peischl et al., 2015)), varying PBL depth, and limited observational data between the lowest altitude transect and the surface (Cambaliza et al., 2014).

### 2.3 Model setups

We use two different models to confine the source area and source origin related to our measurements. With the backward model HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory) we examine the source area of the encountered air masses. With the global/regional nested chemistry climate model MECO\(n\) we simulate the transport of emitted tracers based on different emission inventories to compare simulated GHG enhancements with airborne observations.

#### 2.3.1 Lagrangian dispersion model HYSPLIT

A general description of the HYSPLIT model can be found e.g. in Stein et al. (2015). Here we use meteorological forecast data from the European Centre for Medium-Range Weather Forecasts (ECWMF) because of their high spatial and temporal resolution. Gridded fields are provided hourly with a horizontal resolution of 0.08\(^\circ\) in latitude and 0.125\(^\circ\) in longitude (\(\sim 9\) km × 9 km). The 137 model levels allow a vertical resolution of \(\sim 20\) m near the ground, increasing to about 200 m at 3 km altitude.
We use backward trajectory calculations to reconstruct the pathway of the sampled air masses. Trajectories were started every 10 s from the flight track, while every 15 minutes along the trajectories information about latitude, longitude, height of the trajectory and depth of the model PBL are provided. These air histories can be used to define a so-called back trajectory (BT) footprint, which refers here to a more general definition of an area covered by the air mass within its residence time in the PBL. To do so, we first extract the BT points which were inside the PBL, and then determine their percentage residing in a defined grid box area between the downwind and upwind transects.

2.3.2 On-line coupled atmospheric chemistry model MECO(n) The MECO(n) model couples on-line the global chemistry-climate model EMAC (ECHAM5/MESy Atmospheric Chemistry) with the regional chemistry-climate model COSMO-CLM/MESy in order to achieve a regional refinement (details can be found in Hofmann et al., 2012; Kerkweg and Jöckel, 2012a and 2012b; Mertens et al., 2016). COSMO-CLM (Rockel et al., 2008) is based on the COSMO (Consortium for Small scale Modelling) model and jointly further developed by the CLM-Community. For our campaign, a setup of three simultaneously running COSMO-CLM/MESy instances nested into each other allows a fine horizontal resolution of 0.01° (~1 km × 1 km) over Berlin with 50 vertical model levels and a time step length of 20 s. In the global EMAC model (T42L90MA and 720 s) the prognostic variables temperature, divergence, vorticity and the logarithm of the surface pressure are nudged by Newtonian relaxation towards ECMWF operational analysis data. The COSMO-CLM/MESy nests (i.e. over Europe at 0.44° and 240 s, over central Europe at 0.0625° and 60 s, and over NE Germany at 0.01° and 20 s) are directly driven by lateral and top boundary conditions from EMAC or from the corresponding coarser resolved nesting instance at each of the model time steps. The height of the midpoint of the uppermost grid box is approx. 80 km for EMAC and 22 km for COSMO-CLM/MESy, respectively. A direct nudging of the COSMO-CLM/MESy nests is not performed. Therefore, they can develop their own dynamics to a certain degree.

During the campaign, simulated wind fields and GHG mixing ratios were available from forecast simulations, and after the campaign from one corresponding analysis simulation. Mixing ratios of various GHG tracers have been simulated, where each individual tracer is subject to a specific emission source (or a set of emission sources) from different (individual or combined) inventories (see Table 2). Details about the inventories mentioned below are discussed in Sect. 2.4.

For the “city” tracer (c-CO₂, c-CH₄; see Table 2) we used an emission inventory provided by the city of Berlin (BERLIN inventory), that was gridded to a spatial resolution of 1 km × 1 km. This inventory provides detailed information about emissions from different sectors in Berlin as point, line and area sources and includes both air pollutants and GHG (AVISO GmbH and IE Leipzig, 2016). To account for surrounding emissions, we included strong point sources outside of Berlin, which were reported to the European Pollutant Release and Transfer Register (E-PRTR). For CO₂, eight facilities with combined emissions of 5.7 Mt CO₂ in 2016 were added (which is roughly 1/4⁰ to 1/3⁰ of total Berlin emissions), for CH₄ this applies to two sources: landfill Schoeneicher Plan (south of Berlin) with 1.37 kt CH₄ in 2016 and landfill Neuenhagen (NE of Berlin) with 0.13 kt CH₄ in 2015. These simulated “city” tracers serve as baseline for the later analysis.

We further derived emissions for the “waste water” (w-CH₄) and “landfill” (l-CH₄) tracer according to Sect. 2.4.2, ranging from 1 to 7 kt CH₄ a⁻¹ and from 0.1 to 32 kt CH₄ a⁻¹, respectively. To account not only for point sources outside of Berlin, but also the regional background, we merged the BERLIN inventory into the European CAMS-REG inventory (r-CO₂, r-CH₄). Since total CH₄ emissions for both inventories within Berlin differ by a factor of ~4.5 (see Table 3) it was necessary to scale the BERLIN CH₄ emissions with this factor to ensure that no artificial gradient in the simulated CH₄ mixing ratio is introduced at the boundary between the two merged inventories. For CO₂ no scaling was applied, however, grid boxes at the outermost boundary of BERLIN with very low emissions were removed.

During the model simulation the vertical profiles of all tracers (and several meteorological quantities) have been on-line sampled along the flight tracks (submodel S4D, described by Jöckel et al., 2010). This high-frequency output (at each model time step) is used for comparison.

| Table 2: Overview of the different tracers simulated with MECO(n) and the corresponding emission sources. DOI: https://doi.org/10.1525/elementa.411.t2 |

| Tracer                  | Inventory input                                                                 |
|------------------------|----------------------------------------------------------------------------------|
| c-CO₂ (“city”)         | BERLIN + E-PRTR                                                                 |
| c-CH₄ (“city”)         | BERLIN (~x4.5) + E-PRTR                                                         |
| w-CH₄ (“waste water”)  | Status report of MLUL (2017) combined with EDGAR v4.3.2 waste water emissions from 2012 |
| l-CH₄ (“landfills”)    | CORRECTIV (2016) as input for LandGEM                                           |
| t-CH₄ (“total”)        | BERLIN × 4.5 + E-PRTR + w-CH₄ + l-CH₄                                          |
| r-CO₂ (“regional”)     | BERLIN merged into CAMS-REG                                                       |
| r-CH₄ (“regional”)     | BERLIN (~x4.5) merged into CAMS-REG                                              |

* For a detailed description of the inventories see Sect. 2.4, Figure 1 and Table 3.
Table 3: CO$_2$ and CH$_4$ annual emission fluxes (integrated within the Berlin city boundary in the upper two panels; corrected according to the footprint in the two lower panels) from the global EDGAR, European CAMS-REG and local BERLIN inventory. DOI: https://doi.org/10.1525/elementa.411.t3

| Emission sector                          | EDGAR 2015 | CAMS-REG 2015 | BERLIN 2012 | EDGAR 2015 | CAMS-REG 2015 | BERLIN 2012 |
|------------------------------------------|------------|---------------|-------------|------------|---------------|-------------|
| Emission sector                          | CO$_2$ [Mt a$^{-1}$] | CO$_2$ [%]    |             |            |               |             |
| Total                                    | 24.2       | 22.6          | 16.8        | 100        | 100           | 100         |
| (ff + bf)                                | (22.5 + 1.7) | (15.4 + 7.1)  |             |            |               |             |
| Fuel combustion for the generation of electricity and heat | 19         | 17.9          | 13.7$^b$    | 79$^e$     | 79$^e$        | 82$^e$      |
|                                           | (17.5 + 1.5)$^b$ | (12.5 + 5.4)$^b$ |             |            |               |             |
| Road transport                           | 2.4        | 2.5           | 2.7$^f$     | 10         | 11            | 16$^c$      |
|                                           | (2.3 + 0.1) | (2.4 + 0.1)   |             |            |               |             |
| Others                                   | 2.8        | 2.2           | 0.4         | 12         | 10            | 2           |
|                                           | (2.7 + 0.1) | (0.5 + 1.6)   |             |            |               |             |
| Emission sector                          | CH$_4$ [kt a$^{-1}$] | CH$_4$ [%]    |             |            |               |             |
| Total                                    | 25.7       | 15.4          | 3.4         | 100        | 100           | 100         |
| Waste                                    | 20.3$^d$   | 0.1           | 0.1         | 79$^d$     | 1             | 3           |
| Fuel combustion for the generation of electricity and heat | 2$^b$     | 5.6$^c$       | 0.8$^c$     | 8$^b$      | 36$^c$        | 24$^c$      |
| Fuel exploitation                        | 2.6        | 5.8           | 2.3         | 10         | 38            | 68          |
| Others                                   | 0.8        | 3.9           | 0.2         | 3          | 25            | 5           |
| Footprint corrected total$^b$             | CO$_2$ [Mt a$^{-1}$] | n.a.           |             |            |               |             |
|                                          | 41.9       | 30.6          | n.a.        |            |               |             |
| Footprint corrected total$^b$             | CH$_4$ [kt a$^{-1}$] | n.a.           |             |            |               |             |
|                                          | 94.3       | 21.9          | n.a.        |            |               |             |

$^a$The first column lists the emission fluxes for different emission sectors in Mt or kt per year [a$^{-1}$], the second column shows the relative share in percent. CO$_2$ is split into contributions from fossil fuel and biofuel when possible and indicated by parentheses (ff + bf).

$^b$–$^f$The EDGAR sector definitions follow the IPCC 1996 code, while CAMS-REG and BERLIN are sectored according to SNAP (Selected Nomenclature for Air Pollution) categories.

$^b$Sum of sector energy for buildings, combustion for manufacturing, and power industry.

$^c$Sum of sector SNAP 1 (combustion in energy and transformation industries), SNAP 2 (non-industrial combustion plants) and SNAP 34 (emissions which cannot be clearly attributed to SNAP 3 (combustion in manufacturing industry) or SNAP 4 (production processes)).

$^d$Sum of sector solid waste incineration, solid waste landfills, and waste water treatment.

$^e$Sum of SNAP 1, SNAP 2, SNAP 3, SNAP 4, SNAP 34, and extra category of heating.

$^f$Sum of minor roads and major roads. Emissions for minor roads were estimated based on NO$_x$ emissions from the same inventory using the constant emission ratio for major roads (CO$_2$/NO$_x$ factor of 355.5 ± 45.4 kg CO$_2$/kg NO$_x$; see Kuhlmann et al., 2019a). Emissions for major roads were compiled from earlier inventories. Therefore, road transport emissions might slightly differ from Berlins official numbers.

$^g$The footprint corrected total emissions for CO$_2$ and CH$_4$ are given for the EDGAR and CAMS-REG inventory (for details see Figure 7 and corresponding text).

with the observations. We refrain from computing simulations with the EDGAR emissions, as the inventory is a factor ~10 coarser than the MECO(n) model resolution (~1 km × 1 km). Hence, using such a coarse emission inventory with a much finer model resolution will not add useful information.

2.4 Greenhouse gas emission inventories

2.4.1 Inventorial emissions

In this section we compare three available GHG emission inventories from global to local scales for the city of Berlin and its surrounding area: (a) the global inventory EDGAR v5.0 for the year 2015 with a resolution of 0.1° × 0.1° (Crippa et al., 2019); (b) the European inventory CAMS-REG v1.1 for 2015 with a resolution of 0.0625° × 0.125° (Kuenen et al., 2014); and (c) a local Berlin inventory, referred to as BERLIN, for 2012 (Berliner Emissionskataster v1.0, AVISO GmbH and IE Leipzig, 2016), provided by the Senatsverwaltung für Stadtentwicklung und Umwelt Berlin (A. Kerschbaumer). The latter inventory consists of individual area, point and line sources which for this study have been projected to a 0.01° × 0.01° longitude-latitude grid (~1 km resolution). Parts of this inventory are described by Brunner et al. (2019) and by Kuhlmann et al. (2019a).
Figure 1 shows the CO$_2$ (upper row) and CH$_4$ (lower row) emission maps for the EDGAR (panels a), CAMS-REG (panels b) and BERLIN (panels c) inventories, respectively, along with the city boundary of Berlin. Table 3 lists the estimated CO$_2$ and CH$_4$ emission totals within the city boundary together with the main contributing sectors in detail. The first column presents for each inventory the emissions in Mt CO$_2$ and kt CH$_4$ per year [a$^{-1}$], while the second column lists the relative share of the sectors in percent. CO$_2$ is split into contributions from fossil fuel (ff) and biofuel (bf) for the EDGAR and CAMS-REG inventory (see parentheses in Table 3), while for the BERLIN inventory this information is not available. Total CO$_2$ emissions from Berlin range from 16.8 to 24.2 Mt a$^{-1}$ between different inventories and thus agree within a factor of ~1.4. The CO$_2$ emissions (note that all CO$_2$ maps are plotted in the same colour range in Figure 1) are rather confined to the inner city, since they are closely coupled to fossil fuel consumption related to energy production and transport (Helfter et al., 2011), and thus to areas with high population density and locations of power plants. This is also reflected in the emission sectors, as the majority of emissions (79 to 82%) within the three inventories are related to fuel combustion for the generation of electricity and heat. The emission strength itself, however, is different, while the inventories agree in the numbers for the road transport sector. According to EDGAR, the relative CO$_2$ contribution from the road traffic sector for Berlin (~10%) is slightly lower than for greater Paris (~16%) or London (~22%). This is most likely explainable by the high dominance of point source emissions from power plants located within the city of Berlin. Roughly 94% of point source CO$_2$ emissions reported to E-PRTR in 2016 was released from power plants. In contrast, these emissions only account for zero and ~21% for the inner and outer city of London, respectively, and for ~24% for Paris. Strong CO$_2$ sources outside the city are located at different spots when comparing EDGAR and CAMS-REG.

With respect to CH$_4$, larger discrepancies are present between the inventories; both in the partitioning into different sectors and in the magnitude of emissions (factor ~8). Therefore, each CH$_4$ panel has its own colour scale. In EDGAR, strongest CH$_4$ sources are located outside the city and mainly consist of solid waste landfills and waste water handling. In contrast, emissions reported in CAMS-REG are mainly due to fuel combustion for the generation of electricity and heat and fuel exploitation, with only little contribution of waste treatment and disposal. Hence, the spatial distribution of CH$_4$ emissions is similar to the CAMS-REG CO$_2$ emission distribution. Interestingly, BERLIN also reports almost no emissions from the waste sector, the main contribution of 2.3 kt CH$_4$ a$^{-1}$ results from leakages during natural gas transport in pipelines and during storage in gasometers. According to the report for the Berliner Emissionskataster (AVISO GmbH and IE Leipzig,
waste treatment in municipal sewage-treatment facilities occurs under aerobic conditions (where no CH$_4$ is produced) and composting facilities of organic waste (where CH$_4$ can be produced) are located outside of Berlin and are thus neglected in the inventory. In contrast, EDGAR uses for solid waste disposal and waste water emissions a proxy mainly based on the distribution of urban and rural population (Janssens-Maenhout et al., 2012).

For Table 3 we estimated the emissions in EDGAR and CAMS-REG based on the percentage of the grid cell area being within the city boundary based on OpenStreetMap (2018), which uses an administrative boundary dividing Germany into 16 federal states, including Berlin. The population is less dense in the surrounding state Brandenburg (with 84 inhabitants per km$^2$) compared to Berlin (4055 inhabitants per km$^2$; Strukturatlas Land Brandenburg, 2017). For details see Figure S9 in the supplement. A sensitivity test, where only the grid cells in EDGAR (CAMS-REG) were taken, which contain at least 75% of the city area, reveal that total emission estimates are lower, both for CH$_4$ with ~25% (20%) and for CO$_2$ with ~7% (4%). Nevertheless, the large discrepancies between the CH$_4$ inventories cannot be explained by this, as total CH$_4$ emissions still vary by a factor of ~6.

Member states of the EU further have to report emissions from single industrial facilities to the European Pollutant and Transfer Register (E-PRTR), if their release to air exceeds a threshold of 0.1 Mt a$^{-1}$ for CO$_2$ or 0.1 kt a$^{-1}$ for CH$_4$ (http://prtr.ec.europa.eu/#/pollutantinfo, last access: 18.09.2019). In Figure 1b these point sources are superimposed on the CAMS-REG inventory to point out that locations of hot spots (and their source strengths) do not always match between EDGAR and CAMS-REG, and thus to highlight the importance of an experimental verification of GHG emissions around Berlin.

2.4.2 Additional CH$_4$ emissions from waste water treatment plants and illegal landfills

As described above, CH$_4$ emissions show large discrepancies between the three different inventories, especially with respect to the waste sector. Therefore, we collected additional information on locations of sewage-treatment plants and on unofficial waste deposits, which reflect strong point sources. We derive corresponding CH$_4$ emissions and implement them as separate tracers in the MECO(n) model simulation (see Table 2) to determine their source strengths individually, and to compare them to the other emissions.

Figure 2a shows sewage-treatment facilities superimposed on the EDGAR v5.0 waste water CH$_4$ emission map from 2015. We took information on sewage-treatment plants from a status report of the Ministerium fuer Laendliche Entwicklung, Umwelt und Landwirtschaft (MLUL, 2017) and determined their exact position with Google Earth. Since the report only covers the state of Brandenburg, we added the one sewage-treatment plant within the Berlin city boundary (Ruhleben, Berliner Wasserbetriebe). As obvious in Figure 2a, larger plants surrounding Berlin (they are sized according to their population equivalent) correlate very well with pixels of stronger EDGAR CH$_4$ emissions. Therefore we transferred their emissions and assigned the remaining smaller facilities with an arbitrarily chosen CH$_4$ rate of 1 kt a$^{-1}$ to ensure a visible signal in the model output (note that although

Figure 2: CH$_4$ emissions from sewage-treatment plants and illegal landfills. (a) Sewage-treatment plants (MLUL, 2017) are sized according to their population equivalent and superimposed on the EDGAR v5.0 waste water CH$_4$ emission map. (b) Illegal waste deposits (CORRECTIV, 2016) are sized according to the amount of deposited waste (ranging from 0.06 to 416 kt of waste). Official landfills listed in E-PRTR are not sized. DOI: https://doi.org/10.1525/elementa.411.f2
Figure 2a shows the EDGAR v5.0 waste water map of 2015, the values from 2012 (v4.3.2) were assigned to the Facilities as the model run was not repeated after updating to the most recent inventory v5.0). Landfills are another well-known CH$_4$ emission source, which we consider separately. In Figure 2b official landfills (reported by E-PRTR) are mapped together with illegal waste dumps, which are based on information from the non-profit organization CORRECTIV. Their article reveals that more than 100 illegal landfills in Brandenburg contain at least three million tons of waste; however, the legal disposal would cost more than 320 million Euros (CORRECTIV, 2016). We calculated CH$_4$ emissions using the Landfill Gas Emission Model (LandGEM, 2005, v3.02), provided by the United States Environmental Protection Agency (EPA). Input parameters are the amount of waste and the year of the first deposit as waste is continuously decomposing. If no information on the year was available, we used the most frequently reported (i.e. year 2000). Furthermore, we assumed a constant decay rate ($k = 0.038$) and a degradable organic carbon content ($DOC = 0.2028$) based on moderate climate, and assuming all waste material being from municipal solid waste landfills (EPA, 2010). Estimated CH$_4$ emissions range from 0.002 to 16 kt a$^{-1}$. We do not claim that calculated emissions from sewage treatment and landfills represent true values, however, they serve as input for model sensitivity tests to study the importance of these sectors (see Sect. 3.3.1).

3 Results and discussion

3.1 Flight strategy and mission overview

As discussed in Sect. 2.2 we optimised the mission flight strategy to study GHG fluxes of Berlin using the mass balance approach. Our flight planning mainly relied on the MECO(n) plume forecast of the GHG city tracers c-CO$_2$ and c-CH$_4$ (see Table 2). With horizontal latitude-longitude projections as well as vertical height profiles, we determined the location of the urban pollution plume and the distance at which the plume is supposed to be vertically well mixed. Furthermore, we used the meteogram for Berlin-Tempelhof provided by the German Weather Service (Deutscher Wetterdienst, DWD) on the basis of their ICON (icosahedral Nonhydrostatic) forecast model, as well as animated forecasts based on ECMWF and GFS (Global Forecast System) from windy.com to verify the wind forecasts from MECO(n). Flight plans had to be submitted to air traffic control (ATC) on the day before the flight and were approved (either as submitted or slightly modified) shortly before the flight. However, discrepancies between the previous day forecast and the actual wind conditions, together with the heavy air traffic situation in and around Berlin with its two major airports (Berlin-Tegel and Berlin Schoenefeld), generally hampered the realization of the flight patterns, which were designed for optimized sampling of the in- and outflow.

Five research flights were conducted with the DLR Cessna aircraft between 16th and 26th of July 2018 from the airfield Schoenhagen, located ~40 km southwest of Berlin (see Figure 3). The typical mass balance pattern was roughly perpendicular to the prevailing wind direction, which was mainly from NW to NE. Flights were conducted between ~11:30 and ~15:00 UTC (+2 hours for local time) to ensure a fully developed PBL. To characterise the PBL during each flight, we obtained a vertical profile (upward spiral, typically between 0.3 and 3 km) at the Tempelhofer Feld (TF, south of the city centre). The horizontal extent of Berlin is approximately 35 to 45 km. In order to assure complete sampling of the urban outflow, the length of our
stacked downwind transects (so-called “wall”) was planned to cover more than 80 km distance at a typical downwind distance of roughly 30 to 40 km. Table 4 presents an overview of all research flights including date, flight time, average wind direction, average wind speed, PBL depth (estimated from the meteorological measurements, see Sect. S2) and the general weather situation. Time series for CO₂, CH₄ and flight altitude of each flight can be found in Figure S1, as well as altitude profiles of virtual potential temperature, static temperature, and relative humidity to estimate the PBL depth in Figure S2.

In the following section we apply and discuss the mass balance approach for the case study on July 20th as strong CO₂ and CH₄ signals from urban emissions were distinguishable from the atmospheric background variability together with ideal meteorological conditions prevalent on that day. For the flight on July 24th the limitations of the mass balance approach are discussed in Sect. 3.3.2. We further excluded the remaining three performed flights (on 18th, 23rd and 25th of July) from the mass balance analysis due to the following reasons: Based on Table 4 and Figure S2 it is apparent that the PBL on July 18th and 25th is not clearly separated from the free troposphere and not well-mixed. In addition, the wind speed on July 18th is rather high with more than 8 m s⁻¹ on average. On July 23rd meteorological conditions appeared to be favourable, however, the urban plume is most likely missed, mainly because of the large difference between forecasted and real wind direction (~40°) and the inflexibility to adapt the submitted flight pattern ad hoc due to air traffic control restrictions.

### 3.2 Mass balance flight on July 20th

#### 3.2.1 Flight pattern and airborne observations

On July 20th the research flight was conducted between ~11:30 and ~14:00 UTC. The flight pattern is shown in Figure 4a, the time series of the measured mixing ratios Table 4: Overview of five performed research flights. DOI: https://doi.org/10.1525/elementa.411.t4

| Day of July 2018 (weekday) | Take-off and landing [UTC] | Avg. a wind direction [deg] | Avg. a wind speed [m s⁻¹] | PBL depth [m] | Weather situation |
|---------------------------|---------------------------|----------------------------|-------------------------|---------------|-------------------|
| 18th (Wed.)               | 11:35–14:13              | 304 ± 14                   | 8.5 ± 1.8               | 1079 ± 26     | West of occlusion along border Germany/Poland |
| 20th (Fri.)               | 11:30–14:07              | 299 ± 27                   | 4.8 ± 1.8               | 2737 ± 2     | Ridge of high pressure system, low level clouds |
| 23rd (Mon.)               | 11:37–14:08              | 296 ± 16                   | 5.6 ± 1.4               | 1419 ± 4     | Clear sky         |
| 24th (Tues.)              | 12:12–14:59              | 25 ± 34                    | 3.3 ± 1.3               | 2046 ± 1     | Eastern edge of high pressure system, low level clouds |
| 25th (Wed.)               | 11:28–14:10              | 27 ± 28                    | 3.3 ± 1.1               | 1969 ± 56    | South-eastern edge of high pressure system, low level clouds |

*For the average wind speed and direction the average of the whole flight is considered, excluding winds above the PBL when different.

*PBL depth is estimated from meteorological measurements, see Sect. S2.

*PBL has two layers.

![Figure 4: Flight pattern and time series of July 20th. (a) The downwind wall is colour-coded according to the flown horizontal distance (0 km corresponds to the starting point ‘A’ at 52.41°N and 13.95°E). Backward trajectories (~12 h, in grey) are started from the downwind “wall” at 13 UTC, while every 5th trajectory within the PBL is colour-coded according to its relative time since the calculation start. (b) The grey sections of the altitude curve correspond to the inflow section of the flight; other colours same as in panel (a). DOI: https://doi.org/10.1525/elementa.411.f4](https://doi.org/10.1525/elementa.411.f4)
of CO₂ and CH₄, as well as flight altitude are presented in Figure 4b. Forecasts indicated north-northwesterly winds and a boundary layer height of ~2 km. However, during the flight itself, the wind came from the north-west with 4.8 m s⁻¹ on average. We first headed towards the north (indicated by the black arrow in Figure 4a) to perform the upwind leg roughly at the middle of the PBL (~780 m). This upwind section is shaded grey on the altitude curve visible in Figure 4b. A detailed vertical profile was obtained at the Tempelhofer Feld from ~350 m up to ~3.2 km (with an ascend rate of ~3 m s⁻¹), hereby detecting the lowest CH₄ mixing ratios during the entire flight (note the split CH₄ axis; see also vertical profile in Figure 5c). The in-flight derived PBL height from the vertical profile was then used to adapt the heights of the following downwind transects to roughly 500 m, 1 km and 1.6 km. This so-called downwind “wall” is colour-coded in both panels according to the horizontal flown distance. Sampling of the urban plume started at point ‘A’ (marked with 0 km distance) towards the south-west (indicated with negative distances). Since in-flight observations of atmospheric GHG mixing ratios did not reach background concentration levels as observed at the starting point, we extended the downwind transects in-flight. Due to air space restrictions this was only possible by flying strictly towards the north (indicated with positive distances). Figure 4a also includes HYSPLIT backward trajectories for the last 12 hours, which were started from the entire downwind “wall” at 13 UTC. Colour-coded is (for simplicity) only every 5th trajectory within the PBL to its relative time since its calculation start. The grey colours underneath depict, in addition, also trajectories being outside the PBL. The trajectories reflect the prevalent wind direction and wind speed quite well (ECMWF forecast data and airborne wind measurements reveal Pearson correlation coefficients between R = 0.95 and R = 0.98; for details see Figure S4 and S5) and indicate a steady wind flow within the PBL for at least 6 hours prior to our measurement. Figure 5a presents the measured CO₂ (upper panel) and CH₄ (lower panel) mixing ratios along the horizontal distance at the three different altitudes of our “wall”. The encountered CO₂ plume is detected approximately between ~30 and 15 km, thus in the more northern part of our flight track, whereas the centre of the CH₄ plume extends more to the south-west (roughly between ~50 and 15 km). A similar CO₂ distribution was observed at all altitudes with maximum enhancements of 4 ppm, indicating a generally well-mixed plume. Measured CH₄ mixing ratios in the two lower flight levels are also coherent with enhancements of 21 ppb, but the highest flight level shows a different pattern with a dip in CH₄ between ~20 and 0 km. Differences in the vertical distribution of CO₂ and CH₄ can be explained by a combination of three effects: a) differences in the
spatial distribution of the emissions sources (see Sect. 2.4), b) differences in the emission height, and c) differences in the buoyancy of the gases at their release point. CO$_2$ is largely emitted via stacks, already occurring at a certain elevated height above ground level, which may influence the distribution due to the altitude dependent wind speed and direction (Brunner et al., 2019). Furthermore, CO$_2$ emissions are closely coupled to fossil fuel consumption and thus to higher temperatures during the release, which enhance the buoyancy of the air parcels and the mixing in the vertical. In contrast, CH$_4$ emissions are usually not emitted during exothermic processes, therefore the vertical mixing might be less pronounced. In addition to these broad enhancements, some small-scale enhancements (~3 to 5 km wide peaks) are observed at ~7 km distance (in CO$_2$) and further north at ~11 km (in CH$_4$).

Figure 5b shows the vertical profile obtained at the Tempelhofer Feld (from roughly 12:00 to 12:30 UTC) of virtual potential temperature ($\theta_v$), static temperature (TS) and relative humidity (RH). The PBL depth was determined from that profile to 27.37 ± 2 m (dashed black line), where the uncertainty of 2 m reflects the average of the three different approaches used herein (see Sect. S2 in the supplement). The PBL on July 20th was well-mixed and efficiently capped as seen in the strong gradients in $\theta_v$, TS and RH between the PBL and the free troposphere. The PBL depth also agrees well with aerosol lidar measurements at Leipzig performed within the European Aerosol Research Lidar Network (EARLINET), indicating a PBL depth of roughly 2600 m at 14 UTC (see Figure S3).

3.2.2 Greenhouse gas flux results

Our measurements on July 20th, presented in Figure 5a, show that the urban CO$_2$ plume is well-mixed at the different sampling altitudes between ~500 m and ~1600 m, which is also well represented by the MECO(n) model (see Figure 8c later). The vertical mixing of the CH$_4$ plume is slightly less pronounced, as indicated by the measurements of the highest altitude transect (see Figure 5a and previous Sect. 3.2.1). Based on the simulated vertical GHG distribution within the entire PBL at the downwind transects (see Figure S10 and Sect. S8 in the supplement for further details), we further deduce that the CO$_2$ emissions are also well-mixed above and below our flight altitudes and more specifically, throughout the PBL, where emissions are efficiently trapped by the strong inversions (as illustrated by the meteorological measurements in Figure 5b). We then estimate the emission rate individually for each single transect, similar to the method described by e.g. Karion et al. (2013) and Peischl et al. (2013). Thereby, actual wind (and temperature and pressure) measurements are considered at each single GHG measurement point. The estimated mass flux then is derived as the mean from the three single mass fluxes, which also account for the less homogeneous distribution of the CH$_4$ plume. As introduced in Sect. 2.2 and shown in Eq. (1), uncertainties in the mass flux are influenced by uncertainties in the choice of background determination, wind speed, wind direction and varying PBL depth. Table 5 lists the emission rate estimate and sources of uncertainties, which are discussed in the following.

We illustrate two approaches to obtain the atmospheric background mixing ratio (see Figure 5a). The first approach uses the linear interpolation of atmospheric mixing ratios between the plume edges (indicated by the dashed lines). For this, the plume was defined as when the measured mixing ratios exceeded the inward running mean (interval of ±30 s) plus one standard deviation (only for CO$_2$) for at least five consecutive measurement points. The atmospheric background is then calculated using the average of 60 measurement points outside the plume. One exception is the south-western end of the intermediate CH$_4$ transect (i.e. at negative distances), which is marked in purple. Due to missing observational data in that transect, we did not observe a decrease in CH$_4$ mixing ratios down to lower (and hence, atmospheric background) values as detected for the other two transects (indicated in grey and blue). In this case, we therefore used the mean CH$_4$ background determined from the other two transects. Each altitude transect thereby is treated separately to account for possible vertical atmospheric gradients, and both sides of each transect are considered to capture the horizontal gradients.

In a second approach to obtain the atmospheric background, the mixing ratios measured during the upwind leg (indicated by the light purple line in Figure 5a) were projected on the downwind wall using the calculated HYSPLIT trajectories. It is assumed that the mixing ratios from the upwind leg are valid for the entire PBL (i.e. for all three transects). Using that approach, we still obtain quite good agreement between the CO$_2$ mixing ratios measured on both edges of the downwind plume and in the upwind leg. However, the measured CO$_2$ mixing ratios in the inflow leg are somewhat larger than the linearly interpolated background. For CH$_4$, the slope of the SW to NE gradient is steeper in the inflow leg compared to the slope in the downwind observations. The large impact of the choice of background is reflected in the resulting flux calculations, which differ by nearly 50% in the case of CO$_2$. Using the first approach, we obtain a stronger flux (2.12 t CO$_2$, s$^{-1}$) compared to the second approach (1.39 t CO$_2$, s$^{-1}$). For CH$_4$, the mean fluxes do not differ as much (21%). It is worth noting that the upwind leg was flown between ~11:35 and ~12:00 UTC, while the downwind wall was probed between ~12:30 and ~13:50 UTC. Consequently, with an average measured wind speed of 4.8 m s$^{-1}$ and a distance of ~50 km between the up- and downwind

|          | CO$_2$     | CH$_4$     |
|----------|------------|------------|
| Mass flux | 1.39 ± 0.76 | 5.20 ± 1.70 |
| Uncertainties | [%] | [%] |
| Choice of background | ±52 | ±21 |
| Wind speed and direction | ±15 | ±23 |
| PBL depth variation | ±9 | ±10 |
transects, the air was not sampled in a Lagrangian manner. Such sampling involves first the measurement of an air mass at a specific starting location, and second the re-sampling of the same air mass after it has been transported to a new location. These observations demonstrate the importance of separating the urban plume from enhancements caused by emissions from anthropogenic sources (or natural variability) upstream of the city, even in the case of an apparently relatively isolated city as Berlin (see population density in Figure S9). Upwind of the flight track (to the east and west) two large nature parks are located (Naturpark Westhavelland, Biosphärenreservat Schorfheide-Chorin) having a population density of only 50 to 100 inhabitants per km² (see also Figure S9). With respect to Figure 1, their location is still included in the plots (at ~53°N and 12.8°E; ~53°N and 13.3°E, respectively), however, neither EDGAR nor CAMS-REG indicate any relevant sources of CO₂. For CH₄ (at least in EDGAR) two grid cells in the vicinity are coloured in green, indicating emissions from solid waste disposal.

Similar to Cambaliza et al. (2014), we calculated the influence of the uncertainty of the wind on the estimated flux by Δflux/Δu with Δflux being the change in the flux and Δu being the uncertainty in the perpendicular wind component. The measurement uncertainties in wind direction (±2°) and wind speed (±0.3 m s⁻¹) translate into errors of ±15% in the CO₂ flux and ±23% in the CH₄ flux. On July 20th we only penetrated the free troposphere once, but comparable conditions (i.e. pronounced inversions) were also prominent on July 18th (see Figure S2). On July 18th we performed two vertical profiles separated by approximately two hours and observed only a difference of ~50 m (~2%) in the PBL depth. If we assume that no significant PBL variation (~2%) occurred during the flight on July 20th, the errors are small in the estimated fluxes (<1%). To test the sensitivity of the derived mass fluxes on the PBL depth variability, we calculated the errors for larger variations as well. In the case the PBL depth would change by 5 to 10% (roughly 140 to 270 m), the errors in the estimated fluxes (<10% for the CH₄ flux and <9% for the CO₂ flux) are still smaller than the uncertainty arising from wind speed and wind direction (15% for CO₂ and 23% for CH₄, see Table 5). Entrainment and detrainment from the free troposphere, another possible source of error, can be excluded in this case due to the pronounced inversions at the top of the PBL (see Figure 5b). Considering all sources of uncertainties, our emission estimate (our emission estimate results in) results in 1.39 ± 0.76 t CO₂ s⁻¹ and 5.20 ± 1.70 kg CH₄ s⁻¹ (see Table 5). For this calculation the second approach to obtain the atmospheric background was chosen, hence, important information of the inflow are considered, which obviously should not be neglected in the case of Berlin. However, we also included the first approach (using the edges of the downwind track) as uncertainty estimate, since the use of the up- and downwind transects has the disadvantage, that the air masses were not sampled in a Lagrangian manner.

3.2.3 Discussion on emission fluxes

To be able to compare our flux estimate with the corresponding area of the three emission inventories EDGAR, CAMS-REG and BERLIN (see Sect. 2.4), we first determine a footprint area with HYPLIT. The footprint reflects the percentage of the air mass within the PBL at a specific location (of a defined latitude-longitude grid) between the upwind and the downwind track (see Sect. 2.3.1). To determine the horizontal edges, backward trajectories were released only for GHG enhancements greater than zero after subtracting the upwind mixing ratios from the observed downwind mixing ratios. Figure 6 gives the flight path of July 20th, where the downwind wall is colour-coded according to the GHG enhancement, and the footprint analysis is shaded with

![Figure 6: CO₂ and CH₄ footprint of July 20th. Footprint corresponding to a grid box of 0.03° × 0.065° for (a) the CO₂ and (b) the CH₄ plume detected on July 20th. The purple marker in panel (a) indicates the location of three point sources from E-PRTR, which are located within a 1 km radius next to each other. DOI: https://doi.org/10.1525/elementa.411.f6](https://doi.org/10.1525/elementa.411.f6)
respect to the observed $\text{CO}_2$ and $\text{CH}_4$ plume. The distribution of trajectory points in Figure 6a is relatively homogeneous and indicates that the $\text{CO}_2$ plume covers the entire city. The sharp spikes in $\text{CO}_2$ (see Figure 5a at roughly 7 km distance) are detected directly downwind of three $\text{CO}_2$ point sources listed in E-PRTR: Fels-Werke GmbH, CEMEX Zement GmbH, and Vattenfall Europe New Energy Ecopower GmbH. They are located within a 1 km radius next to each other (marked in orange) and together report emissions of 1.9 Mt $\text{CO}_2$ in 2016. In contrast, the footprint of the $\text{CH}_4$ plume (Figure 6b) indicates that sources in the south-west outside of Berlin contribute to the measured $\text{CH}_4$ enhancement.

In a next step, we scale our flux estimate to a yearly value (regardless of diurnal, weekly or seasonal cycles), the result is shown in Figure 7 with blue bars. Grey bars denote emissions of CAMS-REG, EDGAR and BERLIN within the Berlin city boundary. The striped bars represent emissions from the footprint-corrected inventories of CAMS-REG and EDGAR, i.e. the emissions in the inventory grid cells, which are covered by the footprint area, are added up. Thus, we get 35 to 75% larger emission rates for $\text{CO}_2$ and 40 to 270% larger emission rates for $\text{CH}_4$ if we consider the sources which are located outside the definition of the city boundaries. According to Stohl (1998), trajectories may deviate by 10% of their travelled distance. Thus, the footprint area for this case study might be mislocated by up to ~7.5 km. The resulting deviation in the total inventorial emissions is considered by the error bars in Figure 7.

For the flight on July 20th, our annual $\text{CO}_2$ emission rate of ~44 ± 24 Mt a$^{-1}$ is larger than the CAMS-REG and the EDGAR footprint estimates, but agrees within the error bars. Previous studies showed, that urban $\text{CO}_2$ emissions based on EDGAR (especially related to the road transport sector) are likely overestimated, however these studies only focused on American cities (e.g. Gately et al., 2013; McDonald et al., 2014; Gately and Hutrya, 2017). Gately et al. (2013) pointed out, that based on the road density proxy a downscaling of total US emissions leads to an overestimation of on-road emissions for areas with higher than average road densities (and vice versa). Also in the study by McDonald et al. (2014) vehicle emissions were overestimated by 40 to 80% in Californian cities. We are not aware of similar studies for European cities. While there is a strong seasonality of $\text{CO}_2$ fluxes due to domestic heating (with its emission maximum in winter), the contribution from road traffic (the second most important $\text{CO}_2$ sector for this study according to the inventories, see Table 3) is relatively constant throughout the year (Kuc et al., 2003; Gioli et al., 2012; EDGAR monthly resolved emissions in 2010). As a consequence, inventorial $\text{CO}_2$ fluxes in summer are expected to be lower than presented in Figure 7. Thus, probing Berlin during different seasons would be very valuable.

Hase et al. (2015) reports on urban Berlin emissions apparent in $\text{CO}_2$ signals (column-averaged dry air mole fraction) from five ground-based Bruker EM27/SUN Fourier-Transform infrared (FTIR) spectrometers. Applying a simple dispersion model for three measurement days in June and July 2014 they retrieve a $\text{CO}_2$ source strength of 0.8 t s$^{-1}$. Compared to the estimate of Hase et al. (2015), our flux estimate is almost twice as large, but agrees well within the error estimate. Our stronger emission rate is not surprising, as measurements were performed four years apart and $\text{CO}_2$ emissions have a very strong seasonal and diurnal cycle. In addition, due to the fixed location of FTIR spectrometers, emission rates might have been estimated from a different (possibly smaller) footprint area, which would result in lower emission rates.

In the temperate latitudes the $\text{CO}_2$ uptake of plants usually predominates in spring and summer. A modelling study using synthetic $\text{CO}_2$ data by Mueller et al. (2018) showed, that for the Baltimore-Washington area a substantial fraction of ~35% of the $\text{CO}_2$ background variability is caused by biogenic sources and sinks in July.

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**Figure 7: GHG emission fluxes for the Berlin area.** Estimated mass balance fluxes for $\text{CO}_2$ (left) and $\text{CH}_4$ (right) from the flight on July 20th in blue. Grey bars indicate inventorial fluxes within the Berlin city boundary. Striped bars represent the inventory fluxes enlarged by the area from the corresponding footprint analysis. DOI: https://doi.org/10.1525/elementa.411.f7
Therefore, we simulate the biogenic CO$_2$ flux with the Vegetation Photosynthesis and Respiration Model (VPRM, Mahadevan et al., 2008) coupled with WRF-GHG (Beck et al., 2013) in order to estimate the influence of the photosynthetic uptake and respiration on our assessment of the anthropogenic flux. According to our analysis, the maximum daytime photosynthetic uptake predicted over the footprint area is at most 12% of the estimated anthropogenic flux. Due to that and the fact that the change of the predicted CO$_2$ biogenic signal over the small constrained area is low (well within the observed variability of the signal), we have concluded that the explicit correction for the biogenic influence is not needed. More information can be found in the supplement (see Figure S6 and the discussion therein). Hardiman et al. (2017) simulates the biogenic carbon flux throughout Massachusetts, including the urban area Boston, by using the VPRM model, which was specifically adapted to urban-specific influences by e.g. incorporating higher temperatures and impervious surfaces like pavement and buildings. They conclude that urban vegetation is probably not offsetting a significant fraction of anthropogenic emissions (e.g. only ~2% in Boston), which is consistent with previous studies cited therein.

In contrast to the observed CO$_2$ plume pattern, the CH$_4$ plume extended further to the west over the city limits. Also the XCH$_4$ signal of Hase et al. (2015) did not follow their XCO$_2$ pattern (due to different sources), and a distinct background could not be observed. Compared to the footprint-corrected inventory data, our annual CH$_4$ flux estimate of $164 \pm 54$ kt a$^{-1}$ is tremendously larger (factor $\sim$7) than the CAMS-REG estimate, which has only little contribution of the waste sector. However, it agrees better with EDGAR, which considers larger emission amounts coupled to any form of waste treatment (for source sector partitioning see Table 3). According to Saunois et al. (2016), CH$_4$ produced in waste water is dependent on the amount of organic content in the water itself. The inventories considered in their study (EDGAR v.4.2F2010, GAINS 5a, USEPA 2012) give global emissions from waste water in the range of 9 to 30 Tg in 2005 (3 to 9% of global anthropogenic emissions). For the greater area of Berlin, EDGAR even states that 18% of total emissions are related to waste water handling and 48% to solid waste deposition. In London, waste treatment and disposal is the primary source of CH$_4$ based on the National Atmospheric Emissions Inventory besides natural gas leakage (Pitt et al., 2019). As ethane (C$_2$H$_6$) is co-emitted with CH$_4$ in oil and gas operations, we can use it as a tracer for gas leakages. However, during our mission flights we did not detect any significant variations in the QCLS C$_2$H$_6$ data. When comparing total CH$_4$ emissions from Germany, CAMS-REG is only a factor of 0.4 lower than EDGAR (and both inventories agree for CO$_2$) as both inventories are scaled to fit to the reported country values. As a result, we conclude that the waste sector (SNAP 9) in CAMS-REG is most likely underrepresented in the Berlin area either in terms of strength or in terms of spatial distribution, or a combination of both.

### 3.3 Comparison of GHG measurements with the MECO(n) simulation results

With the high-resolution numerical model MECO(n) of $1 \text{ km} \times 1 \text{ km}$ we simulate CO$_2$ and CH$_4$ mixing ratios based on different source tracers (as described in Sect. 2.3.2). By comparing the model output to our airborne measurements, we aim to better understand the source sectors and regions contributing to the measured pollution plumes. Note that in the MECO(n) model simulations the potential temporal source strength variability (on daily, weekly or seasonal timescales) cannot be captured, as only annual averaged emissions from the prescribed inventories are available.

#### 3.3.1 Underestimated and missing CH$_4$ sources as observed on July 20th

For the flight on July 20$^{th}$ (as described in Sect. 3.2) backward trajectories were started from the location of the CO$_2$ plume. They cover the entire city of Berlin and the estimated CO$_2$ mass flux is in agreement with current inventories. In contrast, backward trajectories released from the broader CH$_4$ plume, which extend over the city boundary, indicate an impact of emissions from further west of the city and a relatively large CH$_4$ emission flux was derived. In a first step we simulate GHG mixing ratios based on the local BERLIN inventory to isolate the urban emissions (called c-CO$_2$ and c-CH$_4$ for “city”, see Table 2). Please note, that due to the model biases (as discussed below) MECO(n) in the applied setup is able to provide spatial distributions for the two GHG, but is limited in the representation of absolute values. In Figure 8 different perspectives are presented. Panels (a) and (b) show the 2D distribution of simulated column-averaged dry air mole fraction at 13 UTC to illustrate the plume extent. Panel (c) depicts on the right axis the simulated GHG mixing ratios (indicated in grey), which have been sampled on-line along the flight track (submodel S4D as described by Jöckel et al., 2010). Together with the measured GHG enhancements on the left axis, the GHG distribution along the horizontal distance of the flight track is shown. The measured enhancements refer to the upward mixing ratios being subtracted from the observed downwind mixing ratios and are indicated in dark red. The average measured wind direction during the flight was $299 \pm 27^\circ$. The MECO(n) results reflect the prevalent wind direction quite well with $305 \pm 36^\circ$, therefore the location of the simulated c-CO$_2$ plume agrees well with the measured CO$_2$ plume. In addition, the shape of the simulated plume is consistent with observations and also the coherent c-CO$_2$ mixing ratios within all heights. However, the maximum enhancements differ by a factor of $\sim$2. This is either the result from underestimated prescribed emissions, or coupled to any form of model biases (or a combination of both). A quantitative comparison of maximum enhancements is difficult due to large differences in simulated wind speed ($+2.1 \text{ m s}^{-1}$) compared to observations, thus reducing the enhancement. Furthermore, differences in the simulated PBL depth (~600 m lower than observed) influence the GHG dilution within the PBL and increase the enhancement,
but also affect the plume width. Emissions from the point sources east of Berlin (see Figure 6a), which are reflected by the sharp CO\textsubscript{2} measurement spikes at \~7 km distance, are clearly visible at \~52.5°N and \~13.8°E, as well as in the simulated c-CO\textsubscript{2} mixing ratio along the flight track.

With respect to simulated c-CH\textsubscript{4} (grey) and measured CH\textsubscript{4} (dark red), it stands out that simulated c-CH\textsubscript{4} enhancements are far too low (factor of \~5), although the BERLIN inventory is already scaled with a factor of \~4.5 (see Table 2) and the observed plume is significantly wider. This implies a huge underestimation of urban source strengths in the BERLIN inventory, or missing CH\textsubscript{4} emissions in the surroundings of Berlin. One should keep in mind that CO\textsubscript{2} observations are reflected by the simulated city emissions quite well. Thus, a number of different tracers were implemented, in order to investigate the origin of the unexpected missing CH\textsubscript{4} sources which are required to reproduce the observed CH\textsubscript{4} pattern.

The outer parts of the plume could not be reproduced by the "regional" tracer r-CH\textsubscript{4} (coupled CAMS-REG inventory; not shown), therefore we did further studies to examine the contribution from emissions of the waste sector to the detected CH\textsubscript{4} plume. Figure 9 depicts simulated CH\textsubscript{4} mole fractions from the tracers related to waste water treatment plants (w-CH\textsubscript{4}, panel a), landfills (l-CH\textsubscript{4}, panel b) and to the combined tracer t-CH\textsubscript{4} (which is "city" plus "waste water" plus "landfills", panel c). Largest waste water facilities (they are sized according to their population equivalent) are located directly outside of the city; smaller facilities are found more frequently in the west and south-west and produce a rather homogeneous CH\textsubscript{4} mole fraction distribution field, despite their nature of point sources. Simulated CH\textsubscript{4} mole fractions from the landfill tracer (information according to CORRECTIV, 2016) indicate more point-like distributions of enhanced CH\textsubscript{4} either from waste-rich dumps (they are sized according to the amount of deposited waste) or from a spatial concentration of several dumps, e.g. seen in the north of Berlin. Figure 9c shows the mole fractions from the combined t-CH\textsubscript{4} tracer. As already stated in Sect. 2.4, we do not claim that our emission rate estimate for waste water and landfills are correct, however, the inhomogeneous CH\textsubscript{4} distribution, resulting from the t-CH\textsubscript{4} tracer, is consistent with the results of Hase et al. (2015), who report a less uniform and more varying CH\textsubscript{4} background compared to CO\textsubscript{2}. The mean mixing ratio from the three altitude transects for each tracer (w-CH\textsubscript{4}, l-CH\textsubscript{4}, and t-CH\textsubscript{4}) is shown in Figure 9d along the horizontal distance of the flight path together with the measured CH\textsubscript{4} enhancement (mean of the lower two flight transects only). Although these model results do not directly reproduce the measured CH\textsubscript{4} plume shape, emissions from waste water plants and landfills contribute (broaden and enhance) to the simulated c-CH\textsubscript{4} plume (see Figure 8c), which considers only the city emissions. These results suggest that for a better understanding of the regional CH\textsubscript{4} budget, emission strengths from these

![Figure 8: Simulated and measured GHG mixing ratios on July 20th. 2D distributions of simulated column-averaged dry air mole fractions of (a) the c-CO\textsubscript{2} and (b) the c-CH\textsubscript{4} tracer at 13 UTC of July 20\textsuperscript{th}. The flight path is shown in black, and the downwind “wall” is colour-coded according to the measured CO\textsubscript{2} and CH\textsubscript{4} enhancement. Panel (c) presents GHG mixing ratios along the horizontal distance (simulated mixing ratios in grey; GHG measurements, averaged on the model time resolution of 20 s, in dark red). DOI: https://doi.org/10.1525/elementa.411.f8](https://doi.org/10.1525/elementa.411.f8)
sources need to be experimentally verified by e.g. a ground based measurement campaign in the greater region of Berlin.

3.3.2 Greenhouse gas plume as observed on July 24th
From the results of Sect. 3.3.1 two hypotheses can be formulated. First, the reported urban CO₂ emissions from Berlin are well constrained, since the simulated c-CO₂ enhancements are in the same order of magnitude as our measured CO₂ enhancements. Second, and in contrast, the reported urban CH₄ emissions are largely underestimated. Based on this single research flight we were able to show, that simulated c-CH₄ enhancements are dramatically smaller than our measured CH₄ enhancements. To analyse the robustness of our hypothesis in more detail, we compare observations from an additional flight performed on July 24th (see Table 4) with the MECO(n) simulated GHG mixing ratios.

The time series of the whole flight (~12:10 to ~15:00 UTC) can be found in Figure S1. Figure 10a shows measured CO₂ (upper panel) and CH₄ (lower panel) mixing ratios of the three downwind transects along the horizontal distance. The flight track is depicted in Figure 10b and c together with the simulated full atmospheric column-averaged dry air mole fractions from the c-CO₂ and c-CH₄ tracer, respectively. The flight pattern is constructed similar to the one on July 20th with an upwind transect (at 796 m) and a vertical profile at the Tempelhofer Feld (~0.3 to ~3.2 km; ascend rate of ~3.1 m s⁻¹). We sampled the plume at three altitudes (494, 827 and 1308 m) starting from 0 km distance towards the east (indicated with positive distances). Instead of forecasted northern winds (E-W alignment of the flight transects), the wind was blowing from the NNE with 3.3 m s⁻¹ on average. To aim for sampling background air, the transects were prolonged towards the NW (negative distance; determined by restrictions from ATC).

As obvious in Figure 10a, CO₂ and CH₄ mixing ratios were enhanced above the background by up to 5 ppm CO₂ and 18 ppb CH₄, respectively. As seen in the comparable CO₂ mixing ratios at all heights, the PBL is generally well-mixed (which is consistent with the meteorological vertical profile, see Figure S2). Measured CH₄ mixing ratios are coherent in the two lower flight transects, however, show a different pattern at the highest altitude with a dip in CH₄ between ~20 and ~30 km (similar to observations on July 20th; see Sect. 3.2.1). Furthermore, the encountered GHG plume can be divided into two parts, with a positive correlation between CO₂ and CH₄ in the middle and eastern part (R = +0.82) and slightly negative correlations in the north-western part (R = −0.20). The dashed lines in Figure 10a indicate the linear interpolated background
mixing ratios based on the edges of the flight track. GHG mixing ratios in the eastern part are lower than in the western part (~1.5 ppm for CO₂ and ~4 to ~6 ppb for CH₄).

We refrain from estimating GHG fluxes based on the mass balance approach for this second case study, because our analyses indicate that due to very low wind speeds emissions accumulated above Berlin during the night of July 24th (see Sect. S5 and S6 in the supplement for more details). Steady winds are, however, essential for the mass balance approach.

The simulated column-averaged dry air mole fractions from the c-CO₂ and c-CH₄ tracer at 14 UTC are presented in Figure 10b and c, respectively. The downwind "wall" is color-coded according to the measured enhancement (here the linear background is subtracted from the observed downwind mixing ratios). Figure 10b indicates that the simulated c-CO₂ plume only captures the eastern part of the detected plume, although the simulated wind direction is similar to the measured wind direction (+2°).

The mixing ratios along the flight track are depicted in Figure 10d (from the c-CH₄ and c-CO₂ tracer in grey on the right axis, from the airborne measurements in dark red on the left axis). This depiction confirms that the eastern part of the CO₂ plume is well captured by c-CO₂, however, the western part is missing. Neither in the EDGAR inventory, nor in E-PRTR any strong source of CO₂ is present west of Berlin (or within western Berlin), which could explain our observations. Also in the simulated mixing ratio from the r-CO₂ tracer (coupled regional CAMS-REG inventory; not shown) we could not reproduce this part of the plume. However, the disagreement in the plume shape possibly might arise from a transport and/or timing bias in the MECO(n) simulation. The time-series of the simulated GHG plume on the 24th of July shows, that the plume meanders from east to west, i.e. already at 16 UTC the plume is located more in the west (see Figure S11 in the supplement). However, the simulated maximum c-CO₂ enhancement of ~2 ppm is 2 to 3 times lower than our measured enhancement of ~5 ppm. MECO(n) simulates a higher PBL depth (~200 m) and overestimates the wind speed (+2.6 m s⁻¹), thus leading to simulated GHG mixing ratios which are much more diluted within the PBL. Although this dilution cannot be properly quantified, we can deduce that the order of magnitude of simulated c-CO₂ mixing ratios is in much better agreement with our measurements than it is the case for CH₄, similar to the
results from the flight on July 20th. The lower panel of Figure 10d indicates that the plume structure of c-CH₄ is well simulated, as well as the coherent mixing ratios at all heights. The maximum simulated enhancement of only ~4 ppb is, however, heavily underestimated (factor 4 to 5).

4 Conclusion
An improved understanding and quantification of the GHG budget is needed for the development of efficient mitigation strategies and for the prediction of future climate impacts. With our study we contribute to the sparse knowledge of GHG emissions from European cities by studying the urban CO₂ and CH₄ distribution downwind of the German capital Berlin. Emission inventories available for Berlin (EDGAR, CAMS-REG and BERLIN) differ by a factor of ~1.4 and ~8 for CO₂ and CH₄, respectively.

In July 2018 we performed in situ CO₂ and CH₄ measurements aboard the DLR Cessna Grand Caravan in the outflow of Berlin to assess the urban GHG emissions from the city. Our measurements showed, that even with sensitive airborne in-situ measurements it remains challenging to detect Berlins GHG emissions, which fits to the observations reported by Hase et al. (2015) who used ground-based FTIR spectrometer measurements, and by Kuhlmann et al. (2019a) who used synthetic CO₂ satellite observations.

The detection and isolation of the urban GHG plume, however, was possible during two research flights performed on July 20th and July 24th. GHG enhancements were observed in different heights of a well-mixed and efficiently capped boundary layer. Using the mass balance approach, we obtained a CO₂ flux of 1.39 ± 0.76 t s⁻¹ for the flight on July 20th, which is in the same order of magnitude as the numbers given in the presented inventories. Sampled air masses homogenously passed the city of Berlin prior to our measurement, where they were enriched with emissions. Simulated citywide CO₂ mixing ratios agree well with our CO₂ measurements in location and shape, while a quantitative comparison of simulated and measured maximum enhancements is hampered due to deviations in the wind speed and PBL depth.

For the same flight we calculated a CH₄ mass flux of 5.20 ± 1.70 kg s⁻¹, which is almost two times larger than the highest reported value in the inventories (i.e. the footprint-corrected estimate from EDGAR). Simulated citywide CH₄ mixing ratios were substantially lower than observed as well, indicating a significant underrepresentation of urban emissions in the local BERLIN and regional CAMS-REG inventory. The CH₄ plume (in contrast to the CO₂ plume) partly originated also from outside of the Berlin city boundary. Our study shows that emissions potentially originate from sources surrounding the city, which are missing in the three investigated inventories. These might include emissions from waste water treatment plants and illegal landfills as indicated by information from other sources. However, for a clear attribution to these sources, a verification of these emissions is required by ground-based measurements and/or further airborne in-situ observations.

Although Berlin can be considered as a relatively isolated city (especially for the densely populated region of Central Europe), we demonstrated the need for precisely determining the inflow GHG mixing ratios and the natural variability of the background concentrations in order to suitably apply the mass balance approach. One possibility to improve the estimation of the background is to sample the upwind and downwind transects in a Lagrangian manner, e.g. also using two aircraft simultaneously. To further improve the knowledge on the regional CH₄ budget in the greater Berlin area, subsequent measurement campaigns are needed with a special focus on emission sources located outside the city boundaries. Furthermore, different seasons should be investigated to reflect the seasonal cycle of the emissions.

Our study showed that bottom-up CO₂ and, in particular, CH₄ emission inventories are subject to large uncertainties at the city scale, even for major cities in highly developed countries like Germany, and that top down emission estimates are an important tool to verify emission estimates and to reveal missing sources in emission inventories.

Data Accessibility Statement
Observational data (https://doi.org/10.5281/zenodo.3707990; Kuhlmann et al., 2020) and simulation results (https://doi.org/10.5281/zenodo.3707990; Mertens and Jöckel, 2020) are publicly available. The MECO(n) model system is part of the Modular Earth Submodel System (MESSy). MESSy is continuously further developed and applied by a consortium of institutions. The usage of MESSy and access to the source code is licenced to all affiliates of institutions which are members of the MESSy Consortium. Institutions can become a member of the MESSy Consortium by signing the MESSy Memorandum of Understanding. The COSMO model, as part of MECO(n), is licenced by the German Weather Service (DWD), or by the COSMO-CLM community. More information can be found on the MESSy Consortium Website (http://www.messy-interface.org). The code used for the present study has been based on MESSy version 2.54.0.

Supplemental files
The supplemental data for this article can be found as follows:

- Figure S1. Time series of CO₂, CH₄ and flight altitude for five research flights during July 2018. DOI: https://doi.org/10.1525/elementa.411.s1
- Figure S2. Vertical profiles of the upward spiral at Tempelhofer Feld. The PBL depth (dashed black line) was determined from virtual potential temperature, static temperature and relative humidity. DOI: https://doi.org/10.1525/elementa.411.s1
- Figure S3. Aerosol lidar measurements at Leipzig. The temporal evolution of the range-corrected signal at 1064 nm is shown. Available at http://polly.tropos.de/?p=lidarzeit&Ort=1&Jahr=2018. Accessed October 04, 2019. Further information can be found in Engelmann et al. (2016) and Baars et al. (2016). DOI: https://doi.org/10.1525/elementa.411.s1
- Figure S4. Time series of forecasted and actual wind direction and wind speed. The whole cam-
campaign period from July 8th to 26th is shown. DOI: https://doi.org/10.1525/elementa.411.s1

• Figure S5. Correlations of wind direction and wind speed between airborne measurement data and ECMWF forecast data. Correlation coefficients are abbreviated with Pr. DOI: https://doi.org/10.1525/elementa.411.s1

• Figure S6. Biogenic CO₂ mixing ratios up- and downwind of Berlin on July 20th. DOI: https://doi.org/10.1525/elementa.411.s1

• Figure S7. Ascents of a captive balloon system located at Tempelhofer Feld on July 24th. Wind measurements were carried out by the DWD (R. Becker, P. Stanislawsky, M. Koßmann) in the framework of the [UC] project. DOI: https://doi.org/10.1525/elementa.411.s1

• Figure S8. Backward trajectory calculations for July 24th. Trajectories were started at 14:30 UTC from the downwind flight track backwards in time for 6 hours. Colour coded is the height of the trajectory according to its relative position. DOI: https://doi.org/10.1525/elementa.411.s1

• Figure S9. Population density for Berlin and the state Brandenburg from 2017. The yellow to red colours indicate the inhabitants per km², the blue dots show numbers for total population in each municipality. The Berlin city boundary (black) was added as well as latitude and longitude labels. Adapted after Strukturatlas Land Brandenburg (2017). DOI: https://doi.org/10.1525/elementa.411.s1

• Figure S10. Curtain of simulated c-CH₄ and c-CO₂ on July 20th. The third crossing of the Berlin plume at ~1600 m altitude is shown from ~13:20 to ~13:35 UTC. DOI: https://doi.org/10.1525/elementa.411.s1

• Figure S11. Time-series of the simulated c-CO₂ plume on July 24th. Shown are snapshots at (a) 14 UTC, which is identical to Figure 10b and (b) at 16 UTC to indicate, that the plume moves towards the west during the afternoon. DOI: https://doi.org/10.1525/elementa.411.s1

• Text S1. Calibration procedure and measurement uncertainty. DOI: https://doi.org/10.1525/elementa.411.s1

• Text S2. Greenhouse gas time series and altitude profiles. DOI: https://doi.org/10.1525/elementa.411.s1

• Text S3. ECMWF forecast data and wind measurement agreement. DOI: https://doi.org/10.1525/elementa.411.s1

• Text S4. Biogenic CO₂ flux. DOI: https://doi.org/10.1525/elementa.411.s1

• Text S5. Wind situation on July 24th. DOI: https://doi.org/10.1525/elementa.411.s1

• Text S6. Backward trajectories on July 24th. DOI: https://doi.org/10.1525/elementa.411.s1

• Text S7. Berlin and its surrounding area. DOI: https://doi.org/10.1525/elementa.411.s1

• Text S8. Simulated vertical GHG distribution within the PBL. DOI: https://doi.org/10.1525/elementa.411.s1

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Competing interests
The authors have no competing interests to declare.

Author contributions
- Contributed to conception and design: TK, AR, HH
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- Contributed to analysis and interpretation of data: TK, MM, HH, MG, GK, RB, AF, PJ, MP, AR
- Drafted and/or revised the article: TK, MM, HH, MG, GK, RB, AF, PJ, MP, AR
- Approved the submitted version for publication: TK, MM, HH, MG, GK, RB, AF, PJ, MP, AR

References
AVISO GmbH and IÉ Leipzig. 2016. Erstellung der Berliner Emissionskataster Industrie, Gebäudeheizung, sonstiger Verkehr, Kleingewässer, sonstige Quellen, Baustellen – Schlussbericht Juni 2016. Techn. rep. Available at https://www.berlin.de/senuk/umwelt/luftqualitaet/de/emissionen/download/Endbericht_Emissionskataster_2015.pdf. Accessed September 30, 2019.
Beck, V, Gerbig, C, Koch, T, Bela, MM, Longo, KM, Freitas, SR, Kaplan, JO, Prigent, C, Bergamaschi, P and Heimann, M. 2013. WRF-Chem simulations in the Amazon region during wet and dry
season transitions. Evaluation of methane models and wetland inundation maps. Atmos Chem Phys 13[16]: 7961–7982. DOI: https://doi.org/10.5194/acp-13-7961-2013

Bergamaschi, P, Karstens, U, Manning, AJ, Saunois, M, Tsuruta, A, Berchet, A, Vermeulen, AT, Arnold, T, Janssens-Maenhout, G, Hammer, S, Levin, I, Schmidt, M, Ramonet, M, Lopez, M, Lavric, J, Aalto, T, Chen, H, Feist, DG, Gerbig, C, Haszpra, L, Hermansen, O, Manca, G, Moncrieff, J, Meinhardt, F, Necki, J, Galkowski, M, O’Doherty, S, Paramonova, N, Scheeren, HA, Steinbacher, M and Dlugokencky, E. 2018. Inverse modelling of European CH4 emissions during 2006–2012 using different inverse models and reassessed atmospheric observations. Atmos Chem Phys 18(2): 901–920. DOI: https://doi.org/10.5194/acp-18-901-2018

Brandt, AR, Heath, GA, Kort, EA, O’Sullivan, F, Pétron, G, Jordaan, SM, Tans, P, Wilcox, J, Gopstein, AM, Arent, D, Wofsy, S, Brown, NJ, Bradley, R, Stucky, GD, Earylide, D and Harriss, R. 2014. Methane leaks from North American natural gas systems. Science 343(6172): 733–735. DOI: https://doi.org/10.1126/science.1247045

Bréon, FM, Broquet, G, Puygrenier, V, Chevallier, F, Xuereb-Remy, I, Ramonet, M, Dieudonné, E, Lopez, M, Schmidt, M, Perrussel, O and Ciais, P. 2013. Observations: Atmosphere and surface. In: Zhai, PM, Fuglestvedt, J, Van der Linden, PJ, Johnson, OR, Canziani, OP, Lebreton, L, McLean, K, Moss, DJ, Péan, C, Plummer, D, Shah, M, van Aalst, M, Barros, VA, Chatterjee, AR, Christensen, NE, Clark, WL, Field, CB, Held, I, Kwon, OP, Laprise, R, Marchant, MR, Maycock, T, Öktem, B, Riedlinger, J, Tokioka, T, Tubiello, FF, Watanabe, S, Zhao, X and Qin, D (eds.), 509–555. Cambridge, MA, USA: Cambridge University Press. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. DOI: https://doi.org/10.1017/CBO9781107415324.007

Bubier, J, Imboden, M, Elkins, JW, Karl, DM, Sweeney, C,扇, Ramonet, M, Metzl, N, Wong, M, Thomas, G and Wallace, DJ. 2006. Surface exchange of carbon dioxide fluxes and source partitioning in urban areas: The case study of Florence, Italy. Environ Pollut 143: 125–131. DOI: https://doi.org/10.1016/j.envpol.2005.04.023

Campa, L, Debussche, R, Naveau, P, Doucet, S, Vannitsem, S and Vrac, M. 2016. An attempt at estimating Paris area CO2 emissions from atmospheric concentration measurements. Atmos Chem Phys 16: 11645–11666. DOI: https://doi.org/10.5194/acp-16-11645-2016

Ciais, P, Fradelizi, M, Denning, AS, Guenther, A, Heinold, B, Janssens-Maenhout, G, Janssens, IJ, Keeling, CD, Kort, EA, Lawrence, DM, Lindberg, SE, Maksyutov, S, Mook, WDJ, Nisbet, RG, Olivier, JGJ, Paris, FR, Permentier, R, Prather, MA, Rödenbeck, C, Sun, Y, Sweeney, C, Verbeeck, K, Zhang, Y, van der Laan-Luijkx, IT and Sarmiento, JL. 2010. Global and European CO2 emissions from inversions of atmospheric CO2 concentrations. Tellus B 62(2): 178–192. DOI: https://doi.org/10.1111/j.1600-0889.2009.00445.x

Ciais, P, Piao, S-L, Wang, T, Peng, S, Huang, J, Razlivanov, I, Miles, NL, Aalto, T, Chen, H, Gerbig, C, Haszpra, L, Hutterli, MA, Huang, J, Razlivanov, I, Miles, NL and Richardson, S. 2015. Quantification and source apportionment of the methane emission flux from the city of Indianapolis. Elementa: Science of the Anthropocene 3: 195–215. DOI: https://doi.org/10.1525/elementa.195

Ciais, P, Piao, S-L, Peng, S, Wang, T, Huang, J, Razlivanov, I, Miles, NL and Richardson, S. 2014. Assessing uncertainties of an aircraft-based mass balance approach for quantifying urban greenhouse gas emissions. Atmos Chem Phys 14: 9029–9050. DOI: https://doi.org/10.5194/acp-14-9029-2014

Chen, H, Winderlich, J, Gerbig, C, Hoefner, A, Reff, C, Crosson, ER, van Pelt, AD, Steinbach, J, Kolle, O, Beck, V, Daube, BC, Gottlieb, EW, Chow, VY, Santoni, GW and Wofsy, SC. 2010. High-accuracy continuous airborne measurements of greenhouse gases (CO2 and CH4) using the cavity ring-down spectroscopy (CRDS) technique. Atmos Meas Tech 3(2): 375–386. DOI: https://doi.org/10.5194/amt-3-375-2010

CORRECTIV. 2016. Available at https://correctiv.org/artikel/2016/03/02/muellparadies-brandenburg. Accessed August 18, 2019.

Crippa, M, Solazzo, E, Huang, G, Giuzziardi, D, Koffi, E, Muntean, M, Schieberle, C, Friedrich, R and Janssens-Maenhout, G. 2019. High resolution temporal profiles in the Emissions Database for Global Atmospheric Research (EDGAR). Nature Scientific Data. Submitted.
United Kingdom and New York, NY, USA: Cambridge University Press. DOI: https://doi.org/10.1017/CBO9781107415324.008

Hase, F, Frey, M, Blumenstock, T, Groß, J, Kiel, M, Kohlepp, R, Mengistu Tsidu, G, Schäfer, K, Sha, MK and Orphal, J. 2015. Application of portable FTIR spectrometers for detecting greenhouse gas emissions of the major city Berlin. *Atmos Meas Tech* **8**(7): 3059–3068. DOI: https://doi.org/10.5194/amt-8-3059-2015

Heimburger, AMF; Harvey, RM, Shepson, PB, Stirm, BH, Gore, C, Turnbull, J, Cambaliza, MOL, Salmon, OE, Kerlo, A-EM, Lavoie, TN, Davis, KJ, Lauvaux, T, Karion, A, Sweeney, C, Brewer, WA, Hardesty, RM and Gurney, KR. 2017. Assessing the optimized precision of the aircraft mass balance method for measurement of urban greenhouse gas emission rates through averaging. *Elementa: Science of the Anthropocene* **5**(26). DOI: https://doi.org/10.1525/elementa.134

Helfter, C, Fumalari, D, Phillips, GJ, Barlow, JF, Wood, CR, Grimmond, CSB and Nemitz, E. 2011. Controls of carbon dioxide concentrations and fluxes above central London. *Atmos Chem Phys* **11**(5): 1913–1928. DOI: https://doi.org/10.5194/acp-11-1913-2011

Helfter, C, Tremper, AH, Halios, CH, Kothis, S, Bjorkegren, A, Grimmond, CSB, Barlow, JF and Nemitz, E. 2016. Spatial and temporal variability of urban fluxes of methane, carbon monoxide and carbon dioxide above London, UK. *Atmos Chem Phys* **16**(16): 10543–10557. DOI: https://doi.org/10.5194/acp-16-10543-2016

Hofmann, C, Kerkweg, A, Wernli, H and Jöckel, P. 2012. The 1-way on-line coupled atmospheric chemistry model system MECO(n) Part 3: Meteorological evaluation of the on-line coupled system. *Geosci Model Dev* **5**: 129–147. DOI: https://doi.org/10.5194/gmd-5-129-2012

Janssens-Maenhout, G, Pagliari, V, Guizzardi, D and Muntean, M. 2012. Global emission inventories in the Emission Database for Global Atmospheric Research (EDGAR). Manual (I) – gridding: EDGAR emissions distribution on global gridmaps. Luxembourg: Publications Office (EUR, Scientific and technical research series, 25785).

Jöckel, P, Kerkweg, A, Pozzer, A, Sander, R, Tost, H, Riede, H, Baumgaertner, A, Gromov, S and Kern, B. 2010. Development cycle 2 of the Modular Earth Submodel System (MESSy2). *Geosci Model Dev* **3**: 717–752. DOI: https://doi.org/10.5194/gmd-3-717-2010

Karion, A, Sweeney, C, Pétron, G, Frost, G, Michael Hardt, R, Kofer, J, Miller, BR, Newberger, T, Wolter, S, Banta, R, Brewer, A, Dlugokencky, E, Lang, P, Montzka, SA, Schnell, R, Tans, P, Trainer, M, Zamora, R and Conley, S. 2013. Methane emissions estimate from airborne measurements over a western United States natural gas field. *Geophys Res Lett* **40**(16): 4393–4397. DOI: https://doi.org/10.1002/grl.50811

Kennedy, C, Demoulin, S and Mohareb, E. 2012. Cities reducing their greenhouse gas emissions. *Energy Policy* **49**: 774–777. DOI: https://doi.org/10.1016/j.enpol.2012.07.030

Kerkweg, A and Jöckel, P. 2012a. The 1-way on-line coupled atmospheric chemistry model system MECO(n) – Part 1: Description of the limited-area atmospheric chemistry model COSMO/MESSy. *Geosci Model Dev* **5**(1): 87–110. DOI: https://doi.org/10.5194/gmd-5-87-2012

Kerkweg, A and Jöckel, P. 2012b. The 1-way on-line coupled atmospheric chemistry model system MECO(n) – Part 2: On-line coupling with the Multi-Model-Driver (MMD). *Geosci Model Dev* **5**(1): 111–128. DOI: https://doi.org/10.5194/gmd-5-111-2012

Kirschke, S, Bousquet, P, Ciais, P, Saunois, M, Canadell, JG, Dlugokencky, EJ, Bergamaschi, P, Bergmann, D, Blake, DR, Bruhwiler, L, Cameron-Smith, P, Castaldi, S, Chevallier, F, Feng, L, Fraser, A, Heimann, M, Hodson, EL, Houweling, S, Josse, B, Fraser, PJ, Krummel, PB, Lamarque, J-F, Langenfelds, RL, Le Quéré, C, Naik, V, O’Doherty, S, Palmer, PI, Pison, I, Plummer, D, Poulter, B, Prinn, RG, Rigby, M, Ringeval, B, Santini, M, Schmidt, M, Shindell, DT, Simpson, JJ, Spahni, R, Steele, LP, Strde, SA, Sudo, K, Szopa, S, van der Werf, GR, Voulgarakis, A, van Weele, M, Weiss, RF, Williams, JE and Zeng, G. 2013. Three decades of global methane sources and sinks. *Nat Geosci* **6**(10): 813–823. DOI: https://doi.org/10.1038/ngeo1955

Klausner, T, Roiger, A, Fiehn, A, Giez, A, Dreiling, V, Zöger, M and Mallaun, C. 2020. Aircraft measurement data obtained within the framework of the 3DO measurement campaign in summer 2018 over Berlin. DOI: https://doi.org/10.5281/zenodo.3706725

Kostinek, J, Roiger, A, Davis, KJ, Sweeney, C, DiGangi, JP, Choi, Y, Baier, B, Hase, F, Groß, J, Eckl, M, Klausner, T and Butz, A. 2019. Adaptation and performance assessment of a quantum and inter-band cascade laser spectrometer for simultaneous airborne in situ observation of CH4, C2H6, CO2, CO and N2O. *Atmos Meas Tech* **12**(3): 1767–1783. DOI: https://doi.org/10.5194/amt-12-1767-2019

Kuc, T, Rozanski, K, Zimnoch, M, Necki, JM and Korus, A. 2003. Anthropogenic emissions of CO2 and CH4 in an urban environment. *Appl Energ* **75**(3–4): 193–203. DOI: https://doi.org/10.1016/S0306-2619(03)00032-1

Kuonen, JPR, Visschedijk, AJH, Jozwicka, M and van der Denier Gon, HAC. 2014. TNO-MACC-II emission inventory, a multi-year (2003–2009) consistent high-resolution European emission inventory for air quality modelling. *Atmos Chem Phys* **14**(20): 10963–10976. DOI: https://doi.org/10.5194/acp-14-10963-2014

Kuhlmann, G, Broquet, G, Marshall, J, Clément, V, Löschner, A, Meijer, Y and Brunner, D. 2019b. Detectability of CO2 emission plumes of cities and...
power plants with the Copernicus Anthropogenic CO₂ Monitoring (CO₂M) mission. *Atmos Meas Tech* **12**: 6695–6719. DOI: https://doi.org/10.5194/amt-12-6695-2019

Kuhlmann, G, Clément, V, Marshall, J, Fuhrer, O, Broquet, G, Schnadt-Poberaj, C, Löschner, A, Meijer, Y and Brunner, D. 2019a. SMARTCARB – Use of Satellite Measurements of Auxiliary Reactive Trace Gases for Fossil Fuel Carbon Dioxide Emission Estimation, Final report of ESA study contract n°4000119599/16/NL/FF/rg. Tech. rep. Dübendorf, Switzerland: Empa, Swiss Federal Laboratories for Materials Science and Technology. Available at: https://www.empa.ch/documents/56101/617885/FR_Smartcarb_final_Jan2019.pdf (Accessed September 23, 2019).

LandGEM. 2005. Available at https://www.epa.gov/catic/clean-air-technology-center-products#software. Accessed August 18, 2019.

Liu, Z, He, C, Zhou, Y and Wu, J. 2014. How much of the world’s land has been urbanized, really? A hierarchical framework for avoiding confusion. *Landscape Ecol* **29**(5): 763–771. DOI: https://doi.org/10.1007/s10804-014-0034-y

Mahadevan, P, Wofsy, SC, Matross, DM, Xiao, X, Dunn, AL, Lin, JC, Gerbig, C, Munger, JW, Chow, VY and Gottlieb, EW. 2008. A satellite-based biosphere parameterization for net ecosystem CO₂ exchange: Vegetation Photosynthesis and Respiration Model (VPRM). *Global Biogeochem Cy* **22**(2). DOI: https://doi.org/10.1029/2006GB002735

Mallau, C, Giez, A and Baumann, R. 2015. Calibration of 3-D wind measurements on a single-engine research aircraft. *Atmos Meas Tech* **8**(8): 3177–3196. DOI: https://doi.org/10.5194/amt-8-3177-2015

Marcotullio, PJ, Sarzynski, A, Albrecht, J, Schulz, N and García, J. 2013. The geography of global urban greenhouse gas emissions: An exploratory analysis. *Climatic Change* **121**(4): 621–634. DOI: https://doi.org/10.1007/s10584-013-0977-z

Mays, KL, Shepson, PB, Stirm, BH, Karion, A, Sweeney, C and Gurney, KR. 2009. Aircraft-based measurements of the carbon footprint of Indianapolis. *Environ Sci Technol* **43**(20): 7816–7823. DOI: https://doi.org/10.1021/es901326b

McDonald, BC, McBride, ZC, Martin, EW and Harley, RA. 2014. High-resolution mapping of motor vehicle carbon dioxide emissions. *J Geophys Res-Atmos* **119**(9): 5283–5298. DOI: https://doi.org/10.1002/2013JD021219

Mertens, M and Jöckel, P. 2020. Data from MECO(n) model simulations on “Urban greenhouse gas emissions from the Berlin area: A case study using airborne CO₂ and CH₄ in situ observations in summer 2018”. DOI: https://doi.org/10.5281/zenodo.3707990

Mertens, M, Kerkweg, A, Jöckel P, Tost, H and Hofmann, C. 2016. The 1-way on-line coupled model system MECO(n) – Part 4: Chemical evaluation (based on MESSy v2.52). *Geosci Model Dev* **9**: 3545–3567. DOI: https://doi.org/10.5194/gmd-9-3545-2016

MLUL. 2017. Available at https://mlul.brandenburg.de/cms/media.php/lbm1.a.3310.de/Lagebericht-Abwasserbeseitigung2017.pdf. Accessed August 18, 2019.

Mueller, K, Yadav, V, Lopez-Coto, I, Karion, A, Gourdji, S, Martin, C and Whetstone, J. 2018. Siting Background Towers to Characterize Incoming Air for Urban Greenhouse Gas Estimation: A Case Study in the Washington, DC/Baltimore Area. *J Geophys Res Atmos* **123**(5): 2910–2926. DOI: https://doi.org/10.1002/2017JD027364

Nisbet, E and Weiss, R. 2010. Atmospheric science: Top-down versus bottom-up. *Science* **328**(5983): 1241–1243. DOI: https://doi.org/10.1126/science.1189936

OpenStreetMap. 2018. Available at http://download.geofabrik.de/europe/germany/berlin.html. Accessed October 02, 2019.

O’Shea, SJ, Allen, G, Fleming, ZL, Bauguitte, SJ-B, Percival, CJ, Gallagher, MW, Lee, J, Helfter, C and Nemitz, E. 2014. Area fluxes of carbon dioxide, methane, and carbon monoxide derived from airborne measurements around Greater London: A case study during summer 2012. *J Geophys Res Atmos* **119**(8): 4940–4952. DOI: https://doi.org/10.1002/2013JD021269

Peischl, J, Ryerson, TB, Aikin, KC, de Gouw, JA, Gilman, JB, Holloway, JS, Lerner, BM, Nadkarni, R, Neuman, JA, Nowak, JB, Trainer, M, Warnke, C and Parrish, DD. 2015. Quantifying atmospheric methane emissions from the Haynesville, Fayetteville, and northeastern Marcellus shale gas production regions. *J Geophys Res Atmos* **120**(5): 2119–2139. DOI: https://doi.org/10.1002/2014JD022697

Peischl, J, Ryerson, TB, Brioude, J, Aikin, KC, Andrews, AE, Atlas, E, Blake, D, Daube, BC, de Gouw, JA, Dlugokencky, E, Frost, GJ, Gentner, DR, Gilman, JB, Goldenstein, AH, Harley, RA, Holloway, JS, Kofler, J, Kuster, WC, Lang, PM, Novelli, PC, Santoni, GW, Trainer, M, Wofsy, SC and Parrish, DD. 2013. Quantifying sources of methane using light alkanes in the Los Angeles basin, California. *J Geophys Res Atmos* **118**(10): 4974–4990. DOI: https://doi.org/10.1002/2012JD023396

Pillai, D, Buchowitz, M, Gerbig, C, Koch, T, Reuter, M, Bovensmann, H, Marshall, J and Burrows, JP. 2016. Tracking city CO₂ emissions from space using a high-resolution inverse modelling approach: A case study for Berlin, Germany. *Atmos Chem Phys* **16**(15): 9591–9610. DOI: https://doi.org/10.5194/acp-16-9591-2016

Pitt, JR, Allen, G, Bauguitte, SJ-B, Gallagher, MW, Lee, JD, Drysdale, W, Nelson, B, Manning, AJ and Palmer, PI. 2019. Assessing London CO₂, CH₄ and CO emissions using aircraft measurements and dispersion modelling. *Atmos Chem Phys* **19**(13): 8931–8945. DOI: https://doi.org/10.5194/acp-19-8931-2019

Plant, G, Kort, EA, Floerchinger, C, Gvakharia, A, Vimont, I and Sweeney, C. 2019. Large fugitive
methane emissions from urban centers along the US East Coast. *Geophys Res Lett* 46(14): 8500–8507. DOI: https://doi.org/10.1029/2019GL082635

Ren, X, Salmon, OE, Hansford, JR, Ahn, D, Hall, D, Benish, SE, Stratton, PR, He, H, Sahu, S, Grimes, C, Heimbürger, AMF, Martin, CR, Cohen, MD, Stunder, B, Salawitch, RJ, Ehrman, SH, Shepson, PB and Dickerson, RR. 2018. Methane Emissions From the Baltimore-Washington Area Based on Airborne Observations: Comparison to Emissions Inventories. *J Geophys Res Atmos* 123(16): 8869–8882. DOI: https://doi.org/10.1002/2018JD028851

Rockel, B, Will, A and Hense, A. 2008. The regional climate model COSMO-CLM (CCLM). *Meteorol Z* 17: 347–348. DOI: https://doi.org/10.1127/0941-2948/2008/0309

Saunois, M, Bousquet, P, Poulter, B, Peregon, A, Ciais, P, Canadell, JG, Dlugokencky, EJ, Etiope, G, Bastviken, D, Houweling, S, Janssens-Maenhout, G, Tubiolo, FN, Castaldi, S, Jackson, RB, Alexe, M, Arora, VK, Beerling, DJ, Bergamaschi, P, Blake, DR, Brailsford, G, Browik, V, Bruhwiler, L, Crevoisier, C, Crill, P, Covey, K, Curry, C, Frankenberger, C, Gedney, N, Högland-Isaksson, L, Ishizawa, M, Itô, A, Joos, F, Kim, H-S, Kleinen, T, Krummel, P, Laitakari, J-P, Leitl, M, Ren, X, Salmon, OE, Hansford, CR, Chen, J-FW, Patra, PK, Peng, C, Peng, S, Peters, GP, Pison, I, Prigent, C, Prinn, R, Ramonet, M, Saito, M, Santini, M, Schroeder, R, Simpson, J, Spahni, R, Steele, P, Takizawa, A, Thornton, BF, Tian, H, Tohjima, Y, Watanabe, M, Yamashita, T, Yilmaz, M, Yoshida, Y, Zhang, B, Zhang, Z and Zhu, Q. 2016. The global methane budget 2000–2012. *Geophys Res Lett* 43(12): 6386–6394. DOI: https://doi.org/10.1002/2016GL068234

Saunois, M, Bousquet, P, Poulter, B, Peregon, A, Ciais, P, Canadell, JG, Dlugokencky, EJ, Etiope, G, Bastviken, D, Houweling, S, Janssens-Maenhout, G, Tubiolo, FN, Castaldi, S, Jackson, RB, Alexe, M, Arora, VK, Beerling, DJ, Bergamaschi, P, Blake, DR, Brailsford, G, Brovkin, V, Bruhwiler, L, Crevoisier, C, Crill, P, Covey, K, Curry, C, Frankenberger, C, Gedney, N, Högland-Isaksson, L, Ishizawa, M, Itô, A, Joos, F, Kim, H-S, Kleinen, T, Krummel, P, Laitakari, J-P, Leitl, M, Ren, X, Salmon, OE, Hansford, CR, Chen, J-FW, Patra, PK, Peng, C, Peng, S, Peters, GP, Pison, I, Prigent, C, Prinn, R, Ramonet, M, Saito, M, Santini, M, Schroeder, R, Simpson, J, Spahni, R, Steele, P, Takizawa, A, Thornton, BF, Tian, H, Tohjima, Y, Watanabe, M, Yamashita, T, Yilmaz, M, Yoshida, Y, Zhang, B, Zhang, Z and Zhu, Q. 2017. Variability and quasi-decadal changes in the methane budget over the period 2000–2012. *Atmos Chem Phys* 17(18): 11135–11161. DOI: https://doi.org/10.5194/acp-17-11135-2017

Scherer, D, Ament, F, Emeis, S, Fehrenbach, U, Leitl, B, Scherber, K, Schneider, C and Vogt, U. 2019a. Three-Dimensional Observation of Atmospheric Processes in Cities. *Meteorol Z* 28(2): 121–138. DOI: https://doi.org/10.1127/metz/2019/0911

Scherer, D, Antretter, F, Bender, S, Cortekar, J, Emeis, S, Fehrenbach, U, Gross, G, Halbig, H, Hasse, J, Maronga, B, Raasch, S and Scherber, K. 2019b. Urban Climate Under Change [UC] – A National Research Programme for Developing a Building-Resolving Atmospheric Model for Entire City Regions. *Meteorol Z* 28(2): 95–104. DOI: https://doi.org/10.1127/metz/2019/0913

Statistisches Bundesamt. 2018. Available at www.destatis.de/DE/Themen/Laender-Regionen/Regionales/Gemeindeverzeichnis/Administrativ/05-staedte.html. Accessed September 25, 2019.

Stein, AF, Draxler, RR, Rolph, GD, Stunder, BJ, Cohen, MD and Ngan, F. 2015. NOAA’s HYSPLIT atmospheric transport and dispersion modeling system. *Bull Amer Meteor Soc* 96: 2059–2077. DOI: https://doi.org/10.1175/BAMS-D-14-00110.1

Stohl, A. 1998. Computation, accuracy and applications of trajectories – A review and bibliography. *Atmospheric Environment* 32(6): 947–966. DOI: https://doi.org/10.1016/S1352-2310(97)00457-3

Strukturatlas Land Brandenburg. 2017. Plot received from an interactive tool on http://strukturatlas.brandenburg.de/. Accessed January 22, 2020.

Turnbull, JC, Karion, A, Davis, K, Lauvaux, T, Miles, NL, Richardson, SJ, Sweeney, C, McKain, K, Lehman, SJ, Gurney, KR, Patarasuk, R, Liang J, Shepson, PB, Heimbürger, A, Harvey, R and Whetstone, J. 2019. Synthesis of Urban CO2 Emission Estimates from Multiple Methods from the Indianapolis Flux Project (INFLUX). *Environmental science & technology* 53(1): 287–295. DOI: https://doi.org/10.1021/acs.est.8b05552

UN. 2018. World Urbanization Prospects 2018. Available at https://population.un.org/wup/Publications/. Accessed September 25, 2019.

UNFCCC. 2015. Paris Agreement, FCCC/CP/2015/L.9/Rev.1. Available at https://unfccc.int/resource/docs/2015/cop21/eng/l09r01.pdf. Accessed September 25, 2019.

Zhao, CL and Tans, PP. 2006. Estimating uncertainty of the WMO mole fraction scale for carbon dioxide in air. *J Geophys Res* 111(D8): 81. DOI: https://doi.org/10.1029/2005JD006003

Zimnoch, M, Necki, J, Chmura, L, Jasek, A, Jelen, D, Gałkowski, M, Kuc, T, Gorczyca, Z, Bartyzel, J and Rozanski, K. 2019. Quantification of carbon dioxide and methane emissions in urban areas: Source apportionment based on atmospheric observations. *Mitig Adapt Strat Gl* 24(6): 1051–1071. DOI: https://doi.org/10.1007/s11027-018-9821-0
