

Response to Referee #2

We would like to thank reviewer #2 for taking the time to review this manuscript and for providing valuable, constructive feedback and corresponding suggestions that helped us to further improve the manuscript.

In this author's comment, all the points raised by the reviewer are copied here one by one and shown in black color, along with the corresponding reply from the authors in blue.

This paper by Tu et al., focus on observations at an industrial park and simulations of Xining's emissions using portable Fourier transform spectrometer and TROPOMI observations. The topic is interesting and falls into the scope of ACP. I have some major comments that may improve the quality of this paper.

Major concerns:

May-June may be too short to represent the whole year, and in winter there are coal-burning period for heating. Do the authors have longer time observations? Please at least add some discussions on this time coverage influences.

We thank the referee for raising this important point. Our study is indeed based on a three-week intensive campaign, and the number of valid observation days was further reduced due to unfavorable weather conditions.

We acknowledge that the limited time coverage may not fully capture the seasonal variability, especially during the winter heating period. We have now included a discussion in the revised manuscript addressing this limitation and its possible influence on the representativeness of our results:

“The observed discrepancies compared with inventories may be attributed to differences in temporal coverage, methodological approaches, and potential changes in emission patterns over time. Additionally, it should be noted that the field campaign spanned only three weeks from May to June, which mainly represents early summer. During other seasons, such as summer or winter, when photosynthesis activities or coal burning for heating is more prevalent, the $\Delta XCO/\Delta XCO_2$ ratios and associated CO_2 emissions may differ. A longer period of ground-based observations and running several spectrometers upwind and downwind may improve our results. Our findings so far demonstrate the potential of the EM27/SUN spectrometer as a promising tool for comprehensively evaluating greenhouse gas (GHG) and air pollutant emissions in urban areas (Che et al., 2022b; Lee et al., 2024).”

CAMS resolution and emissions information may be too sparse to include local emission areas and may not be appropriate for the comparison.

CAMS inventory has a relatively high spatial resolution of $0.1^\circ \times 0.1^\circ$, which allows for reasonably fine-scale emission estimates. Our comparison is based on TROPOMI CO data over a regional area, making the datasets generally comparable.

As also noted by another referee, we have revised the collocation criteria in the updated manuscript to use the CAMS data from the nearest grid cells to the location of the EM27/SUN instrument. The relevant figures and text have been updated accordingly.

I suggest the authors include analyses and comparisons with open accessed inventory (e.g. MEIC). And add some discussions on the difference between inventory and inversions.

We have added discussion about MEIC inventory to section 3.5. A figure presenting emission from this study and different inventories has been added to the manuscript:

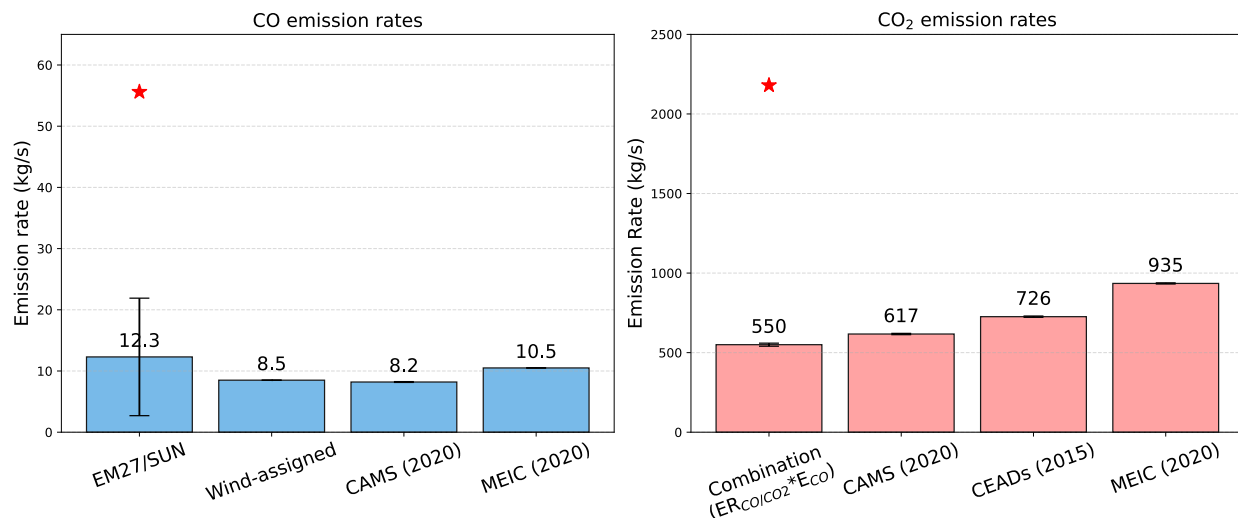


Figure 5: CO and CO₂ emissions from this study and different inventories. The red start symbols represent the highest value derived from EM27/SUN observations.

More discussions are also added in the manuscript:

“We estimate an average CO₂ emission rate of approximately 550 kg/s, which aligns well with the CAMS-GLOB-ANT (617 kg/s for 2020) though lower than the Carbon Emission Accounts and Datasets (CEADs) (726 kg/s for 2015) (“Methodology and applications of city level co₂ emission accounts in China,” 2017; Shan et al., 2018) and MEIC (935 kg/s for 2020) estimates. The data also reveal strong daily fluctuations in emissions. The peak event was observed on May 27, which exhibited a maximum $\Delta X_{CO}:\Delta X_{CO_2}$ ratio of 40.08 ($R^2 = 0.8544$). This ratio translates to a maximum CO emission rate of 55.6 kg/s and a concurrent maximum CO₂ emission rate of 2180 kg/s.

Additionally, the CAMS and MEIC inventories show similar CO/CO₂ emission ratios of 0.021 and 0.018, respectively. As detailed in Section 3.3, both TROPOMI and CAMS underestimates the atmospheric CO column by a factor of approximately 1.6. When we correct for this bias by scaling the TROPOMI-derived emission and CAMS inventory, the resulting emission ratio increases to 0.034. This corrected value aligns closely with our ground-based observed $\Delta X_{CO}:\Delta X_{CO_2}$ enhancement ratio of 0.035 ppb/ppb.”

Spatial distributions associated with the TROPOMI data, simulations and inversions are needed to improve the content of this paper.

Thank the referee for this suggestion. In section 3.4, we have addressed this by applying a multi-year inversion of emissions based on TROPOMI data, using a dispersion model coupled with a wind-assigned anomaly method.

Our method uses a dispersion model driven by wind fields and a priori emissions to simulate plume enhancements. A wind-assigned anomaly technique is then applied to both the TROPOMI data and the model simulations. This technique calculates the difference in enhancements under opposing wind conditions, effectively removing background bias. The final emission inversion is derived by scaling the a priori emissions to minimize the difference between the modeled and observed anomalies. We have added the explanation of this approach in Section 2.6:

“2.6 Dispersion model and wind-assigned anomaly method

For a single point source, the total emission is calculated by multiplying the measured total column enhancement (ΔCO) by the area of the affected plume (Babenhauserheide et al., 2020). This plume area is modeled as an evenly distributed cone, representing the long-term averaged dispersion (Tu et al., 2022a). The relationship is given by the following equation:

$$\varepsilon = \Delta CO \times d \times v \times \partial \quad \text{Eq. 1}$$

where ΔCO represents the enhanced CO column observed at the downwind site, d is the distance from the source to the measuring site and v is the wind speed.

To estimated averaged emissions from satellite observations over a region, the wind-assigned method was applied (Tu et al., 2022a, 2022b, 2023, 2024b). This technique fits the anomalies between the satellite observations and the dispersion model by analyzing enhancements under opposing wind sectors. Specifically, the wind-assigned anomaly is defined as the difference in observed enhancements between two opposite wind fields (e.g., E: 0°–180° and W: 180°–360°). A key advantage of this approach is that it inherently eliminates the uncertainty associated with background concentration calculations for long-lived gases like CO, thereby significantly improving the reliability of the resulting emission estimates.”

Besides the CO and CO₂ emissions rates, the CH₄ emissions rates are also important.

We appreciate the referee’s comment and agree that CH₄ emissions are indeed important. Ground-based FTIR measurements did capture CH₄ concentrations. However, we found that the ΔXCH_4 does not exhibit a consistent correlation with ΔXCO or ΔXCO_2 (Figure 1), unlike the more stable relationship between ΔXCO and ΔXCO_2 . This consistent correlation between CO and CO₂ suggests co-emission, aiding the reliability of CO₂ emission estimates from CO. In contrast, the variable correlation for CH₄ introduces greater uncertainty in estimating its emissions using the same methodology applied to CO. Longer observation periods may help improve these correlations and refine CH₄ emission estimates. Additionally, the weaker correlation between ΔXCH_4 and the other species may indicate that CH₄ is not significantly co-emitted with CO and CO₂.

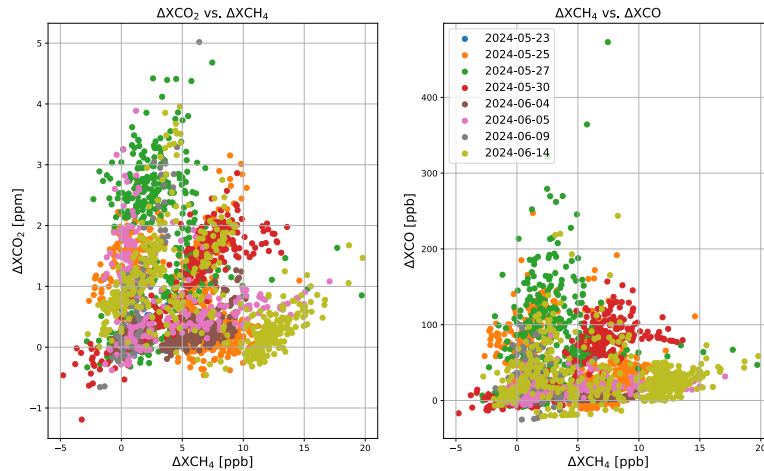


Figure 1: correlation between ΔXCO and ΔXCH_4 (left), and between ΔXCH_4 and ΔXCO (right).

Additionally, the availability of TROPOMI XCH_4 data in this region is limited (see Figure 2), with only about 3000 observations collected over five years. This relatively small dataset also makes it challenging to estimate CH_4 emissions accurately from the TROPOMI dataset in this region.

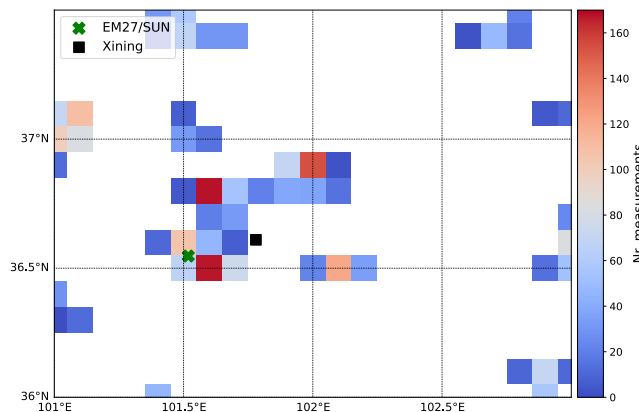


Figure 2 total number of measurements in each grid during May 2018 – May 2024.

It is also important to note that the primary focus of this study is on connecting satellite and ground-based remote sensing observations, specifically by estimating CO_2 emissions from the ground-based observed $\Delta XCO/\Delta XCO_2$ ratio particularly when CO_2 observations are sparse. As such, CH_4 emissions are not addressed in this analysis.

Minor comments:

Add serial numbers to the subFigures in Fig.1, and the font in subFigure2 is too small and difficult to read.

Thanks. Figure 1 has been updated.

line159: Does this sentence means that CO and CO_2 come from different sources?

For better clarification, we have revised the sentence to:

“The enhancement of XCO and XCO₂ ratio ($\Delta XCO:\Delta XCO_2$, see section 3.5) exhibited slopes of 14.43 ppb/ppm before noon and 4.76 ppb/ppm in the late afternoon. Both values were significantly lower than those observed under easterly wind conditions. This suggests that the CO and CO₂ emissions in the western regions originate from different combustion processes or source types compared to those in the east.”

Add more descriptions for Fig.2 (a,b,c). And for Figure3, do data from TROPOMI (5.5 km × 7 km) and COCCON (point) have comparable spatial representativeness? What processing methods were applied? These should be explicitly stated in the Methods and in discussions.

We thank the referee for this suggestion. We have expanded the discussion on Fig2 in section 3.1.

COCCON is a network of ground-based remote sensing FTIR spectrometers that supplements the existing TCCON stations. Like TCCON, COCCON provides column-averaged concentrations that are directly representative of the local conditions above the measurement site. To ensure a robust comparison between the point measurements from COCCON and the integrated area measurements from TROPOMI, it is crucial to apply appropriate spatial and temporal collocation criterion. This method is well-established in the literature for satellite validation (e.g., Klappenbach et al 2015, Velazco et al 2019, Tu et al 2020, Knapp et al 2021, Alberti et al 2022, Sha et al 2024).

Various studies have applied different spatiotemporal criteria based on the characteristics of the satellite data and ground-based measurements. For example, Klappenbach et al. (2015) used a 5° latitudinal/longitudinal radius and a 4-hour temporal window for GOSAT overpasses. More stringent criteria have been used in subsequent studies, including a 100 km – 200km spatial radius and a ±1 to ±2 hour temporal window for GOSAT (Velazco et al 2019) and TROPOMI (Tu et al 2020, Sha et al 2025).

In this study, to ensure sufficient data pairs for robust validation, we applied the following collocation criteria:

- Spatial: a 200 km radius for XCH₄ and 100 km radius for XCO.
- Temporal: a ±2h window around the COCCON measurements to align with TROPOMI overpasses.

These criteria are stated in section 3.2 of the manuscript.

lines 193-195: Why not match the COCCON data with the grid scale of CAMS? At distances beyond 20km or even 50km, and the factors influencing observations or forecast results are local emission sources and atmospheric transport processes.

We thank the referee for this comment. The CAMS forecast data have a spatial resolution of 0.1°×0.1°. We have revised the collocation criteria to use the CAMS data from the nearest grid cells in which the EM27/SUN instrument was located. The relevant figures and text have been updated throughout the manuscript.

Figure4b: The data points are overly clustered. It is recommended to reduce the range of the x-y axes, for instance to 1870-1950. And other subplots also need to be improved for this aspect.

Thanks. We have updated this figure.

Figure4c: The legend should not overlay the data plots.

Thanks. We have updated this figure.

Line 205: To what extent is this underestimation a result of observation? Have you considered spatial representativeness inconsistency as a potential source?

This underestimation in satellite observations might due to errors at higher altitude

Line 217: Enhanced relative to what?

The enhanced CO column is relative to the background, i.e., representing the emitted CO.

lines 225-227: The definitions of background CO concentration and ΔXCO should be provided when these terms were firstly appeared.

Thank the referee. We have provided the definitions when these terms were firstly appeared in the manuscript.

Line 241-242: Has the higher emissions led to the observed concentration peak?

The referee is right that the higher emissions contribute to observed peak on this short time ($\sim 1h$). To observe concentration peaks is also largely due to the wind direction. Peaks are easily observed when observe site is exactly in the downwind of the sources and the wind is steady.

12 Why only analyze the CO emission and the relation of ΔXCO and ΔXCO_2 ? How about CH_4 ?

Thank the referee for raising this point. The XCO and XCO₂ enhancements show a clear correlation, reflecting their co-emission from fossil fuel combustion and biomass burning, especially in urban region. However, we did not find a consistent correlation between CH_4 and either CO₂ or CO (as discussed in the major concerns), suggesting that CH_4 emissions in the study region are influenced by additional sources beyond combustion.

13 Please have the manuscript polished again for grammar and spellings.

Thanks. We have tried our best to modify the manuscript.

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