Point-to-point responses

We appreciate the careful read-through and constructive comments from both reviewers. The reviewers' comments are copied in **black** with our point-to-point responses in **blue** and revised text in **red** or copied.

Response to reviewer #1:

General comments:

- Interesting and relevant addition to existing literature.
- Comprehensive description of the methodology, including many relevant figures detailing steps in the process. Clearly a substantial effort was put into the work.
- Discussion of the results (section 3.2) is somewhat limited in depth compared to the rest of the study.
- Good discussion of the limitations. However, one potentially relevant limitation is not covered (see major specific comments).
- Overall, paper is well written. The writing in some sections could still be improved by a thorough read through (e.g. section 4.3).

We truly thank reviewer #1 for the recognition and constructive comments and attempted to revise the manuscript by

1) adding sensitivity analyses to examine how changes in hourly emissions between OCO-3 and TROPOMI overpass times may affect the enhancement ratio. We now analyze the column data from TCCON at Caltech
2) clarifying the relevant text to avoid confusion, especially the important point on the interpretation of combustion efficiency that varies significantly with regions, technologies, and the type of activities.

Major specific comments:

The authors combine satellite data on XCO and XCO2 that are collected at different overpass times. The authors have identified 3 factors that may complicate the combination of this data into accurate ER_{CO} values, which are discussed at length and properly considered in the data processing and interpretation. *However, it appears one potentially relevant factor is missing, namely the timing of the CO and CO2 emissions themselves. While possibly less relevant for industrial sources, other emission sources, such as road transport and residential combustion, may follow a specific temporal pattern where a time difference of 1- 3 hours could lead to a mismatch in observed XCO and XCO2 even when wind conditions remain relatively constant.* Example: XCO2 is observed during rush hour and XCO two hours later. This could add to the uncertainty in derived ER_{CO} values when overpass time differences are larger but remain < 3 hours.

We agree with reviewer #1 that the mismatch in the timing of CO or CO_2 emissions themselves may influence the ER_{CO}, although the overpass time difference has been limited to 1-3 hours. One can likely rely on bottom-up emission inventories or prior assumptions to infer hourly emission patterns. However, because emissions or combustion characteristics are what we are solving for, it is quite challenging to properly account for such mismatch WITHOUT involving prior assumptions towards emissions themselves. Such hourly emission variations depend on the relative contributions from individual emission sectors for a given city.

We have now carried out a sensitivity analysis to investigate how such a mismatch in emission timing may affect the ER_{CO}, or more generally the high-frequency variability of ERCO (as reviewer #2 also mentioned). Thanks to the TCCON network that provides high-frequency XCO and XCO_2 measurements (TCCON 2022). We utilized the latest GGG2020 version with several upgrades including a much-improved prior profile. Because XCO and XCO_2 are simultaneously retrieved, we can assume that atmospheric transport associated with two species are the same and their enhancement ratio fully reflected the emission characteristics once species-specific averaging kernel profiles are corrected for following Appendix A2 in Hedelius et al. (2018).

Here we reported the estimated enhancement ratio at Caltech with background observations from another TCCON site outside the LA basin (see figure and figure caption shown below). Because the temporal frequency of the Caltech TCCON site may not perfectly match that of the background TCCON site, we first averaged observations from both sites to each 20-min interval and calculated the enhancements and enhancement ratio. Indeed - the variation in ER_{CO} can be large throughout the day. Note that such variations may reflect not only the change in FF emissions (e.g., due to traffic) but also measurement noise (given these high-frequency data) as well as changes in the meteorology throughout the day (ocean versus mountain flow for LA). Nevertheless, changes in ER_{CO} between OCO-3 and TROPOMI times inferred from concurrent TCCON measurements appear to be small.

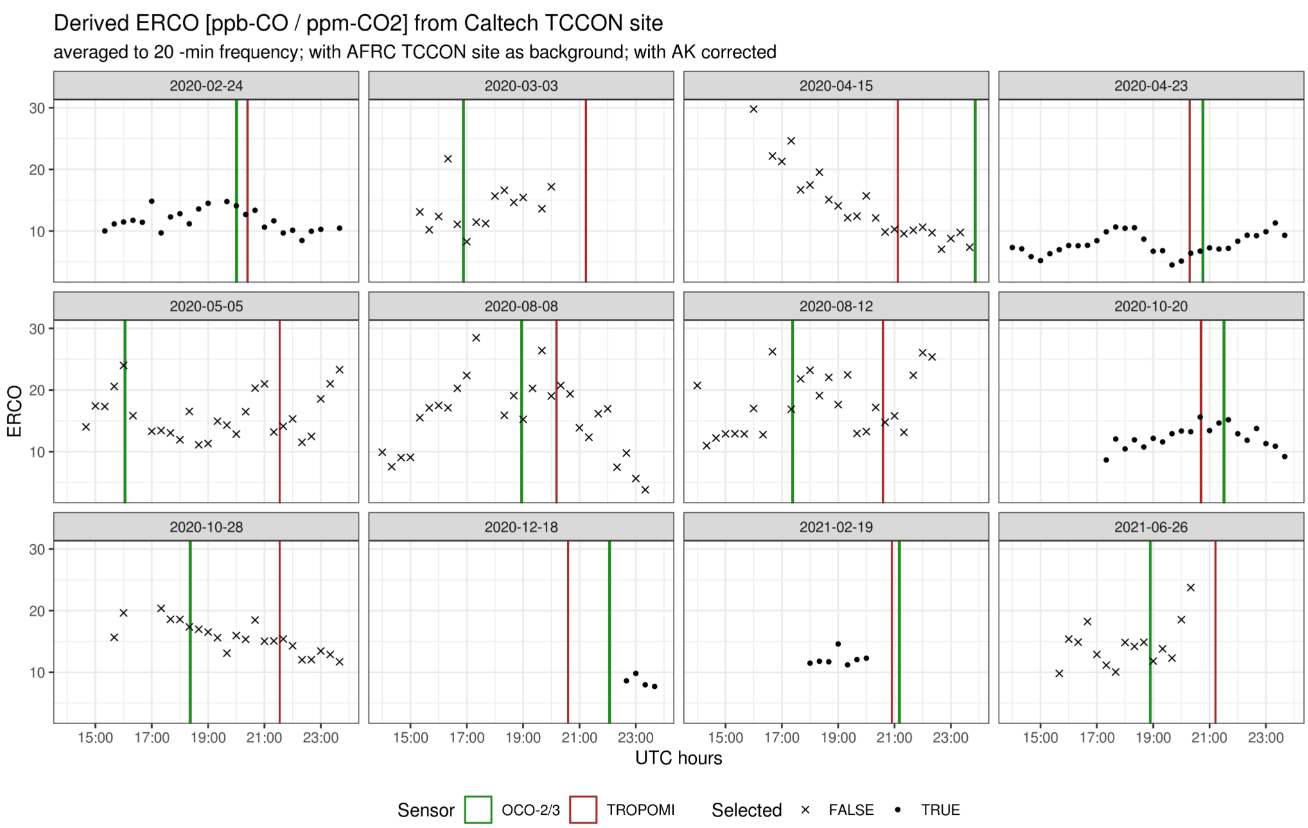


Fig. S8 - Time series of observed ERCO at the California Institute of Technology (Caltech) TCCON site (Wennberg et al. 2017) on the OCO-3 overpass dates till June 2021. The background is defined using the NASA Armstrong Flight Research Center (AFRC) site near Lancaster, California (Iraci et al., 2022). Column enhancements with corrections of TCCON averaging kernel are calculated following Hedelius et al. (2018). The overpasses that went into the final result are shown in solid black dots, while the remaining overpasses with significant plume shift between OCO-3 and TROPOMI overpass times are shown in black crosses. The vertical lines indicate the OCO-3 (green) or TROPOMI (red) overpass times in UTC times. The day of the week for each date is shown as follows: Feb 24, 2020 (Mon), Mar 3 (Tues), Apr 15 (Wed), Apr 23 (Thurs), May

5 (Tues), Aug 8 (Sat), Aug 12 (Wed), Oct 20 (Tues), Oct 28 (Wed), Dec 18 (Fri), Feb 19, 2021 (Fri), and June 26, 2021 (Sat). Note that no qualified data exists during the overpass time of OCO-3 or TROPOMI on April 1, 2020. The TCCON data were obtained from the TCCON Data Archive hosted by CaltechDATA at https://tccondata.org. We thank Laura T. Iraci and Coleen M. Roehl for preparing the TCCON data for these two sites.

Even though many more analyses related to the hourly or seasonal ER_{CO} can be drawn from these TCCON observations, they are out of the main goal of the current manuscript in using space-based observations. Since these TCCON and spatially resolved satellite observations (OCO-3-TROPOMI) may have a different emphasis on the emission signals within the LA basin, we tried to avoid unnecessary direct comparisons of their derived ER_{CO} and decided to leave the analysis in the supplementary material but added some discussions in

Sect. 2.1.3:

3. *Overpass times, meteorological conditions, and emission variations:* As a result of the overpass time difference between sensors, variations in meteorological conditions (e.g., wind direction and speed) can lead to changes in urban plume shapes detected by the two sensors as they pass by. We dealt with changes in wind speed and wind direction separately. The former is resolved by the "scaling factor" inferred from an atmospheric transport model and the latter undergoes manual evaluations (**Sect. 3.1**). Besides, CO and CO_2 emissions themselves can vary over the course of a day, e.g., driven by road transportation and residential sectors. Given the overpass time difference between sensors, it is likely that such mismatch in the timing of CO versus CO_2 emissions may affect the observed ER_{CO}.

Sect. 3.1:

Besides changes in wind directions, CO and CO_2 emissions themselves can vary across daytime hours, likely driven by the road transportation and residential sectors. As a result, variations in the derived ER_{CO} across multiple overpasses may reflect not only the variation in combustion efficiencies but also the mismatch in the emission timing. LA may be one of the cities with more distinct daytime changes in emissions compared to industry-centered cities. Fortunately, based on a supplementary sensitivity analysis using measurements from the Total Carbon Column Observing Network in Pasadena (TCCON, Wennberg et al., 2017), by limiting satellite overpasses to those with a smaller time difference, ER_{CO} appear to be less variable (**Supplementary Fig. S8**). Future geostationary satellite monitoring NO_x (e.g., TEMPO, Chance et al., 2022) may provide better guidance towards the hourly pattern in urban emissions, especially from the traffic sector with more daytime fluctuations, which have been discovered using surface monitoring networks (e.g., over Chicago; de Foy, 2018).

And Sect 4.1:

"If TROPOMI pixel sizes are relatively large (i.e., non-nadir observations) or the wind is steadier, this dt constraint may be relaxed, as long as emissions for a specific city is less driven by sectors with noticeable diurnal cycle (e.g., road transportation)."

Reference:

Hedelius, J. K., Liu, J., Oda, T., Maksyutov, S., Roehl, C. M., Iraci, L. T., Podolske, J. R., Hillyard, P. W., Liang, J., Gurney, K. R., Wunch, D., and Wennberg, P. O.: Southern California megacity CO2, CH4, and CO flux estimates using ground- and space-based remote sensing and a Lagrangian model, Atmos. Chem. Phys., 18, 16271–16291, https://doi.org/10.5194/acp-18-16271-2018, 2018.

Wennberg, P. O., D. Wunch, C. Roehl , J.-F. Blavier, G. C. Toon, N. Allen. 2017. TCCON data from California Institute of Technology, Pasadena, California, USA, Release GGG2020R0. TCCON data

archive, hosted by CaltechDATA, California Institute of Technology, Pasadena, CA, U.S.A. https://doi.org/10.14291/tccon.ggg2020.pasadena01.R0

Iraci, L., J. Podolske, C. Roehl, P. O. Wennberg, J.-F. Blavier, N. Allen, D. Wunch, G. Osterman. 2022. TCCON data from Armstrong Flight Research Center, Edwards, CA, USA, Release GGG2020R0. TCCON data archive, hosted by CaltechDATA, California Institute of Technology, Pasadena, CA, U.S.A. https://doi.org/10.14291/tccon.ggg2020.edwards01.R0

Minor specific comments:

Abstract

15: “After removing those cases...” is the exclusion of cases only on the basis of a t > 3 hours’ time difference or also separately on significant wind direction or plume shape changes?

We examined the plume shape for every overpass (e.g., Figure 5) and remove those whose plume shapes differ significantly (i.e., those overpasses with an asterisk in Figure 8). Thus, it is possible that cases with *dt* > 3 hours are included in the final results because the plume shift is negligible compared to the TROPOMI footprint size. A time difference of 3 hours is summarized here to provide a very conservative threshold if one would like to examine OCO-3 and TROPOMI overpasses for other places.

18-19: This statement seems incorrect. If I understand correctly, a low ER translates to a higher combustion efficiency (… of heavy industry in LA compared to the city-wide value). In the comparison between cases, it would be preferable to talk either of the combustion efficiency or the ER, and not compare the combustion efficiency with the ER as this may lead to confusion.

We apologize for the careless mistake and now just use ERCO when comparing ERs from heavy industry versus the whole city. Right - lower ERCO corresponds to higher combustion efficiency. We attached the relevant text here:

“Results suggest that ER_{CO} impacted by the heavy industry in Los Angeles is slightly lower than the overall city-wide value (< 10 ppb-CO / ppm-CO_2).”

1 Introduction

37: This statement is a bit tricky. In my opinion, for greenhouse gas emissions, the only key solution would be avoiding combustion of fossil fuels altogether. For some air pollutants such as NOx, higher thermodynamic efficiencies may actually be accompanied with higher emissions. For many air pollutants, abatement technologies such as particulate filters of catalytic converters are more successful in reducing emissions than increasing the combustion efficiency.

We agree with the reviewer and have now revised the relevant text as follows:

“Given the co-benefits of GHG reduction and improved air quality at various scales (Zhang et al., 2017), controlling the consumption of fossil fuels altogether is the key.”

2 Data and methodology

Figure 2: “FF XCO_2 enhancements...”. Have the concentrations in these images indeed been corrected for the background/non-FF sources?

The constant background values like Xbg_{co2} and Xbg_{co} in Equations 1 & 2 were subtracted from the total columns, but not the gradients from non-FF sources (i.e., delta X-terms). $XCO_{2.ff}$ enhancements with corrections for non-FF sources were displayed in **Fig. S5a**. To improve the consistency, we have now changed **Fig. 2** to further exclude the non-FF gradient following Equations 1 & 2 and added the $XCO_{2.ff}$ with or without non-FF sources in **Fig. S5** to show the differences. Here are the updated figures for LA on Feb 24, 2020.

Fig. 2

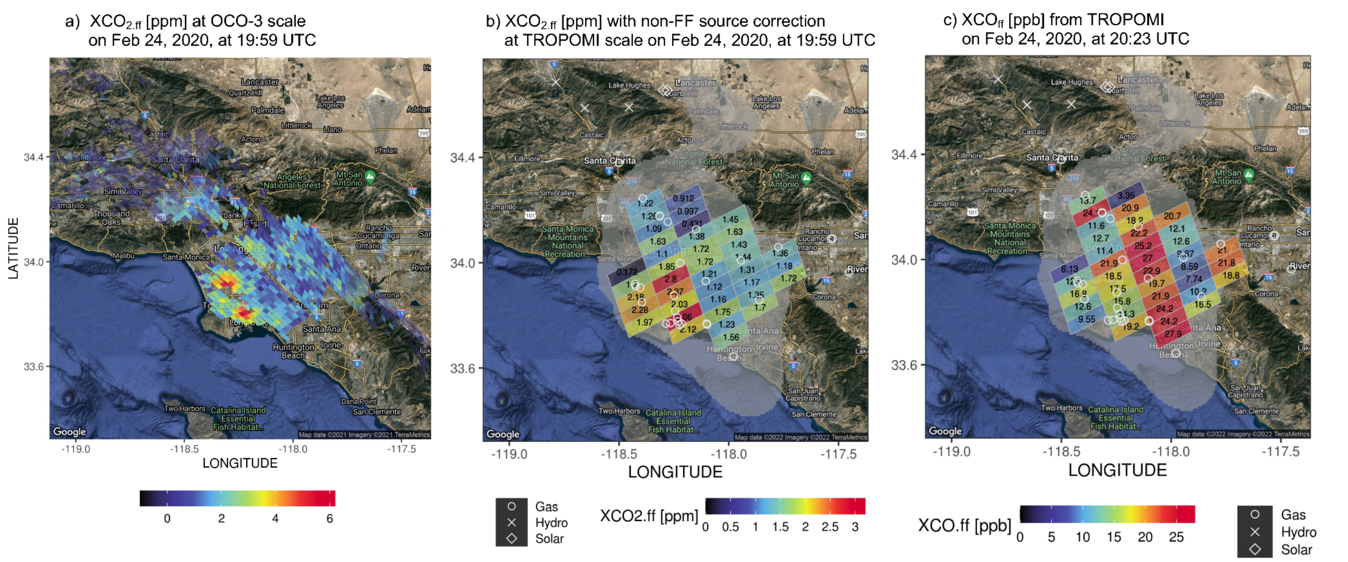
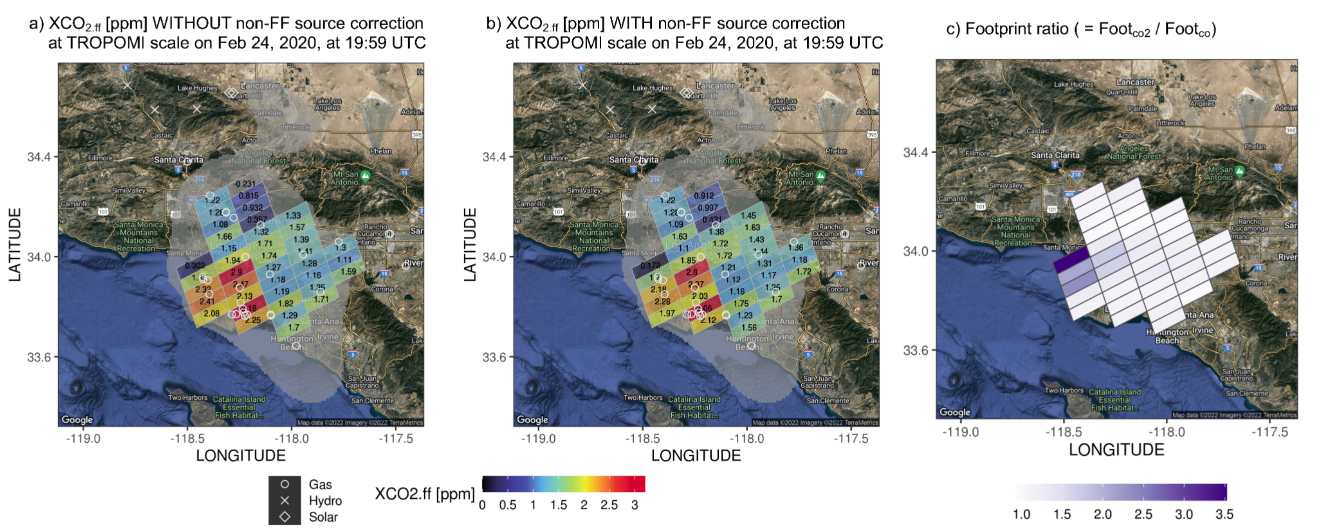


Fig. S5 In general, $XCO_{2.ff}$ within the urban plume with biospheric corrections appears to be slightly smaller than those without the biospheric corrections, as explained in the main text.



216-217: None of these 3 methods involve prior information on emissions, correct?

Correct - this statement has now been removed. For clarification,

- The first statistical method utilized standard deviation or percentiles to derive the background purely from observed XCO_2. So, no use of prior emissions.

- The second method uses modeled initial conditions from atmospheric transport models and global CO_2 fields. Global CO_2 fields may involve prior assumptions on emissions.
- The third method first considered atmospheric transport and identified the background region (but not too far from the target city) and then calculated the median of the observed XCO_2 in the background region as the background. An assumption on the rough spatial extent of the city emission (e.g., a rectangle).

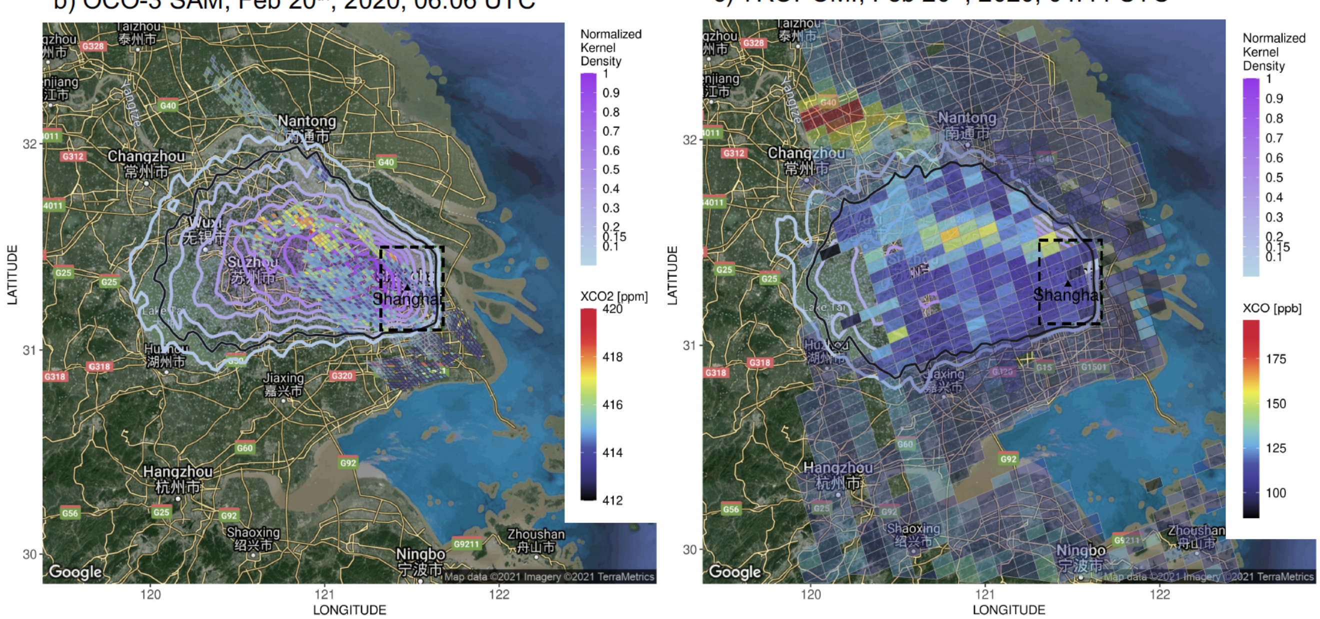
231-232: How does this method exclude observations elevated by another city? Simply by spatially limiting the plume definition sufficiently to avoid interference from another city?

This is a good point. Yes - the spatial limiting of the plume domain is realized by the particle distribution and the normalized kernel density function is sufficient to avoid interference from another city in most cases. Also, we only selected the soundings over regions from which the wind is blowing (e.g., to the south or east). With the median statistics for the background value, high XCO or XCO_2 values affected by a potential nearby city would play a minor impact on the background.

It is possible to conduct manual checks by releasing particles from a nearby city (e.g., the city of Nantong to the north of Shanghai) and outlining a similar urban plume for a nearby city. Alternatively, if several cities are so lumped together (e.g., three major cities in the Pearl River Delta in China, Ye et al., 2020), the entire metropolitan area can be treated as a whole entity for enhancement or emission estimates. Nevertheless, we may argue that the current approach is sufficient to minimize the influence of background from another city.

b) OCO-3 SAM, Feb 20th, 2020, 06:06 UTC

c) TROPOMI, Feb 20th, 2020, 04:44 UTC



Ref: Ye, X., Lauvaux, T., Kort, E. A., Oda, T., Feng, S., Lin, J. C., Yang, E. G., and Wu, D.: Constraining Fossil Fuel CO2 Emissions From Urban Area Using OCO-2 Observations of Total Column CO2, Journal of Geophysical Research: Atmospheres, 125, e2019JD030 528, 2020.

3 Results

Section 3.1: Here a discussion of a potential interference of changes in the emission of CO2 and CO in the target area between two overpasses that may be up to 3 hours apart would seem appropriate (see major comments).

As explained above, we have added an additional analysis and discussion in Sect. 3.1 (also pasted below) to investigate the impact of changes in hourly emission of CO_2 and CO onto observed ER_{CO}

Besides changes in wind directions, CO and CO_2 emissions themselves can vary across daytime hours, likely driven by the road transportation and residential sectors. As a result, variations in the derived ER_{CO} across multiple overpasses may reflect not only the variation in combustion efficiencies but also the mismatch in the emission timing. LA may be one of the cities with more distinct daytime changes in emissions compared to industry-centered cities. Fortunately, based on a supplementary sensitivity analysis using measurements from the Total Carbon Column Observing Network in Pasadena (TCCON, Wennberg et al., 2017), by limiting satellite overpasses to those with a smaller time difference, ER_{CO} appear to be less variable (**Supplementary Fig. S8**). Future geostationary satellite monitoring NO_x (e.g., TEMPO, Chance et al., 2022) may provide better guidance towards the hourly pattern in urban emissions, especially from the traffic sector with more daytime fluctuations, which have been discovered using surface monitoring networks (e.g., over Chicago; de Foy, 2018).

319-320: Not clear why the effect of biogenic and pyrogenic contributions itself is limited by removing overpasses interfered by wind shift.

We apologize for the confusing text that sounds like we suggested certain causality between the two. The effect of non-FF contributions will not be minimized by removing overpasses with significant plume shifts. The non-FF contributions were only estimated for tracks with insignificant plume shits, which turns out to be minimal (but may not be small for all overpasses and all cities). To clarify, we have modified the text to be:

> "For the final 24 overpasses we selected, temporal variations in the emission pattern and urban-background gradients in biogenic/pyrogenic contributions play minor roles in overpass- or city-level ERs."

348-349: Urban-background gradients in biogenic anomalies would not change FF (fossil fuel) enhancement by definition.

Right - the non-FF sources (namely their urban-background gradients) will not alter FF enhancements since FF enhancements have already been accounted for (Eqs. 1&2). We just would like to clarify that our definition for the background is quite unique, that is a constant value for a given satellite swath. Such *constant background* is derived based on observations of soundings often over the rural areas. While using such constant background to derive the enhancements may fail to account for the urban-rural or urban-background gradients in non-FF sources. We have now revised the text as:

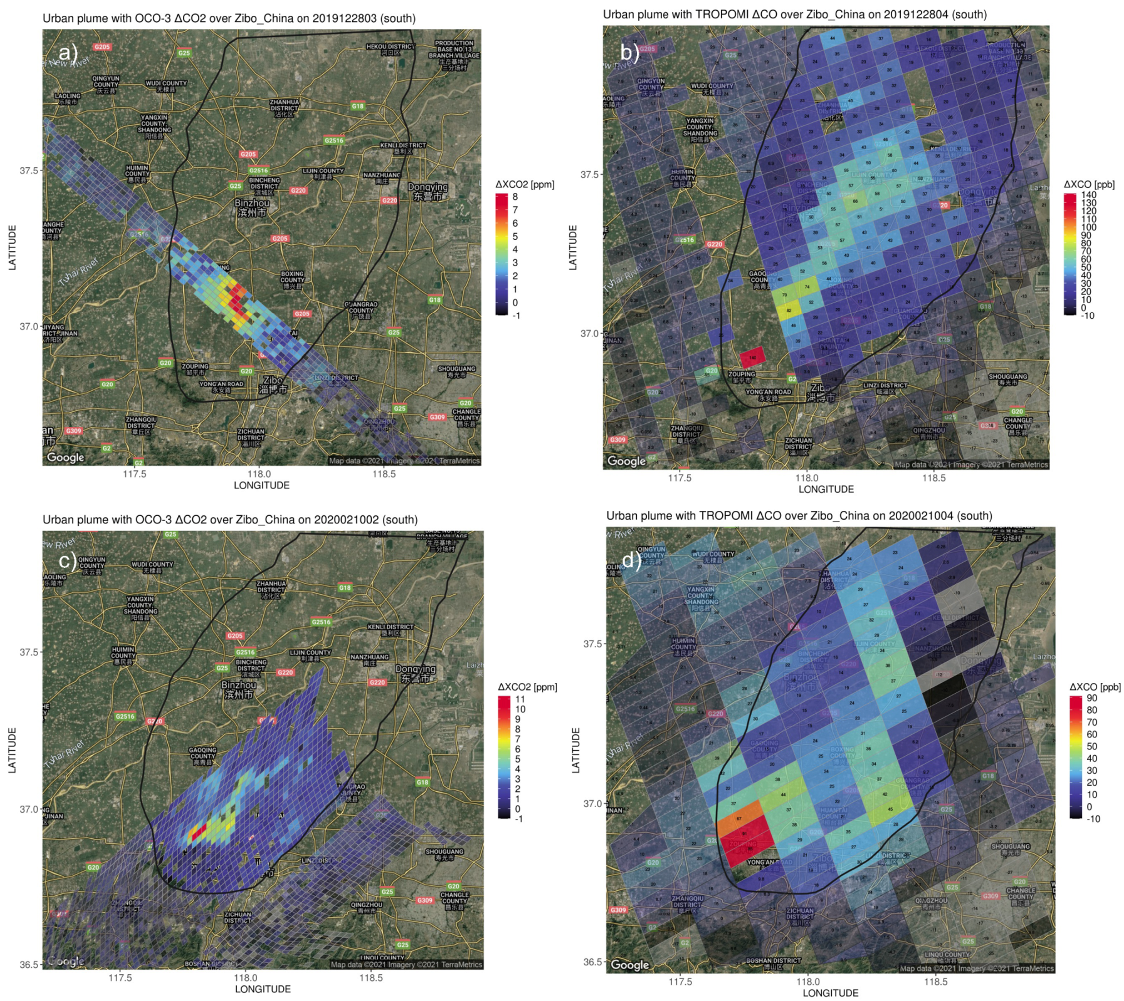
> "As explained in **Sect. 2.2.2**, urban-background gradients in these biogenic anomalies (i.e., δX_{bio}) were used to correct the constant background X_{bg} (Eq. 1)."

$$\langle E_{\mathrm{CO_2,s}}\rangle = \frac{\mathrm{X_{ffCO_2,s}}}{\langle \mathrm{XF_{CO_2,s}}\rangle} = \frac{\mathrm{X_{obsCO_2,s}} - \mathrm{X_{bgCO_2}} - \delta \mathrm{X_{bioCO_2,s}} - \delta \mathrm{X_{bbCO_2,s}}}{\iint \mathrm{XF_{CO_2,s}(x,y)\,dx\,dy}}. \tag{1}$$

$$\langle E_{\mathrm{CO,s}}\rangle = \frac{\mathrm{X_{ffCO,s}}}{\langle \mathrm{XF_{CO,s}}\rangle} = \frac{\mathrm{X_{obsCO,s}} - \mathrm{X_{bgCO}} - \delta \mathrm{X_{bbCO,s}}}{\iint \mathrm{XF_{CO,s}(x,y)\,dx\,dy}}. \tag{2}$$

382: While it is clear for the enhancements, why the lockdown should influence ER_{CO} in a specific direction is not directly clear. Perhaps comment on a potential mechanism here. Also, for Zibo the enhancements on 2020-02-11 appear larger than on 2019-12-28.

These are good questions. According to the new figure shown below, it seems that XCO2 enhancements for 2020-02-10 are larger than those for 2019-12-28, while XCO enhancements for 2020-02-10 are generally smaller than those for 2019-12-28. The wind directions between two cases are very similar (implied by the urban plumes in solid black curves). The much larger XCO2 enhancements for 2020-02-10 may be due to differences in sampling locations (e.g., 2019-12-28 case is farther away from the city source). Additionally, because 2019-12-28 is not a SAM and there are some missing TROPOMI polygons, only a few TROPOMI polygons (that overlap with the narrow swath of OCO) are used for calculating ER_{CO}. Thus, ECO_2 in **Fig. 6** for 2020-02-10 appears to be larger than that for 2019-12-28.



Considering differences in the satellite sampling (i.e., locations and available numbers of soundings), we may argue that evaluating ER_{CO} between overpasses would be more robust in evaluating FF enhancements between overpasses. This is also true for our meteorology-normalized emissions (E = XFF / XFootprint) since the model-based meteorology (X-STILT footprint) may be associated with transport uncertainties.

We suspect the drop and then rise in ER_{CO} is tied to the power generation and metal industry around Zibo. Zibo with its neighborhood city Zouping is responsible for ⅛ of the total coal consumption of its entire Shandong province in 2017. According to the global power plant dataset (WRI, 2018, new **Table 1** showen below), the total power generation capacity is around 9720 mW (100% fueled by coal), which is more likely to support the local metal industry (e.g., electrolytic aluminum and iron and steel), as the city itself is small and not heavily populated. Weiqiao Pioneering Group locating in the area has become the world's largest aluminum producer but "used relatively inefficient subcritical steam generators" (https://chinadialogue.net/en/energy/10040-coal-power-and-privilege-china-s-problem-with-industry-owned-generators/). Combining all these info, we suspect that those industries and the nearby power plants may be partially shut down during the COVID lockdown in 2020, which correspond to lower ER_{CO} or higher combustion efficiency.

We do acknowledge that it may be easier to interpret temporal changes in combustion characteristics for these less-populated but industry-centered cities (e.g. Zibo or Baotou), than for megacties with diverse sectoral signals (e.g., Shanghai or LA).

Here are the revised discussions on Zibo:

City	Total power capacity (MW) and by fuel types	Key industry OR annual crude steel capacity (kt yr^{-1})
Los Angeles	5,808 MW (95.6% fueled by gas; 0% by coal)	refinery, shipping
Shanghai	16,031 MW (75.2% fueled by coal; 24.4% by gas)	iron & steel (25,099 kt yr^{-1})
Baotou	6,470 MW (100% fueled by coal)	iron & steel (12,619 kt yr^{-1})
Zibo (w/ Zouping)	9,720 MW (100% fueled by coal)	electrolytic aluminum; iron & steel (2,532 kt yr^{-1})

Table 1. A summary of total power generation capacity (Global Power Plant Dateset by World Resources Institute, 2018) and information on heavy industry including annual crude iron capacity (GID, Wang et al., 2019). Power plants are selected from a 0.5° × 0.5 °region around each city with percentage generated by the main fuel types.

Zibo with the nearby county-level city Zouping accounts for over 1/8 of the total coal consumption of Shandong province in 2017. The coal-fired power plants in the area contribute to a total power generation capacity of 9,720 MW (**Table 1**), which are likely to support local metal industries especially the production of electrolytic aluminum (world's top producers). The maximum XCO_2 enhancement per OCO-3 sounding can even reach up to 10 ppm for a few overpasses (not shown). Interestingly, ER_{CO} for Zibo first declined from 10.1±1.1 mmol $mol^{-1}$to 6.1±0.6 mmol mol^{-1}during Feb 2020 and gradually increased back to 18.2±1.1 mmol $mol^{-1}$by June 2020 (**Fig. 6b**). Such temporal variations in ER_{CO} agree nicely with the timing of the initial phase of COVID-19 lockdown in China (i.e., Feb to May in 2020) (e.g., Laughner et al., 2021). We suspect changes in ER_{CO} could be driven by the partial shut-down and re-opening of the multiple coal-fired power plants and metal industries in the area.

392-394: "In contrast to LA, ...". It is suggested here that wind speeds and direction do not change dramatically for LA, while the opposite appears to be suggested in line 386.

Lines 392-394 and line 386 may appear to be conflicting but they were referring to two different aspects regarding wind conditions. To clarify, the statement of "relatively small wind changes over LA compared to Shanghai" around lines 392-394 was referring to the changes in wind vectors across overpass dates/seasons (e.g., Feb, April, Dec). While "the dramatic wind changes over LA" around line 386 described the changes in wind vectors between OCO and TROPOMI overpass times but for one overpass. To avoid confusion, we now modified both sentences.

Line 392-394: "In contrast to LA, where urban plumes are usually well-constrained with the basin, wind speeds and directions vary across different overpasses over Shanghai —i.e., southeasterly wind on Feb 4 and Feb 20, 2020; southwesterly wind on Feb 24, 2020, and Feb 19, 2021; and northerly wind on April 23 and Dec 30, 2020.

Line 386: Although OCO-3 has sampled the Los Angeles basin dozens of times to date, many overpasses did not pass the quality check (i.e., QF) and were removed from the final result due to their noticeable shifts in urban plumes between two overpass times (e.g., March 3, April 15, and May 5, 2020 for LA; discussed in Sect. 4.1).

425: This is an important point for interpreting the results in terms of "combustion efficiency". While in many industrial processes, hydrocarbons are combusted with the intention of achieving as high as possible combustion efficiencies, in iron/steel production, CO actually serves an important function during the process. While an iron/steel plant will aim at burning as much as possible the CO before release to the atmosphere, it still is a tricky comparison with other sectors under the concept of 'combustion efficiency'. Perhaps a short mention of this would be appropriate.

We agree that some explanations of different industrial activities are needed. Here is the paragraph on acknowledging the uniqueness in iron/steel production, compared to other industrial processes:

"We acknowledged that although many iron/steel plants may aim at combusting as much CO as possible before releasing CO into the atmosphere, the indispensable role CO played in the iron/steel industry makes it unique when assessing its ER_{CO} and combustion efficiency among various industrial processes."

4 Discussion

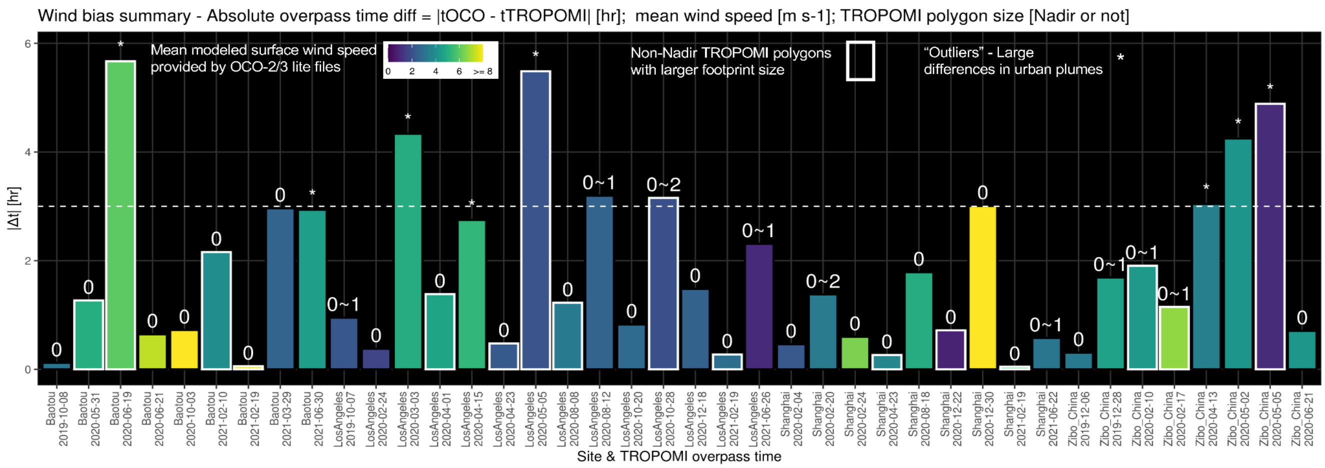
459-460: "...constraint can be relaxed". This statement may need to be reconsidered when also taking into account the changes in emissions of (FF) CO2 and CO during the day (see major comments).

We agree with the reviewer and have modified the text as follows:

"If TROPOMI pixel sizes are relatively large (i.e., non-nadir observations) or the wind is steadier, this dt constraint may be relaxed, as long as emissions for a specific city is less driven by sectors with noticeable diurnal cycle (e.g., road transportation)."

461-462: This sentence is not clear to me. The text appears to suggest that the plumes during the overpasses on May 31, 2020 are sufficiently similar. Still, in figure 8 it is marked as an outlier (*). This also makes the relation to the next sentence (461-462) unclear.

We apologize for the confusing tilted label on the x-axis; but if looking closer, the May 31, 2020, one is the second bar from the left and associated with the “0” on the top of the bar. We now adjusted the orientation of these x-labels.



497: I had to reread the paragraph on VOC’s several times and I’m still not really sure what is the conclusion on the relevance for the present work. Please consider guiding the reader a bit more.

We have now rewritten the paragraph as:

Another factor that we did not explicitly account for is the secondary CO production from both anthropogenic and biogenic VOCs (AVOCs, BVOCs). Under a cascade of reactions in favorable conditions, VOCs emitted from the upwind source location are oxidized to CO at various rates, which result in possible higher CO at the downwind sounding location and a divergence between enhancement ratios and emission ratios. As BVOCs are usually associated with shorter lifetimes compared to many AVOCs (e.g., Surl et al., 2018), we discuss BVOCs and AVOCs separately. BVOCs can contribute significantly to the total CO source at the regional scale especially during growing seasons (e.g., Miller et al., 2008; Hudman et al., 2008; Gonzalez et al., 2021). However, since BVOCs like biogenic CO_2 come mainly from rural areas outside the city, by subtracting localized CO background using CO observations outside the urban plume, the impact from BVOCs on the derivation of CO enhancements would be minimized. The lifetime of most AVOCs remains long enough, except for a few species including alkenes (Surl et al., 2018). Without a good observational constraint of the VOC composition and group-specific emissions for different cities around the globe over the years, it would be challenging to accurately quantify the impact on atmospheric XCO and ER_{CO} due to AVOCs emitted from urban areas or specifically from industrial areas. More future efforts regarding urban VOCs may include 1) exploring what good proxies can be measured from space that well represent the bulk AVOC characteristics (e.g., formaldehyde, Zhu et al., 2014) and 2) interpreting such observations, e.g., by utilizing chemical transport models for source attribution (Gonzalez et al., 2021). Note that the noise/uncertainty in current daily TROPOMI formaldehyde observations may be too large for daily resolved analyses.

516: “using the industry emissions from EDGAR”. Does this comparison indeed only consider industry emissions or also the other sectors (e.g. those shown in Fig S11d-f)?

To clarify, we performed two separate sets of simulations, 1) using total emissions from sectors related to fossil fuel burning from EDGAR (in **Fig. S11b**), and 2) using emissions from separate sectors from

EDGAR (in **Fig. S11d-f**). For the second set of simulations, we have only shown the modeled enhancements from the four major emission sectors, namely 'industry', 'on-road', 'power', and 'resident' as labeled in the initial **Fig S11d-f**. Several non-FF sectors (e.g., AGS - agricultural soils; SWD - solid waste; AWB - agricultural waste burning; and FFF - fossil fuel fires) had not been chosen.

Below are the FF sub-sectors from EDGAR we selected and how we modified them.

 # ENE: Power industry ***-> renamed as 'power' (Fig. S11e)***

 # TRO: Road transportation ***-> renamed as 'on-road' (Fig. S11d)***

 # RCO: Energy for buildings ***-> renamed as 'resident' (Fig. S11f)***

Subsectors that were ***combined into one sector as "industry" (Fig. S11c)***

 # IND: Combustion for manufacturing

 # CHE: Chemical processes

 # NMM: Non-metallic minerals production

 # NFE: Non-ferrous metals production

 # IRO: Iron and steel production

The following sub-sectors are associated with very small emissions for our cities and were not shown in Fig S11 previously.

 # REF_TRF: Oil refineries and Transformation industry

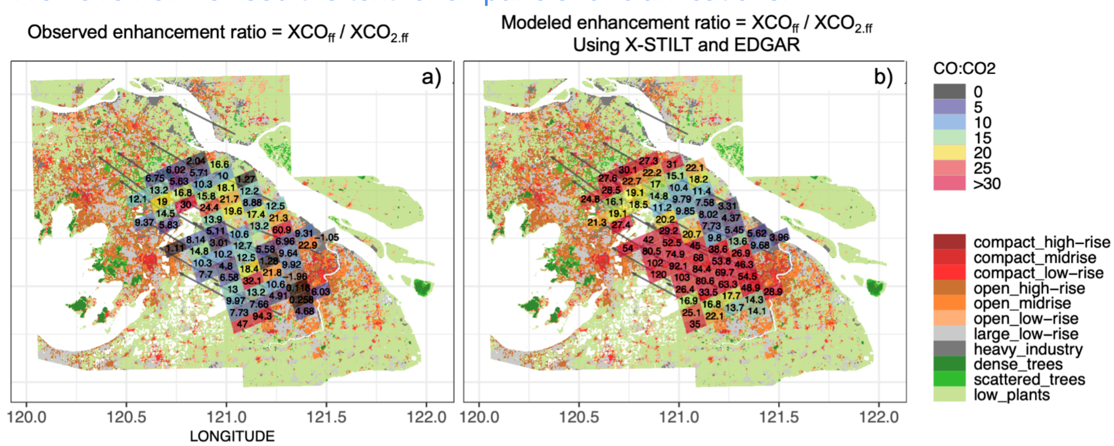
 # PRO: Fuel exploitation

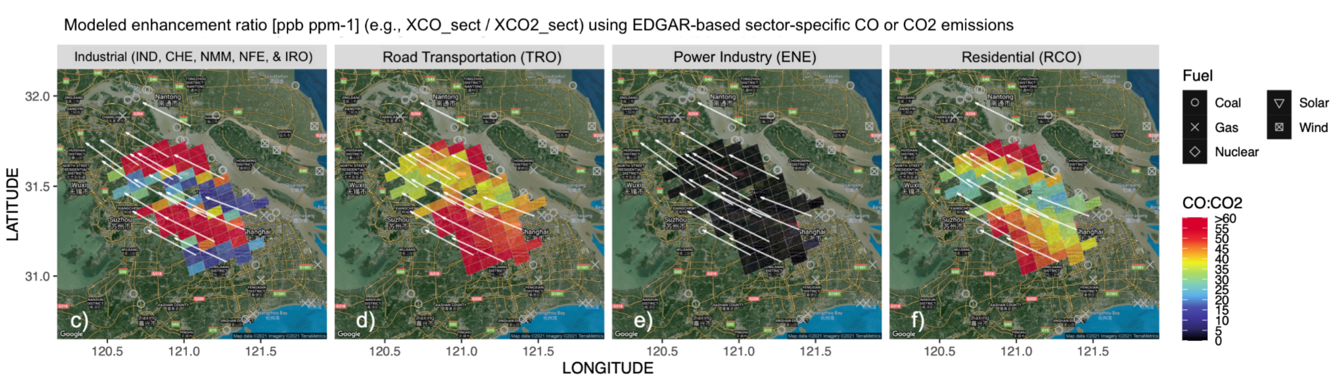
 # TNR: Other transportation sources

 # NEU: Non-energy use of fuels

 # PRU_SOL: Solvents and products use

We have now revised the text for all panels for clarifications:

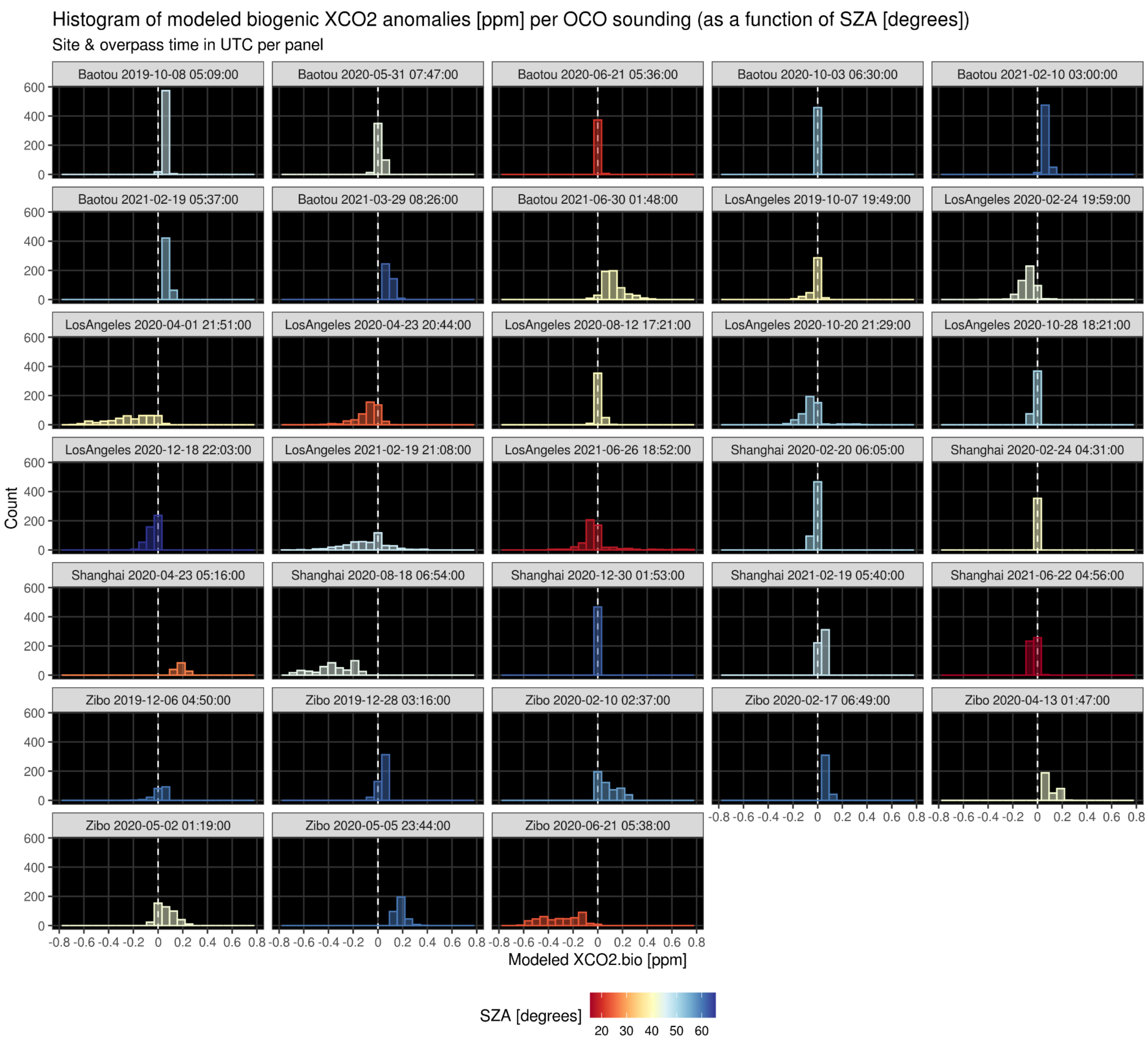




Supplementary information

Figure S7: The figure does not show the overpass time, that could for example explain the difference between the lower right figures (Zibo 2020-05-05 and Zibo 2020-06-21). Perhaps the colour can be used to indicate overpass time instead of the month (which is already shown above each graph).

We agree that adding overpass time is a good idea because overpass hours and seasons determine the net positive or negative anomalies from the nearby biosphere. We have now modified **Fig. S7** and its associated caption by including the solar zenith angle (SZA) as an indicator for biogenic contributions (see below). For example, for Zibo, contributions from the biosphere are almost all positive on May 05, 2020, at 23 UTC (= 7 am local time) while contributions are almost all negative on June 21, 2020, at 05 UTC (1 pm local time). More generally, biogenic anomalies are more positive when overpass times are in early mornings (high SZA) or in wintertime, due to accumulative influences from net positive biospheric fluxes.



Technical corrections:

Abstract

3: Consider using “combustion efficiency” consistently instead of “burning efficiency”.

We have now replaced all “burning efficiency” with “combustion efficiency” throughout the manuscript.

1 Introduction

43-45: “Benefit ... reported”. Please check the sentence structure, it is not fully clear.

We agree that this sentence sounded awkward and have reworded it:

“The commonly used approach in estimating combustion efficiency is to combine atmospheric observations of multiple trace gases and report the ratio of the total or excess measured concentrations (above a defined background value) between tracers (Silva and Arellano, 2017; Reuter et al., 2019; Park et al., 2021). Such tracer-to-tracer ratio calculation has the benefit that errors in describing the atmospheric transport that carries tracers to the measurement site can be cancelled.

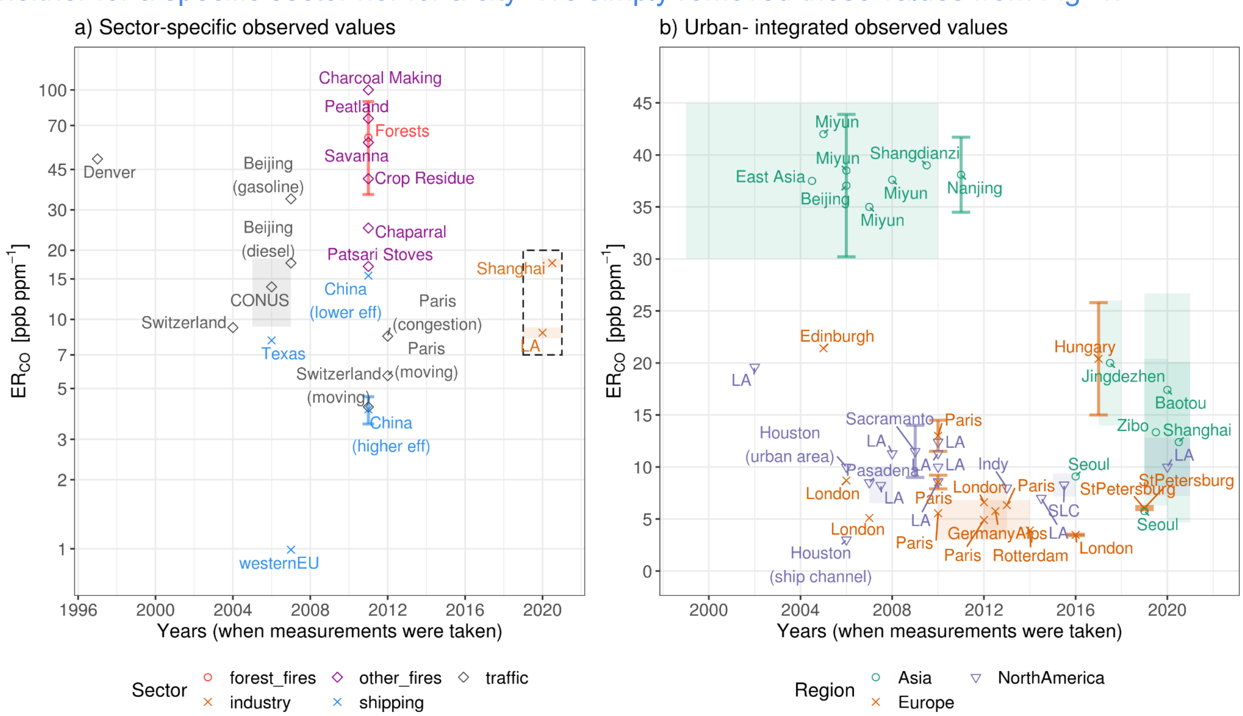
50: “be difficult.” some words are missing. Perhaps “...be difficult to identify”?
56: “diagnosed”. consider using a different term, e.g. “analysed”.

All corrected.

Figure 1:

- What sector(s) is “biofuel + ff” referring to? All anthropogenic combustion sectors?
- In fig.1b the vertical (minor) grid lines fall in between years. The figure may be easier to read if these lines coincide with the years.

Indeed, “biofuel + fossil fuels” is referring to all possible anthropogenic combustion sectors, thereby neither for a specific sector nor for a city. We simply removed those values from Fig. 1:



65-66: Not clear what is meant by "...the generalization and representation of gridded ERs." Also, the next few sentences could be written a bit more clearly.

> To clarify, we were trying to state that large variation in ERs makes it difficult to map ERs for the entire globe. For example, adopting one universal number per sector may even be insufficient to account for the contrast in ERs across regions or years. The reason we brought it up is that most gridded emission inventories involve prior assumptions of emission factors. The relevant text has now been rewritten as:
>
> When estimating fossil fuel emissions from a bottom-up perspective, most inventories rely on activity data and may involve prior knowledge of emission factors (Gurney et al., 2019; Solazzo et al., 2021). One notable example is Hestia, a high-resolution inventory for the US, which estimates CO_2 emissions of non-point sources based up CO emissions from the National Emission Inventory with EFs and carefully evaluates their adopted EFs (Gurney et al., 2019). However, when constructing emission inventories across regions/nations, the large variability in ERs across combustion processes, sectors, years, and regions (as seen in **Fig. 1a**) makes the choice of EFs extremely challenging. Accurate bottom-up emission estimates require accurate activity data and EF_X that naturally vary with combustion conditions (e.g., temperature, fuel load, oxygen level) and are generally not well known especially over data-scarce regions. To our knowledge, only a few global inventories, such as the Emissions Database for Global Atmospheric Research (EDGAR, Solazzo et al., 2021), offer global anthropogenic CO and CO_2 emissions. Considering the challenge in approximating ERs, certain knowledge derived from atmospheric observations may 1) complement inventory-based ERs (e.g., $CO:NO_x$ ratio in Lama et al., 2020) and 2) facilitate the emission constraint for a desired gas usually with relatively larger uncertainties (Wunch et al., 2009; Palmer et al., 2006; Wang et al., 2009; Brioude et al., 2012; Nathan et al., 2018). Such prior achievements motivate us to examine ERs using satellite observations of multiple tracers.

2 Data and methodology

107: “and surrounded coal-fired” —> "surrounding"? or "presence of multiple coal-fired power plants"
109: “that is the spatial extent” —> “that is in the spatial extent”
111: “that estimated” —> “that are estimated”
111: “FF enhancements” this term is a bit vague.
116: "we illustrate how much ERs ... can be extracted." What does this mean?
124: “north hemisphere”. This term appears to be more commonly spelled as “northern hemisphere”.
163: “interfere the calculation” —> “interfere with the calculation”
218: “swaths of stretching” —> “swaths stretching”
220: “have been emphasized” —> “has been emphasized”

> Lines 107-220 have been corrected as suggested by the reviewer. For the definition of “FF enhancements”, we added a note for clarification:
>
> “Since we do not differentiate emission signals due to biofuel or fossil fuel (FF) combustion, the term “FF enhancements” is simply referred to *column enhancements induced by any anthropogenic combustion processes* from the target city.”

3 Results

375: No need to present two decimals for the crude iron production.

> Corrected.

4 Discussion

451: “considered ~~for~~ when”
456: “Interfering the” —> interfering with the”.
456-457: *Please rephrase, as the lack of concurrence is not itself inducing the wind directional shift.*
464: “future geostationary satellite” singular or plural?
464: What is meant by “spontaneously” here?
468: “a urban” —> “an urban”
512: “efficiency can” —> “efficiency, which can”
513-514, 521: “inventory” —> “inventories”
Section 4.3. the readability of the text in this section could be improved by a thorough read through

All corrected. Line 456-457 is now reworded as “The biggest challenge affecting the robust spatially-resolved ER estimates is the shift in wind directions across different satellite overpass times.” We have revised the text (please refer to **Sect. 4.3 and 4.4** in the revised version for more details).

5 Conclusion

545: “industry-related ER_CO slightly lower”—> “industry-related ER_CO are slightly lower”

Corrected.

Supplementary information

Figure S10: “distribution is stewed towards” —> “distribution is skewed towards”

Corrected. We truly thank the reviewer for the careful read-through!

Point-to-point responses

We appreciate the careful read-through and constructive comments from both reviewers. The reviewers' comments are copied in **black** with our point-to-point responses in **blue** and revised text in **red** or copied.

Response to Reviewer #2:

The study presents a technically detailed implementation of a Lagrangian model adapted for use with column data, X-STILT, focusing on emission ratios of CO:CO2 on sub-city scales using XCO measurements from TROPOMI and XCO2 measurements from OCO-2 and OCO-3. The level of technical detail is quite good (with a few exceptions, see comments below), and it is nice to see that the adapted X-STILT code has been made publicly available. The approach is interesting, particularly in its attempt to not make use of emissions inventories as part of the prior information. However some potential sources of error have been overlooked, particularly with respect to the high- frequency variability of the CO:CO2 ratio, which seems to be assumed to be constant during the time between measurements. This is not something that the current study can really correct, but it needs to be included in the discussion as a clear limitation of the current results. Despite these limitations, the study is certainly appropriate for publication in ACP once these concerns are addressed. A good proof-reading of the paper is also needed before resubmission: while it was almost always clear what was meant, and the paper was well-written and easy to follow, there were lots of missing articles etc. that the native speakers among the co-authors could clear up quickly.

We truly thank Julia Marshall for the detailed and constructive comments and have tried to address the main concerns about the high-frequency variability in ER_{CO}.

Major concerns:

The authors have gone to great pains to try to correct for temporal shifts between the measurements, considering the impact of the different meteorological conditions and averaging kernels. *What was not taken into account is changes in the CO:CO_2 ratio over the course of a day.* While this may not be as critical for heavy industry and power generation, other sectors (e.g. traffic) have highly heterogenous emission ratios in time, depending also on traffic patterns. Having the XCO and XCO_2 measurements offset by even a couple of hours complicates this approach considerably, and might also cloud the proposed analysis of temporal trends in the emission ratio over the year. That does not mean that nothing can be learnt from this approach, only that this neglected error source needs to be explicitly described. In any case, GeoCarb data will make such analyses considerably easier in the future.

We agree with Julia that high-frequency variability in CO or CO_2 emissions may influence the ER_{CO}, although the overpass time difference has been limited to 1-3 hours. One can likely rely on bottom-up emission inventories or prior assumptions to infer hourly emission patterns. However, because emissions or combustion characteristics are what we are solving for, it is quite challenging to properly account for such mismatch WITHOUT involving prior assumptions towards emissions themselves. Such hourly emission variations depend on the relative contributions from individual emission sectors for a given city.

We have now carried out a sensitivity analysis to investigate how such a mismatch in emission timing may affect the ER_{CO}, or more generally the high-frequency variability of ERCO (as reviewer #2 also mentioned). Thanks to the TCCON network that provides high-frequency XCO and XCO_2 measurements (TCCON 2022). We utilized the latest GGG2020 version with several upgrades including a much-improved prior profile. Because XCO and XCO_2 are simultaneously retrieved, we can assume that atmospheric transport associated with two species are the same and their enhancement ratio fully reflected the emission characteristics once species-specific averaging kernel profiles are corrected for following Appendix A2 in Hedelius et al. (2018).

Here we reported the estimated enhancement ratio at Caltech with background observations from another TCCON site outside the LA basin (see figure and figure caption shown below). Because the temporal frequency of the Caltech TCCON site may not perfectly match that of the background TCCON site, we first averaged observations from both sites to each 20-min interval and calculated the enhancements and enhancement ratio. Indeed - the variation in ER_{CO} can be large throughout the day. Note that such variations may reflect not only the change in FF emissions (e.g., due to traffic) but also measurement noise (given these high-frequency data) as well as changes in the meteorology throughout the day (ocean versus mountain flow for LA). Nevertheless, changes in ER_{CO} between OCO-3 and TROPOMI times inferred from concurrent TCCON measurements appear to be small.

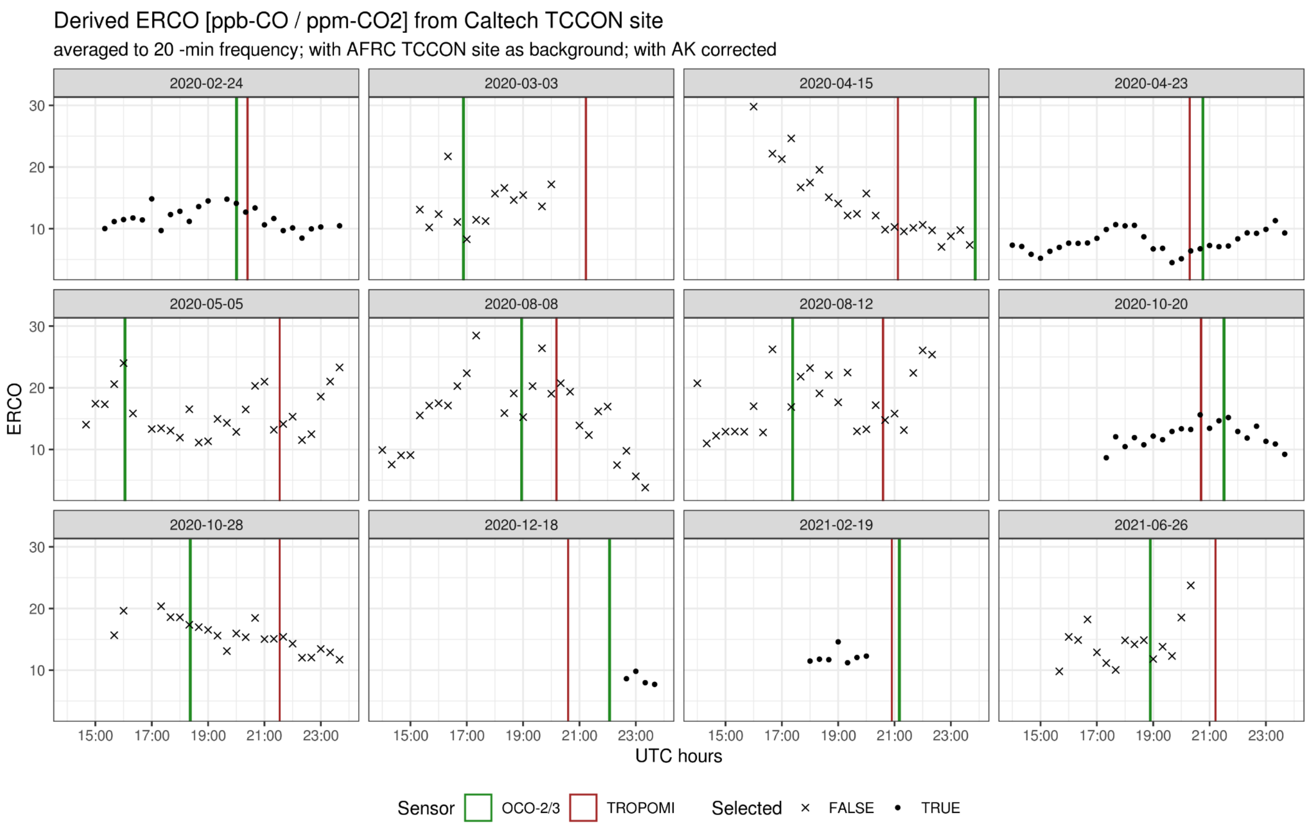


Fig. S8 - Time series of observed ERCO at the California Institute of Technology (Caltech) TCCON site (Wennberg et al. 2017) on the OCO-3 overpass dates till June 2021. The background is defined using the NASA Armstrong Flight Research Center (AFRC) site near Lancaster, California (Iraci et al., 2022). Column enhancements with corrections of TCCON averaging kernel are calculated following Hedelius et al. (2018). The overpasses that went into the final result are shown in solid black dots, while the remaining overpasses with significant plume shift between OCO-3 and TROPOMI overpass times are shown in black crosses. The vertical lines indicate the OCO-3 (green) or TROPOMI (red) overpass times in UTC times. The day of the week for each date is shown as follows: Feb 24, 2020 (Mon), Mar 3 (Tues), Apr 15 (Wed), Apr 23 (Thurs), May 5 (Tues), Aug 8 (Sat), Aug 12 (Wed), Oct 20 (Tues), Oct 28 (Wed), Dec 18 (Fri), Feb 19, 2021 (Fri), and June 26, 2021 (Sat). Note that no qualified data exists during the overpass time of OCO-3 or TROPOMI on April 1, 2020. The TCCON data were obtained from the TCCON Data Archive hosted by CaltechDATA at https://tccondata.org. We thank Laura T. Iraci and Coleen M. Roehl for preparing the TCCON data for these two sites.

Even though many more analyses related to the hourly or seasonal ER_{CO} can be drawn from these TCCON observations, they are out of the main goal of the current manuscript in using space-based observations. Since these TCCON and spatially resolved satellite observations (OCO-3-TROPOMI) may have a different emphasis on the emission signals within the LA basin, we tried to avoid unnecessary direct comparisons of their derived ER_{CO} and decided to leave the analysis in the supplementary material but added some discussions in

Sect. 2.1.3:

3. *Overpass times, meteorological conditions, and emission variations:* As a result of the overpass time difference between sensors, variations in meteorological conditions (e.g., wind direction and speed) can lead to changes in urban plume shapes detected by the two sensors as they pass by. We dealt with changes in wind speed and wind direction separately. The former is resolved by the "scaling factor" inferred from an atmospheric transport model and the latter undergoes manual evaluations (**Sect. 3.1**). Besides, CO and CO_2 emissions themselves can vary over the course of a day, e.g., driven by road transportation and residential sectors. Given the overpass time difference between sensors, it is likely that such mismatch in the timing of CO versus CO_2 emissions may affect the observed ER_{CO}.

Sect. 3.1:

Besides changes in wind directions, CO and CO_2 emissions themselves can vary across daytime hours, likely driven by the road transportation and residential sectors. As a result, variations in the derived ER_{CO} across multiple overpasses may reflect not only the variation in combustion efficiencies but also the mismatch in the emission timing. LA may be one of the cities with more distinct daytime changes in emissions compared to industry-centered cities. Fortunately, based on a supplementary sensitivity analysis using measurements from the Total Carbon Column Observing Network in Pasadena (TCCON, Wennberg et al., 2017), by limiting satellite overpasses to those with a smaller time difference, ER_{CO} appear to be less variable (**Supplementary Fig. S8**). Future geostationary satellite monitoring NO_x (e.g., TEMPO, Chance et al., 2022) may provide better guidance towards the hourly pattern in urban emissions, especially from the traffic sector with more daytime fluctuations, which have been discovered using surface monitoring networks (e.g., over Chicago; de Foy, 2018).

And Sect 4.1:

"If TROPOMI pixel sizes are relatively large (i.e., non-nadir observations) or the wind is steadier, this dt constraint may be relaxed, as long as emissions for a specific city is less driven by sectors with noticeable diurnal cycle (e.g., road transportation)."

Reference:

Hedelius, J. K., Liu, J., Oda, T., Maksyutov, S., Roehl, C. M., Iraci, L. T., Podolske, J. R., Hillyard, P. W., Liang, J., Gurney, K. R., Wunch, D., and Wennberg, P. O.: Southern California megacity CO2, CH4, and CO flux estimates using ground- and space-based remote sensing and a Lagrangian model, Atmos. Chem. Phys., 18, 16271–16291, https://doi.org/10.5194/acp-18-16271-2018, 2018.

Wennberg, P. O., D. Wunch, C. Roehl , J.-F. Blavier, G. C. Toon, N. Allen. 2017. TCCON data from California Institute of Technology, Pasadena, California, USA, Release GGG2020R0. TCCON data archive, hosted by CaltechDATA, California Institute of Technology, Pasadena, CA, U.S.A. https://doi.org/10.14291/tccon.ggg2020.pasadena01.R0

Iraci, L., J. Podolske, C. Roehl, P. O. Wennberg, J.-F. Blavier, N. Allen, D. Wunch, G. Osterman. 2022. TCCON data from Armstrong Flight Research Center, Edwards, CA, USA, Release GGG2020R0.

TCCON data archive, hosted by CaltechDATA, California Institute of Technology, Pasadena, CA, U.S.A. https://doi.org/10.14291/tccon.ggg2020.edwards01.R0

Minor concerns:

What the authors mean by "FF" needs to be made clear. The implication is that the emission signature of fossil fuels is being measured directly, which is clearly not the case. Emission from combustion, sure. There is no capacity to separate e.g. biofuel from fossil fuels in this approach, and this needs to be made clear.

We agree that FF enhancements need to be more precisely defined. It is challenging to isolate the biofuel signals from fossil fuel signals by only using limited atmospheric observations. Nevertheless, the proportion of biofuel combustion would likely be small compared to fossil fuel combustion. We now added a note for clarifications in the second paragraph of Sect. 2:

"Since we do not differentiate emission signals due to biofuel or fossil fuel (FF) combustion, the term "FF enhancements" is simply referred to *column enhancements induced by any anthropogenic combustion processes* from the target city."

L37: While this is true for some air pollutants such as CO, this is not true for many CO2. In fact, increasing efficiency during combustion activities increases the amount of CO_2 and NO_x emitted (unless the latter is scrubbed) (e.g. Lama et al., 2020). The key to reducing emissions of greenhouse gases is to reduce combustion, period.

We agree with the reviewer that the initial statement was inaccurate. As reviewer #1 and previous studies have also mentioned, NO_x emissions will be higher during efficient combustion with high temperatures. We have now taken this point into account and modified the text as:

"Given the co-benefit between GHG reduction and improved air quality at various scales (e.g., Zhang et al., 2017), controlling the consumption of fossil fuels altogether is the key."

L223-224: Why use a rectangle as the source function? Was this to be "independent of emission inventories", while still knowing that emissions are locating within the city?

A city is simply represented by a rectangle that outlines the broader boundary for possible emissions. The assumption of a rectangle shape is somewhat limited by the model code. Ideally, the spatial extent of urban emissions could be any shape but will then involve more info about the spatial distribution of emissions. To determine a rectangle that covers a wide area, we looked at google map images or urban land cover maps from WUDAPT (if applicable).

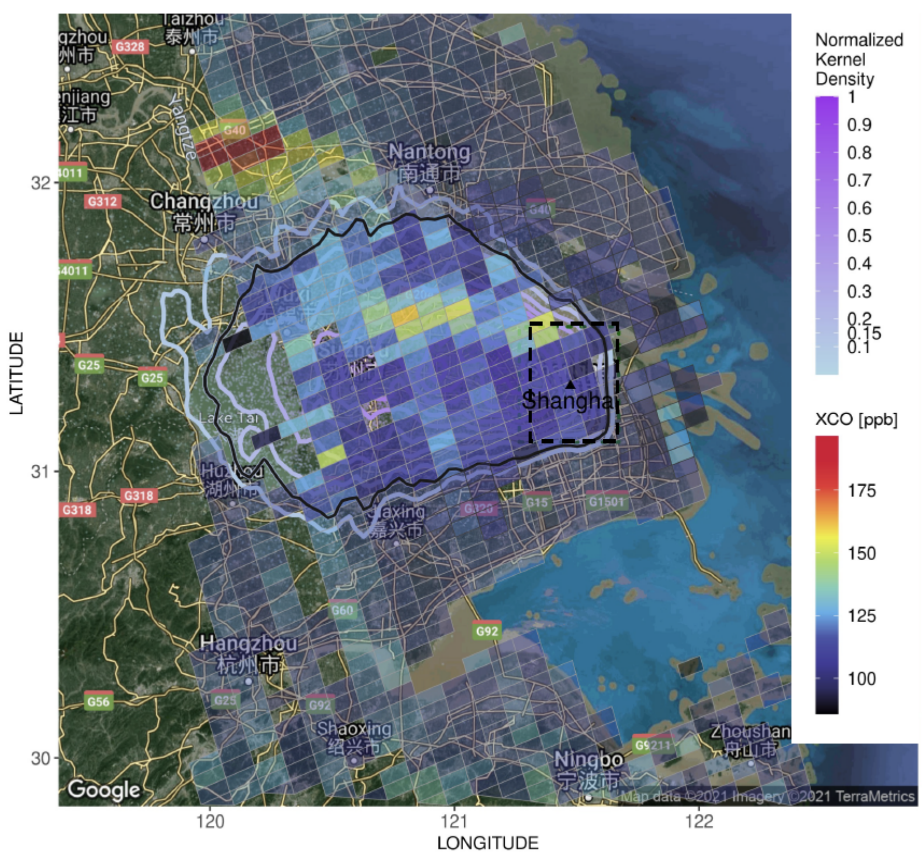
We may argue that it is fair to assume FF emissions are very likely to be located within the city and such an assumption may not contradict the statement of "independent of emission inventories". Emission inventories provide very specific information about the magnitude and spatial distribution of FF emissions. One can think of the other end of the spectrum: we release air parcels from the individual sources according to emission inventories in the cities (e.g., more air parcels are released from the grid with larger emissions) and watch how those air parcels move in space and time. Compared to such an approach, our assumption and approach involve very little info from inventories (no info about specific emission locations nor magnitudes).

Lastly, we would stress that the idea is more about estimating the probability of urban plume realized by the particle distribution during overpass time and the quantitative expression via 2D kernel density.

L231: Regarding the second point about excluding observations elevated by another city: yes, this excludes enhancements outside the model-defined urban plume, *but what about enhancements from other cities that might also be contributing to enhancements within the urban plume?* This seems to be the case for the XCO values shown in Figure 3c. Is there any way to correct for these values? Especially given that there do not seem to be XCO2 values over the same area?

Enhancements within the urban plume might be influenced by other cities. It is possible to calculate the FF enhancements due to other cities' emissions, for example, using footprint and prior emissions. Or, one can perform the full atmospheric inversion to optimize emissions from not only Shanghai but also nearby cities. But, such calculations or inversion will involve prior assumptions, which is less wanted for this work. Alternatively, we could further limit the spatial extent of the urban plume by choosing a higher kernel density. However, we may argue that the current setting is sufficient for the cases we examined. As the reviewer mentioned, there seem to be no XCO_2 values in those regions. Since ER_{CO} was only calculated when OCO-3 XCO_2 is available, there seems to be a lower priority for such corrections.

c) TROPOMI, Feb 20th, 2020, 04:44 UTC



L235: Is there any way to mark on here which soundings were used to define the background? Some more quantification of the information would also be useful here. Which latitude range was used to define the background for the OCO-2 swath? In Figure 3a I don't see any soundings outside the urban plume region… For 3b it is somewhat clearer, aided by the coastal cut-off (although point sources near the coast are clearly in the “background”, but how the area would be defined for Figure 3c is really vague. With this level of detail, the approach would be hard to replicate.

The background soundings/polygons were actually highlighted with a black outline (e.g., polygons to the east of the urban plume), while other soundings/polygons were outlined in white. However, the color seemed to be difficult to observe as the reviewer pointed out. We have now added several notes in the figure caption to guide readers.

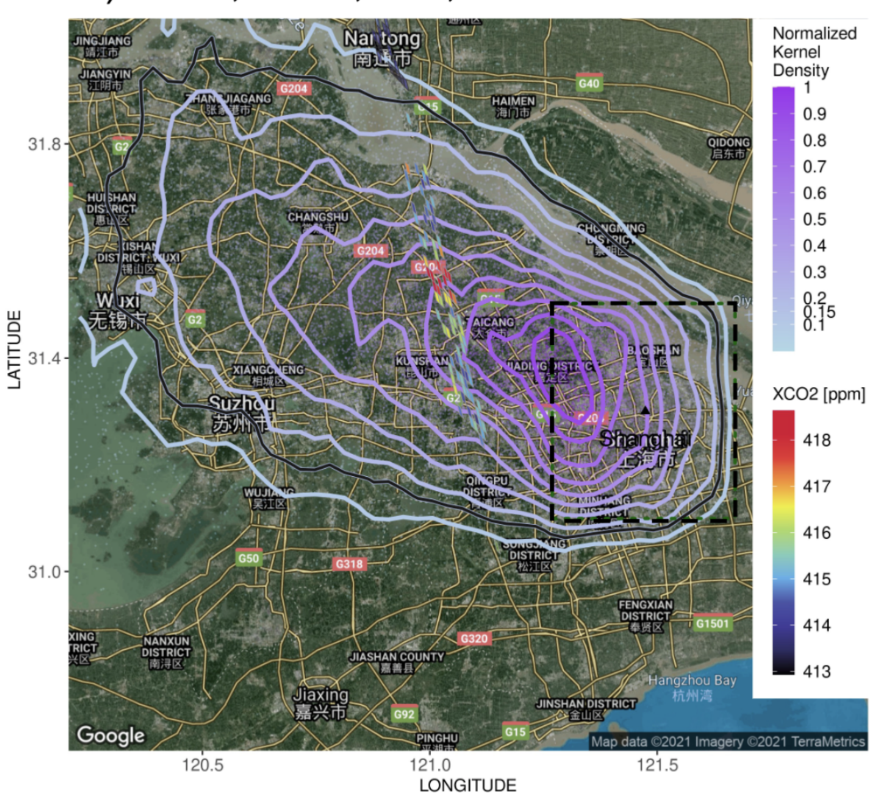
For clarification, we allowed for a 0.1degree “buffer area” outside the urban plume (to further avoid including some enhanced values, even though a random wind error has been included to generate the urban plume). The background range/domain is further extended up to 0.3 degrees in either longitudinal or latitudinal direction depending on the wind direction. We also tested a background range of 0.5 degrees, but the resultant background values did not change much. The background extension range is a free parameter in the X-STILT background r code, where the user can test their own values for their targets.

Here are more details and explanations regarding the reviewer’s individual questions.

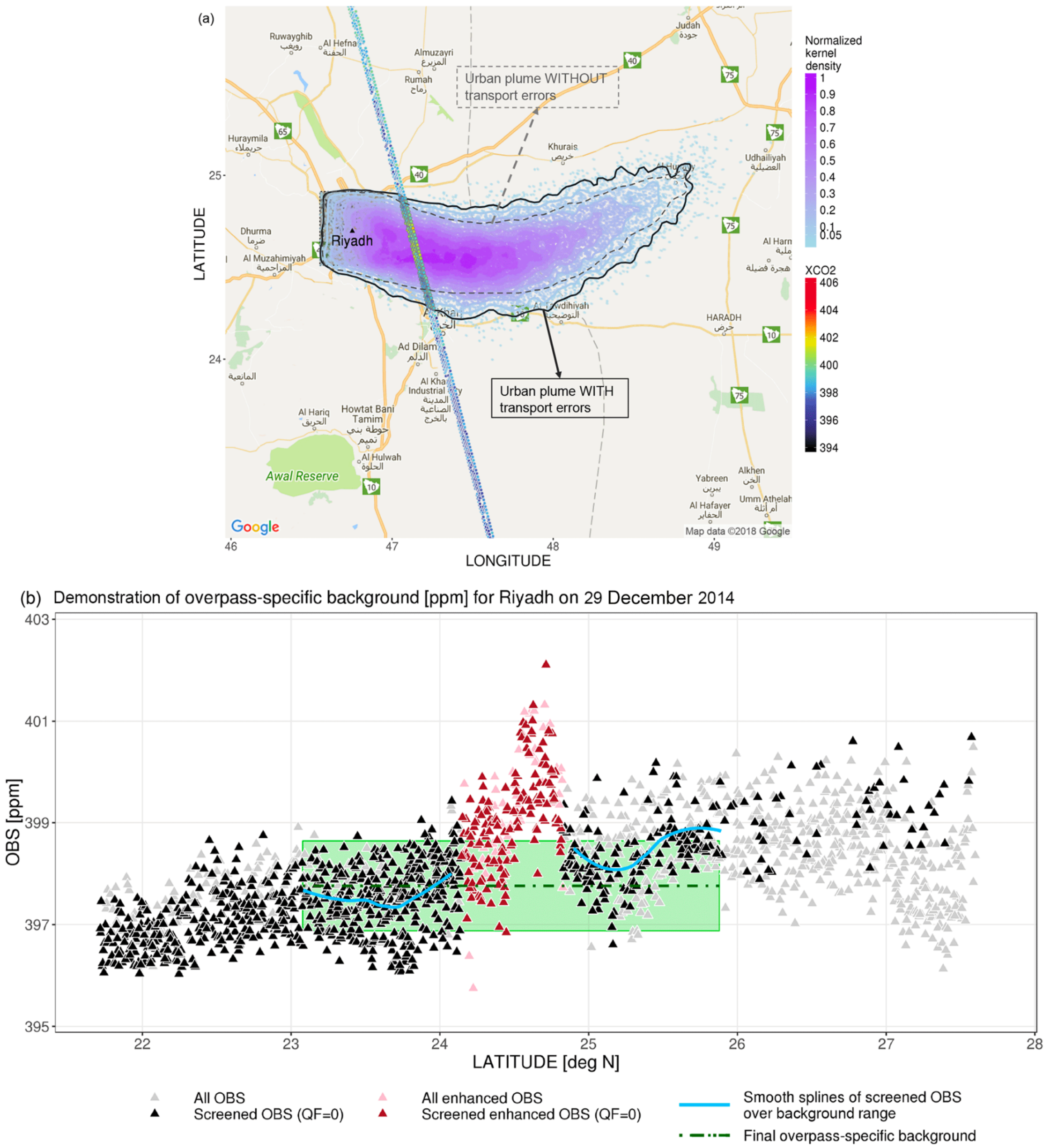
- For Fig. 3a, there were a couple of soundings to the north outside the urban plume (around 121.0 degE, 31.9 degN). Because those background soundings are tilted with relatively lower values (displayed in dark blue), they could be hard to identify. We have now added a note in the figure caption to guide readers.

  Ideally, we would choose OCO-2 soundings to the east, or the south of the urban plume given the southeasterly wind on Feb 4th, 2020, but the narrow swath of OCO-2 makes it impossible. Hence, we had to treat either the northern or southern soundings as background soundings.

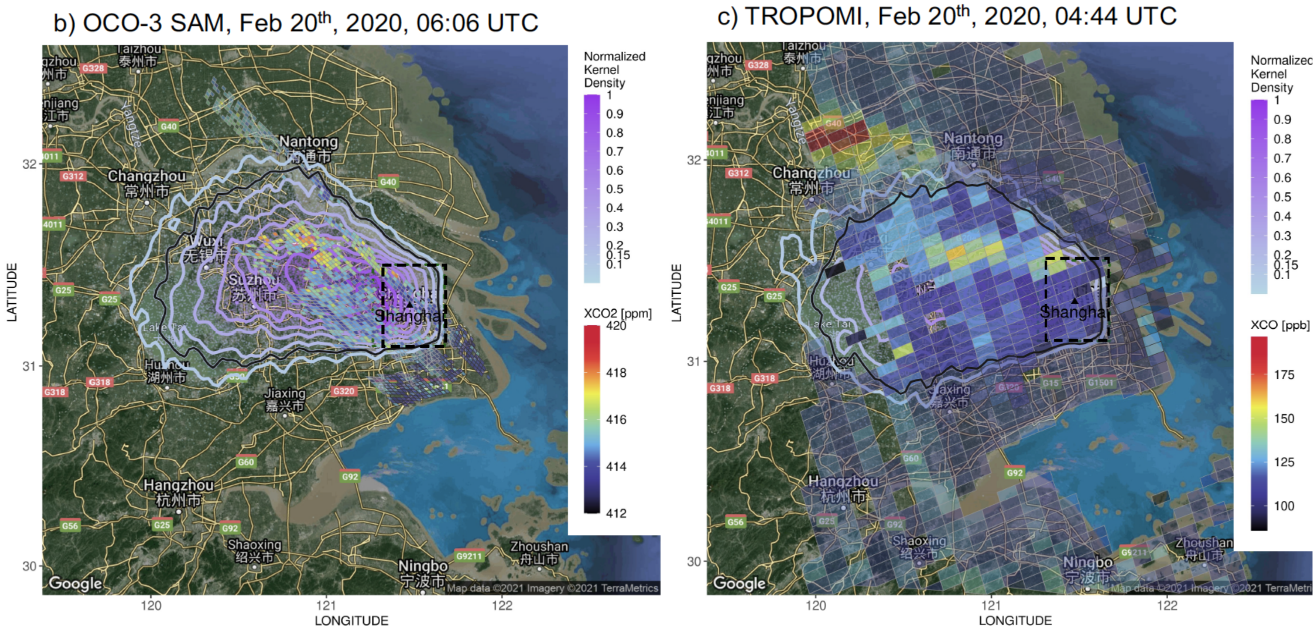
a) OCO-2, Feb 4th, 2020, 05:11 UTC



As explained in Wu et al. (2018), we showed a better demonstration of the background using OCO-2 data over Riyadh (https://gmd.copernicus.org/articles/11/4843/2018/#&gid=1&pid=1, see figure copied below). A wider latitude range of 1 degree was chosen in that Riyadh case because Riyadh is a relatively isolated city as we discussed in that paper. While for Shanghai (surrounded by other Chinese cities), we limited the background range to 0.3 degrees to avoid potential influence from a nearby city and tested an alternative of 0.5 degrees for the impact.



- For Fig. 3b (in the figure below), the background soundings were also highlighted in a darker color to the south of the urban plume (latitude < 31 degN). Individual swaths have their own slightly different background values. One may also see there is a small gap between the urban plume in the solid black curve and the darker soundings with latitudes < 31 degN, corresponding to the "buffer area" mentioned earlier.



- For Fig. 3c, the background soundings are highlighted with a black outline rather than a white outline for other soundings outside the urban plume. And, the soundings within the urban plume are outlined in grey. As explained above, the longitude range of the background region extends from around 121.7 degE toward the ocean (~ 0.3 degrees along longitude). The latitude range of the background region has the same range as that of the urban plume (i.e., from ~30.8degN to 31.9degN). These explanations have been added to the figure caption to guide readers.

Section 2.2.4: It's unclear how the observation uncertainty is computed here. Each sounding has a reported uncertainty that comes with the data product – is this the retrieval error? The "measurement noise" seems to be equated with the standard deviation of the retrievals within a TROPOMI footprint. But this isn't really what is usually meant by measurement noise.

We apologize for the lack of details on uncertainty calculations. Yes - each sounding is associated with a reported retrieval posterior XCO_2 uncertainty from Level 2 files. We agree with the reviewer that the terminology of "measurement noise" is not that accurate. As investigated by Worden et al. (2017), the standard deviation (SD) of retrieved XCO_2 values contains contributions from several possible sources, including 1) natural variability, 2) measurement noise and 3) a slowly varying bias (Sect. 3 of https://authors.library.caltech.edu/85361/1/amt-10-2759-2017.pdf).

The SD of XCO_2 retrieval is still needed and better be termed as "uncertainties due to binning", since we had to reconcile the resolution difference between two satellites, by taking the average of observed $XCO_{2.ff}$ enhancements from a few OCO-2/3 soundings within a given TROPOMI polygon to arrive at the averaged $XCO_{2.ff}$ enhancement at the TROPOMI scale. This "binning or average uncertainty source" was also reported in Table 1 by Wu et al. (2018).

It is not completely clear how these two types of errors (one per OCO-2 sounding and one per larger TROPOMI sampling) are combined, only that the retrieval errors are aggregated in a "standard- deviation-of-mean" manner, which sounds like it's being divided by the square root of the number of soundings. *This assumes that the measurements and their errors are independent of one another, which is not the case.*

This almost certainly underestimates the measurement uncertainty on the X_{ffCO2} term. A formula here is certainly needed.

We agreed that the lack of error covariance between every OCO-3 sounding within a TROPOMI grid may underestimate the observational uncertainty for the FFCO2 enhancement part. We have now performed additional variogram analyses to obtain a typical error correlation length scale (Lx, km) for all the overpasses per city. There were spatial gaps between OCO-3 soundings for some overpasses, leading to a higher correlation length scale. Thus, we adopted the minimum Lx among all overpasses as the final Lx per city (~normally around 7 to 15 km when using 4 to 6 bins). Below is an example of the variogram analysis to *investigate how retrieval errors correlate in space*.

Interestingly, including the extra covariance term of the retrieval errors leads to a small impact on the total observational error variance, because the overall observational error variance is often dominated by the background error component.

To address the reviewer's comments on error analysis, we have rewritten Sect. 2.2.4 and added Fig S4 for the variogram analysis:

2.2.4 Uncertainty sources

The uncertainty related to emissions should contain uncertainties from 1) the atmospheric transport (i.e., column footprints), 2) observations, and 3) non-FF sources and sinks, according to **Eqs. 1 or 2**. We neglect uncertainties from column footprints assuming no transport bias exists during either OCO or TROPOMI overpass time. The urban-background gradient in non-FF fluxes remains very small compared to FF enhancements (**Sect. 3.1**).

We estimated uncertainties of observed FF enhancements following **Eq. 5**. As previously described, observations from a few screened OCO soundings ($\sim$ 5 to 28 OCO soundings depending on the TROPOMI footprint size) are averaged to arrive at mean XCO_2 at the TROPOMI scale. Due to such averaging/binning process, the XCO_2 uncertainty due to binning is considered using the standard deviation of XCO_2 observations ($\sigma^2_{\varepsilon,bin}$ in **Eq. 5**) within a TROPOMI polygon.

$$\sigma^2_{\varepsilon,obs} = \sigma^2_{\varepsilon,bin} + \sigma^2_{\varepsilon,bg} + \sigma^2_{\varepsilon,retrv}. \quad (5)$$

$\sigma^2_{\varepsilon,bin}$ is not required for estimating XCO uncertainty. Background uncertainty ($\sigma^2_{\varepsilon,bg}$) contains both the retrieval error and the variability of column observations (as standard deviation) within background regions.

The retrieval uncertainty ($\sigma^2_{\varepsilon,retrv}$) of XCO is available for each TROPOMI sounding, whereas that of XCO_2 is reported for individual OCO-2/3 sounding (as read from Level 2 Lite files), which need to be aggregated at the TROPOMI scale. Due to possible correlations in retrieval errors between nearby OCO soundings, we estimated the error correlation length scale (L_x) using exponential variograms as demonstrated in **Supplementary Fig. S4**. Within a TROPOMI polygon that contains N numbers of OCO soundings, an error variance-covariance matrix with a dimension of $N \times N$ is constructed with its diagonal elements filled with OCO sounding-specific retrieval error variance. Then, L_x is used to form the normalized covariance matrix, i.e., $exp(-\frac{D(S_i,S_j)}{L_x})$ where $D(S_i, S_j)$ denotes the distance between each two OCO soundings ($1 \leq i < j \leq N$). Lastly,

the sum of all elements in the error covariance matrix (both variance and covariance elements) is divided by N^2 to obtain one $\sigma^2_{\varepsilon,retrv}$ per TROPOMI grid. As a result, the overall uncertainty of FF enhancement per sounding is often predominated by the background error component.

Here is the new Fig. S4 for an example of the variogram analysis to examine horizontal correlation of the retrieval XCO_2 errors (not retrieved XCO_2 values for clarification):

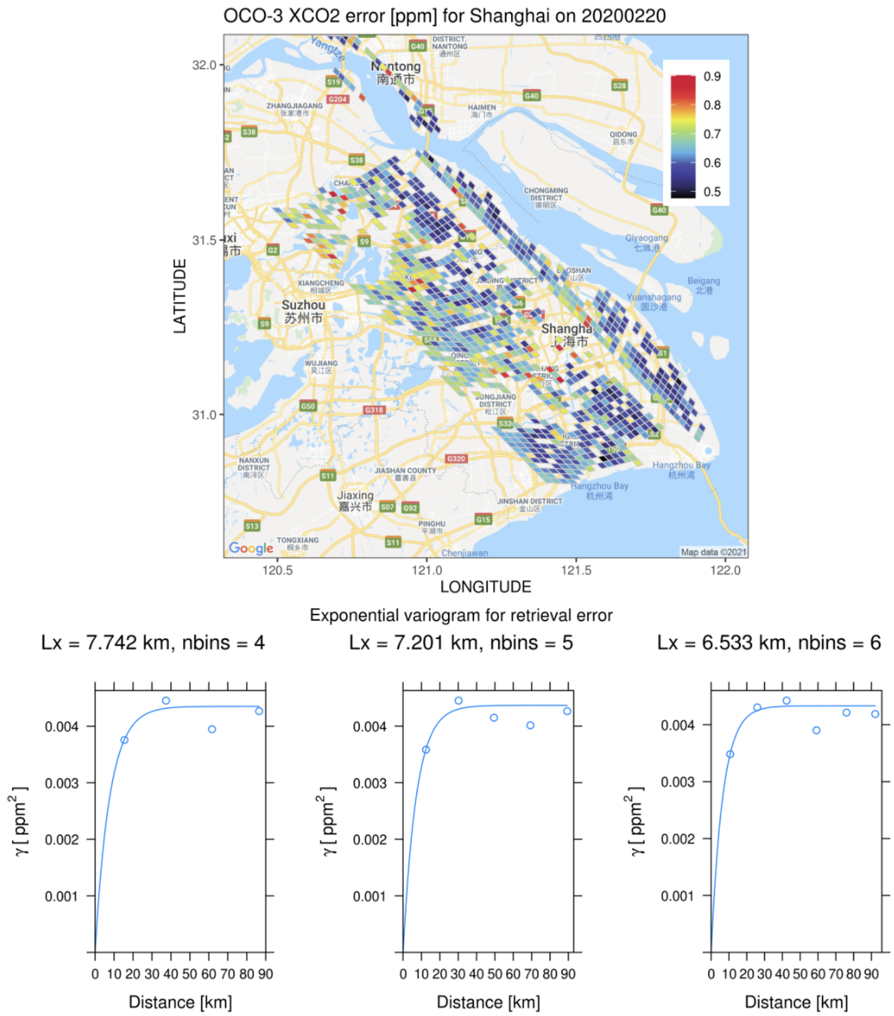


Figure S4. Upper Panel: An example of the sounding-specific XCO_2 retrieval errors from L2 files over Shanghai on Feb 20, 2020. **Lower Panels**: Exponential variogram analyses of retrieval errors and resultant error correlation length scale (L_x, km) with 4, 5, or 6 bins.

Technical/language comments:

L43-45: I'm confused by this sentence. Maybe should "Benefit" be "Benefiting"? In any case, the sentence should be rewritten to make it clearer, or even split into two sentences, starting with the second half, i.e. something like "The ratio… between tracers is reported. This has the benefit that errors … cancel out."

The text has been reworded as -

"The commonly-used approach in estimating combustion efficiency is to combine atmospheric observations of multiple trace gases and to report the ratio of the total or excess measured concentrations (above a defined background value) between tracers \citep{Silva2017,Reuter2019,Park2021ratio}. Such tracer-to-tracer ratio calculation has the benefit that errors in describing the atmospheric transport that carries tracers to the measurement site can be canceled."

L46: would remove “their”

L50: …difficult to detect?
L53-54: Rewrite this, something like: “Given its much longer lifetime, CO is much easier to interpret…” The “on the other hand” doesn’t fit here as written.
Figure 1 caption: “The x-axis indicates…”
L82: such gradient -> either “such a gradient” or “this gradient”
L87-88: in vertical -> in the vertical
L89: by the gaps in prior literature -> by gaps in the existing literature L92: adopt or adapt?
L97: quantify accurately -> accurately quantify
L101: implication and limitation -> implications and limitations
L107: surrounded -> surrounding
L109: urban plume that is the spatial extent -> urban plume, defined as the area
L111: that estimated -> that are estimated
L113: requires estimate -> requires an estimate
L116: would remove “much”
L155: accounted for AK -> accounted for the AKs?
L156: As result -> As a result
L160: evaluations -> evaluation
L163: interfere -> interfere with; also: more explanations in Sect. -> for more explanation, see Sect.

> We appreciate the careful read-through and detailed comments from the reviewer. Grammatical issues associated with **L46 to L163** have been corrected as suggested.

Figure 2 caption: The citation for the Google Maps data in the last sentence doesn’t sound quite right – please check what it’s supposed to be (i.e. adopted the Google Maps what?)

> Revised as: “The underlying hybrid maps were created using the ggmap library in R with the hybrid view of Google Maps over LA (copyright: Map data \copyright 2021 Imagery \copyright 2021 TerraMetrics).”

L166: and atmospheric transport model -> and an atmospheric transport model
L175: for sounding-specific -> for the sounding-specific

> Both corrected.

L178-181: I think I understand what is meant here, but it’s a bit hard to parse. When I hear “pathways” I’m thinking of chemical reactions, and I’m not sure what is meant by an air parcel being “tied” to somewhere, or correction terms being “attached” (perhaps “applied” would fit better)? In any case this should be rewritten for clarity.

> We have changed “pathways” to “routes”. Here is the revised sentence: “From a Lagrangian viewpoint, the air parcels arriving at an urban sounding might be traced back to different origins from the air parcels arriving at a rural sounding, meaning observations at the two soundings may be influenced differently by the surrounding biosphere.”

L193: corresponding for -> corresponding to

L196: wind condition -> wind conditions

L200: by sounding -> by the sounding

L201: Because AK -> Because the AK
L204: If -> By

All corrected.

L208-209: I guess the meteorological conditions and the AK profile are specific to the sounding, not just the sensor?

Correct - we have changed to "sounding-specific".

L209: condition -> conditions; L216: combines-> combine

Corrected.

L216-217: This seems a bit backwards – isn't the first method more independent of information about emissions, unlike the two modelling-based approaches?

Correct - this statement has now been removed. For clarification,

- The first statistical method utilized standard deviation or percentiles to derive the background purely from observed XCO_2. So, no use of prior emissions.
- The second method uses modeled initial conditions from atmospheric transport models and global CO_2 fields. Global CO_2 fields may involve prior assumptions on emissions.
- The third method first considered atmospheric transport and identified the background region (but not too far from the target city) and then calculated the median of the observed XCO_2 in the background region as the background. An assumption on the rough spatial extent of the city emission (e.g., a rectangle). Please refer to our response above regarding this assumption.

L218-219: improve it over what?

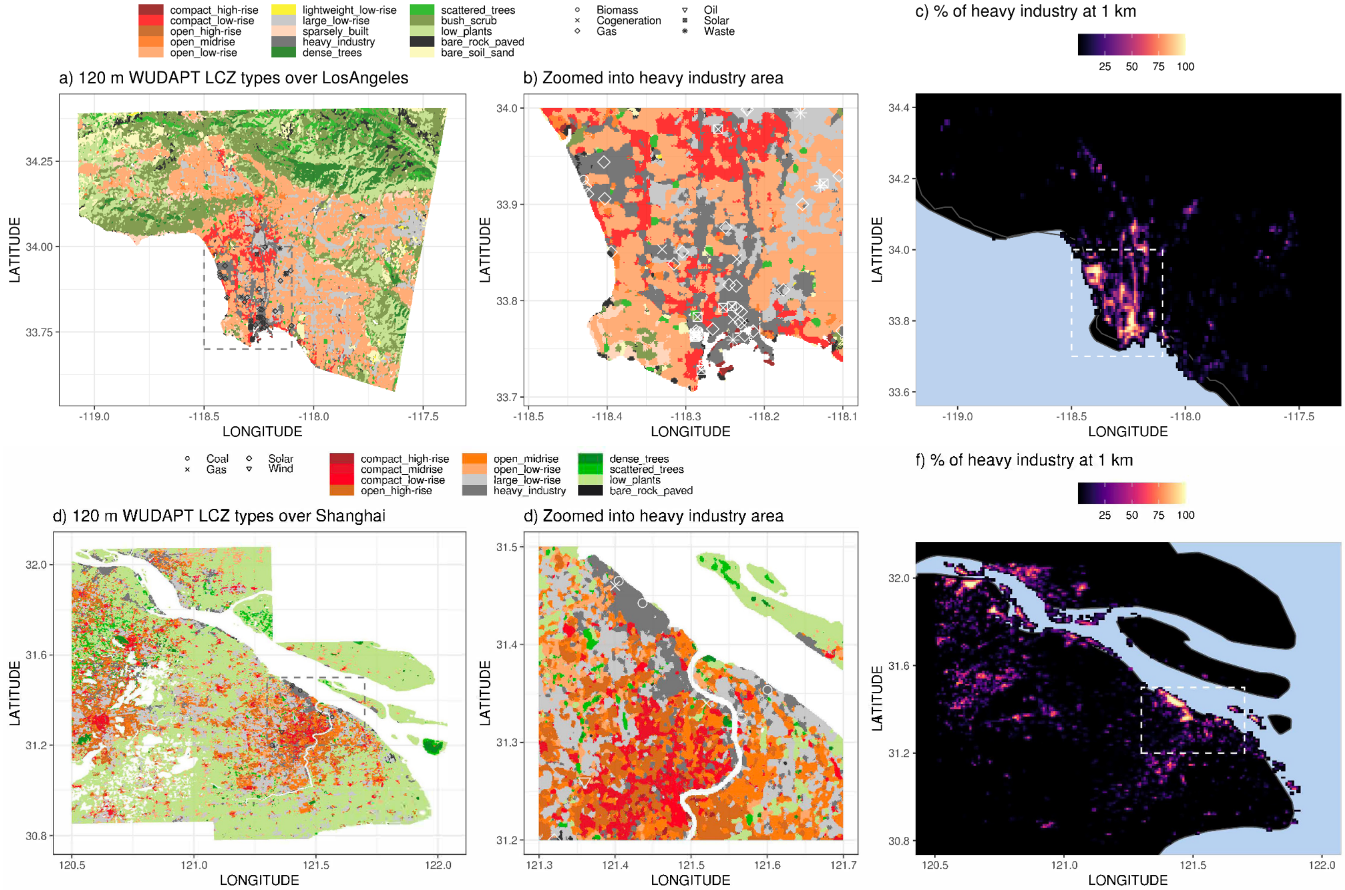
Worded as - "The broader spatial coverage and multiple swaths stretching out of the city domain of OCO-3 SAMs improve the determination of the background terms by introducing spatial variations in the background, compared to the narrow swath of OCO-2."

L222: soundings within -> soundings as within
L258-262: I would split this into two sentences.
L265: lasts for -> ranges from; L267: are -> is
L273: Observed uncertainty of XCO2 are -> Observation uncertainty of XCO2 is

All corrected.

Figure 4: Please add a coastal outline in panels c and f to make it easier to interpret. (Masking water would also be an option.); Figure 4 caption: lightgray should be two words.

Corrected. Here is the updated Figure 4 with the ocean colored in light blue:



L301: I think a word is missing. Maybe: “Those industry coverage *maps* are then convolved…”

Corrected.

L311: I would remove “originating” here, it is more confusing than helpful.

Corrected.

L313: When you write “too low valid soundings” do you mean “too few valid soundings” or “too low enhancements”?

Corrected - too few valid soundings.

L313-314: remove “the few”

L319: interfered by wind shift -> affected by shifting winds

L329-330: Recommended change: “Again, the colored contours and curves in Fig. 5 indicate neither the intensity of concentrations nor flux fields (as no prior emissions are used), but rather the likelihood of urban plumes determined solely by atmospheric dispersion.”

All corrected. It reads much better.

L332: but problematic -> but becomes problematic

L335: remove “cases”

Figure 5: in figure label, it should be “caution” rather than “cautions”

Corrected.

L364: I would suggest replacing “atmospheric movement” with “transport”. Also, something seems to have gone wrong with many of the subscripts in this paragraph (e.g. ECO instead of E$_{CO}$ in LaTex syntax).

Subscripts of E_{CO} and E_{CO_2} have been corrected.

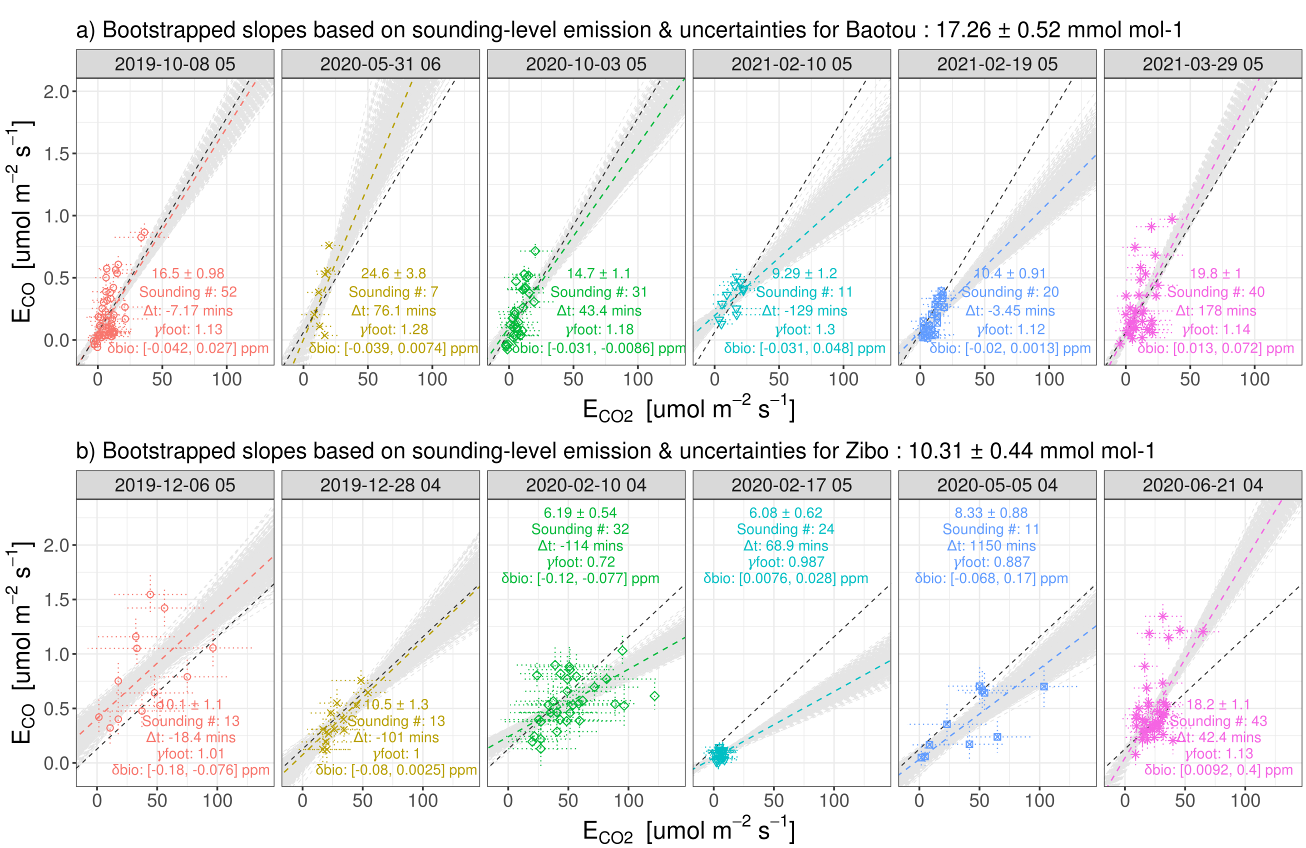
Figure 6 caption: The second sentence and the second-last sentence seem to repeat the same information. Remove one? Also, it is mentioned here that only regression lines with positive slopes were chosen from the Monte Carlo experiment. What proportion of these lines needed to be removed?

Caption corrected. Yes - the Monte Carlo analysis will remove bootstrapped linear fits with negative slopes, assuming CO and CO_2 emissions are positively correlated at the city scale (when sufficient soundings are being considered). Since it is a random process, the exact proportion of lines with negative slopes may vary each time. We added a note in the figure caption of Fig. 6:

> “Only bootstrapped regression lines with positive slopes were chosen from the Monte Carlo experiment (dashed gray lines; ~98.4% and 93.3% of the total 6,000 bootstrapped lines for Baotou and Zibo, respectively).”

Figure 6: the \gamma bio term shown on the plots should be in units of ppm, right? Also, is this somehow different from the \delta X_{bio} discussed previously? If not, please make this consistent.

We modified the two figures and text to remove \gamma bio, to reduce the number of terminologies introduced in the paper. Instead, we now printed the range of \delta X_{bio} in the interval format (i.e., min and max values of the urban-rural gradient in biogenic anomalies estimated at the TROPOMI scale, in ppm). Here is an example of Zibo and Baotou cases (please see the last row of text in each panel):



L385: remove second comma, also change “them” to “these overpasses”.
L392: in time -> in the time
L406: tend -> tends
L408 (and elsewhere): perhaps “industry-dominated” might be more appropriate in some places than “industry-dominant”?
L432: i.e., less -> i.e., those less
L452: the Solar-Induced -> remove “the”
L455: function -> functional; L456: either “interfering” -> “interfering with” or “interfering” -> “affecting”; “wind directional shift induced by” -> “the shift in wind direction due to”
L464: spontaneously -> simultaneously
L464: do you mean “a future geostationary satellite (i.e. GeoCarb)” or “future geostationary satellites”?

All corrected. We meant “future geostationary satellites”.

Figure 8 caption: Text could use a bit of work. Suggestion: “indicate the urban plumes between two times differ significantly that a simple plume rotation fails to fix” -> “indicate that that urban plumes between the two overpass times differ so much that they cannot be brought into agreement with a simple plume rotation”. Also: what is the meaning of 0~1 and 0~2? Are they shifted or not?

Corrected. We added a note in the caption to explain “0~1”: “The number on each bar (e.g., 0~1) denotes the number of TROPOMI polygons needed to be shifted to align the urban plumes at two times. For example, 0~1 means that TROPOMI polygons over certain locations are shifted by one grid.”

L479: begun -> began; L483: take -> takes
L492: to coarser -> to the coarser
L494: may locate -> may be located
L500: against OH -> against the OH
L503: contributed to 21.2% -> contributed 21.2%
L503: but negligible -> but a negligible
L504: season, the -> season, and as such the
L504: remove “likely”, “encapsulated” -> “included”, “yield” -> “have”
L506-507: “…whether … remains unclear” -> “it is unclear whether AVOCs… of interest.”
L512: “can help” -> “to”
L513-514, and L521: inventory -> inventories
L515: footprint -> footprints
L520:
L527: in informing locations of -> to provide information about
All corrected.

Supplement:

Figure S1 caption: average -> averaged

Corrected.

Figure S3 caption: unique to each satellite sounding given unique -> unique to each satellite sounding, giving a unique; Figure S3 caption: Column footprint -> The column footprint

Corrected.

Figure S4 caption: “these resultant normalized fraction Pind(x, y) informs the influence on the observation at a given sounding (white rectangle) due to heavy industry. Lighter the color, stronger impact from heavy industry in LA.” -> “these resultant normalized fractions Pind(x, y) show the influence of heavy industry on a given sounding (white rectangle). The lighter the color, the stronger impact from heavy industry.”

Corrected.

Figure S5 caption: “of urban-background” -> “of the urban-background”; “Since biogenic” -> “Since the biogenic”; “Two sets of footprint totals” -> “The two sets of footprints”; “difference in AK” –> “difference in the AKs”; “Higher the footprint ratio, larger the discrepancies” -> “The higher the footprint ratio, the larger the discrepancy”

Corrected.

Figure S6 caption: “during two” -> “during the two”

Corrected.

Figure S7 (and others): Is the date format in the plots (YYYYMMDD) consistent with the Copernicus guidelines? Figure S7 caption: “close to the noon, daytime carbon sink dominant leading” -> “close to noon, the daytime carbon sink dominates, leading”; “nighttime respiration dominant,” -> “respiration dominates the biogenic fluxes,” (I would remove “nighttime” because winter cases are also mentioned.)

Fig. S7 has been revised to show the dependence of overpass time/hour, according to a minor comment from reviewer #1. And the date format has been modified -

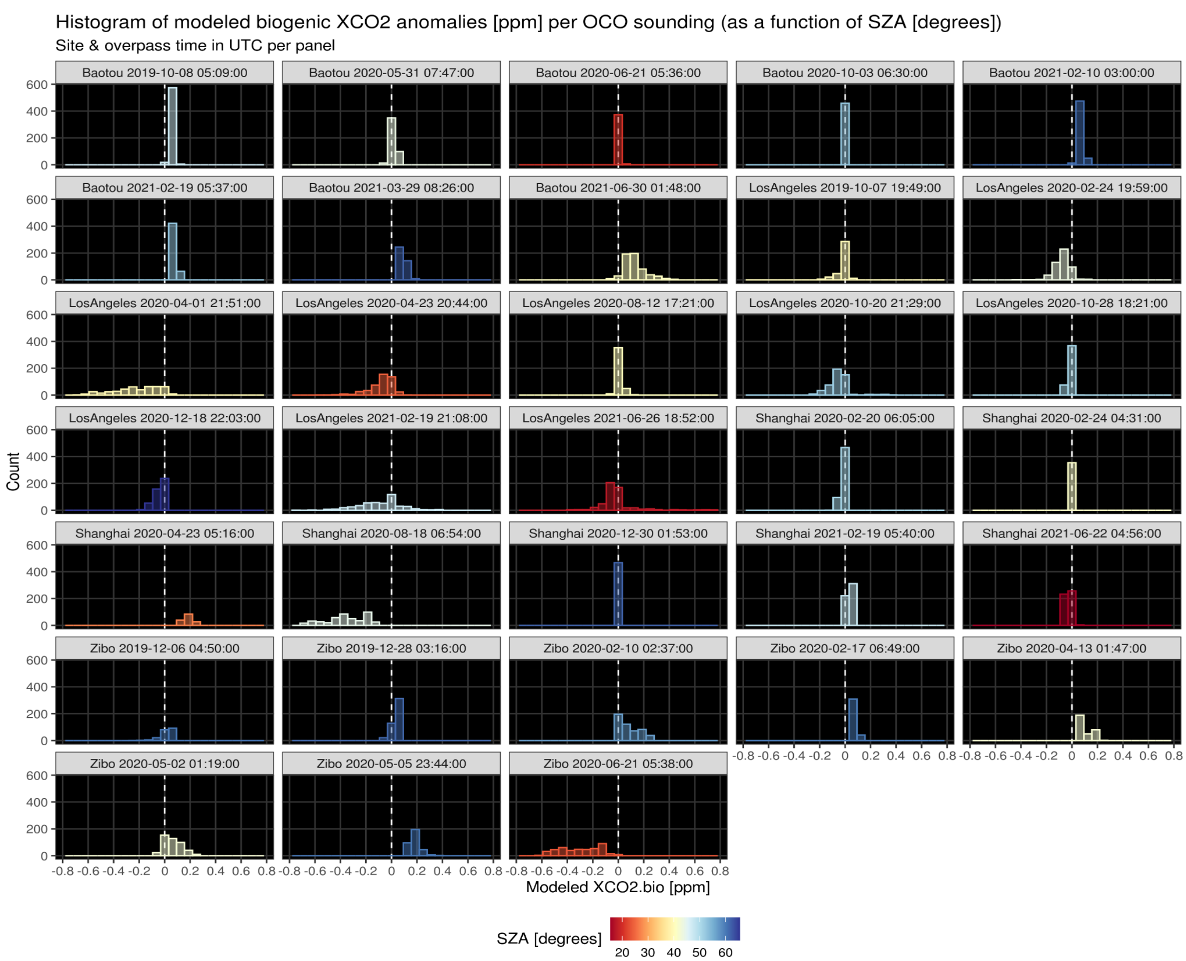


Figure S9 caption: “with or without the account of the urban-rural biogenic gradient over Zibo on June 21, 2020” -> “over Zibo on June 21, 2020, with and without taking the urban-rural biogenic gradient into account”; “light grey shading denote” -> “light grey shading denotes”; “Such positive gradient” -> “Such positive gradients”

Corrected.

Figure S10 title: A bit awkward, would suggest “Log-normal distributions of ERCO [ppb-CO / ppm/CO2]; Figure S10 caption: “on log-normal” -> “on a log-normal”; “stewed” -> “skewed”; Also, aren’t the values skewed towards the higher end (i.e. positive skew or right-skewed) if the mean is higher than the median?

Right - right-skewed when mean is larger than median. The title and caption have been corrected.

Figure S11: Should the title for panels c) through f) be Sectoral CR_CO or ER_CO? Also, I guess panel f) should be marked “residential” rather than “resident”? (Perhaps “road traffic” might also be better for d)…)

Right - those panels c) through f) are simulated enhancement ratios using EDGAR-based sectoral emissions. Just for clarification, here are the FF sub-sectors from EDGAR we selected and how we modified them.

 # ENE: Power industry ***-> renamed as ‘power’ (Fig. S11e)***
 # TRO: Road transportation ***-> renamed as ‘on-road’ (Fig. S11d)***
 # RCO: Energy for buildings ***-> renamed as ‘resident’ (Fig. S11f)***

Subsectors that were ***combined into one sector as “industry” (Fig. S11c)***
 # IND: Combustion for manufacturing
 # CHE: Chemical processes
 # NMM: Non-metallic minerals production
 # NFE: Non-ferrous metals production;
 # IRO: Iron and steel production

We have now modified panel titles for the bottom panels.