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 pasted.

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.

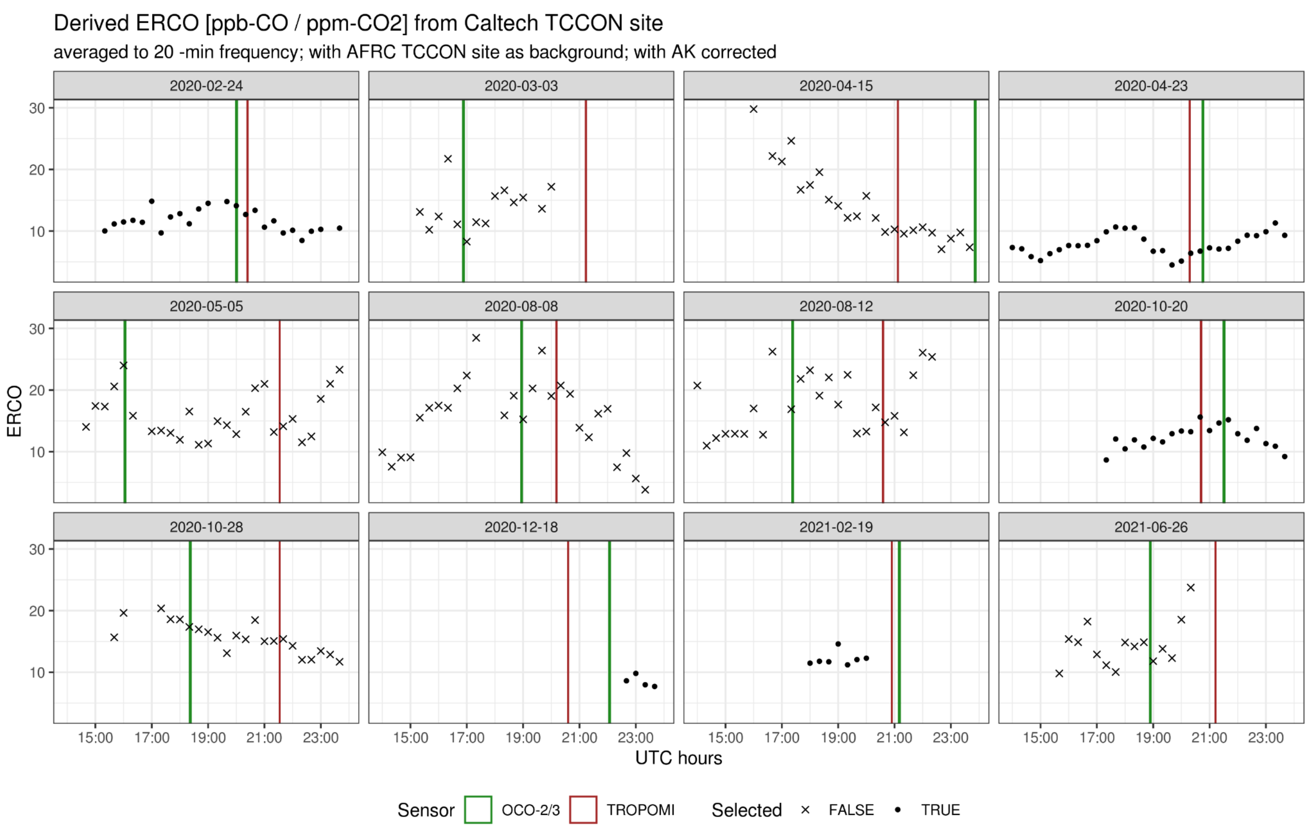


Fig. S8 - Time series of observed ERCO at the California Institute of Technology (Caltech) TCCON site (Wennberg et al. 2017) on the OCO-3 overpass dates till June 2021. The background is defined using the NASA Armstrong Flight Research Center (AFRC) site near Lancaster, California (Iraci et al., 2022). 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

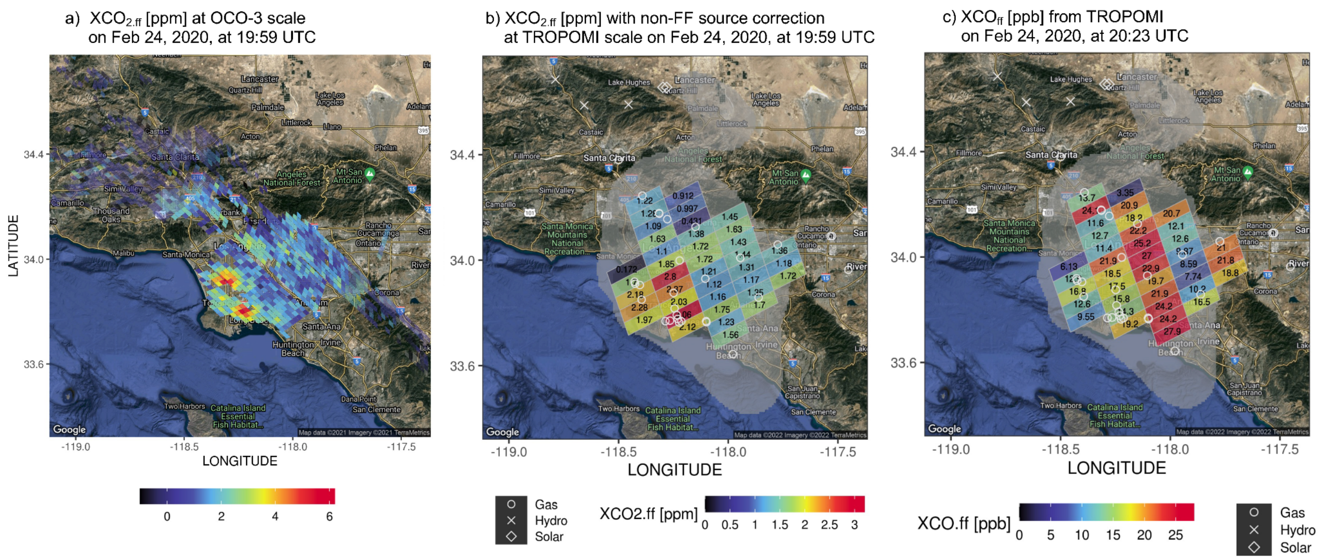
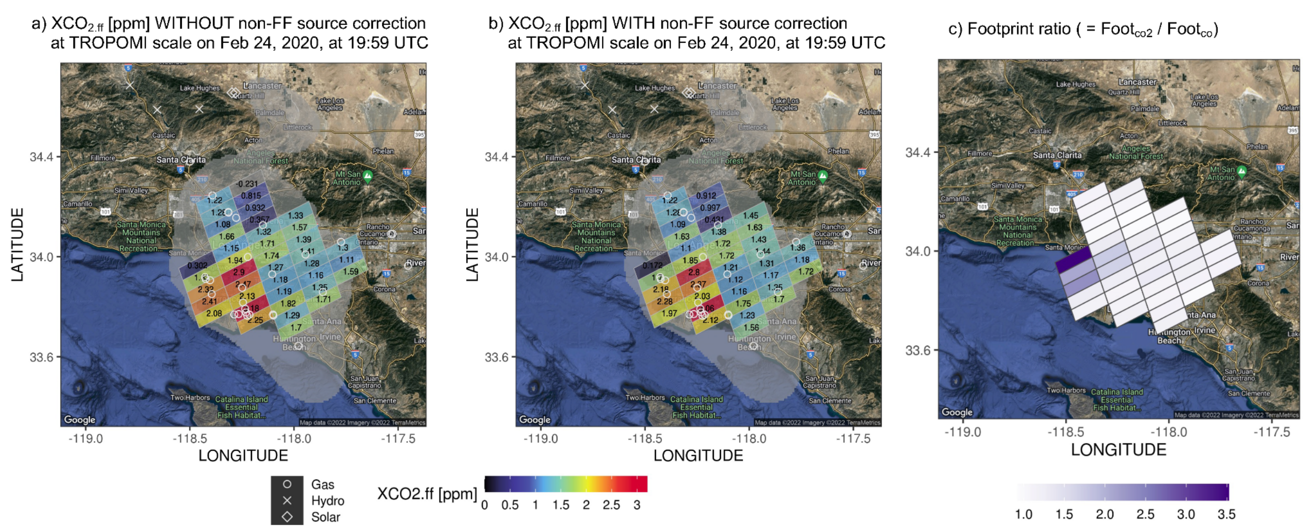


Fig. S5 In general, $XCO_{2.ff}$ within the urban plume with biospheric corrections appears to be slightly smaller than those without the biospheric corrections, as explained in the main text.



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

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

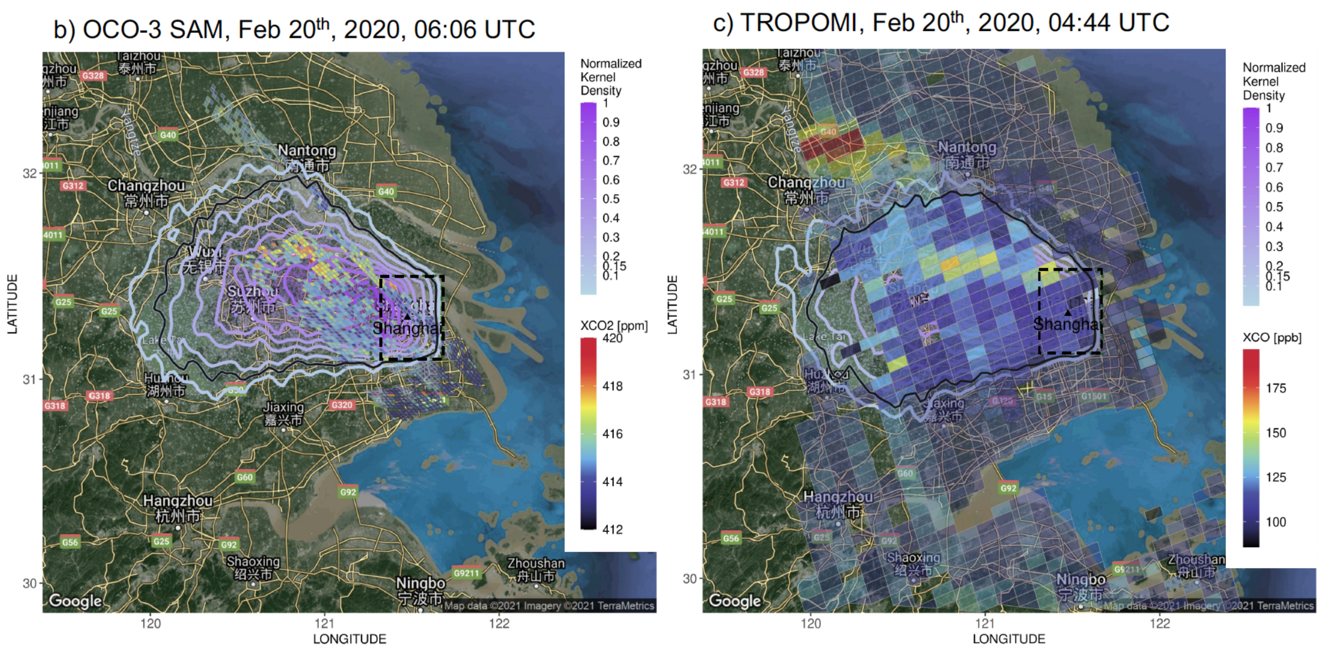
- The first statistical method utilized standard deviation or percentiles to derive the background purely from observed XCO_2. So, no use of prior emissions.

- 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.



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:

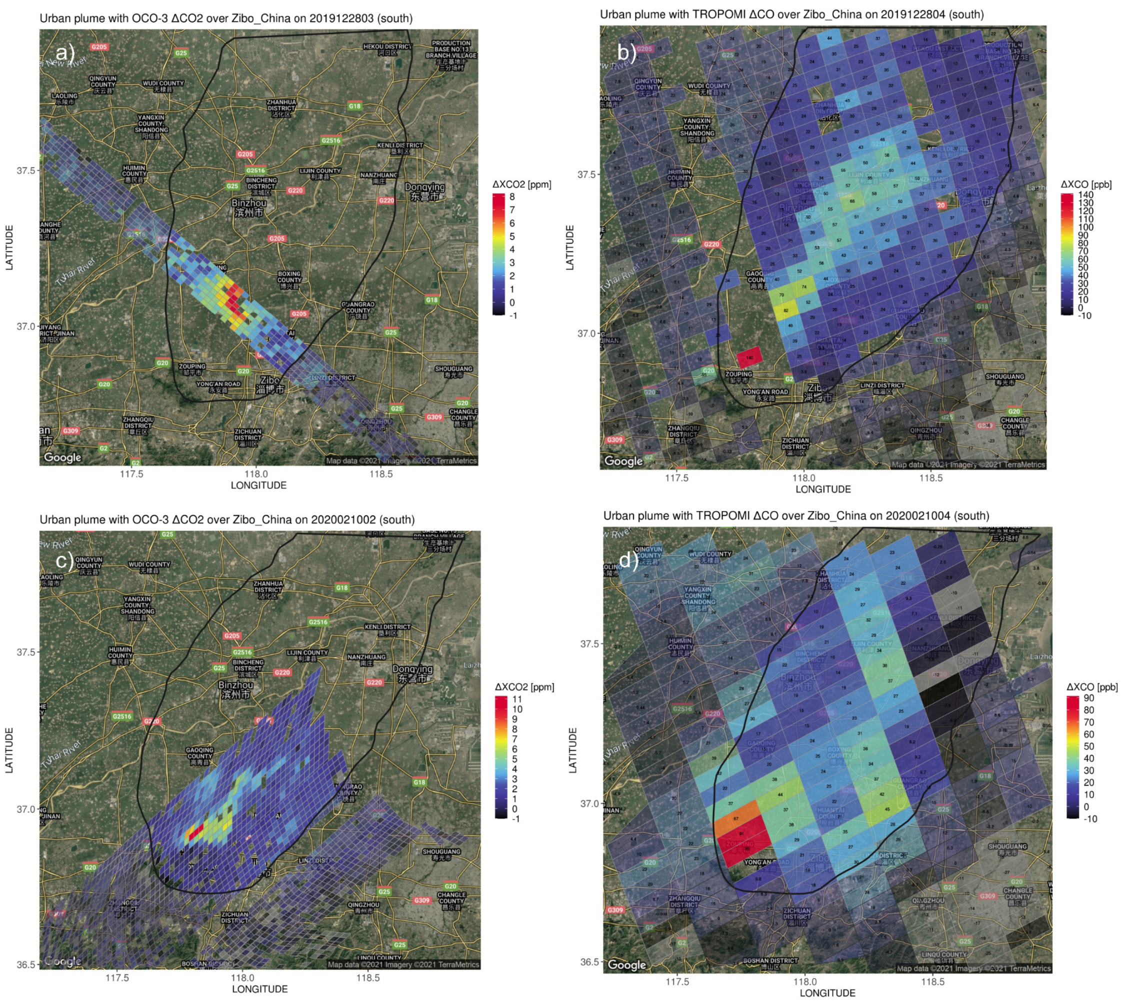
"As explained in **Sect. 2.2.2**, urban-background gradients in these biogenic anomalies (i.e., δX_{bio}) were used to correct the constant background X_{bg} (Eq. 1)."

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

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

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 $0.5° \times 0.5°$ 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

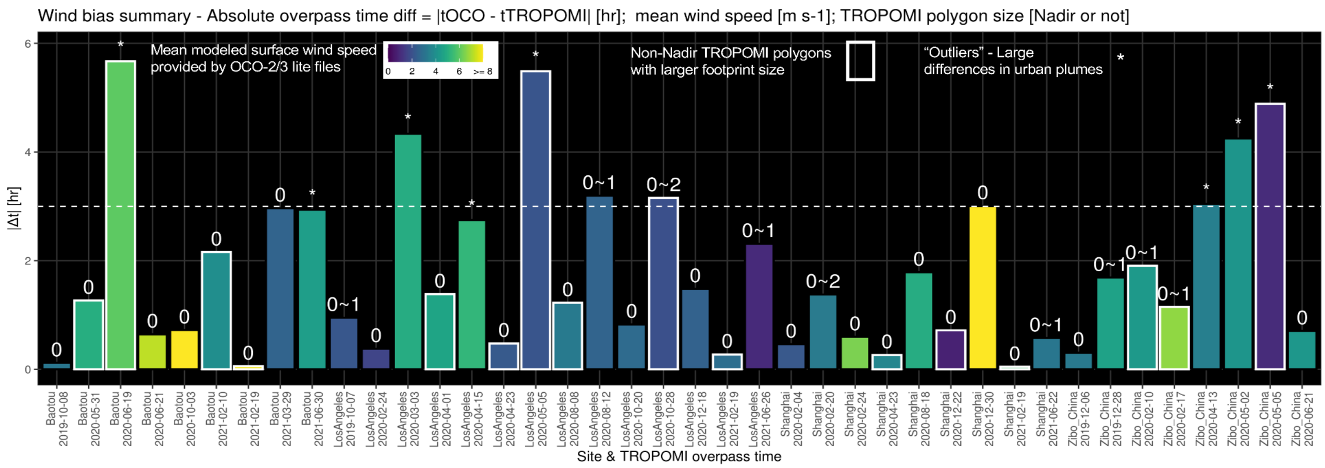
459-460: “...constraint can be relaxed”. This statement may need to be reconsidered when also taking into account the changes in emissions of (FF) CO2 and CO during the day (see major comments).

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

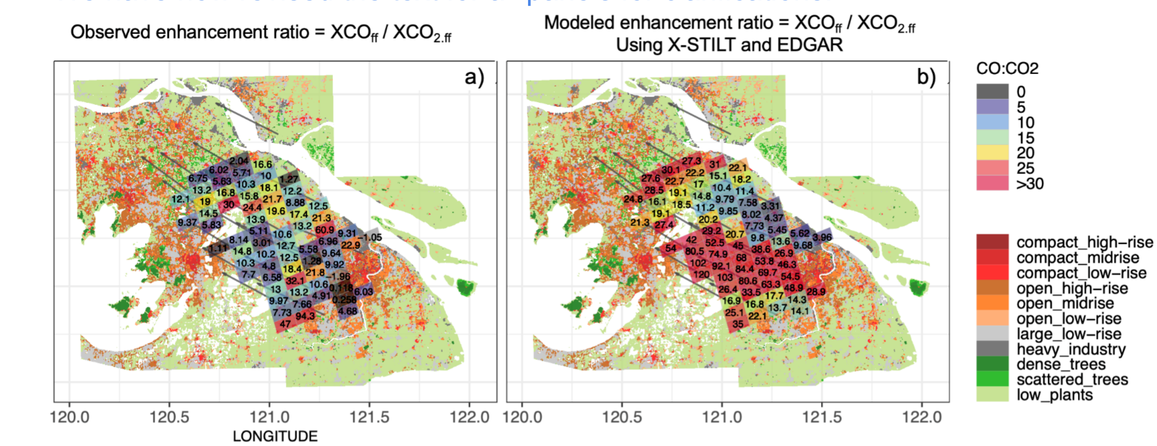
 # PRO: Fuel exploitation

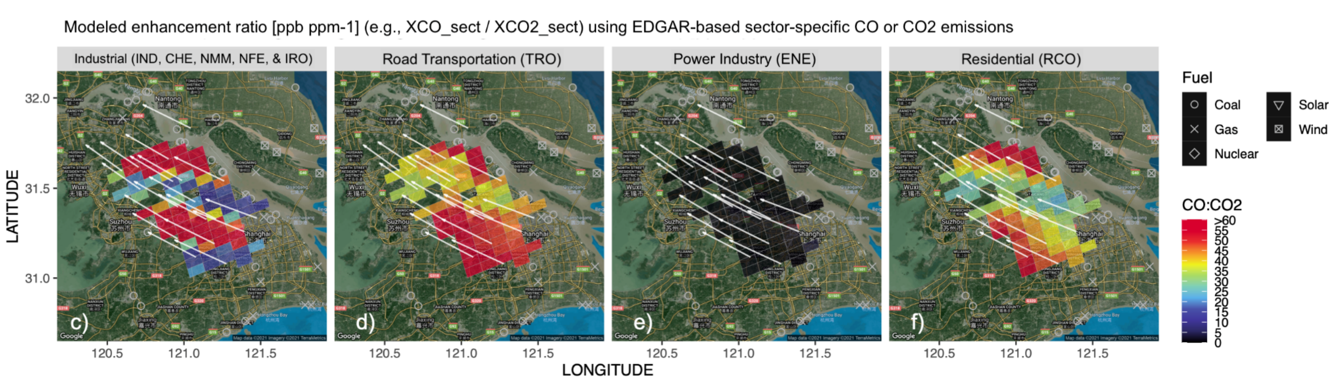
 # TNR: Other transportation sources

 # NEU: Non-energy use of fuels

 # PRU_SOL: Solvents and products use

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

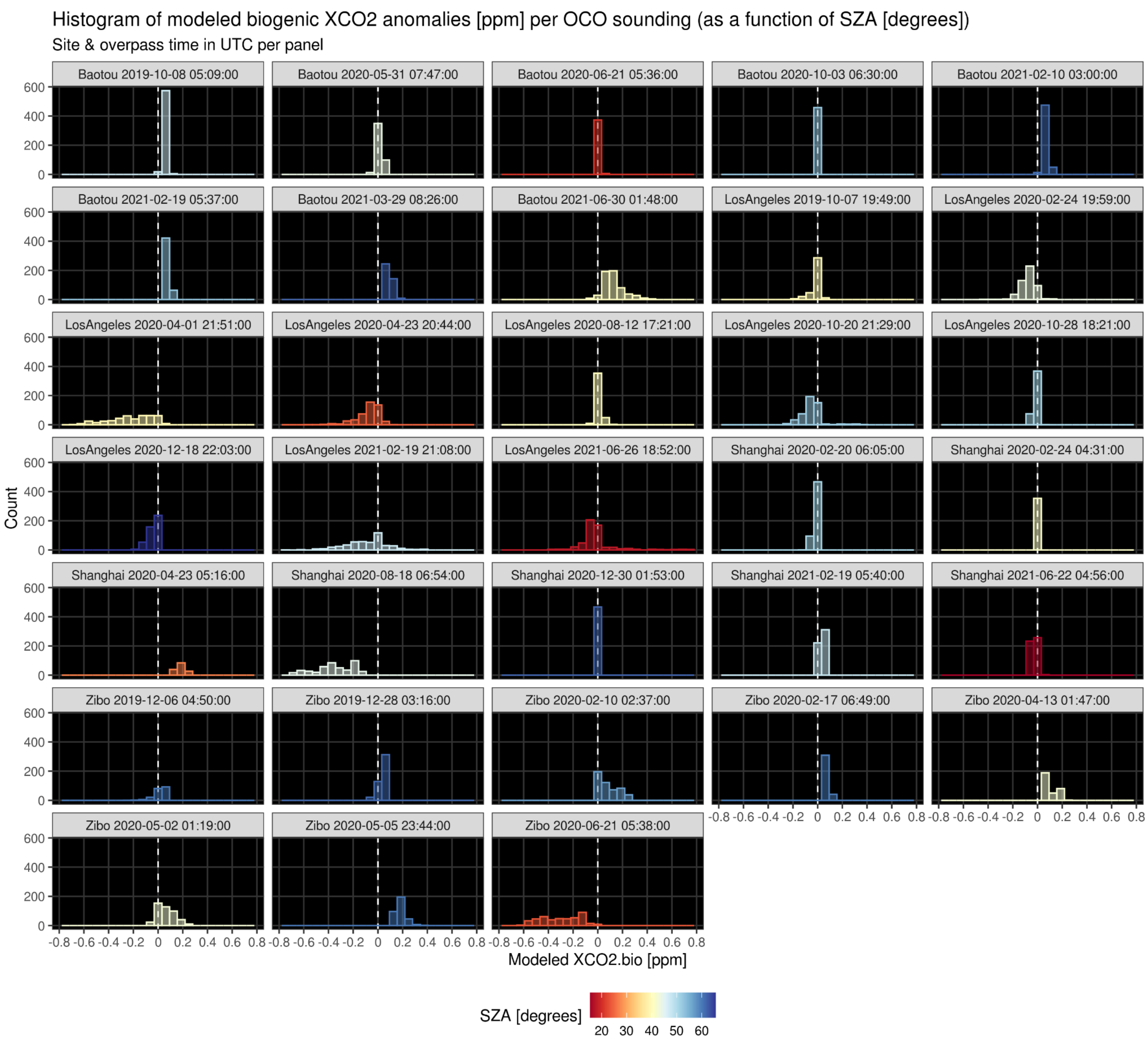




Supplementary information

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”?

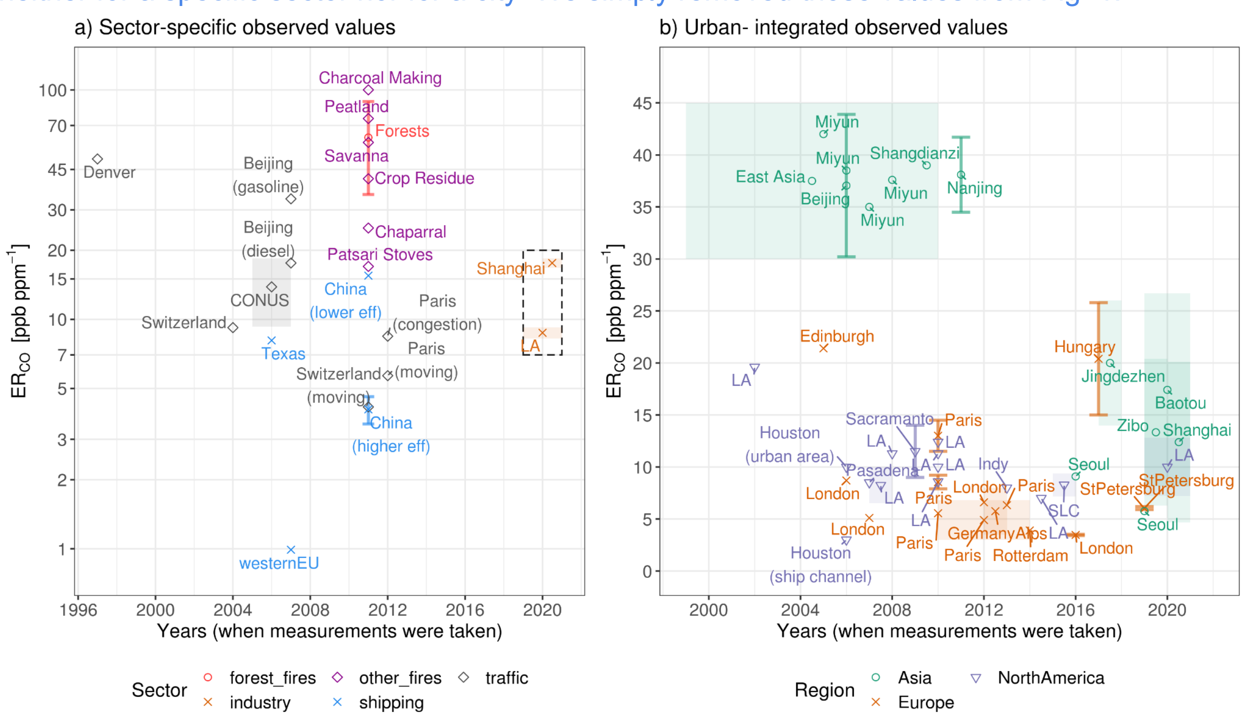
56: “diagnosed”. consider using a different term, e.g. “analysed”.

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!