Reply to interactive comments

Arne Babenhauserheide[1,*]**, Frank Hase**[1]**, and Isamu Morino**[2]

[1]IMK-ASF, Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany
[2]National Institute for Environmental Studies (NIES), Tsukuba, Japan
[*]now at Disy Informationssysteme GmbH, Karlsruhe, Germany

Correspondence: Arne Babenhauserheide <arne_bab@web.de>

1 Introduction

Thank you for the in-depth review of our publication! (First author speaking). I am sorry, that the response was delayed for so long. There were far more technical and organizatorial hurdles than expected after I changed from University to working in industry. My deepest gratitude also to our editor at AMT for her patience.

2 Answers to RC 1

Reviewer 1 gave an in-depth review, with solid and practical advice to improve our work. Our heartfelt thanks for the time and effort invested. Answers to the comments are provided inline.

2.1 General comments

"*This paper discusses a first order estimate of carbon dioxide (CO2) emissions from the greater Tokyo area using total column measurements of CO2 at one location, namely the Tsukuba Total Carbon Column Observing Network (TCCON) site. The authors derive this flux estimate by first assuming typical annual and diurnal trends are what would be observed without local/regional anthropogenic emissions. Then they use radiosondes to get an average wind speed and direction of the layer of atmosphere enhanced by local emissions. Next they assumed an emissions area defining Tokyo as an arc of a circle with the center at the Tsukuba site, and the angles were based on wind direction and typical enhancements. Finally, they assumed uniform emissions then used this assumption with wind speed to infer a flux.*"

"*Studies like this are very important, as there are few such studies estimating fluxes using total column measurements to fully estimate a CO2 flux. It is well suited for AMT. The authors made good progress on a difficult problem – namely, how to estimate fluxes when few data are available. However, I have concerns with the stated and unstated assumptions, as well as with the general methodology. Further, the flux estimate is 5x that from the Bureau of the Environment Tokyo which suggests the un- certainties are very large. It is important to improve accuracy in these studies as much as is reasonably possible because inaccuracies could lead to false interpretation and perhaps improper carbon pricing by policymakers. Hence even though I think with major revisions this study could be useful to the community I have many comments.*"

Important to note is here that we do not try to get the best possible accuracy, but rather the best accuracy which can be reached while only using directly measured data with readily understandable methodologies. As we note in section 8 (Conclusions and

Outlook), the accuracy can be increased a lot by using wind fields from meteorological models, and there are already systems which use this approach for ground-based in-situ data. This paper shows the accuracy that we reached with just one single ground-based total-column site.

2.2 Specific comments

"*1. Title: Clarify that the estimates are 1) of net CO2 (not all GHG and not just FF), 2) from the *greater* Tokyo area. Also 3) these are not "direct" as wind measurements were required.*"

We adjusted the title to be more precise:

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

"*2. Abstract: Needs more detail. Specify the FTIR is not in situ. Define TCCON. Briefly describe methodology (2-4 sentences). Compare with literature estimates.*"

The abstract is now expanded. It includes a short definition of FTIR which should clear up that these are not derived from in-situ measurements of the local air parcel but rather from the total column of air, include a short description of the methodology and comparison with literature:

We present a simple 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.

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 by 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 emissions from the area of Tokyo to be $86 \pm 33 \frac{MtC}{\mathbf{year}}$ 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.

Our estimates are factor two larger than estimates using the ODIAC emission inventory, 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 estimates are much larger than the emissions of Tokyo given by the Bureau of the Environment Tokyo in 2010, which likely stems from different definitions of the source area that cannot be reconciled with this measurement setup.

"*3. p 1, line 13-16: Only some of these references appear relevant, and there are some the authors should consider including that include a CO2 flux estimate. I suggest omitting the following: Bovensmann (only an OSSE), Hakkarainen (includes enhance- ments only, not a flux), Hammerling (simulation), Hakkarainen (duplicate), Butz (did not use satellite data), Frey (only an instrumental study), Chen (estimated CH4, not CO2 flux). Consider including: Liu et al, 2017 (estimates of CO2 fluxes from different regions using OCO-2 data, doi: 10.1126/science.aam5690), Nassar et al 2017 (CO2 estimates from power plants using OCO-2 data, doi: 10.1002/2017GL074702), Ye et al 2017 (constraining urban CO2 emissions with OCO-2 data, doi: 10.5194/acp-2017- 1022), Irana et al 2018 (CO2 fluxes from a peatland using column measurements, doi: 10.1038/s41598-018-26477-3), Hedelius et al 2018 (CO2 fluxes from a California re- gion using column measurements, doi: 10.5194/acp-2018-517), Vogel et al 2018 (CO2 fluxes from Paris using column measurements, doi: 10.5194/acp-2018-595), Wu et al 2018 (made simulation of CO2 columns for OCO-2 using a new modelling tool, did not compute flux but still may be useful, doi: 10.5194/gmd-2018-123).*"

Thank you for the references. We removed Bovensmann and the duplicate and shifted Butz 2016 to the ground-based emission estimates where it should have been. From the suggested references we added Nassar2017, Ye2017, Hedelius2018, and Vogel2019, and skipped Liu et al, 2017, because it does does not estimate fossil fuel emissions, and Wu et al 2018 for the same reason that we removed Bovensmann (they did not derive emissions from data).

"*4. p 2, line 1-2: provide a little more detail on TCCON here. How accurate and precise is it? Where do the data come from?*"

This is described in section 2 (Observations). We added a reference.

"*5. p 2, line 6: When I think "inexpensive," I think of BEACON (Shusterman et al, 2016 doi: 10.5194/acp-16-13449-2016), with a price of 5500 USD per sensor. Surely a TCCON site costs more than this otherwise there would be more around Tokyo? I would also expect the cost of one-time radiosondes to add up to more than this. Same comment for p 14, line 6 – are the "affordable" mobile spectrometers really less than 5500 USD?*"

The qualifier inexpensive is in comparison to satellite missions, with a cost in the hundreds of millions of dollars. To make it clearer, the text now reads:

This method provides an approach to estimate city emissions which is inexpensive when compared to satellite missions while 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.

The cost of a mobile spectrometer is on the order of $100k, but we cannot give exact prices, since they are set by the producers.

"*6. p 2, line 10: Better to cite the original reference where the stated instrument- to-instrument bias is 0.3 ppm (Messerschmidt et al, 2010, doi: 10.1111/j.1600- 0889.2010.00491.x).*"

Messerschmidt is more direct, yes. Switched and moved to **less than 0.3 $\mathbf{ppm}$**, because Messerschmidt et al reported 0.27 ppm. Thank you!

"*7. p 2, line 13 – 0.2% is 0.8 ppm of CO2*"

Precision has to be 0.1%, adjusted.

"*8. p2, line 16-17 – This sentence can be omitted, as the dates are already listed on line 6*"

Removed the dates from the introduction to make them easier to find in the Observations section.

"*9. p2, line 18-19 – I checked this website, and the information does not seem "es- sential" to this paper. It is mostly interesting photos. This sentence can be omitted or should be reworded.*"

Reworded as **Further information**.

"*10. Throughout – please incorporate the footnotes into the main text (see https://www.atmospheric-measurement- techniques.net/for_authors/manuscript_preparation.html). For example, move data references to the "Code and data availability."*"

The reference to the TCCON data portal was moved to "Code and data availability".

"*11. Section 3/4 – Put the discussion of mean wind speed/direction first because it seems important to the discussion of removing trends, especially the discussion of air origins.*"

The supporting graphics for the origin of the air requires information from the detrending, therefore the section needs to come after the detrending. However the detrending section now starts with an explanation about the approach of segmenting by wind direction. This should make it easier to follow in the original order.

"*12. Remove parenthetical comments about granularity. From the trendline in Fig 1 it is clear that the polynomial fits finer and coarser time periods.*"

These parenthetical notes are included to make it easier for people with experience in direct monthly or hourly binning and comparison to get an understanding for the effect of fitting with the polynomials. Therefore these are important to make the publication easier to understand.

"*13. Section 3: I am concerned with these fits in general. Why are they polynomials instead of the typical sines and cosines (e.g., Thoning 1989, doi: 10.1029/JD094iD06p08549). There is even a Python package to do this: ccgfilt (ftp://ftp.cmdl.noaa.gov/user/thoning/ccgcrv/). Why are these fits done in 2 parts (over- all linear then annual polynomial), rather than one unified fit? Why were these degrees chosen (the higher order, the better the fit should be)? How are you sure the diurnal fits are really background and not a measurement artifact (the R2 is very low)? How are you sure there is not a persistent enhancement in column CO2 due to Tokyo emissions that would get fit out and hence bias your enhancements? How do these "background measurements" or fits compare to similar measurements (TCCON or satellite) away from urban areas nearby?*"

The degrees were chosen to fit not only this site, but measurements from all TCCON sites.

Polynomials are chosen because these are the easiest way to fit which can be reproduced with every evaluation system. The goal of this publication is to show that the data from TCCON sites can already be evaluated with a simple, easy to reproduce way. Even slightly more complex evaluation methods make it harder to follow the evaluation, radically reducing the number of people who might try such an evaluation themselves.

However I now repeated the full evaluation pipeline with ccgfilt as fit method. There is one difference: ccgfilt always fits against all data instead of fitting the trend against background directions and the rest against all data. This is because separating out these two aspects resulted in large artifacts of the fit. Which makes the point again that keeping to simpler methods is benefitial here. The resulting graphs of this ccgfilt-evaluation are available in the auxiliary material. In short: The estimated emissions are about 10% larger than with the polynomial fit, clearly within the uncertainty.

However this difference between the fit methods is not seen in the scatter of the residuals, therefore I added it as part of the discussion of uncertainties.

To go towards more advanced evaluations it would be best to directly use a global wind-field driven inverse model like CarbonTracker or TM5-4DVar or one of the other inverse GHG flux estimation models, and cleanly assimilate TCCON measurements with full simulated wind-fields and good priors. This publication here is intended to show that in the absence of such a more powerful method simple ways can already yield results.

A persistent enhancement could show up, if the wind direction would change much faster than the transport into the wind direction. For Tokyo at the wind speeds evaluated ($5 - 15\frac{m}{s}$), this is not the case. It is however one of the possible reasons for the lower differences at low wind speeds. In Figure 3 you can see a persistent enhancement from all wind directions with wind speeds up to $3\frac{m}{s}$.

"*14. p 4, line 12: State which supplemental figure number.*"

Thank you for the catch!

"*15. p 5, line 4: This looks like a 2D heatmap of averages values per bin, rather than counts per bin (i.e., it does not look like a histogram. . .)*"

You're right. A more correct term is hexbin averages. Histogram is now replaced by **following the hexbin averages shown in the right panel**.

"*16. p 5, line 7-8: These last 2 sentences are confusing, please clarify.*"

The sentences have been cleared up:

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). This split is purely for visualization: in the following calculations and graphs, negative and positive residuals are used together.

"*17. p 5, Fig 3: It would be useful to include a variant of the statement from p 14, line 22-24 here.*"

We added such a statement: **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.**

"*18. Section 4.1: General convention is to only number subsections if there is more than one. Also, please clarify throughout what the winds data source was. It seems like there were 3: ground-based from the measurement site (?), HYSPLIT, and radiosondes*"

The subsection is no longer numbered. The text is adjusted to ensure that origin of the different wind speeds discussed is always clear.

Wind speeds which enter further calculations are only from the Tateno radiosondes and the TCCON ground data.

"*19. p 6, line 10: A few sentences to a paragraph should be written to describe how HYSPLIT was run. Were these forward or backward trajectories? What heights were they released from? What gridded model was used with them? How long were these runs? How many days were these runs for? Consider adding a figure to show the results. Those in the SM are a start, but are in my opinion over too short a time period and with too coarse a model.*"

This was now clarified: **To calculate the required height of this profile, forward trajectories from Tokyo for 5 to 15 hours were calculated with HYSPLIT (Stein et al., 2015) using READY (Rolph et al., 2017), accessed via the HYSPLIT-WEB online service from NOAA[1] as described in the auxiliary material. 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 the auxiliary material**

"*20. p 6, line 10: Please have a separate comment to indicate Ready was also used for the Rolph 2017 reference.*"

This is now noted explicitly.

"*21. p 6, line 11: Include Tateno on a map and/or include coordinates.*"

Coordinates are now included: Latitude 36.06° N, Longitude 140.13° E.

"*22. p 6, line 16: Is "average wind speed in the profile" defined as that from the surface to 1 km, 2 km, or the highest sonde point?*"

It is averaged between 31m and 1000m. This is now more clearly noted. **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.**

"*23. p 6, line 22: This statement does not appear to be in this reference.*"

The citation is now removed (it was duplicated from the previous, so no information is lost), since citing here was misleading. The text was adapted to be clearer: **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**.

"*24. Fig 4: This figure is slightly deceptive as a ratio is plotted, so 1/10 is equivalent to as much scaling as 10x. This makes it look like there are more high outliers than low ones. Consider plotting on a logarithmic scale instead.*"

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

A logarithmic scale would place higher focus between 0 and 1, which would also be misleading. The advantage of the linear scale is here, that the high outliers can be clipped to the right and that linear scales are typically easier to read.

Logarithmic scale was tried, but linear scale was clearer.

2.2.1 Section 5

"*25. Section 5: I personally had difficulty understanding this section and hence the validity. It should be rewritten in a stepwise, building fashion with explanations and assumptions stated throughout. Specifically*"

1. "*a. Watch units, and keep them consistent with names. P6L2 should units be gCO2/m2? P6L3 area is in m2, so use a different letter besides A for m2/s. P6L6 "area" (m^2) should be "distance" (m) here. P7L8 "t" usually refers to time. It would also be helpful to include units for everything.*"

2. "*b. Try to be consistent with past literature notation. P7L9 I confused $\bar{\Delta}_{CO_2}$ as just the average column CO2 enhancement, but here it refers to the average column enhancement multiplied by wind speed.*"

   This is now replaced by just $\bar{\Delta}$, since it is the direct result from the fitting and integration which is then used to derive quantities to compare with other publications.

3. "*c. Eliminate subscripts where possible. E.g., $v_{wind} -> v$, $E_m -> DeltaC$, $S_T -> ForE$, $v_{alpha} -> v(alpha)$, $g_{a,t}$, $l -> g$, $p_t -> p$ (or p(t) if time is important). These are just examples, choose notation that suits you and is in line with the literature. In some cases the authors could argue that one or two letter subscripts are useful, but to me subscripts have been used in excess.*"

   - $A_{aff} \Rightarrow \mathcal{A}$
   - $g_{a,t} \Rightarrow g$

4. "*d. Proceed in a stepwise fashion, and state assumptions along the way. It seems Eq. 4 should be first, and should probably be split up. It would also be helpful to list limits of the integral, and not recycle alpha (this is one case where I think $alpha_0 and alpha_1$ would be acceptable).*"

   The equations are now restructured.

5. "*e. Consider placing P7L17-27 and the footnote in a Table. Units could be included in this table.*"

   These equations and definitions are now in the table "Units and definitions".

6. "*f. Equation 5 – is water not important? Also, this term is duplicated on line 24.*"

   TCCON measures the dry air mole fraction, so water is not important here.

7. "*g. Equation 7 belongs in Section 3. Pick units and stick with them for this fit. Here the "m" terms are in g/m2, but Figure 1 is in ppm. As currently described in P9L11-16 it seems circular (Eq 7 requires 5 & 6, but 5 & 6 are not applied until after 7).*"

   Figure 1 is given in ppm, because the orders of magnitude of ppm for XCO_2 are well known. This makes it easier to understand the fitting procedure.

   However the same does not apply to the derivation of amounts of emitted CO_2, therefore choosing different units is warranted.

8. "*h. p 9, line 26 – This is mislabeled as "gravity," but is actually acceleration due to gravity (m/s^2). Gravity is a force (kg m/s^2).*"

   Thank you for the catch! This is fixed.

"*27. p 8, Fig 5: What are the bin sizes for the mean values?*"

They are 1 degree, as noted in the calculations. This is now also noted in the caption.

"*28. p 9, line 5: What are the coordinate of the palace? Why was this location chosen?*"

The coordinates are 35.6825°N 139.7521°E, chosen because it is a clear landmark which lies between the densely populated area and the powerplants on the other side of Tokyo bay.

However reinvestigating the location uncovered a mistake in the calculation, because the distance between the palace and Tsukuba station is not 65 km but only 52 km. Since this distance is a linear factor in the equations, the result changes from 86 ± 41 MtC/a to 69 ± 33 MtC/a.

"*29. p 10: I think equations 10-12 can be omitted, and the result of Eq. 12 simply appended to Eq. 9.*"

With the restructuring the equations should be clearer.

"*30. p 10 Equations 13-15 and p 11 Equation 17: these seem to be using multiple definitions of "degrees." Gridded emission inventories (e.g., ODIAC) are expressed on grids of global latitude and longitude degrees. Here the degrees refer to the angles around the TCCON site out to an unspecified distance. These should be omitted or properly converted to emissions per latitude/longitude degree box instead.*"

The ODIAC dataset used is given in grids of 1x1km, so there should be no confusion. Degree here always means degree by angle.

"*31. p 10, line 20-22: The TIMES product provided by Nassar et al (2013) is on a 0.25-degree grid. What grid area was selected here to represent Tokyo?*"

The Tokyo area is assembled following the wind-direction to avoid the problems with comparing Tokyo city emission data.

"*32. p 11, line 23: It seems like this estimate would be more representative of actual emissions, and is better aligned with other estimates anyways. Consider listing it in the abstract and placing it in the Conclusions section instead (or in addition to).*"

This estimate relies model data, which goes counter to the goal of working directly from data.

Also it is unclear whether these scaling factors apply on the fine scale of emissions seen here. Therefore these belong in the uncertainty discussion, but are not the main result.

"*33. p 11: The "background" could also be biased (see my previous comment), and this uncertainty should be included. What is the accuracy of the winds? Why are the uncertainties added directly instead of summed in quadrature as is standard for Gaussian uncertainties? Are there uncertainties from the unstated assumption of uniform column sensitivity (i.e., averaging kernels equal to unity)? This last one is a common oversight, but needs to be discussed in remote sensing studies.*"

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 similar, because they 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%. See for example https://tccon-wiki.caltech.edu/Sites/Lamont/Averaging_Kernels

These 10% are also part of the scatter of the data. Systematic effects could be a slightly higher sensitivity in the morning and evening, but that's also the time with the least amount data.

The uncertainties for the distance are now included with correct error propagation (summed in quadrature).

"*34. p 12, Fig 6: Draw enclosed boundaries representing 1) the extent of the Tokyo area using your circle arc definition, for 2) the ODIAC area summed, and for 3) the Bureau of the Environment Tokyo definition if available.*"

Since the Bureau of the Environment Tokyo definition is not given and the boundaries used in this publication are given in Figure 3, the uncertainty definition now rather states that the evaluation **also includes emissions from Kanagawa, Saitama, and Chiba, the prefectures around Toko which are part of the greater Tokyo area.**.

"*35. p 13, line 2: which version of ODIAC was used? Also, ODIAC is an abbreviation, so it should be defined and capitalized.*"

The exact version is given in the bibliography. Now there's also an explicit version in the text.

"*36. p 13, Equation 18: Why is the full area summed, and then the background sub- tracted? Why isn't just the area from non-background (i.e., source) directions added?*"

This is done to simulate the measurements. It is what the method would see if the reality were exactly like ODIAC and the measurement would measure emissions perfectly as the source.

"*37. p 13, line 9-10: A wind direction difference by layer should be added as a subplot to Figure 4 to support this claim.*"

This is added now. It shows the scatter of the wind direction against the ground, as well as the ekman spiral.

"*38. p 13, line 17: I do not understand the last part of this sentence, please rephrase.*"

Part of this discrepancy cannot be reconciled, because the method shown in this paper cannot limit the emission aggregation parallel to the wind direction and has around 30° uncertainty of the direction, so it also includes emissions from Kanagawa, Saitama, and Chiba, the prefectures around Toko which are part of the greater Tokyo area.

"*39. p 14, line 7: Please change to a reference that includes a decade of CO2 column measurements from the mobile spectrometers instead.*"

This does not exist yet.

"*40. p 14, line 11-16: It seems like this includes contradicting statements. First it is stated that uncertainties could be reduced, but at the end it is implied that the simpler evaluation cannot reduce uncertainties. If the uncertainty can be reduced,* *it should be. If it can (likely) only be reduced by using a more complete and hence complex model, this should be stated instead of the first sentence.*"

This is now reformulated: **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, 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 (i.e. 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.**

"*41. p 14, line 18: Turner et al (2016, doi: 10.5194/acp-16-13465-2016) would be an appropriate reference for increasing network density to improve spatial understanding of emissions. It seems Hase et al (2015) also made some progress towards this.*"

That's a nice paper — thank you!

"*42. p 14, line 20: I do not understand this claim. Please rephrase and/or provide a reference.*"

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

"*43. Consider including an Author Contribution section (strangely stated as optional in the *.tex template, stated as required online).*"

I see it now — thank you!

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 evaluaton, calculated the results, and wrote most of the manuscript

"*44. References – In several cases a discussions paper is cited, when a peer-reviewed version is available. Of the articles I would not exclude these are Massart 2014, van der Laan-Luijkx 2017, 2014b.*"

Thank you!

"*45. p 18, line 35: Is more information available for this reference? A doi? A url?*"

Yes, there are several publications listed on , but the reference is not as strong as it should be, so I removed it.

2.3 Technical comments

"*p 1, line 17,20: Should "i.e." (in other words) be "e.g." (for example) here?*"

Yes.

"*p 1, line 19: *historically* short mission times. (Some satellites like GOSAT have been in orbit nearly 10 years! Though GOSAT-2 has been in orbit less than 1 week)*"

I consider 10 years as short.

"*p 4, line 4: there's -> there is*" thank you.

"*p 4, line 14: over *data from* all TCCON*" done.

"*p 6, line 2: source *angles* of Tokyo*" It actually is the source of Tokyo. I'm clearing it up as "emission source".

"*p 6, line 10: *HYSPLIT* (define on first use also)*" done.

"*p 8, line 2: the source of Tokyo -> the CO2 flux from Tokyo*" I'm using "the carbon source". It is clear in the context.

"*p 11, line 2: distribution *of distances* of*" done.

"*p 11, line 5-6: I see no reason why "from Tokyo area" should be italicized (same with p 13, lines 3-4)*" This is highlighted, because it is a central concept in the manuscript.

"*p 14, line 3: megatons carbon per year -> MtC/yr (or megatonnes carbon per year)*" megatonnes it is.

"*p 14, line 19: lower -> finer (?)*" It means less fine. Switching to "coarser".

2.4 supplemental comments

"*S1. The SM includes two parts, (1) scripts to reproduce work, and (2) figures to sup- port the main text as needed. Because most readers will only be interested in (2), the material besides the most relevant *.pdf (emissions-tokyo-auxilliary.pdf) should be moved into a subdirectory.*"

“*S2. SM Fig. 1: I actually disagree that there is only a weak correlation between windspeed and time of day. In the morning the direction appears to predominately be from 3̃00, and then it moves towards 1̃20 in the afternoon. Also, where did these data come from? A meteorological station near the TCCON site? What is the quality? Is the banding at 100, 200, and 300 real or an artifact? There also appears to be a lot of null data at 0 degrees, likely from when the sensor was not moving.*”

The banding seems to be real: there are three dominant wind directions. For data before 10:00 local time and after 16:00 local time there is a correlation, but most data we have lies between the two points in time. It is from the TCCON station.

“*S3. Section 2: This appears to be unfinished.*”

This was removed now.

“*S4. Section 3: availble -> available*” thanks!

“*S5. Section 4: More detail is needed in the text on how HYSPLIT was run. Also, the current model (GDAS1) is 1 degree, which seems too coarse to support claims of understanding vertical transport within 65 km. I cannot see the concentric circle labels, but it appears this plot only goes out to 20 km.*”

Yes, the first figure shows that there are weather patterns during which 5 hours are too short. The other two figures show that between 10 and 15 hours suffice to reach 1000m height.

At $5 fracms$ 5 hours should already suffice for 75 km transport, for example from Tokyo to Tsukuba.

The information to run this are given in the picture itself.

“*S6. Figure 3: I agree that scientific presentations could use more humour, especially in talks and posters. However, I think for this more formal scientific article the fig- ure should be recreated without the doodles though indeed the resemblance is there. Besides, such references can lead to bizarre dreams, obscure fevers, and such knowl- edge was dangerous to Professor Angell. The caption is also unnecessarily verbose (same for Fig. 2, 4).*”

It is true that professor Angell was endangered by his discoveries. It is, however, also true that his discoveries outlived him. True not in the strict sense from other sections in this response but in the strictly fictional sense, therefore we cleared the doodles from the figure. Thank you for your diligence to check our auxiliary material in detail!

The verbosity of the captions is required to get permission to use the diagrams, therefore we cannot change this.

3 Answers to RC 2

Reviewer 2 gave very constructive criticism and helped in improving the clarity of the writing. Our deepest thanks! Answers to the specific comments are inline.

3.1 Major Comments

“*1. Daily variability - I am not convinced that there is structure in the spray of data for Figure 2, particularly as the R 2 is very low for the curve fit. Could a box and whisker plot be overlaid on the inset to convince the reader there is structure? Care needs to be taken to avoid adding uncertainty or structure where there is none. Do the emis- sion values change if a daily cycle is not included? The daily variability shown is not consistent with the daily variability described in Nassar et al. (2013).* ”

The uncertainty of the emission estimate is higher when ignoring the daily cycle.

Section 2 of the auxiliary material now includes a box and whisker plot for the re-evaluation with sines and cosines. It shows the daily cycle of the residuals separated by direction: from outside Tokyo area and from inside Tokyo area. It shows that without daily cycle correction, the residuals from outside Tokyo area become lower over the day, such that values from different times of day could not be aggregated without the correction.

This shows that the daily cycle is most likely due to the actually increasing concentrations from the city from night to day, at 50km distance and wind speeds between 5 and 15 m/s delayed by 1-4 hours. The change is inverted from what Nassar2013 showed because without daily cycle correction the fitting also fits against city emissions.

“*2. Please expand on the one sentence at the end of Section 3 (pg 4, lines 13-14) that suggests why the seasonal and daily cycles were empirically chosen. If I interpret correctly, all the TCCON stations were used to determine the best "global fitting proce- dure". However, I disagree that all the TCCON stations will exhibit the same seasonal and daily variability, as it depends on their latitude, proximity to sources and type of sources impacting the sites. Consequently the curves to describe this variability might be different between stations. Therefore, I think it would be more valuable to describe the procedure to choose the optimal seasonal and daily curves for each site separately.* ”

The description is now expanded to **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.**.

The additional evaluation in the auxiliary material using sines and cosines via ccgfilt does not need a choice of polynomial degree. It gives results comparable to the polynomial fit. However the goal of this publication is to be as simple as possible, which is more easily achieved with polynomial fits. Therefore this additional evaluation is only in the auxiliary material.

The advantage of a global choice of the degree is that it makes it easy to compare the results between different sites and to directly apply the procedure to other sites.

"*3. Is it reasonable to assume divergence perpendicular to wind direction does not occur (pg 8, line 10), but vertical diffusion occurs to complete mixing (pg 6, line 21)? It would also be instructive to provide an example of A_{eff} along one wind trajectory. Also, a diagram to help describe the "spread" would be valuable for visualization (e.g. Figure 1, below). The arc length (A to B, green dashed curve in Figure 1) may overestimate the spread of Tokyo impacting Tsukuba. I suggest that using the cosine rule could be appropriate to estimate the Tokyo cross section measured by Tsukuba, which would in this case give 74.56 km (A to B, red line in Figure 1). Otherwise, please explain why the arc length is a better approximation of the spread.* "

The arc length is a better approximation, because it gives a constant scaling at each angle. However the uncertainty in the distance of the center of emissions is much larger than the difference between the two methods, so this distinction does not significantly change the outcome.

Note that as also written in the reply to RC1, the distance between TCCON site and the center of emission was mis-measured in the original manuscript. Re-measuring the distance using the coordinates of the TCCON site on Google maps gave a distance of only 52 km, which yields a orthogonal spread of 64km.

"*4. Instead of average column wind speed (pg 8, line 6), partial columns could be used, seeing as the scaling factors have been determined (Figure 4). Partial columns could also account for the rotation of the wind direction at higher altitudes (suggested on pg 13, line 9).* "

Partial columns would be an option, but would also complicate the evaluation. To give a robust improvement, it would also need better knowledge about the vertial distribution of emissions from Tokyo. This approach would therefore be better suited when using a full transport model as done in different inverse models for flux calculation.

Nevertheless this was now added as concrete option for improvement in the outlook: "correcting for the wind direction at different altitudes **by using partial columns**".

3.2 Minor Comments

"*I) Abstract mentions "greenhouse gas emissions", but here only CO 2 is the focus. In general, be consistent with using CO 2 emissions, because "carbon emissions" could mean total carbon emissions from CO 2 , methane, CO, VOCs, etc. unless otherwise defined. Also, I suggest you identify a "statistical-based approach" in the abstract.* "

We also looked at results using this same approach for methane and CO, showing that the method could also be used for other species than CO_2. This is now shown in section 3 of the auxiliary material.

"*II) Introduction*"

"*Pg 1, line 13: Clarify "fossil fuel burning emitters" to link this to Megacities like Tokyo. Also, what is the main contributor for Tokyo, vehicles, energy generation, or something else? Are these energy generation centers (e.g. coal-fired power plants) located within or outside the city? These explanations will prime the reader for why your method will work for a city like Tokyo.*"

This reads better, thank you! "The carbon dioxide footprint of large scale fossil fuel burning emitters **like powerplants or heating and personal transport in megacities,** has been retrieved from satellite ...".

Also the emission types are now noted in the last paragraph of the introduction as **The main emission sources of Tokyo are Transport, Residential and industry, with about half the emissions coming from the large coal and gas fired powerplants on the east side of Tokyo Bay, south-east of Tokyo city (Bureau of the Environment Tokyo, 2010).**.

"*Pg 1, line 20: The "long term changes" are not addressed in this paper, although I was expecting it from this introduction. Perhaps bring in a comment to imply that with longer measurements than at Tsukuba currently, long-term changes in emissions can be investigated.*"

Thank you for that catch, and thank you for your help to improve the writing. The introduction now ends with **This publication shows that emissions can be estimated from four years of data. Continued measurements will allow tracking the change in emissions.**.

"*Pg 2, line 1: Link the benefits of TCCON to the problems you have described previ- ously. That is, what does the "highly accurate, precise, multi-year total column" allow you to do?* "

This is now added to the the introduction, too: **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.**

"*III) Observations* "

“*Pg 2, line 9: What is mean by "best" - best precision? highest time resolution?* ”

They are the most precise and accurate. The paragraph is more precise now:

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

“*Pg 2, line 14: More information about the constant scaling factors (for what? why are they sometimes necessary?) and why you can ignore them.* ”

These scaling factors cannot be ignored, because the direct measurement of the wind speed is done close to the ground, but the wind speed are higher in the higher parts of the column. The effective wind speed section now starts with an intro to make this clearer:

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.

“*IV) Removing trend and annual cycles*
Figure 1: The yellow and magenta vertical lines are not necessary now the bound- aries are explained in the text. Also, are the data displayed day averages, individual measurements, etc.?”

The lines are not necessary but they give a visual indication of the bounds.

The data are individual measurements. This is now made clear in the text.

“*Pg 4, line 8: Add in how and why the degree 3 and degree 6 were chosen here, and why they are more appropriate than harmonics (which reflect orbital characteristics of seasons and days). Perhaps move lines 13-14 to here.* ”

This is now made clear via **Polynomials are used in this estimation to make the method as easy to implement as possible. The auxiliary material provides results from an alternate implementation using harmonics instead which gives comparable results.**

“*Pg 4, lines 9-12: Clarify the postulation about wind direction and daily cycle and the impact on the analysis.* ”

This is now made clearer via **since uncorrelated differences get reduced in statistical aggregation**.

“*V) Directional Dependence*
Figure 3: Some of the caption information could be moved to the main text. ”

This would be possible, but the explanations are so closely tied to the graph that they would be harder to follow if they were moved away from teh graph.

“*From pg 5, line 3 onwards I was unsure whether only the enhanced CO 2 concentra- tions were investigated further. If not, please clarify why the data is separated and then recombined.* ”

This is now cleared up via **The data in Figure 3 is separated into positive and negative to ease identification of the limits for emissions from Tokyo area. The quantitative evaluation uses both positive and negative residuals.**.

“*Pg 6, line 11: What does "most parcels in the lowest 2 km" mean for your analysis, e.g. does it support that any enhance-ments from those wind directions are likely due to Tokyo?* ”

This means that it suffices to use wind speed measurements within this range.

This is now clearer via **Therefore calculating the effective air speed of the column with enhanced concentrations only requires wind speed measurements in this part of the atmosphere.**.

“*Pg 6, lines 11-15: The radiosonde data information would be better in the observa- tions section. Also, the relevance of the radiosonde data is unclear, e.g. was it used to produce Figure 4?* ”

Yes. This is now noted explicitly: **Figure 4 visualizes the variability of the wind speed profile weighted by atmospheric pressure by aggregating the radiosonde data measured at the Tateno site.**.

“*Pg 6, line 19: What exactly is the volume of air assumed to contain enhanced CO 2 ? Is it the 0-1000 m described by Figure 4, or is it up to 2 km as described by the trajec- tories (line 11)?* ”

It is the 0-1000m. To make this clearer, the text now always uses the 0-1000m.

“*Pg 6, line 25: Is it possible to quantify the uncertainty due to mixing by comparing your uniform CO 2 assumption with an assumed vertical gradient?* ”

Now with the approach we are using. This would require aircraft profiles within the “exhaust plume” of Tokyo, which are not available. However this is also a good point for possible future work. It is now added to the outlook as **A better classification of uncertainty due to the assumption of uniform vertical distribution could be given by measuring highly resolved vertical profiles by aircraft downwind of Tokyo.**.

“*VI) Estimated source*
Figure 5: The vertical lines are very hard to see. Also, should it be "outside the urban influence" instead of "outside the background limits"?”

The lines are now wider and the limits are given as numbers, too. It means “outside the background limits”, because this is the definition of the background limits in the code.

"*The discussion on pg 9, lines 5-6 about vertical divergence is a little disjointed and requires clarification as well as including a relevance statement.* "

This sentence is gone now, since it repeated information already given in the effective wind speed section. Thank you!

"*Reorder pg 9, lines 12-16 to describe equations in the order they appear.* "

The calculations are re-ordered and clearer.

"*Pg 9, line 10: State the mean enhancement (126.4) here so the reader doesn't have to search.* "

Done - thank you!

"*Equation 9, What is t CO 2 ? Elsewhere you have use "t" for time. Also, there is an extra dot at the front of the equation.* "

t is tonnes. The use of t as index has been removed.

"*Pg 10, line 9: How does calculating 1 degree steps in the wind angle at Tsukuba mean you can compare with a gridded inventory? More description on this comparison methodology is necessary* "

This is now made clearer in section 7: **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):**.

"*VII) Estimating Uncertainties*
Pg 11, line 13: How are changing the "center of mass" calculations performed? Does the Tsukuba to Tokyo distance change? "

The distance in itself does not change, but it is unclear where the center of mass is located exactly. There are no calculations done, because that would require relying on another model which we want to avoid in this publication.

"*VIII) Comparisons*
Figure 6: Label the axes. Add boundaries showing summed regions for Tokyo and Background."

The axes are labelled now. However adding boundaries is problematic due to technical problems with the gdal library.

"*Figure 7: Need to add a second y-axis for the residuals from Figure 5.* "

The y-axis is the same for both datasets, therefore it should not need another y-axis.

"*Pg 13, line 12: Clarify what is meant by "every reproduction".* "

This is clarified now: "but that would then require every person reproducing the estimates from this study to run such a transport".

"*IX) Conclusions*
I suggest to mention that TCCON simultaneously measures other gases, so this method has the potential to be applied to other species."

This is added now: **and that this method can also be used to analyze other greenhouse gases measured by the TCCON network, including methane and carbon monoxide.**

"*Pg 14, lines 22-23: There is also potential drawdown from about 180 degrees at high wind speeds (> 40 m/s) from the forested peninsula.*"

Yes, but these are few datapoints.

3.3 Technical Corrections

"*Pg 2, Line 7: Do you mean this statistical method is "suitable for applying to long-term monitoring observations"?* "

We mean "suitable as a means to monitor emissions over longer time periods".

"*Pg 2, Line 9: TCCON currently provides the best* "

this is fixed: provides the most precise and accurate …

"*Pg 2, Line 14: this study -> our study* " changed.

"*Figure 2: Legend and text in Section 3 says the daily cycle is degree 3, Figure caption says degree 4.* "

Thank you! It's degree 3.

"*Pg 4, Line 8: The long-term trend is fitted* "

"*Pg 4, Line 12: supplement Fig. 1 of this paper* "

"*Pg 5, Line 3: actual extent of Tokyo impacted wind directions.* " applied.

"*Pg 6, Line 2: source of Tokyo (described further in section 5).* " thank you!

"*Figure 4, caption: including Ijima (2016). -> from the Ijima (2016) dataset.* " applied.

"*Pg 8, line 6: average column wind speed within the boundary layer (0-2000 m), v wind .* " looks good, tank you!

"*Pg 9, line 8: t aff -> A aff* " good catch!

"*Pg 11, line 3: ofd istances -> of distances* " fixed.

"*Pg 11, line 23: should the uncertainty be 33 not 38?* " yes, other fixes during review reduced it, though.

"*Pg 13, line 9: from the ODIAC model to*" switched to uppercase, but not adding model, since the expanded name already includes "inventory" and is noted before the abbreviation.

Thank you again for your review! It helped to improve the manuscript a lot!

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

Arne Babenhauserheide[1,*], Frank Hase[1], and Isamu Morino[2]

[1]IMK-ASF, Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany
[2]National Institute for Environmental Studies (NIES), Tsukuba, Japan
[*]now at Disy Informationssysteme GmbH, Karlsruhe, Germany

Correspondence to: Arne Babenhauserheide (arne_bab@web.de)

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 ~~$86 \pm 33 \frac{MtC}{year}$~~ $69 \pm 21 \pm 6 \frac{MtC}{year}$ 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.

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.

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 ~~(UNFCCC secretariat, 2015)~~ "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 ~~to reduce emissionsand stimulate research requires~~ 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 ~~(Bovensmann et al., 2010; Hakkarainen et al., 2016; Hammerling et al., 2012; Hakkarainen et~~ ~~and via~~ (Hakkarainen et al., 2016; Hammerling et al., 2012; Ichii et al., 2017; Deng et al., 2014; Nassar et al., 2017; Ye et al., 2017; Hedel and from ground based differential measurements using multiple mobile total column instruments ~~(Frey et al., 2015; Hase et al., 2015; Chen~~ (Frey et al., 2015; Hase et al., 2015; Chen et al., 2016; Butz et al., 2016; Vogel 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 ~~(i.e. van der Laan-Luijkx et al., 2017; Basu et al., 2011; Babenhauserheide et al., 2015; van der Velde et al., 2014b; M~~ (e.g. van der Laan-Luijkx et al., 2017; Basu et al., 2011; Babenhauserheide et al., 2015; van der Velde et al., 2014b; Meesters et al., 2012) , 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 ~~(i.e. Andres et al., 2011; Bureau of the Environment Tokyo, 2010; Peters and van der Laan-Luijkx, 2012; van der Velde et al., 2014a; Le Q~~ (e.g. Andres et al., 2011; Bureau of the Environment Tokyo, 2010; van der Velde et al., 2014a; Le Quéré et al., 2015, 2016).

The Total Carbon Column Observing Network (TCCON, Wunch et al., 2011; Toon et al., 2009), 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 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)~~using~~. 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 To . 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 ~~(2011-08-04 – 2016-03-30)~~ by the TCCON site at Tsukuba, Japan, along with radiosonde measurements of daily local wind profiles.

This method provides an ~~inexpensive~~ approach to estimate city emissions which is inexpensive when compared to satellite missions while 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 ~~provides the currently best~~ 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 ~~0.2~~0.3 ppm ~~(Wunch et al., 2011)~~ (Messerschmidt et al., 2010).

The stations of the TCCON-network measure the absorption of CO_2 ~~in the solar spectrum~~ and other molecular species using the sun as background radiation source (Wunch et al., 2011). Dividing ~~this absorption by the absorption of~~ the retrieved column amount of the target species by the co-observed column amount 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 ~~0.2ppm (0.2%)~~ 0.1% by Messerschmidt et al. (2011). Since ~~this~~ our study restricts itself to a single station ~~it can ignore constant scaling factors~~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.

~~This~~ 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 [1] ~~used in this study~~ 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. ~~Essential~~ Further information about the TCCON site in Tsukuba is available from the TCCON wiki[1] . [1] In addition to concentration data of trace gases, the station provides wind direction and speed measured at the rooftop of the observatory.

[1] ~~TCCON data is publicly available from~~

[1] ~~The TCCON wiki is located at~~

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

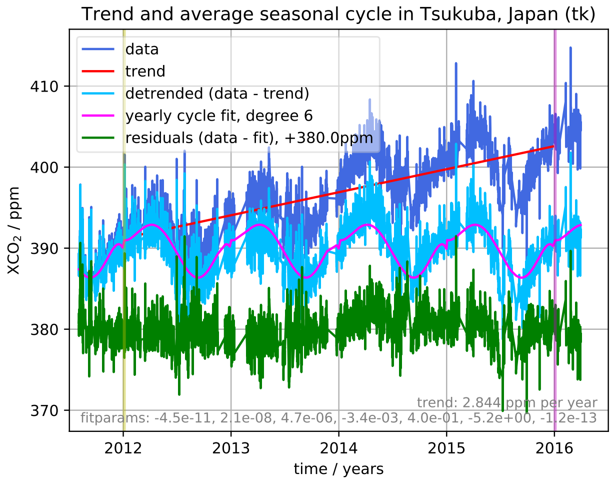

Figure 1. Detrending and deseasonalization of the XCO_2 total column measurements in Tsukuba, Japan. The data shown are individual measurements. The trend (shown as a red "trend" line) is removed with a linear least squares fit to the data from background directions between 2012-01-01 and 2016-01-01 (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.

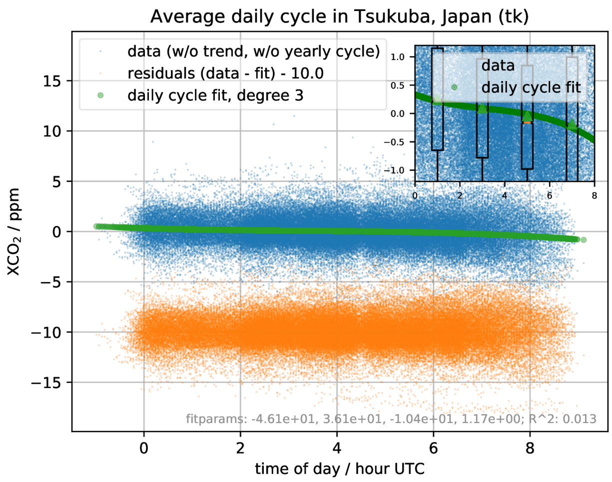

Figure 2. To remove a potential bias from correlation of wind direction ~~and~~ with daytime which would couple the signal from photosynthesis and respiration, the daily cycle is removed by fitting and subtracting a polynomial of degree ~~4.~~ 3. Degree ~~4~~ 3 was chosen empirically.

3 Removing trend and natural cycles

~~To estimate the~~ The approach chosen in this paper to estimate the CO_2 emissions of Tokyo ~~directly from the measurements~~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 ~~subject to strong~~ 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 ~~’s~~ 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 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 ~~time of day~~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 is available in the supplement of this paper). The programs are provided in the supplement of this paper~~; 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.~~ 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.

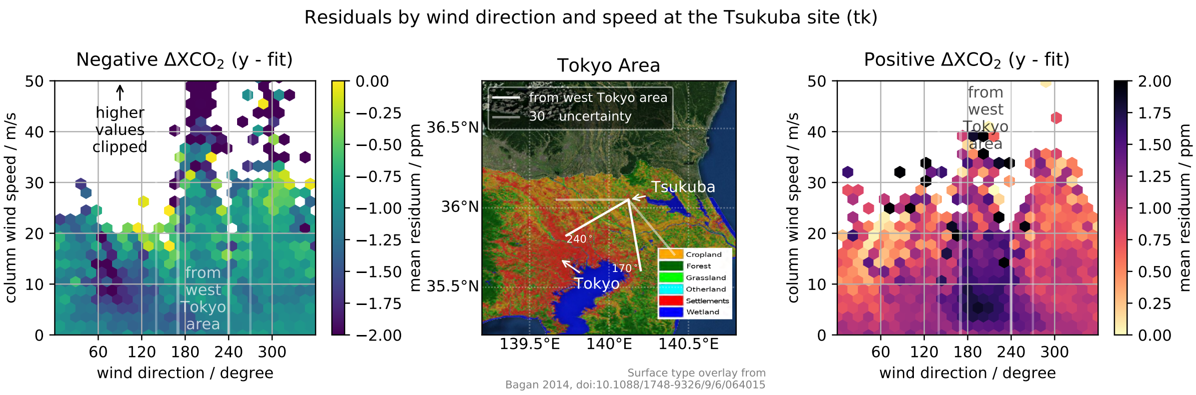

Figure 3. The **center graph** shows a map of Tokyo and its surroundings, retrieved from the ArcGIS REST service[2] with an overlay indicating the surface type. The colors in the overlay visualize land-use and settlement density (taken from Bagan and Yamagata, 2014). It clearly shows decreasing population 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 ~~form~~ 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. ~~Splitting the~~ Displaying residuals ~~into positive and negative values is done to simplify~~ which are lower than zero in a different graph than residuals which are higher than zero aids visual detection of emissions~~without interference from effective~~, because it separates the features of CO_2 sinks (lower than zero) from CO_2 sources (higher than zero). ~~In~~ The strongly negative values on the ~~following~~left graph at a wind direction around 60° might be due to biospheric drawdown of CO_2 by woodland, ~~detailed treatment~~ but since the focus of this publication are the ~~residuals uses both positive~~ 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 ~~2D~~

[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.

~~histogram~~ hexbin averages shown in the right panel in Figure 3. The data in Figure 3 is separated into positive and negative to ease identification of the limits for emissions from Tokyo area. The quantitative evaluation uses both positive and negative residuals. Within these directional delimiters, the interval with wind speeds between 5 and 15 ms^{-1} contains only bins with an enhanced concentration of CO_2. Perfect definition of these limits ~~isn't~~ is not possible in the 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).

4.1 ~~Effective wind speed~~

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 $\cancel{v_{wind}}$ v can be replaced by the density weighted average wind speed profile within the boundary layer~~, as seen in~~. To calculate the required altitude extension of this profile, forward trajectories from Tokyo ~~, calculated with hysplit (Rolph et al., 2017; Stein et al., 2015). These trajectories~~ for 5 to 15 hours were calculated with the HYbrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT, Stein et al., 2015) using 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. 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 ~~2km~~ 1000m of the atmosphere. ~~Radiosonde~~ Therefore calculating the effective air 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 available from radiosondes. Specifically, 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

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

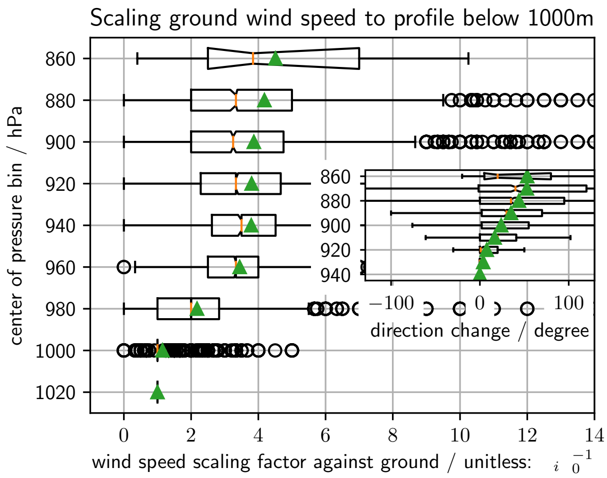

Figure 4. Wind speed profile statistics up to 1000 m at Tateno, Japan using data from 2009 to 2016, ~~including~~ from the Ijima (2016) dataset. Values are calculated by dividing the wind speed at a given pressure by the wind speed at the lowest level. The boxplots show the median (red line) and the 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.

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 and Hase (2018).

Figure 4 visualizes the variability of the ~~profile~~ wind speed profile weighted by atmospheric pressure by aggregating 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 and Hase, 2018) but provide a significant source of uncertainty, since their use rests on the assumption ~~on~~ of uniform mixing of the carbon emissions across the boundary layer. ~~Forward~~ The forward trajectory calculations with HYSPLIT ~~(Stein et al., 2015)~~ 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.

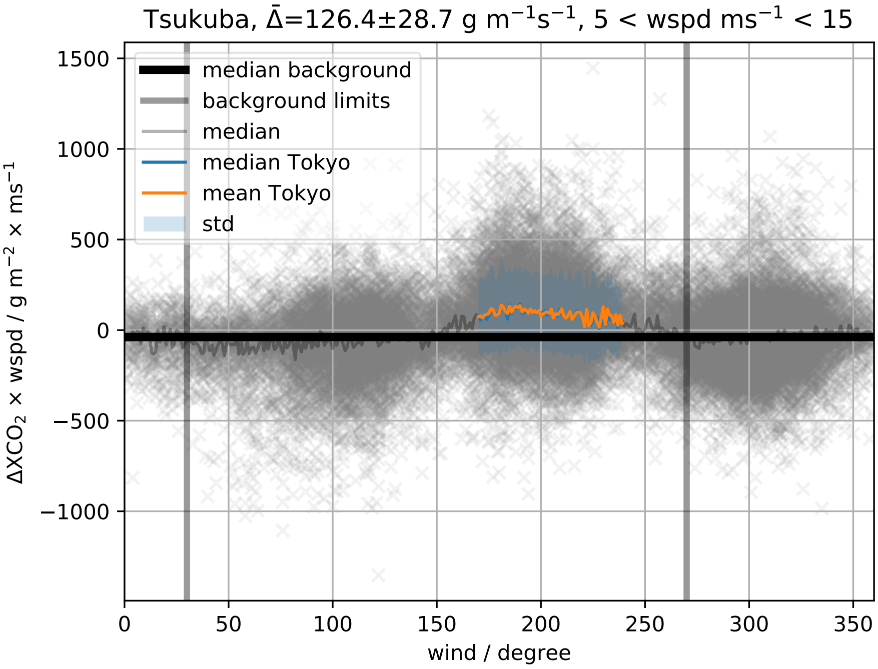

Figure 5. Residuals multiplied by wind speed plotted against the wind direction for scaled wind speeds between 5 ms^{-1} and 15 ms^{-1}; measured by the TCCON site in Tsukuba, Japan. The mean enhancement ~~$\bar{\Delta}$~~ $\bar{\Delta}$, the mean Tokyo and median Tokyo and the std are calculated for 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 a_t.

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

$$S_T = E_m \cdot A_{\mathit{aff}} \mathcal{A} \tag{1}$$

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	$m_{CO_2,\text{col}}$	$= \frac{M_{CO_2}}{M_{\text{air}}} \frac{m_{\text{air,col}}}{f_{col}} \cdot C_{CO_2,t,\text{col}}$
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)
enhancement:	E_m	$= R_{\text{from Tokyo area}} - \text{median}\,(R_{\text{from background}})$
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)
tracer mass	$m_{\text{gas,col}}[g]$,	
total column dry air mass	$m_{\text{air,col}}[g]$,	
column concentration	$C_{CO_2,t,\text{col}}$,	
acceleration due to gravity	$g[\frac{m}{s^2}]$	(from TCCON apriori),
pressure	$p[\frac{g}{ms^2}]$.	

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

$$A_{aff}\mathcal{A} = v_{\text{wind}} \cdot s_{\perp\perp}, \tag{2}$$

this source can be derived from the mean total column enhancement of XCO_2 $\bar{\Delta} = 126 \pm 29 \frac{g_{CO_2}}{ms}$ 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 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 ~~$s_{\text{Tsukuba–Tokyo}} \approx 65\text{km}$~~ $s_{\text{Tsukuba–Tokyo}} \approx 52\text{km}$. The city center of Tokyo was chosen to be at the palace ~~. Due to measuring total columns, vertical divergence only affects the height of the enhancement in the column and therefore the wind speed of air with enhanced concentrations at the measurement location~~(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 ~~79.4 km~~ $2\pi \cdot 52km \cdot \frac{70°}{360°} = 64km$.

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

$$E_m \cancel{A_{aff}} A = \int_{\cancel{\alpha}\alpha_0}^{\alpha_1} E_{m,\alpha} A_{aff,\alpha} d\alpha = \frac{s_\perp}{\Delta\alpha} \cdot \int_{\cancel{\alpha}\alpha_0}^{\alpha_1} E_{m,\alpha} v_{\text{wind},\cancel{\alpha}\alpha} d\alpha = s_\perp \cancel{\Delta_{CO_2}} \bar{\Delta} \tag{5}$$

~~Here $\bar{\Delta}_{CO_2}$ denotes the mean enhancement as depicted in Figure 5.~~

~~$\bar{\Delta}_{CO_2}$ is calculated from the XCO₂ residuals. The residuals are calculated by subtracting the trend and fits to the yearly and daily cycle following equation ?? as described in section 3. To calculate $\bar{\Delta}_{CO_2}$, the median total column residual from background wind directions (chosen as 310° to 50°, using 0° as from north by meteorological conventions) is subtracted from the target direction residuals (see equation ??), then the result is multiplied with the wind speed during the time of measurement. Finally it is converted from measured total column concentration $C_{CO_2,t,\text{col}}$ to total column mass $m_{CO_2,\text{col}}$ using equations ?? and ??.~~

$$\begin{aligned}
\text{unit air column mass:} \quad & m_{\text{air,col}} && = \frac{p_t}{g_{a_t,t}} \cdot 10^5 \left[\frac{g}{m^2}\right] \\
CO_2 \text{ column mass:} \quad & m_{CO_2,\text{col}} && = \frac{M_{CO_2}}{M_{\text{air}}} \frac{m_{\text{air,col}}}{f_{col}} \cdot C_{CO_2,t,\text{col}} \\
\text{total column residuum:} \quad & R && = m_{CO_2,\text{col}} - m_{CO_2,\text{col},\textbf{seasonal cycle fit}} - m_{CO_2,\text{col},\textbf{daily cycle fit}}; \text{ (described in section 3)} \\
\text{enhancement:} \quad & E_m && = R_{\text{from Tokyo area}} - \text{median}\left(R_{\text{from background}}\right)
\end{aligned}$$

~~at time t, level l and daily apriori a_t[4] with~~

~~tracer mass $m_{\text{gas,col}}$, total column air mass $m_{\text{air,col}}$, column concentration $C_{CO_2,t,\text{col}}$, molar masses $M_{CO_2} = 44.0\frac{g}{mol}$ and $M_{\text{air}=28.9\frac{g}{mol}}$, gravity $g_{a_t,t}$ (from TCCON apriori), pressure p_t.~~

~~[4] $f_{col} = 0.9975$ is a column mass correction following Bannon et al. (1997)~~

~~From~~ Therefore the source of Tokyo can be derived from the mean enhancement ~~$\bar{\Delta}_{CO_2}$ the source of Tokyo is derived~~ $\bar{\Delta}$ as

$$.S_T = \bar{\Delta}_{CO_2} \cdot s_\perp = 126 \pm 29 \frac{g_{CO_2}}{ms} \cdot 7940064000m = 10.08.1 \pm 2.31.9 \frac{t_{CO_2}}{s} \tag{6}$$

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:

$$S_{T,\mathrm{C,yearly}} = \bar{\Delta}_{CO_2} \frac{M_C}{M_{CO_2}} s_\perp \cdot \frac{s}{\mathrm{year}} \tag{7}$$

$$= 126 \pm 29 \frac{g_{CO_2}}{ms} \cdot \frac{12}{44} \left[\frac{g_C}{g_{CO_2}} \frac{g}{ms} \right] \cdot 7940064000m \cdot 31557600 \frac{s}{\mathrm{year}} \tag{8}$$

$$= 8669 \pm 20 \frac{Mt_C}{\mathrm{year}} 16 \frac{MtC}{\mathrm{year}} \tag{9}$$

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):

$$S_{\tau,\mathrm{CO_2,average,deg,monthly}} = \bar{\Delta}_{CO_2} \frac{M_C}{M_{CO_2}} s_\perp \cdot \frac{s}{\mathrm{month} \cdot \mathrm{degree}} \tag{10}$$

$$= 126 \pm 29 \left[\frac{g_{CO_2}}{ms} \right] \cdot \frac{12}{44} \left[\frac{g_C}{g_{CO_2}} \right] \cdot 1134914m \cdot 2592000 \frac{s}{\mathrm{month} \cdot \mathrm{degree}} \tag{11}$$

$$= 10181 \pm 23 \frac{kt_C}{\mathrm{month} \cdot \mathrm{degree}} 19 \frac{ktC}{\mathrm{month} \cdot \mathrm{degree}} \tag{12}$$

6 Estimating uncertainties

In addition to the statistical uncertainty and the uncertainty of the wind profile discussed in section 4.1, the estimated emission depends on the ~~selected~~ assumed extent of Tokyo area and is limited by the unknown actual distribution ~~ofd istances of~~ 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 ~~symmetric~~ a distribution of emission strengths ~~parallel to~~ 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)~~ Oda and Maksyutov (2011, 2016) show a similar extension towards the south~~east~~, and the power plants are southward of the palace. ~~If the~~ Assuming an uncertainty of 10 km for the distance between the "center of mass" ~~of the emissions were located 10 km further away from the site, this would yield a 15% increase of the estimate, in absolute numbers~~ ~~$\pm 13 \frac{MtC}{\text{year}}$~~. ~~This uncertainty always needs to be taken into account.~~ and the measurement site increases the uncertainty:

$$S_T = \bar{\Delta} \cdot s_\perp = \bar{\Delta} \cdot 2\pi \cdot 52 \pm 10 km \cdot \frac{70°}{360°} = \bar{\Delta} \cdot 64 \pm 12.3 km \tag{13}$$

$$= 126 \pm 29 \frac{g_{CO_2}}{ms} \cdot 64000 \pm 12300 m = 8.1 \pm 2.4 \frac{t_{CO_2}}{s} \tag{14}$$

$$\Rightarrow 69 \pm 21 \frac{MtC}{\text{year}} \tag{15}$$

$$\Rightarrow 81 \pm 24 \frac{ktC}{\text{year,degree}} \tag{16}$$

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 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, this calculation yields $\bar{\Delta} = 137 \pm 31 \frac{g_{CO_2}}{ms}$ as source instead of the $126 \pm 29 \frac{g_{CO_2}}{ms}$ found with polynomial fits. This corresponds to a relative difference of 8.7% which is not captured by the internal variability of the residuals. For $69 MtC$, the absolute difference is $\pm 6 MtC$. 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

$$\text{Yearly carbon emissions of Tokyo:} \quad S_{T,\mathrm{C,yearly}} = 8669 \pm 2016 \pm 135 \pm 6 \frac{MtC}{\text{year}} \tag{17}$$

$$\text{Monthly carbon dioxide emissions of Tokyo:} \quad S_{\tau,\mathrm{CO_2,average,deg,monthly}} = 10181 \pm 2318 \pm 156 \pm 7 \frac{ktC}{\text{month} \cdot \text{degree}} \tag{18}$$

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 ~~$74 \pm 38 \frac{MtC}{\text{year}}$~~ $59 \pm 18 \pm 6 \frac{MtC}{\text{year}}$.

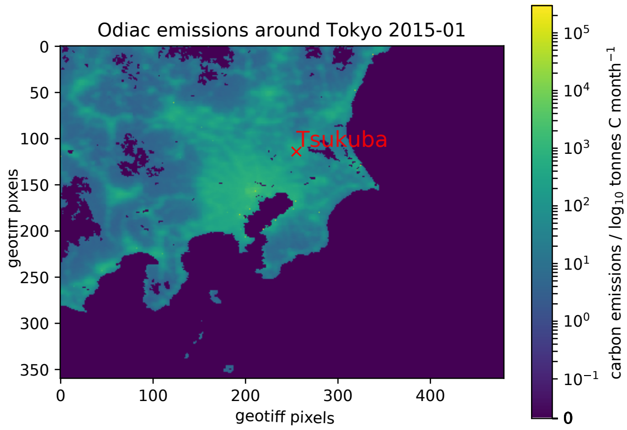

Figure 6. ~~Odiac~~ ODIAC carbon emissions per ~~1km×1km~~ 1km × 1km in pixel for January 2015 in $\log_{10}$ scale. This graph is created directly from the 1km×1km ~~Odiac~~ ODIAC dataset ~~(Oda and Maksyutov, 2011)~~ (Oda and Maksyutov, 2011, 2016) to visualize its structure. The unit is tonne Carbon/cell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

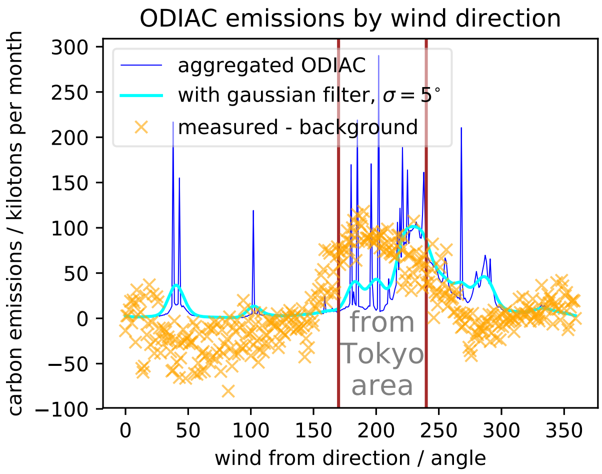

Figure 7. Sum of ~~Odiac~~ ODIAC carbon emissions by direction as seen from Tsukuba, Japan. The "aggregated ~~odiac~~ODIAC" emissions show emissions per direction from beginning of 2011 to end 2016. The gaussian filter data uses a moving average with gaussian weight to estimate signals measured at a 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 ~~Odiac emission inventory (Oda and Maksyutov, 2011),~~[4] Open-Data Inventory for Anthropogenic Carbon dioxide emission inventory (ODIAC, Oda and Maksyutov, 2011) in version ODIAC2016 (Oda and Maksyutov,

[4] ~~Odiac data retrieved from~~

, using the regional slice shown in Figure 6. Measurements are simulated from ~~Odiac by summing over all emissions~~ 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 ~~as (emissions by angle are shown in Figure 7), and then subtracting~~ 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):

$$\frac{1}{5}\sum_{t=2011-01}^{2015-12}\left(\sum_{\alpha}^{\text{Tokyo}} E_{\underline{odiac,t,\alpha}\,ODIAC,t,\alpha} - \sum_{\beta}^{\text{bg}} E_{\underline{odiac,t,\beta}\,ODIAC,t,\beta}\right) = 40.4\frac{MtC}{\text{year}}, \tag{19}$$

which is around ~~half~~ 60% of the emissions estimated in this paper from TCCON measurement data and within two standard deviations ($\cancel{2\sigma}\sigma$) of the estimated emissions. With the scaling for the time of day of the measurement, ~~Odiac~~ 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 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 ~~reproduction~~ person reproducing the estimates from our study to run such a transport, which would defeat the purpose of ~~this~~ 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 ~~larger~~ large discrepancy to our results. This discrepancy ~~which likely stems~~ could stem from different definitions for the source area. Part of this discrepancy cannot be reconciled, because the method shown in this paper cannot limit the emission aggregation parallel to the wind direction and has around 30° uncertainty of the direction, so it also includes emissions from Kanagawa, Saitama, and Chiba, the prefectures around Toko which are part of the greater Tokyo area.

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 ~~86 ± 33 megatons~~ 69 ± 21 ± 6 mega-tonnes carbon per year found for Tokyo has less than ~~40~~50% uncertainty despite our intentionally chosen 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) 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. This can complement global source and sink estimates and improve acceptance of carbon trading programs by enabling independent verification of findings.

~~The~~ Significant reduction of the uncertainties in these estimates ~~could be reduced further by~~ without adding more measurement stations would require taking into account more detailed wind fields from meteorological models, ~~by~~ correcting for the wind direction at different altitudes ~~and by~~ 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 ~~(i.e. van der Laan-Luijkx et al., 2017; Riddick et al., 2~~ ~~, therefore we restrict ourselves~~ with biosphere models (e.g. van der Laan-Luijkx et al., 2017; Riddick et al., 2017; Massart et al., 2014; Ba , therefore this implementation keeps close to the simpler evaluation which allows ~~us to stick closely to~~ 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 could be given by measuring highly resolved vertical profiles by 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 ~~lower~~ coarser spatial resolution as well as long term datasets. ~~The provided yearly emission estimates~~

To reduce 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 by calculating the diurnal scaling of the emission source from CO_2 concentration measurements of an in-situ instrument or to take 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.

We conclude that long-term ground-based measurements of column-averaged greenhouse gas abundances with sufficient accuracy for detecting the signals of local emission sources are a 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. See the README in the auxiliary material for usage information. The 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/ccgcrv/.

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 they are working on other projects with ground-based total column measurement instruments.

Acknowledgements. Large parts of the inspiration for this method of evaluation and of the boldness to keep it simple are due to our treasured colleague Dr. Friedrich Klappenbach (especially his evaluation of CO_2 in Klappenbach et al., 2015) and the simple estimate of effective boundary layer wind speed from radiosonde data was suggested by Dr. Bernhard Vogel. Matthias Frey contributed insights into differential measurements of the Tokyo source using multiple portable spectrometers, as well as fruitful discussions about these evaluations. Support for this study was provided by the Bundesministerium für Bildung und Forschung (BMBF) through the ROMIC project, with funding for initial work provided by the Emmy-Noether program of the Deutsche Forschungsgemeinschaft (DFG) through grant BU2599/1-1 (RemoteC).

The article processing charges for this open-access publication have been covered by a Research Centre of the Helmholtz Association.

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