Second reply to interactive comments

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1 Introduction

Thank you for the review of the revised manuscript.

2 Answers to RC1

As preface: The updated supplement seems to not have been referenced strongly enough. It is hosted on zenodo to make it easier to find and reference directly as data or code.

It is and has been available at https://doi.org/10.5281/zenodo.3395421 There is an updated version which re-organizes the folder structure such that the most important files are shown first: https://doi.org/10.5281/zenodo.3397464

However the link to it was only at two places in the manuscript while the other places just referenced it as auxiliary material. This is changed to be more obvious. Now every section that references the auxiliary material includes a reference to the zenodo-upload with DOI.

2.1 Answers to individual comments

“*The authors made a nice start towards going through my numerous initial comments. However, some of these were only glanced over or skipped - especially some of the more scientifically important comments (highlighted below with **). Sometimes good responses were given, but then changes were not reflected in the manuscript. In addition the updated manuscript and auxiliary material do not always match what is in the responses. Again, I will reiterate my initial review that I think this is a fair start to a difficult problem but needs work especially on comments I already made.*”

“*Please recheck:*”

“*2) The changes in the response do not match the changes in the manuscript.*”

The factor two was adjusted to 1.7 after fixing the error found thanks to your comments. I’m sorry that I missed copying this change into the reply.

“*There are 2 uncertainties on the fluxes, please provide a short description why in the abstract so people do not need to read the full article.*”

This is provided now.

"*I think most global inventory (e.g., ODIAC) developers would generally agree that comparing global inventories with local flux estimates is generally not an ideal comparison.*"

In the general case I would agree, but in the specific case, this is the best data available, and while this is a local flux estimate, it is averaged over 4 years, so it it is more fit for comparison with a global inventory than for example a short-term measurement campaign.

"*Pisso et al. (2019, doi: 10.1186/s13021-019-0118-8) should be referenced somewhere in this paper and compared with (does not need to be the abstract).*"

Thank you for the reference![1]

Pisso et al. estimate the emissions of Tokyo City as 22 MtC/y (80 $MtCO_2$/y) and of the Tokyo metropolitan area as 151 MtC/y (554$MtCO_2$/y). With $69 \pm 21 \pm 6$ MtC/y ($253 \pm 77 \pm 22$ $MtCO_2$) this study lies between those values, which is to be expected because the definition of the metropolitan area used in Pisso et al. includes all the fluxes from the prefectures Kanagawa, Saitama, and Chiba, while this study only includes part of these fluxes. Pisso et al. also compare against several other datasets using their source definition.

This comparison is now included in the manuscript. While they include uncertainties in the figure, these are not given in the table, therefore I leave them out in the comparison in this manuscript. It is encouraging to see that the uncertainties found in this work are larger but in a similar order of magnitude as those found by Pisso et al. running a full inversion model with priors. This shows that there is value in simpler calculations based purely on direct measurements.

"*The authors should include a caveat in the abstract that matches 2.1 in the response. Namely this study was not designed to have the best accuracy using all possible measurements, but rather is just a starting estimate from 1 ground site and should not be interpreted as being unbiased with a high accuracy (e.g., for policy makers).*"

This is now added:

The goal of this study is not to calculate the best possible estimate of CO_2 emissions, but to describe a simple method which can be replicated easily and uses only observation-data.

"*3) Again, I don't know why Frey et al. 2015 (instrumental), Chen at al. 2016 (CH4 not CO2), and Butz et al. 2016 (not fossil fuel) are included as references here. If the authors insist on including these only marginally related citations, they should put them in a different location, for example in the conclusions section where mobile spectrometers are mentioned. In that case the authors may also consider citing Luther et al., 2019 (CH4 from coal mines, doi: 10.5194/amt-12-5217-2019), Frey et al. 2019 (also instrumental, doi: 10.5194/amt-12-1513-2019), Viatte et al. 2017 (similar to Chen, but used a different inversion method doi: 10.5194/acp-17-7509-2017).*"

With Frey et al. 2019 in the outlook, the initial Frey et al. 2015 is no longer required for reference of the instruments, so it is removed. Viatte and Luther are now added as references of differential measurements.

The method shown here is not limited to CO_2, and there is a strong CH_4 signal near Tokyo, too, but that is out of scope for this manuscript.

"*5) I still don't think "inexpensive" is a word that should be used here. Smaller satellites can have a cost of less than CAD 10 million (e.g., GHGSat Claire: https://spectrum.ieee.org/energy/environment/private-satellite-to-track-carbon-polluters). Besides, the scope of satellite missions and ground-based missions is often very different which makes a simple cost comparison difficult or not particularly useful.*"

It is clear that the scope of satellite missions and ground-based missions is different. Satellites can measure in regions unreachable by ground-based measurements, and they provide comparable global coverage.

However satellites are much harder to calibrate and have lower precision, while with mobile spectrometers a network of ground-based sites can be kept in calibration.

For 10 million CAD it would be possible to install more than 50 ground-based sites which could then operate for over 20 years while the typical lifetime of a satellite is still around 5 years. So even with the cheaper satellite, there is a large gap in the cost.

Satellites cannot replace ground-based measurements, just like ground-based measurements cannot replace satellites. Omitting these measurements limits the precision that CO_2 emission calculations can achieve.

"*12) I still think it strange to say the granularity around the end/beginning of the years where there is a sharp dip is the same as everywhere else in the fits.*"

That's why it's written as "roughly". The reason for adding the granularity is to make this more approachable to people used to granularity as measure.

"*13) a) It seems perhaps the original supplement was uploaded again, so I am unable to see the results of the ccgfilt. To me as a reviewer the current method with the multiple steps is actually more convoluted.*"

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[1] Since my contract ended in 2017 I cannot follow newer publications very well, so I'm happy to get information about more recent publications. That is also why this discussion phase is taking so long: I have to fit scientific work between a full-time job and family.

See the zenodo link in the preface.

"*b) **Degrees of polynomials – but why 6? Why not 4, or 8? Or 15? I'm guessing you had some sort of adjusted R^2 you analyzed to reduce overfitting? Or was it more just qualitative as it sounds on page 5 of the manuscript? The beginning/end of year looks over fit.*"

It was qualitative, checked over all the TCCON sites, as described in the manuscript. With lower degrees the residuals were far bigger, with higher degrees the overfitting became worse.

"*c) **Could you comment on the very low R^2 for the diurnal fit in the manuscript?*"

The R^2 is low, because there is very high daytime independent varibility. The difference between air from Tokyo und from background directions is much higher than the difference between morning and noon.

"*d) **The claim that both "background" and "enhancements" can be measured from one site is one of the key requirements of this study. If the background values are not in any sort of agreement with those more removed from Tokyo (but at a similar latitude and not too far away) then the flux bias could be quite large.*"

The background values are calculated from two distinct wind directions, one where air is transported over the mountains and one where air is transported over the ocean. Compared to the enhancements from Tokyo area, the median values of these measurements are in good agreement.

"*19) I still think it would be nice for the reader to not have to turn to the auxiliary material to see the model used was GDAS 1 degree, and starting heights varied from 20-30 meters. That way those familiar with HYSPLIT will know it is just a basic estimate (coarse grid for scale of interest, and trying to model winds close to the ground), hence why the radiosondes were critical.*"

I agree that it could help to illustrate why HYSPLIT was used, but I also think that the added complexity in the text would outweight the advantages. The HISPLIT runs are used to verify that the profile height of radiosonde data suffices for these calculations. They do not enter the calculations otherwise.

"*24) So low outliers are not clipped to the left? I do not understand why having a greater focus between 0 and 1 is less valid. If a linear scale is insisted upon, why not make the ratio the opposite way?*"

Low outliers are not clipped: the scaling factors cannot be lower than zero. I did not say that having a greater focus between 0 and 1 is less valid, just that we had to choose between different representations and this is the one which is the easiest to understand. The goal of this manuscript is to show a simple calculation which is easy to re-implement. Part of this goal is to make plots easy to understand for people without scientifc background.

The median lines in the plots show an unbiased estimate of the wind scaling.

To avoid making either high or low outliers more visible in the plots, we could divide the values at each height by the median, but doing so would make the plot much harder to understand, which would undermine the purpose of this manuscript.

"*25) **This is better, but there are still major issues. a) Please include a citation for your chosen notation. It seems almost arbitrary. See for example Chen et al., 2016.*"

While I agree that this would be nice, it is not feasible at this stage.

"*b) Again, I'm concerned that units were not thoroughly checked or at least labelled. I may have been off on page numbering, but I don't think that's an excuse for units not working out or at least not matching. Units are not listed for all terms in the table. Please use kg instead of g for mass.*"

Units are now listed for all terms. g is used instead of kg to unify the units starting from the molar mass (which is given in g/mol).

"*c) The 0.9975 factor from Bannon needs more of an explanation in this manuscript.*"

added "to adjust for curved geometries".

"*d) TCCON measuring a dry-air mole fraction was precisely my point. By not including a water correction term, the "unit air column mass" is now based on a whole or "wet" air fraction rather than a dry one, which will bias "CO2 column mass." See Appendix A in Wunch et al. 2011 (doi: 10.5194/acp-11-12317-2011).*"

You are right: I missed converting the pressure from wet air to dry air, as written above. This is now corrected: It caused underestimation of the emissions by about 0.2%. The correction is visible in the diagrams.

"*e) If you insist on using different units, why not also show the actual plots where you calculated "R"? Do you do the subtraction in ppm or in kg/m2? What is written in the text still does not agree with what is in the table.*"

The actual plots are shown, but they directly calculate $\hat{\Delta}$ which is then converted to $E_m\mathcal{A}$ as described in the text.

"*26) **No response?*"

This got lost during answering. I'm sorry for that. See the answer to 25d. Thank you for persisting!

"*26. p 8, line 9: What is the importance of the "perpendicular spread"? Generally, I would think of estimating fluxes using the wind speed divided by the transect across the emitting region to get a residence time (e.g, Eq. 2 in Viatte et al, 2017 doi: 10.5194/acp- 17-7509-2017) multiplied by the total region area assuming constant emissions. How do you reconcile this difference?*"

The perpendicular spread is just the total area divided by the transect.

“*28) **This choice of location seems somewhat arbitrary. Not to mention the calculations seem very sensitive to this arbitrary choice.*”

The choice of location is somewhat arbitrary, because the actual distribution of emissions is not known. This is reflected in the uncertainty estimate by assuming an uncertainty of 10km for the distance.

“*29) This looks like actually almost no change was made at all. I don’t see the necessity of dedicating 3 equations to a unit conversion.*”

The flow of equations was inverted and more parts were moved into the table of units to make them easier to follow.

“*30) ODIAC is on a 30 arc second grid (see page 545 of Oda et al. 2011 doi: 10.5194/acp-11-543-2011), not a perfect distance grid. To reduce any possibility of misinterpretation you could you specify these are angular degrees.*”

This is noted now in the caption of the graph.

“*31) Why is this response not incorporated into the manuscript? Can you show the TIMES grids selected on a map?*”

Because this was already present in the text. I now added the explicit information about angular degrees in the caption of Figure 7.

“*32) But as you have admitted, the data you work with only makes measurements during daytime hours. This biases the result if daytime emissions are different from nighttime (which they almost certainly are).*”

Yes, and this is described at the end of the section about uncertainty estimation. But there are no direct measurements from the area that we could use, therefore this is not taken as the final estimate.

“*33) a) Accuracy of background?*”

This is now noted as a potential source of uncertainty but not added, because it is small compared to the uncertainty due to the scatter of the data. A systematic bias could be due to forests in background locations, but this is small compared to the emissions of Tokyo and reduced further, because the wind travels only a limited distance over land. For sites with a large forest in one direction but none close to the city, this would have to be taken into account.

This is now noted.

“*b) Why didn’t the rest of this discussion make it into the manuscript?*”

Because it does not provide added uncertainty. I have now added the answer to the discussion of uncertainties.

“*34) What about #2?*”

2 is sampled as shown in figure 3, therefore it is not repeated. I now added an explicit reference to figure 3.

“*39) If this does not yet exist, then this claim should not yet be here. A better citation may be Frey et al., 2019 (doi: 10.5194/amt-12-1513-2019) which at least has a 3.5 year dataset.*”

I switched to that publication.

“*The supplement does not actually appear to be updated.*”

As noted in the preface, the supplement was updated. It just moved to another hoster, so it was not uploaded in the copernicus interface.

3 Answers to RC2

Thank you for also reviewing the updated version of our manuscript!

“*Review for Net CO2 Fossil Fuel Emissions of Tokyo estimated directly from measurements of the Tsukuba TCCON site and radiosondes” by Babenhauserheide, A, Hase, F. and Morino, I.*”

“*The manuscript presents an approximation method for determining greenhouse gas emissions from large cities close to a TCCON measurement station. A general algorithm is developed for Tokyo that the authors suggest can be applied to other cities. Comments from the first review round have been adequately addressed. In particular, the methodology description section has greatly improved. Also, the expanded conclusion section now has a stronger connection to the rest of the paper. I have some very minor additional comments below, to be addressed prior to the manuscript being accepted for publication.*”

“*MinorComments: 1. Pg 7, ln 1-3: I disagree that only enhanced concentrations are seen between 5 and 15 m/s between 170º and 240º. The left plot of Figure 3 shows negative* Δ*XCO2 of between -0.75 and -1.00 ppm. Additionally, Pg 7, line 1 explains that both positive and negative residuals are used. Figure 5 also shows negative and positive enhancements in the "Tokyo" influenced directions. Please clarify what is meant by "only bins with an enhanced concentration" means.*”

This is now more precise:

Within these directional delimiters, all bins in the interval with wind speeds between 5 and 15 ms^{-1} contains enhanced concentrations of CO$_2$

“*2. Figure 4: Please separate out the wind speed and wind direction plots into two plots 4 (a) and 4 (b). Also, the caption mentions 1000 m altitude, so please also show the altitude in meters as well as pressure levels.*”

The graph is now split into two images.

The ground-pressure changes with every sonde, so the height cannot be given together with pressure in this aggregated graph.

"*3. I am confused about the height relating to the average column wind speed. On pg 10, ln 2, it states (0-2000 m). However, Pg7, ln 24 and ln 31 says the upper limit is 1000 m. Please clarify which it is or why it changes?*"

It is 1000m; this change was missed. It is now adjusted. Thank you!

"*Technical Corrections: 1. Pg 9, ln 7: apriori -> a priori*"

Fixed.

"*2. Pg 11: Change* A_{aff} *in line 1 and Eq 5.*"

Also fixed, thank you!

References

Net CO_2 Fossil Fuel Emissions of Tokyo estimated directly from measurements of the Tsukuba TCCON site and radiosondes

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Abstract. We present a simple statistical approach for estimating the greenhouse gas emissions of large cities using accurate long-term data of column-averaged greenhouse gas abundances collected by a nearby FTIR (Fourier Transform InfraRed) spectrometer. This approach is then used to estimate carbon dioxide emissions from Tokyo.

FTIR measurements by the Total Carbon Column Observing Network (TCCON) derive gas abundances by quantitative spectral analysis of molecular absorption bands observed in near-infrared solar absorption spectra. Consequently these measurements only include daytime data.

The emissions of Tokyo are derived by binning measurements according to wind direction and subtracting measurements of wind fields from outside Tokyo area from measurements of wind fields from inside Tokyo area.

We estimate the average yearly carbon dioxide emissions from the area of Tokyo to be ~~$69 \pm 21 \pm 6 \frac{MtC}{\text{year}}$~~ $70 \pm 21 \pm 6 \frac{MtC}{\text{year}}$ between 2011 and 2016, calculated using only measurements from the TCCON site in Tsukuba (north-east of Tokyo) and wind-speed data from nearby radiosondes at Tateno. The uncertainties are estimated from the distribution of values and uncertainties of parameters (±21) and from the differences between fitting residuals with polynomials or with sines and cosines (±6).

Our estimates are factor 1.7 higher than estimates using the Open-Data Inventory for Anthropogenic Carbon dioxide emission inventory (ODIAC), but when results are scaled by the expected daily cycle of emissions, measurements simulated from ODIAC data are within the uncertainty of our results.

The goal of this study is not to calculate the best possible estimate of CO_2 emissions, but to describe a simple method which can be replicated easily and uses only observation-data.

1 Introduction

Anthropogenic emissions of carbon dioxide are the strongest long-term control on global climate (Collins et al., 2013, Figure 12.3, page 1046), and the Paris agreement “recognizes the important role of providing incentives for emission reduction activities, including tools such as domestic policies and carbon pricing“ (UNFCCC secretariat, 2015). Implementing carbon pricing policies is widely regarded as an effective tool for reducing emissions. Such measures also motivate the development of new approaches for accurate measurements of carbon emissions (Kunreuther et al., 2014, ch. 2.6.4 and 2.6.5, pp. 181ff).

The carbon dioxide footprint of large scale fossil fuel burning emitters like power plants or heating and personal transport in mega cities ~~,~~ has been retrieved from satellite ~~(Hakkarainen et al., 2016; Hammerling et al., 2012; Ichii et al., 2017; Deng et al., 2014; Nass~~ (Hakkarainen et al., 2016; Hammerling et al., 2012; Ichii et al., 2017; Deng et al., 2014; Nassar et al., 2017; Hedelius et al., 2018) and from ground based differential measurements using multiple mobile total column instruments ~~(Frey et al., 2015; Hase et al., 2015; Che~~ (Hase et al., 2015; Chen et al., 2016; Butz et al., 2017; Viatte et al., 2017; Vogel et al., 2019; Luther et al., 2019). Inverse modelling allows coupling in-situ measurements (which only capture enhancements in mixing ratio close to the ground) with atmospheric transport for similar investigations ~~(e.g. van der Laan-Luijkx et al., 2017; Basu et al., 2011; Babenhauserheide et al., 2015; van der~~ (e.g. Basu et al., 2011; Meesters et al., 2012; van der Velde et al., 2014b; Babenhauserheide et al., 2015; van der Laan-Luijkx et al., 2017) , but due to short mission times of satellites and differential measurement campaigns and high uncertainties when using in-situ data, long term changes in emissions are typically derived from economic fossil fuel and energy consumption data ~~(e.g. Andres et al., 2011; Bureau of the Environment Tokyo, 2010; van der Velde et al., 2014a; Le Quéré et al., 2015, 2016)~~(e.g. Bureau of .

The Total Carbon Column Observing Network ~~(TCCON, Wunch et al., 2011; Toon et al., 2009)~~(TCCON, Toon et al., 2009; Wunch et al , described in section 2, provides highly accurate and precise total column measurements of carbon dioxide mixing ratios with multi-year records of consistently derived data.

The aim of our study is to provide an estimate of the ~~Tokyo~~ CO_2 emissions of Tokyo, Japan, by correlating measured XCO_2 with wind speed and direction, resulting in a measurement-driven approach to derive the annual carbon dioxide emissions of Tokyo city (Japan). The main emission sources of Tokyo are Transport, Residential and industry, with about half the emissions coming from the large coal and gas fired power plants on the east side of Tokyo Bay, south-east of Tokyo city (Bureau of the Environment Tokyo, 2010). The quality of the data and long time series of available data enables inferring fluxes from the measurements by statistical matching of measurements to wind directions without being dominated by measurement noise. We use four years of measurements by the TCCON site at Tsukuba, Japan, along with radiosonde measurements of daily local wind profiles.

This method provides an approach to estimate city emissions which is inexpensive when compared to satellite missions while being easy to reproduce and to establish, and is suitable for long-term monitoring. Compared to still cheaper in-situ measurements it gives the advantage of directly measuring all emissions from a city in the air column, while ground-based in-situ measurements only capture emissions in the lowermost part of the air profile.

This publication shows that emissions can be estimated from four years of data. Continued measurements will allow tracking the change in emissions.

2 Observations

The column data from TCCON currently provides the most precise and accurate remote-sensing measurements of the column averaged CO_2 abundances. The average station-to-station bias is less than 0.3 ppm (Messerschmidt et al., 2010).

The stations of the TCCON-network measure the absorption of CO_2 and other molecular species using the sun as background radiation source (Wunch et al., 2011). Dividing the retrieved column amount of the target species by the co-observed column amount of O_2 yields a pressure-independent measure for the concentration of carbon dioxide in the dry atmospheric column (XCO_2). The precision of these measurements is better than 0.1% by ~~Messerschmidt et al. (2011)~~(Messerschmidt et al., 2011) . Since our study restricts itself to a single station and the uncertainty budget is dominated by other factors, we can ignore any potential minor calibration bias of the selected station or the whole network.

~~our~~ Our study uses the current dataset of column-averaged carbon dioxide abundances generated with GGG2014 from solar absorption spectra recorded at the Tsukuba TCCON station, Japan (Ohyama et al., 2009; Morino et al., 2016). Publicly available data from Tsukuba at the TCCON data site used in our study (referenced from section 8, *code and data availability*) extends from 2011-08-04 to 2016-03-30. The coordinates of the Tsukuba TCCON site are 140.12° East and 36.05° North, the altitude is 31m. Further information about the TCCON site in Tsukuba is available from the TCCON wiki. [1] In addition to concentration data of trace gases, the station provides wind direction and speed measured at the rooftop of the observatory.

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[1] The Tsukuba site of the TCCON wiki is located at https://tccon-wiki.caltech.edu/Sites/Tsukuba

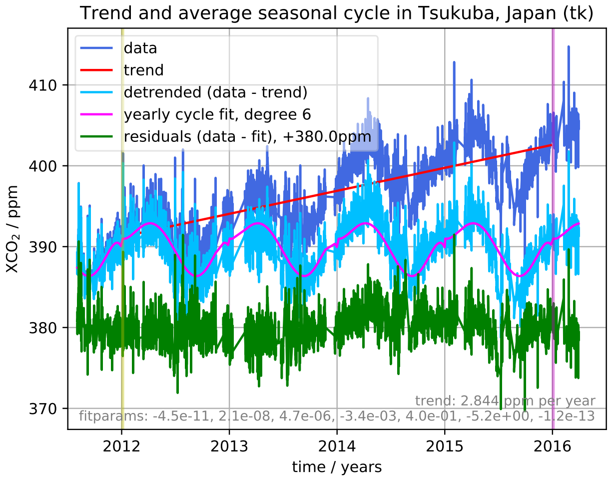


Figure 1. Detrending and deseasonalization of the XCO_2 total column measurements in Tsukuba, Japan. The data shown are individual measurements. The trend (shown as a red “trend” line) is removed with a linear least squares fit to the data from background directions between 2012-01-01 and 2016-01-01 (denoted by the yellow and magenta vertical lines), the seasonal cycle from the signal due to photosynthesis, respiration and decay (shown as “yearly cycle fit, degree 6”) is removed by fitting a polynomial of degree 6 to the combined yearly cycles of the detrended data. Degree 6 was chosen empirically to minimize structure in the residuum over all TCCON sites.

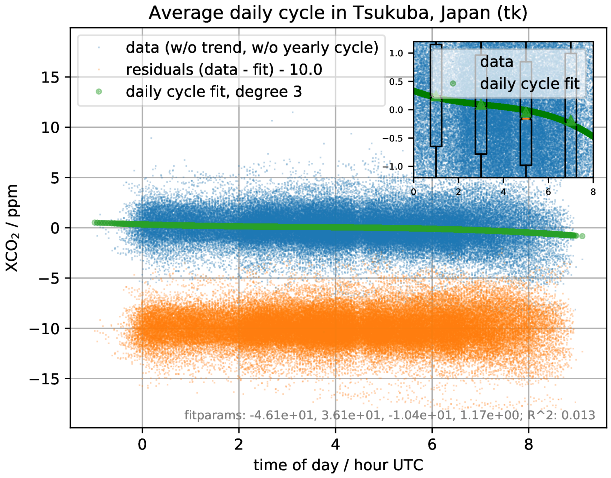


Figure 2. To remove a potential bias from correlation of wind direction with daytime which would couple in the signal from photosynthesis and respiration, the daily cycle is removed by fitting and subtracting a polynomial of degree 3. Degree 3 was chosen empirically. The fit has low R^2, because the data is dominated by noise.

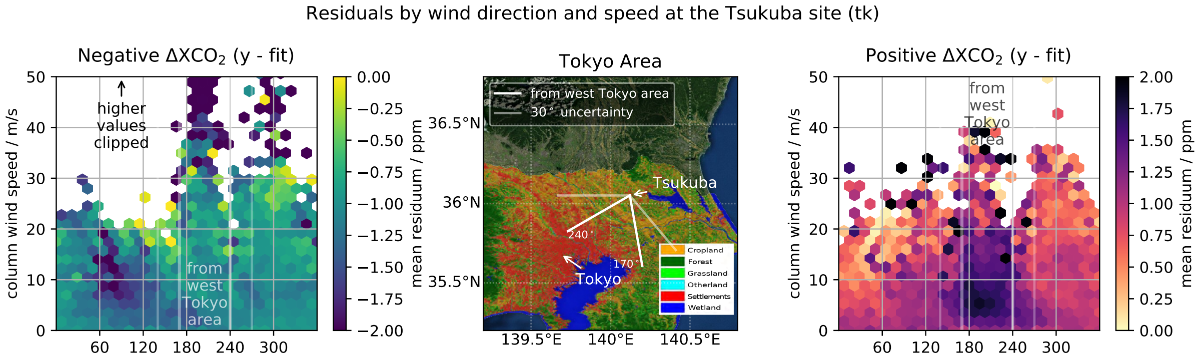
3 Removing trend and natural cycles

The approach chosen in this paper to estimate the CO_2 emissions of Tokyo is to separate CO_2 measurements by the wind direction for which they were measured. To make measurements from different wind directions comparable, they must be made accessible to ~~a~~ simple statistical analysis, therefore the first step is to remove trends as well as yearly and daily cycles.

Column averaged atmospheric CO_2 abundances are dominated by seasonal variations and a yearly rise of about 2.0 ppm per year (Hartmann et al., 2013, page 167 in section 2.2.1.1.1). Additionally there is an average daily cycle of about 0.3 ppm in the densely measured daytime between 2:00 UTC and 7:00 UTC (local time between 11:00 and 16:00 GMT+9). To allow direct comparisons of values from different times of year and times of day, these cycles are removed by fitting and subtracting polynomials from the data: linear for the trend, degree 6 for the yearly cycle (roughly equivalent to bi-monthly granularity) and degree 3 (roughly 3-hour granularity) for the daily cycle.

Polynomials are used in this estimation to make the method as easy to implement as possible. Section 2 of the auxiliary material of our study (Babenhauserheide et al., 2020) provides results from an alternate implementation using harmonics instead which gives comparable results.

The trend is fitted against measurements from background directions, but the yearly and daily cycle is fitted against measurements from all directions, so the fitting might remove a certain amount of the actual annual and daily cycle of emissions. However, the impact of this fitting in final estimates is limited to wind directions correlated with the cycle, since uncorrelated differences get reduced in statistical aggregation. Such a correlation between wind direction and the time of day exists, but mainly outside the densely measured daytime; a graph verifying this and the programs applied for the data analysis are available in section 1 of the auxiliary material of our study. Fitting the yearly cycle only against background directions creates artifacts, therefore this was avoided. Figure 1 and 2 show the fits and residuals resulting from the process. The calculations only use data provided directly from the TCCON network. The degrees of the fits were chosen empirically (by manual adjustment) to minimize the residuals over data from all TCCON sites available in 2016: polynomial fits with degrees between 3 and 9 were tested for the yearly cycle and the residuals checked for all TCCON sites. Higher degrees than 6 increased artifacts, lower degrees increased the overall size of residuals.



Surface type overlay from Bagan 2014, doi:10.1088/1748-9326/9/6/064015

Figure 3. The **center graph** shows a map of Tokyo and its surroundings, retrieved from ~~the~~ an ArcGIS REST service[2] with an overlay indicating the surface type. The colors in the overlay visualize land-use and settlement density (taken from Bagan and Yamagata, 2014). It clearly shows decreasing population density with distance from Tokyo city, along with the long tail of Tokyo settlements towards the north west. Close by Tokyo Bay in the lower center of the map, at the south east perimeter of Tokyo and on the opposite shore, there are multiple coal and gas power plants. Tokyo city center and the position of the TCCON site in Tsukuba are marked along with white lines which define an opening angle for incoming wind at Tsukuba which is interpreted as coming from Tokyo Area, along with additional widening by 30° as estimate of the actual origin of transported CO_2 arriving at Tsukuba from the given wind direction. These white lines denoting the incoming wind angle limits are reproduced in the **right graph** and the **left graph** as delimiting directions in which the wind blows from west Tokyo area. The **right** graph shows the **positive** half of the residuals from Figure 2, binned by wind direction and -strength. The color represents the mean value of the positive residuals within the bin. The **left** graph shows the **negative** half of the residuals from Figure 2, binned by wind direction and -strength. The color represents the mean value of the negative residuals within the bin. The black arrow at the upper edge of the left graph indicates that values for wind speeds above 50 m/s have been left out to focus on the area between 5 and 15 m/s used in the later evaluation. Displaying residuals which are lower than zero in a different graph than residuals which are higher than zero aids visual detection of emissions, because it separates the features of CO_2 sinks (lower than zero) from CO_2 sources (higher than zero). The strongly negative values on the left graph at a wind direction around 60° might be due to biospheric drawdown of CO_2 by woodland, but since the focus of this publication are the emissions from Tokyo, those values will not be evaluated further here. The split of the dataset applied here is purely for visualization: in the following calculations and graphs, negative and positive residuals are used together.

4 Directional dependence of remaining differences

To calculate the carbon source of Tokyo, the residuals generated by applying the procedures described in section 3 are binned by wind direction and speed, as shown in Figure 3. A major source of uncertainty in this endeavor is the actual extent of Tokyo in wind directions. This extent was chosen as 170° to 240° as seen from the TCCON site in Tsukuba, following the hexbin averages shown in the right panel in Figure 3. The data in Figure 3 is separated into positive and negative to ease identification

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[2] retrieved as EPSG:4301 using a ESRI_Imagery_World_2D request to server.arcgisonline.com/ArcGIS via the basemap library in matplotlib (Hunter, 2007) as described at basemaptutorial.readthedocs.io/en/latest/backgrounds.html#arcgisimage. Used with permission (Permission for publication of this graph under creativecommons attribution license granted by Esri). Copyright (c) (2017) Esri, ArcGIS. All rights reserved.

of the limits for emissions from Tokyo area. The quantitative evaluation uses both positive and negative residuals. Within these directional delimiters, all bins in the interval with wind speeds between 5 and 15 ms^{-1} ~~contains only bins with an enhanced concentration~~ contain enhanced concentrations of CO$_2$. Perfect definition of these limits is not possible in the ~~Scheme~~ scheme presented here, because the area can only be delimited orthogonal to the wind direction measured in Tsukuba. In parallel direction the only limit are changes in wind direction over time: if wind speed is low enough that on average a direction change occurs before the air reaches Tsukuba, then concentration measurements from background locations and from Tokyo average out. This is indicated by the weaker enhancement seen for wind speeds below 5 ms^{-1}.

Using ΔXCO$_2$, a measure proportional to the carbon dioxide column enhancement, and the effective wind speed ascribed to these enhancements from the direction of Tokyo allows estimating the emission source of Tokyo (described further in section 5). However, the directly measured wind speed which is provided by the TCCON network only provides an approximate indication of the effective wind speed and direction in the altitude range carrying the enhanced carbon dioxide (similar to the effects discussed by Chen et al., 2016, for differential measurements of the emissions). For this study, the required effective wind speed is estimated from radiosonde data.

Effective wind speed

The wind speed at the station is measured close to the ground. The effective speed of the air column however depends on the wind speed higher up in the atmosphere. Estimating the wind speed of air with enhanced carbon dioxide concentrations due to emissions from Tokyo therefore requires taking the difference in height of the measured concentrations and the measured wind speed into account. To this end, the ground wind speed v can be replaced by the density weighted average wind speed profile within the boundary layer. To calculate the required altitude extension of this profile, forward trajectories from Tokyo for 5 to 15 hours were calculated with the HYbrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT, Stein et al., 2015) using the Real-time Environmental Applications and Display sYstem (READY, Rolph et al., 2017), accessed via the HYSPLIT-WEB online service from NOAA[3] as described in the auxiliary material (Babenhauserheide et al., 2020). Since the calculations in this publication only use data from measurements with wind speeds of at least $5ms^{-1}$, 5 hours suffice for all Trajectories originating in Tokyo to reach Tsukuba. All the parameters used are contained in the graphs in section 5 of the auxiliary material. The HYSPLIT profiles show that most air parcels from Tokyo arriving at Tsukuba are contained within the lowest 1000m of the atmosphere. Therefore calculating the effective ~~air~~ wind speed of the column with enhanced concentrations only requires wind speed measurements in this part of the atmosphere.

Direct measurements of the wind speed profile ~~is~~ are available from radiosondes. ~~Specifically, radiosonde~~ Radiosonde data from Tateno, Japan, Prefecture Ibaraki, Latitude 36.06° N, Longitude 140.13° E, Altitude 27 m, situated close to Tsukuba station, provides 7 years of measurements from 2009 to 2016. The data was retrieved from the Atmospheric Soundings site at University of Wyoming (http://weather.uwyo.edu/upperair/sounding.html). One example of these data sets is by Ijima (2016). Further details are available in the auxiliary material, provided as Babenhauserheide et al. (2020).

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[3] The HYSPLIT-WEB online service is available at https://ready.arl.noaa.gov/HYSPLIT.php.

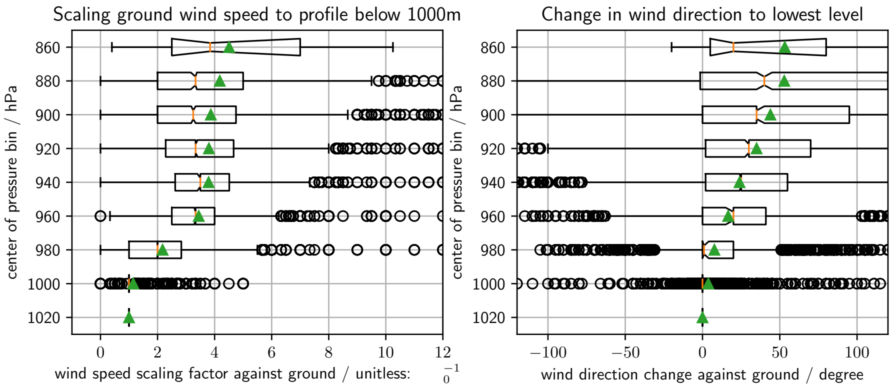
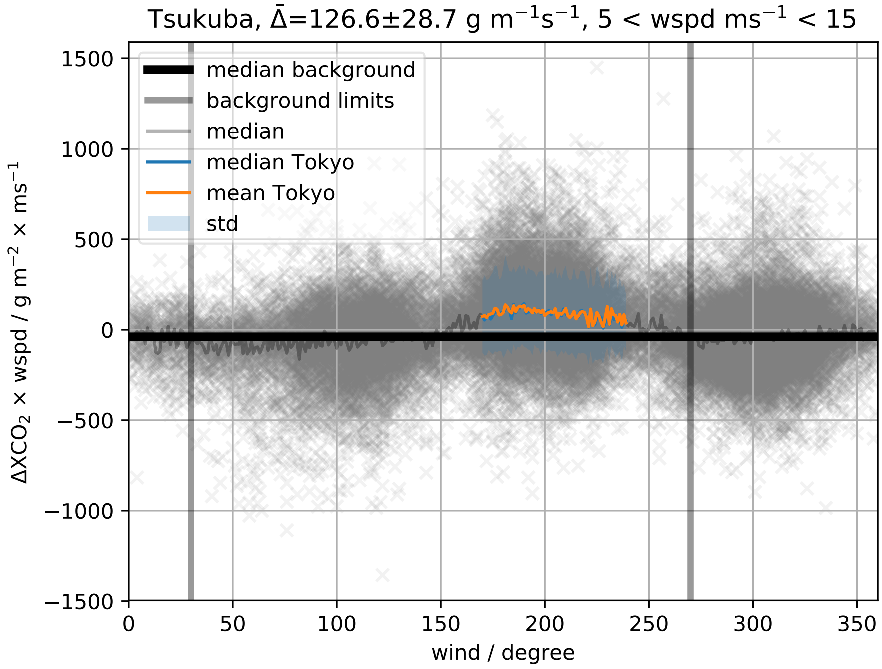


Figure 4. Wind speed profile statistics (**and**) left wind direction statistics (**right**) up to 1000 m at Tateno, Japan using data from 2009 to 2016, from the Ijima (2016) dataset. Values are calculated by dividing the wind speed at a given pressure by the wind speed at the lowest level. The boxplots show the median (red line) and the mean (green triangle). 50% of values are within the box, the whiskers include 95% of the values and the rest is shown as outliers (black circles). The notch in the box shows the uncertainty of the median calculated via resampling.

Figure 4 visualizes the variability of the wind speed profile weighted by atmospheric pressure ~~by aggregating~~ from the radiosonde data measured at the Tateno site. The average wind speed in the profile with a lower limit of 31m and the upper limit of 1000m is used to derive daily scaling factors from the ground wind speed to the average profile wind speed. These scaling factors are applied to the ground wind speed measured at the TCCON site in Tsukuba to estimate the effective wind speed of the volume of air with enhanced carbon dioxide concentration in the total column.

These scaling factors are provided in the auxiliary material ~~(Babenhauserheide et al., 2020)~~ but provide a significant source of uncertainty, since their use rests on the assumption of uniform mixing of the carbon emissions across the boundary layer. The forward trajectory calculations with HYSPLIT provided in the auxiliary material suggest that 50 km transport distance suffices for particles to reach the top of the boundary layer, but they do not prove that this suffices to generate a uniform CO_2 mixing ratio. Therefore, as also seen by Chen et al. (2016), the unknown actual transport pathway of emitted CO_2 to the measurement location is a significant source of uncertainty of the results.



Tsukuba, $\bar{\Delta}$=126.6±28.7 g $m^{-1}s^{-1}$, 5 < wspd ms^{-1} < 15

Figure 5. Residuals multiplied by wind speed plotted against the wind direction for scaled wind speeds between 5 ms^{-1} and 15 ms^{-1}; measured by the TCCON site in Tsukuba, Japan. The mean enhancement $\bar{\Delta}$, the mean Tokyo and median Tokyo and the std are calculated for the directions defined as from Tokyo in section 3. The median background is calculated from the residuals outside the background limits (lower than 30° or higher than 270°; limts drawn as vertical lines). The bin size is 1 degree.

5 Estimated carbon source of Tokyo

Figure 5 shows the data used to calculate $\bar{\Delta}$, the mean total column enhancement of XCO_2. $\bar{\Delta}$ is derived from the XCO_2 residuals, the result of subtracting the trend and fits to the yearly and daily cycle as described in section 3: The median total column residual from background wind directions (chosen as 270° to 30°, using 0° as from north, clockwise, following meteorological conventions) is subtracted from the target direction residuals, then the result is multiplied with the wind speed during the time of measurement. Finally it is converted from measured total column concentration $C_{CO_2,t,\mathrm{col}}$ to total column mass $m_{CO_2,\mathrm{col}}$ using equations from table 1 at time t ~~, level l and daily apriori α_t~~and angle α.

To calculate the carbon source of Tokyo S_T, the measured total column enhancement E_m (in $[g_{CO_2}]$) needs to be multiplied with the *area affected per second* by the emission source from within Tokyo area, $\mathcal{A}$ (in $\left[\frac{m^2}{s}\right]$):

$$S_T = E_m \cdot \mathcal{A} \qquad (1)$$

Table 1. Units and definitions

unit air column mass:	$m_{\text{air,col}}$	$= \frac{p}{g} \cdot 10^5 \left[\frac{g}{m^2}\right]$
CO_2 column mass	$m_{CO_2,\text{col}}$	~~$= \frac{M_{CO_2}}{M_{\text{air}}} \frac{m_{\text{air,col}}}{f_{col}} \cdot C_{CO_2,t,\text{col}}$~~ $= \frac{M_{CO_2}}{M_{\text{air}}} \frac{m_{\text{air,col}}}{f_{col}} \cdot C_{CO_2,t,\text{col}} \left[\frac{g}{m^2}\right]$
total column residuum:	R	~~$= m_{CO_2,\text{col}} - m_{CO_2,\text{col,}\textbf{seasonal cycle fit}} - m_{CO_2,\text{col,}\textbf{daily cycle fit}}$; (described in section 3)~~ $= m$
enhancement:	E_m	~~$= R_{\text{from Tokyo area}} - \text{median}\left(R_{\text{from background}}\right)$~~ $= R_{\text{from Tokyo area}} - \text{median}\left(R_{\text{from background}}\right) \Big[\frac{g}{m}$
molar mass of CO_2	M_{CO_2}	$= 44.0 \frac{g}{mol}$
molar mass of dry air	M_{air}	$= 28.9 \frac{g}{mol}$,
column mass correction	f_{col}	= 0.9975, following Bannon et al. (1997) to adjust for curved geometry
tracer mass	$m_{\text{gas,col}}[g]$,	
total column dry air mass	$m_{\text{air,col}}[g]$,	
column concentration	~~$C_{CO_2,t,\text{col}}$~~ $C_{CO_2,t,\text{col}}\,[ppm]$,	
acceleration due to gravity	$g[\frac{m}{s^2}]$	(from TCCON ~~aprioria~~ a priori),
pressure	$p[\frac{g}{ms^2}]$.	(with wet air to dry-air correction)
perpendicular spread	$s_\perp\,[m]$	

By separating the affected area per second $\mathcal{A}$ into the wind speed of the volume of air with enhanced concentrations at the measurement location ~~,~~ v (approximately the average column wind speed within the boundary layer (~~0-2000 m)~~0–1000 m)), v ~~,~~ and the spread of the Tokyo area perpendicular to the wind speed $s_\perp$,

$$\mathcal{A} = v \cdot s_\perp, \tag{2}$$

this source can be derived from the mean total column enhancement of XCO$_2$ ~~$\bar{\Delta} = 126 \pm 29 \frac{g_{CO_2}}{ms}$~~ $\bar{\Delta} = 127 \pm 29 \frac{g_{CO_2}}{ms}$ shown in Figure 5 via

$$S_T = E_m \mathcal{A} \approx s_\perp \bar{\Delta} \tag{3}$$

The perpendicular spread $s_\perp$ is calculated by assuming that total columns of carbon dioxide from Tokyo area are transported to the measurement location without effective divergence perpendicular to the wind direction and assuming ~~approximately~~ roughly circular city structure. Therefore this spread can be approximated from the distance between Tokyo city center and the TCCON measurement site in Tsukuba:

$$s_\perp \approx 2\pi \cdot s_{\text{Tsukuba–Tokyo}} \cdot \frac{\Delta\alpha}{360°} \tag{4}$$

with $\Delta\alpha$ the opening angle of the limits of wind directions associated with Tokyo and $s_{\text{Tsukuba–Tokyo}} \approx 52$km. The city center of Tokyo was chosen to be at the palace (35.6825°N 139.7521°E), between the densely populated area and the power plants on the other side of Tokyo bay. Treating 170° to 240° as wind direction coming from Tokyo, this yields a perpendicular spread of ~~$2\pi\cdot 52km\cdot\frac{70^\circ}{360^\circ}=64km$.~~ $2\pi\cdot 52km\cdot\frac{70^\circ}{360^\circ}=64km=64000m$. The choice of the palace is arbitrary, because the actual "center of mass" of the emissions of Tokyo is unknown. Section 6 estimates the uncertainty due to this arbitrary choice.

For the approximation in equation 3, the angle-integrated ~~$E_m\Delta A_{aff}$~~ $E_m\mathcal{A}$ is collected into contributions from different wind directions as shown in equation 5:

$$E_m\mathcal{A}=\int\limits_{\alpha_0}^{\alpha_1}E_{m,\alpha}\underline{A_{aff,\alpha}}\mathcal{A}_\alpha d\alpha=\frac{s_\perp}{\Delta\alpha}\cdot\int\limits_{\alpha_0}^{\alpha_1}E_{m,\alpha}v_\alpha d\alpha=s_\perp\bar{\Delta} \tag{5}$$

Therefore the source of Tokyo can be derived from the mean enhancement $\bar{\Delta}$ as

$$S_T=\bar{\Delta}\cdot s_\perp=\underline{126}127\pm 29\frac{g_{CO_2}}{ms}\cdot 64000m=8.1\pm 1.9\frac{t_{CO_2}}{s} \tag{6}$$

The given uncertainty is taken from the standard deviation as shown in Figure 5.

For comparison with city emission inventories, the CO_2 source is scaled to yearly carbon emissions:

$$\begin{aligned}
S_{T,\mathrm{C,yearly}} &= \bar{\Delta}\frac{M_C}{M_{CO_2}}s_\perp\cdot\frac{s}{\mathrm{year}} && (7)\\
&= \underline{126}127\pm 29\cdot\frac{12}{44}\left[\frac{g}{ms}\right]\cdot 64000m\cdot 31557600\frac{s}{\mathrm{year}} && (8)\\
&= \underline{69}70\pm 16\frac{MtC}{\mathrm{year}} && (9)
\end{aligned}$$

For comparison with gridded emission inventories in section 7, the CO_2 emissions are scaled to average monthly carbon emissions per wind direction (in 1° steps):

$$\begin{aligned}
S_{T,\mathrm{CO_2,average,deg,monthly}} &= \bar{\Delta}\frac{M_C}{M_{CO_2}}s_\perp\cdot\frac{s}{\mathrm{month}\cdot\mathrm{degree}} && (10)\\
&= \underline{126}127\pm 29\left[\frac{g_{CO_2}}{ms}\right]\cdot\frac{12}{44}\left[\frac{g_C}{g_{CO_2}}\right]\cdot 914m\cdot 2592000\frac{s}{\mathrm{month}\cdot\mathrm{degree}} && (11)\\
&= \underline{81}82\pm 19\frac{ktC}{\mathrm{month}\cdot\mathrm{degree}} && (12)
\end{aligned}$$

6 Estimating uncertainties

In addition to the statistical uncertainty and the uncertainty of the wind profile discussed in section 4, the estimated emission depends on the assumed extent of Tokyo area and is limited by the unknown actual distribution of distances of emission sources from the measurement site at Tsukuba.

Choosing different opening angles for air *from Tokyo area* yields a yearly emission range from $54.0 \pm 7.4 MtC\text{year}^{-1}$ when choosing air *from Tokyo area* between 180° and 220° up to $93 \pm 35 MtC\text{year}^{-1}$ when choosing air *from Tokyo area* between 150° and 260°. This uncertainty also plays a role in comparisons, if the actual wind direction higher up in the atmosphere is not distributed symmetrically around the wind direction at ground.

The distance of emission sources from the TCCON site in Tsukuba affects the estimated spread of the emission region perpendicular to the wind direction. This calculation assumes a distribution of emission strengths along the wind direction symmetric around a center given by the distance. This assumption is plausible, since the most densely populated region of Tokyo extends to the north west towards the prefecture of Saitama. However Bagan and Yamagata (2014) and Oda and Maksyutov (2011, 2016) show a similar extension towards the south, and the power plants are southward of the palace. Assuming an uncertainty of 10 km for the distance between the "center of mass" and the measurement site increases the uncertainty:

$$\begin{aligned} S_T &= \bar{\Delta} \cdot s_\perp = \bar{\Delta} \cdot 2\pi \cdot 52 \pm 10km \cdot \frac{70°}{360°} = \bar{\Delta} \cdot 64 \pm 12.3km && (13) \\ &= 126127 \pm 29 \frac{g_{CO_2}}{ms} \cdot 64000 \pm 12300m = 8.1 \pm 2.4 \frac{t_{CO_2}}{s} && (14) \\ &\Rightarrow 6970 \pm 21 \frac{MtC}{\text{year}} && (15) \\ &\Rightarrow 8182 \pm 24 \frac{ktC}{\text{year,degree}} && (16) \end{aligned}$$

This uncertainty needs to be taken into account, but can only be estimated. It gives a contribution of $\pm 5 \frac{MtC}{\text{year}}$.

The ground wind speed from TCCON-data also varies by around 30%, but this is part of the scatter in the data, so it is already averaged and reported. The scaling factors are calculated daily, so their uncertainty is part of the scatter in the data, too. Within the relevant pressure region here (860hPa and more), the TCCON averaging kernels ~~can vary by around 10%, which is also part of the scatt~~ effects could be a slightly higher sensitivity in the morning and evening, but that's also the time with the least amount of data.

Another source of uncertainty is the accuracy of the background. A bias in the background translatesinto a bias of the result. For sites with a large forest in one direction but none close to the city, this would have to be taken into account.

The fitting procedure can affect the outcome. Repeating the same calculations with a different fitting procedure based on sines and cosines (e.g. Thoning et al., 1989), implemented using the ccgfilt library (referenced in section 8) gives an idea of the impact of the fitting. As shown in section 2 of the auxiliary material (Babenhauserheide et al., 2020), this calculation yields ~~$\bar{\Delta} = 137 \pm 31 \frac{g_{CO_2}}{ms}$~~ $\bar{\Delta} = 138 \pm 31 \frac{g_{CO_2}}{ms}$ as source instead of the ~~$126 \pm 29 \frac{g_{CO_2}}{ms}$~~ $127 \pm 29 \frac{g_{CO_2}}{ms}$ found with polynomial fits. This corresponds to a relative difference of ~~8.7~~8.3% which is not captured by the internal variability of the residuals. For

~~$69MtC$~~$70MtC$, the absolute difference is $\pm 6MtC$. The structure of this error is unknown, though, therefore it is ~~only~~ shown separately.

Consequently the most robust estimate of the emissions of Tokyo is

$$\text{Yearly carbon emissions of Tokyo:} \qquad S_{T,\text{C,yearly}} = \cancel{69}70 \pm 16 \pm 5 \pm 6 \frac{MtC}{\text{year}} \tag{17}$$

$$\text{Monthly carbon dioxide emissions of Tokyo:} \qquad S_{T,\text{CO}_2\text{,average,deg,monthly}} = \cancel{81}82 \pm 18 \pm 6 \pm 7 \frac{ktC}{\text{month} \cdot \text{degree}} \tag{18}$$

These values provide an estimate of the source of Tokyo calculated directly from measurements. The measurements are only conducted in the hours of day between 0 UTC and 8 UTC, though, and the fossil fuel source of Tokyo might be different during nighttime due to reduced human activity. Nassar et al. (2013) provide hourly scaling factors for fluxes for global models. In the measurement interval these scaling factors are 1.09, 1.11, 1.13, 1.16, 1.16, 1.18, 1.20, 1.21 and 1.188 which gives an average factor of 1.16, with the standard deviation given as 0.15. Dividing the fluxes by 1.16 gives an estimate of the fluxes which would be derived from measurements around the day. This would result in a total emission estimate of ~~$59 \pm 18 \pm 6 \frac{MtC}{\text{year}}$~~$60 \pm 18 \pm 6 \frac{MtC}{\text{year}}$.

$$\text{Daily cycle corrected yearly emissions:} \qquad \hat{S}_{T,\text{C,yearly}} = 59 \pm 18 \pm 6 \frac{MtC}{\text{year}} \tag{19}$$

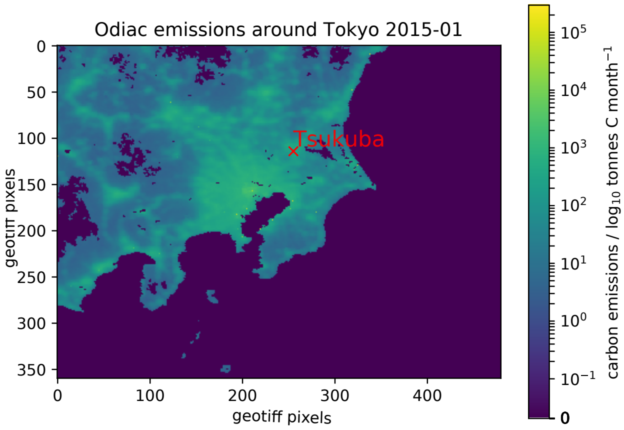


Figure 6. ODIAC carbon emissions per 1km × 1km in pixel for January 2015 in $\log_{10}$ scale. This graph is created directly from the 1km×1km ODIAC dataset (Oda and Maksyutov, 2011, 2016) to visualize its structure. The model emissions are shown for the same area visualized in the middle panel of figure 3. The unit is ~~tonne Carbon~~metric ton carbon/cell and month as described in the readme at db.cger.nies.go.jp/dataset/ODIAC/readme/readme_2016_20170202.txt.

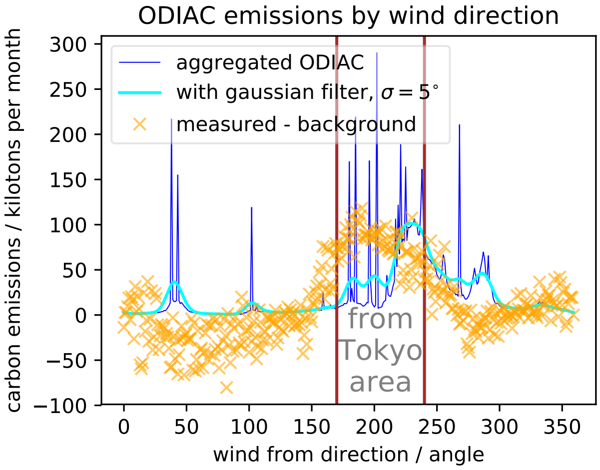


Figure 7. Sum of ODIAC carbon emissions by direction in angular degrees as seen from Tsukuba, Japan. The “aggregated ODIAC” emissions show emissions per direction from beginning of 2011 to end 2016. The gaussian filter data uses a moving average ~~with gaussian weight~~ to estimate signals measured at a distance. The measured dataset shows the median residuals from figure 5 for comparison.

7 Comparison with other Datasets

To compare the results with the high-resolution Open-Data Inventory for Anthropogenic Carbon dioxide emission ~~inventory~~ (ODIAC, Oda and Maksyutov, 2011) in version ODIAC2016 (Oda and Maksyutov, 2016), using the regional slice shown in

Figure 6~~. Measurements~~, measurements are simulated from ODIAC by summing emissions by direction as seen from the position of Tsukuba station. For total emissions, all emissions within the arc spanned by the limits of *from Tokyo area* from 2011 to 2016 are aggregated, then the sum of the emissions *from background directions* is subtracted. Emissions aggregated for each 1° angle segment are shown in Figure 7):

$$\frac{1}{5}\sum_{t=2011-01}^{2015-12}\left(\sum_{\alpha}^{\text{Tokyo}} E_{ODIAC,t,\alpha} - \sum_{\beta}^{\text{bg}} E_{ODIAC,t,\beta}\right) = 40.4\frac{MtC}{\text{year}}, \tag{20}$$

which is around 60% of the emissions estimated in this paper from TCCON measurement data and within two standard deviations (σ) of the estimated emissions. With the scaling for the time of day of the measurement, ODIAC results lie within one standard deviation of the estimate in this paper.

The peak of the distribution of emissions (within "from Tokyo area") is shifted about 30° counterclockwise from model to measurements. This is within the expected ~~shifts~~ changes due to the typical shift in wind direction between measurements conducted close to the ground and measurements higher up in the planetary boundary layer (Ekman, 1905). These discrepancies could be corrected ~~for~~ by using more complex atmospheric transport, but that would ~~then~~ require every person reproducing the estimates from our study to run such a transport, which would defeat the purpose of our study, namely to provide an easily reusable approach for estimating city emissions.

The economic data published by the Bureau of the Environment Tokyo (2010) report emissions of $57.7\frac{MtCO_2}{\text{year}}$ in the fiscal year 2006 for the Tokyo Metropolitan Area. This is equivalent to $15.7\frac{MtC}{\text{year}}$ and shows a large discrepancy to our results. This discrepancy could stem from different definitions for the source area. Part of this discrepancy cannot be reconciled, because the method shown in this ~~paper~~ study cannot limit the emission aggregation parallel to the wind direction and has around 30° uncertainty of the direction, so it also includes some emissions from Kanagawa, Saitama, and Chiba, the prefectures around ~~Toko~~ Tokyo which are part of the greater Tokyo area.

Pisso et al. (2019) estimate the emissions of Tokyo City as 22 MtC/y (80 MtCO$_2$/y) and of the Tokyo metropolitan area as 151 MtC/y (554MtCO$_2$/y). With $70 \pm 21 \pm 6$ MtC/y ($256 \pm 77 \pm 22$ MtCO$_2$) this study lies between those values, which is to be expected because the definition of the metropolitan area used in Pisso et al. (2019) includes all the fluxes from the prefectures Kanagawa, Saitama, and Chiba, while this study only includes part of these fluxes. Pisso et al. (2019) also compare against several other datasets with their source definition.

8 Conclusions and Outlook

We find that a single multi-year dataset of precise column measurements provides valuable insights into the carbon emissions of city-scale emitters. The estimated emissions of ~~69 ± 21 ± 6~~ 70 ± 21 ± 6 mega-tonnes carbon per year found for Tokyo has less than 50% uncertainty despite our ~~intentionally chosen~~ intentional constraint to use only a basic evaluation scheme which can be repeated on any personal computer with publicly available data. While the operation of a TCCON station is a major effort, a decade of CO_2 column measurements of comparable quality can be conducted with affordable and easier to operate mobile spectrometers ~~(see for example Frey et al., 2015)~~ (see for example Frey et al., 2019) which opens an avenue for every country to measure and evaluate emissions of mega cities: Placing a single total column measurement site in the vicinity of ~~a major city allows estimating its emissions~~ major cities can make it possible to estimate their emissions purely from these measurements. This can complement global source and sink estimates and improve acceptance of carbon trading programs by enabling independent verification of findings.

Significant reduction of the uncertainties in these estimates without adding more measurement stations would require taking into account more detailed wind fields from meteorological models, correcting for the wind direction at different altitudes by using partial columns, more detailed correction for expected CO_2 takeup from the biosphere by wind direction, or correcting for the diurnal cycle of fossil fuel emissions. These corrections are already taken into account in source-sink estimates based on inverse modelling of atmospheric transport with biosphere models (e.g. van der Laan-Luijkx et al., 2017; Riddick et al., 2017; Massart et al., 2014; Basu et al., 2013), therefore this implementation keeps close to the simpler evaluation which allows staying closer to easily accessible data which keeps our findings easy to replicate. A better classification of uncertainty due to the assumption of uniform vertical distribution of the emitted CO_2 could be given by measuring highly resolved vertical profiles ~~by~~ from aircraft downwind of Tokyo.

Further uncertainty reductions can be achieved by establishing several observing sites within and around the source area (Hase et al., 2015; Turner et al., 2016, e.g.). This approach also provides information about the spatial structure of emissions and can be used in focused measurement campaigns to obtain constraints for evaluation of measurements with coarser spatial resolution as well as long term datasets. It would eliminate most of the uncertainty in the mean distance between measurement site and emitters.

~~To reduce~~ Reduction of the bias due to measuring only during daytime~~similar to the approach shown at the end of 6~~, while keeping close to direct measurements, ~~our study could be improved~~ could be achieved by calculating the diurnal scaling of the emission source from CO_2 concentration measurements of an in-situ instrument or ~~to take~~ by taking moonlight measurements (Buschmann et al., 2017).

To complete this outlook, we would like to suggest that the negative values seen on the left graph of Figure 3 at a wind direction around 60° indicate that it might also be possible to detect biospheric drawdown of CO_2 by woodland with just a single total column instrument, and that this method can also be used to analyze other greenhouse gases measured by the TCCON network, including methane and carbon monoxide. Residuals for methane are shown in figure 6 of the auxiliary material.

We conclude that long-term ground-based measurements of column-averaged greenhouse gas abundances with sufficient ~~accuracy~~ precision for detecting the signals of local emission sources are ~~a~~ an effective and cost-efficient approach to improve our knowledge about sources and sinks of greenhouse gases.

Code and data availability. All code used and pre-processed data in JSON format (as described in RFC 7159) are available in the auxiliary material (Babenhauserheide et al., 2020). See the README in the auxiliary material for usage information. All non-included data is publicly available from the TCCON data portal (tccondata.org), from the ODIAC project odiac.org, and from the Atmospheric Soundings site at University of Wyoming (weather.uwyo.edu/upperair/sounding.html). The ccgfilt library is available from NOAA via ftp://ftp.cmdl.noaa.gov/user/thoning/ccgc

Author contributions. Isamu Morino provided the TCCON-Data at Tsukuba station and helped to interpret it, Frank Hase helped finding working approaches for the evaluation and improving the manuscript, Arne Babenhauserheide implemented the evaluation, calculated the results, and wrote most of the manuscript.

Competing interests. The authors have no competing financial interests, but Frank Hase and Isamu Morino are working on other projects with ground-based total column measurement instruments.

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