This study uses continuously measured methane measurements at three tower locations in and around Hangzhou, China, to investigate temporal variations of emissions, especially from the treatment of waste. The authors use the WRF-STILT (Weather Research and Forecasting-Stochastic Time-Inverted Lagrangian Transport) model combined with a Bayesian inversion framework to compare the data driven results with the prior emissions inventory. They conclude that emissions have been overestimated for the city of Hangzhou and that there is a seasonality to the emissions that can only be explained by the waste treatment sector.

This topic is very timely and important for understanding the influence of climate change on emissions of this high global warming potential pollutant, but several issues in this paper need clarification before publication.

Thanks so much for these detailed suggestions. All points have been addressed below (review query in Italic; author response in blue). Changes to the text in the manuscript have been marked in blue.

Of the three sites, it appears that only one is in the city (Hangzhou), and one is on a relatively remote mountain (Damingshan). Is the third site, Linan, in a suburb or also background region, as stated on line 177? If this is true, then there is only one site that is truly relevant to determining emissions from the city, since the other two are described as background sites. However, background values are taken from much more remote sites. There can be significant sources between the very remote sites and the urban region being studied, including large cities such as nearby Shanghai between Hangzhou and TAP and RYO, the latter being used almost always as background. The footprint for the Damingshan site is only slightly influenced by emissions in the urban core.

For all three sites used in this study, CH_4 concentration at Hangzhou site was used to constrain emissions for Hangzhou city, and CH_4 concentration at Linan site was used to constrain emissions for much larger regions as Zhejiang province or Yangtze River Delta Area. The reason of choosing two sites in the emission constraint are mainly based on simulated enhancement contributions from different regions. The explanations are also displayed on lines 399-404 as "We further calculated anthropogenic contributions from Hangzhou city (excluding wetland because of coarser spatial resolution for Hangzhou city) and other provinces, which were 158.4 ppb at Hangzhou site, 30.7 ppb at Linan site, and 10.1 ppb at Damingshan site, respectively. And they accounted for 69.3%, 34.0%, and 16.9% of total anthropogenic enhancements at corresponding sites. These results indicate the CH_4 observations at Hangzhou site, which is located at the core urban region, was more influenced by local emissions (mainly for waste treatment and will be discussed later) and contain much higher enhancements than other two sites. The relative contributions from different regions also imply that the observations at Linan and Damingshan sites can present CH_4 emissions of much larger region as Zhejiang province or YRD area than Hangzhou city (Figure 4e)."

The reason to use different background sites at the edge of simulation domain instead of Damingshan site have been explained on lines 210-221 as "Note some previous studies of city scale greenhouse gas concentration observation networks chose sites at the edge of urban borders as background in emission inversion system (i.e. Indianapolis, U.S.A., Miles et al., (2017); Los Angeles, U.S.A., Verhulst et al., (2017); Washington, DC-Baltimore, U.S.A., Lopez-Coto et al., (2020); Paris, France, Lian et al., (2021)), but we chose to use five CH_4 background sites as the potential background to be selected including UUM, TAP, YRO, YON and WLG site (Figure 1a), which were much further than the

observations at Damingshan site. This strategy is based on following three reasons: (1) our footprint domain is much larger than Hangzhou city and these five sites are also located close to the edge of model domain; (2) CH_4 concentrations within Hangzhou city will be influenced by seasonal varied monsoon and the monthly varied wind directions will lead to obvious changes of CH_4 background than only at Damingshan site; (3) our model setups can partition CH_4 enhancements from within Hangzhou city and other regions."

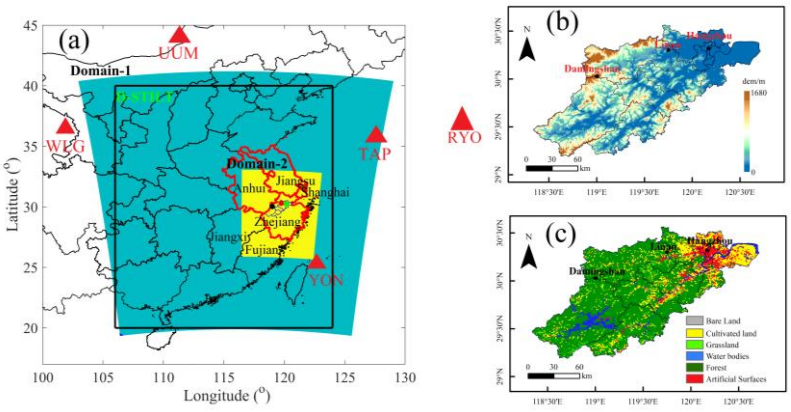


Figure 1. (a) WRF-STILT model domain setups, three CH_4 concentration observation sites in Hangzhou city, and five CH_4 background sites, note the green, red and black dots represent locations for Hangzhou site, Linan site and Damingshan site, respectively, Yangtze River Delta regions is displayed in red boundary, back rectangle represents domain in STILT model, (b) geophysical height within Hangzhou city, (c) land surface categories in Hangzhou city.

Table S1. The choice of CH_4 background based on simulated monthly footprint, 'Y' indicates concentration at this background site (or averages of both) will be used as CH_4 background for this month.

Sites	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
TAP												
YON								Y				
RYO		Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
WLG												
UUM	Y	Y										

We agree that the simulated enhancements using more remote sites as background contain contributions from other cities as Shanghai, our emission constraint results can also represent Hangzhou city based on the following two reasons: (1) the contribution from Hangzhou city accounted for majority of all enhancement (~70%) for CH_4 observations at Hangzhou site; (2) the *posteriori* SFs of the monthly scaling factors mainly represent temporal variations and revised the *a priori* bias, these SFs should be reasonable for a much larger regions and represent the common bias in *a priori* EDGAR inventory.

What emissions did you use for the prior? It seems like you used the EDGAR v6.0 inventory for anthropogenic sources (except rice patties) and WetCHARTs for wetland emissions, including from rice patties. Please state explicitly how you calculated the prior – "a priori" is not mentioned in the WRF-STILT model setup section.

The EDGAR v6.0 and WetCHARTs are used as *a priori* emissions, we revised the sentence as "The most recent inventory of Emission Database for Global Atmospheric Research (EDGAR v6.0), which has 20 categories, and WetCHARTs ensemble mean were used as the *a priori* anthropogenic and natural CH_4 emissions." on lines 247-249.

A major assumption of the paper is that waste treatment is the dominant source of emissions and the other anthropogenic sources do not contribute to the seasonality of the observed CH_4 measurements. What you show in Figure 4d is that waste treatment contributes most to the CH_4 signal, but the other sources are also important. Perhaps you can show a map of the locations of the anthropogenic sources – power plants (especially natural gas powered), landfills, wastewater treatment plants, distribution lines for natural gas, refineries, dairies, rice paddies – especially close to the urban center. Enlarge the urban center to show locations. I am not convinced that you have enough information to discount the influence of other CH4 emissions sources or to characterize the sources in the urban center with only one site, especially when the reader does not know the sources in the region or the general seasonal wind patterns. A measurement that you might consider for the future is ethane, since fossil-fuel-derived CH4 contains measurable C2H6, whereas biological sources (including waste treatment and wetlands) do not. Seasonality due to fossil CH4 is observed in cities, even as far south as Los Angeles. Is rice cultivation seasonal – should you expect some seasonality from this sector?

Here as displayed in Figures 7a, S7, CH_4 emissions from waste treatment, and RCO (energy for building) dominated the seasonal variations of CH_4 emissions. But contrary to waste treatment, our constrained results indicated the *posteriori* RCO CH_4 emissions did not have obvious difference with *a priori* emissions

We added more description on 475-477 as "These *posteriori* SFs for the rest anthropogenic categories and wetland indicated much smaller bias than waste treatment. The monthly *posteriori* SFs for PRO and RCO also illustrated obvious seasonal variations, but were still smaller than the *a priori* seasonality in inventory (Figure S7)."

We double checked the locations of some CH_4 emitters as landfills, resident area and RCO (energy for building), they are located in the similar locations as EDGAR, indicating overall good representativity of main anthropogenic CH_4 emissions in Hangzhou city. We agree that more tracers as ethane (C2H6) is a good tool to separate CH_4 emissions from biological and fossil CH_4 emissions, we added this suggestion on lines as "We will use multiple years' CH_4 concentration to quantify the influence of new technology and other meteorological variables on waste treatment CH_4 emissions in our following study, and we suggest other tracers (i.e. ethane, $^{14}CH_4$) are also important to separate CH_4 emissions from biological and fossil CH_4 emissions.". on lines 682-686.

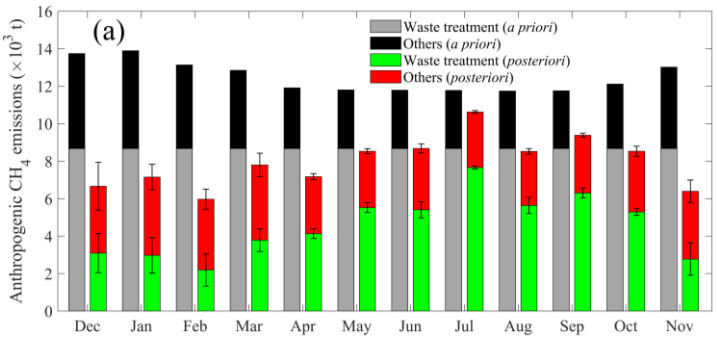


Figure 7. (a) Monthly anthropogenic (excluding agricultural soil) CH_4 emissions for *a priori* and *posteriori* emissions for Hangzhou city,

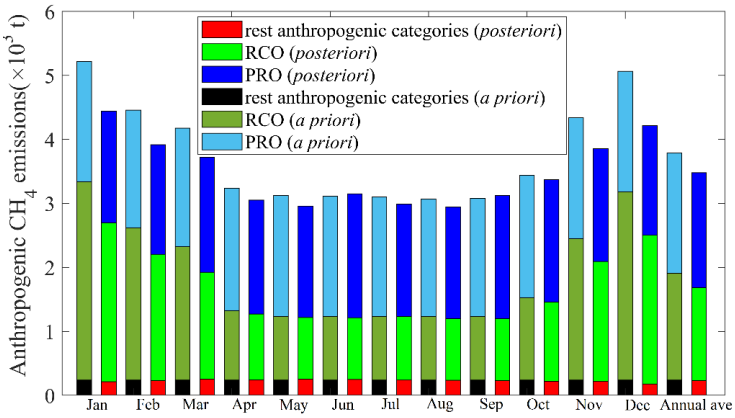


Figure S7. Comparisons of anthropogenic CH_4 emissions between *a priori* and *posteriori* results, PRO: fuel exploitation, RCO: energy for building, the rest anthropogenic emissions: excluding waste treatment, PRO, RCO and agricultural soil.

This paper uses all of the diurnal cycle of the measurements. I definitely agree that emissions at night are not captured if only afternoon measurements are used, as is commonly done. However, one reason most investigations don't use the entire 24-hour record is that WRF does not do a good job with the transport parameters at night, specifically the planetary boundary layer height (PBLH). It is very important to get this right for modeling to produce meaningful results. You don't show how your model performed for this critical parameter. Can you show how the modelled PBLH compares with measurements, even if only a limited number of measurements are available?

We contacted with local meteorological office, but they said there are not available PBLH observations during study period. But we can raise other supporting evidence for the PBLH simulations by citing one of our previous study (Huang et al., 2021), Huang et al. (2021) used the same physical schemes as this study and conducted in Nanjing city from years 2017 to 2018, which is located in the same Domain 2 and vary close to Hangzhou city. Their study found high consistence between observed and simulated PBLH in winter as displayed in following figure.

We cited this reference on lines 440-445 "Note PBLH simulations are important in evaluating model performance, we did not have direct PBLH observations to evaluate model performance, but our previous study used the same physical and PBLH schemes as this study, which was conducted in

Nanjing city in the same Domain 2 and vary close to Hangzhou city. The study found high consistence between observed and simulated PBLH in winter (Huang et al., 2021)."

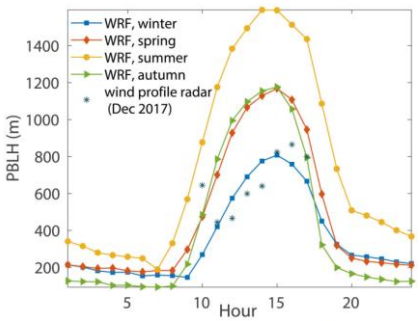


Fig. S7. Diurnal mean planetary boundary-layer height (PBLH).

More detailed comments follow:

Abstract: mention the types of waste included in this study

Thanks so much for pointing it out, we have added "(including solid waste landfills, solid waste incineration and sewage)" on lines 36-37 for clarification.

Line 72: Out of curiosity, what are the top five anthropogenic sources of CH_4 in China?

The top five anthropogenic sources in China are fuel exploitation (coal+natural gas+oil), livestock, rice paddies, waste treatment, and household use, the top four (fuel exploitation, livestock, rice paddies and waste treatment) of them accounted nearly 90% of national total anthropogenic CH_4 emissions.

Line 91: USEPA

Thanks for pointing out this typo, we have changed "USPA" with "USEPA".

Line 106: replace "absence" with "omission"

Done as suggested, we have replaced it with "omission".

Lines 143-145: City-scale studies have not focused on waste treatment sources because there are many sources, as in Hangzhou. Yadav et al. (2019; JGR Atmospheres) were able to see the effects of the closure of a landfill in the Los Angeles, CA area that was included in the prior inventory and not seen in the modelled results.

Done as suggested, we have revised this sentence as "And to our best knowledge, there is few tower-based observation inversion studies which focuses on waste treatment emissions at city scale or much larger regional scales especially in China. Only one study in Los Angeles, U.S.A. used tower-based CH_4 concentration and found the influence of landfill site closure on CH_4 emissions, which was not included in *a priori* inventory (Yadav et al., 2019)." on lines 149-153.

Yadav, V., Duren, R., Mueller, K., Verhulst, K. R., Nehrkorn, T., and Kim, Jet., Spatio-temporally resolved methane fluxes fromthe Los Angeles megacity J. Geophys. Res. Atmos. 124, 5131–5148 (2019).

Pages 6-7: In the description of the sites, please summarize the regional, seasonal wind patterns and any differences between the sites.

Done as suggested, we added more description for clarification as "Based on the wind direction for three sites, there are not obvious difference of seasonal wind direction patterns among them. The prevailing wind direction from October to February was from the north, which changed to east from February to May and then changed to south during the monsoon in summer." on lines 189-192.

Lines 188-190: How frequently were standards run? What uncertainty, including both precision and accuracy, did you assign for the measurements?

The analyzer was calibrated every 6 hours with the measured precision and accuracy within 2 ppb and 1 ppb, respectively. These uncertainties were pretty small when compared with background and we have considered these uncertainties in the Bayesian inversion. We revised it on lines 198-200 as "two different standard gas was measured every 6 hours and a linear two-point fit was used to calibrate observations, with the precision and accuracy of 2 ppb and 1 ppb.".

Line 238: What does "fuel exploitation from coal, oil, and natural gas" include? Extraction, transportation, refining, distribution, and combustion, or some subset of these?

The fuel exploitation from coal, oil, and natural gas in EDGAR contains all related processes as extraction, transportation, refining, distribution as list in IPCC database (https://www.ipcc-nggip.iges.or.jp/EFDB/find_ef.php). But such category is minimum in our study region when compared with other sources. We revised it as "PRO (all processes related to fuel exploitation from coal, oil, and natural gas)" on lines 258.

Line 239: How and where is the energy for buildings generated? E.g., natural gas power plants in the suburbs, coal burned in the buildings, …?

Here in Hangzhou, we think the energy for buildings mainly contains nature gas escape from household use. we revised this sentence as "RCO (energy for buildings, mainly containing nature gas escape from household use)" on lines 259-260 for clarification.

Lines 241-245: Is 0.5° high enough spatial resolution for your study region?

Here considering the WetCHARTs simulations have been widely used as CH_4 emissions from wetland in many previous studies, and wetland CH_4 emissions was pretty small compared with other CH_4 emissions. Hence we decided to use 0.5° WetCHARTs emissions。

Line 287: reference for CCGCRV? Thoning et al., 1989, JGR 94, 8549-8565; https://gml.noaa.gov/ccgg/mbl/crvfit/crvfit.html

Done as suggested, we added this reference.

Thoning, K. W., Tans, P. P., and Komhyr, W. D.: Atmospheric carbon dioxide at Mauna Loa observatory 2. Analysis of the NOAA/GMCC data, 1974–1985, J. Geophys. Res.-Atmos., 94, 8549–8565, https://doi.org/10.1029/JD094iD06p08549, 1989.

Line 295: Is it meaningful to give an annual average when 1-2 months are missing data?

Done as suggested, we added the calculated uncertainty and revised it as "(annual mean of 2013.4±(3) ppb, where the uncertainty is calculated when assuming the missing data in September and October varied between August and November)," on lines 321-322.

Line 295: replace "variations" with "trends"

Done as suggested, we replaced "variations" with "trends".

Line 296: What are the "similar atmospheric transport processes?" Summarize seasonal wind direction and speed patterns.

Considering the wind speed at different height should change largely, here the "similar atmospheric transport processes" mainly means the wind directions and synoptic process (i.e. monsoon). We have added more clarification with "such as synoptic process (i.e. monsoon) and seasonal changing wind direction as summarized above." On lines 324-325.

Line 309: replace "YON" with "TAP"

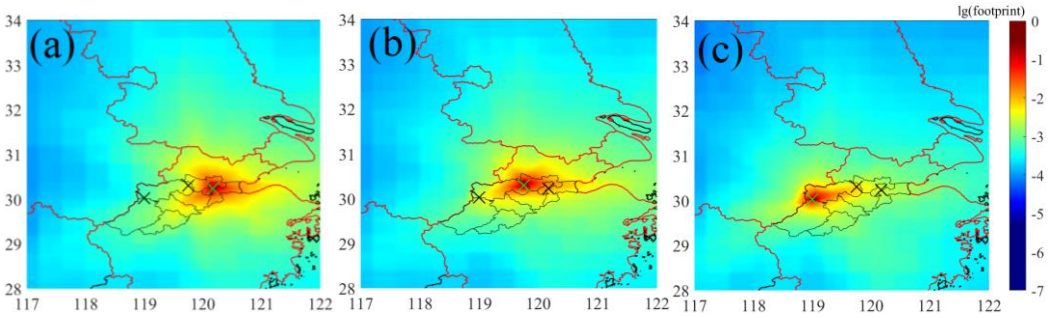
Done as suggested and thanks so much for catching up this typo.

Line 310: replace "temporal" with "spatial"

Done as suggested.

Lines 320-321: Figure 3 does not show significant differences in the size of the footprints at the different sites. You might want to expand the scale to show this.

Done as suggested, we have expanded the scale as displayed below.



Lines 323-326: Cities shown significant diurnal variation in PBLH.

Done as suggested, we revised this sentence as "and it will show significant diurnal variation in PBLH, especially have higher nighttime PBLH"

Line 345: Not sure what you mean by "amplitudes" here – amplitude of the seasonal variations? I don't see obvious differences. The absolute average abundances are different.

Done as suggested, we changed "amplitudes" with "relative variations".

Line 348: The simulated data for Linan actually approximate the observations very well!

Done as suggested, we changed this sentence as "We found the simulations at Linan site shows overall good agreement with observation, but still with slight overestimation from January to April and underestimation from May to September." on lines 383-384.

Line 364: It is very much to be expected that the Hangzhou site is more influenced by local emissions since it is in the urban core. What are the major emitters within 5-10 km of the site?

It seems the major emissions within 5-10 km of Hangzhou site are waste treatments. We revised this sentence as "was more influenced by local emissions (mainly for waste treatment and will be discussed later)" on lines 400-401.

Lines 366-368: How did you show that the Linan and Damingshan sites are influenced by emissions from a much larger region? The footprints don't indicate this.

The simulated CH_4 enhancement contributions from Hangzhou city was the highest (69.3%) than Lian (34.0%) and Damingshan (13.0%), indicating the rest contributions are from much further regions.

We revised this sentence as "The relative contributions from different regions also imply that the observations at Linan and Damingshan sites can present CH_4 emissions of much larger region as Zhejiang province or YRD area than Hangzhou city (Figure 4e)." on lines 402-404 for more clarification.

Lines 375-378: Can you give a reference for the statement that waste treatment emissions are larger during the daytime than at night?

Here we mean the waste treatment CH_4 emissions are sensitive to temperature, where the large diurnal variations and seasonal variations of temperature can lead to large variations of its emissions. We double checked the references and have not found the direct diurnal observations as eddy covariance. Hence, we deleted "are larger during the daytime than at night" and revised this sentence as "which should be highly sensitive to temperature and indicates obvious diurnal and seasonal patterns (Mønster et al., 2019; Kumar et al., 2022)." on lines 412-414.

Line 420: Emissions from waste treatment dominated the total CH_4 and the seasonal pattern, but you do show significant seasonal variations for the other anthropogenic sources in Figure 7a. Can you split those up at all? Can you say anything about the natural gas distribution infrastructure – more leaks in winter than summer, …?

The monthly variations of *a priori* EDGAR emissions was mainly driven by RCO (Energy for buildings), which changed from the highest 22% in winter to lowest ~8% in summer. Such information indicates the *a priori* inventory assigned more leaks from natural gas distribution infrastructure in winter than in summer.

To make clarification, we added "The seasonality in *a priori* EDGAR inventory was mainly dominated by RCO (Energy for buildings), with proportions to total anthropogenic emissions changed from the highest 22% in winter to lowest ~8% in summer. Such information indicates the *a priori* inventory assigned more leaks from natural gas distribution infrastructure in winter than in summer." on lines 459-462.

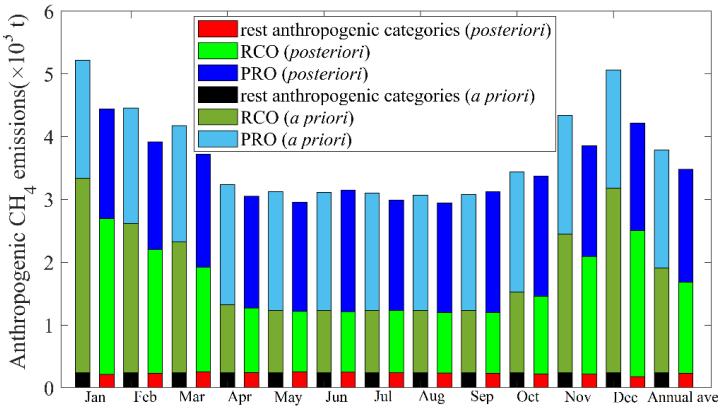


Figure S7. Comparisons of anthropogenic CH_4 emissions between *a priori* and *posteriori* results, PRO: fuel exploitation, RCO: energy for building, the rest anthropogenic emissions: excluding waste treatment, PRO, RCO and agricultural soil.

we also added one more figure (Figure S7) in supplementary file, and added more discussions as "Besides, the annual mean *posteriori* SFs varied between 0.87 and 0.94 for rest total anthropogenic categories (excluding agricultural soil), and were 0.97 for PRO (fuel exploitation) and 0.91 for RCO (energy for building), respectively; the annual mean *posteriori* SFs and were 1.05 and 1.05 for wetland (including agricultural soil and natural wetland). These *posteriori* SFs for the rest anthropogenic categories and wetland indicated much smaller bias than waste treatment. The monthly *posteriori* SFs for PRO and RCO also illustrated obvious seasonal variations, but were still smaller than the *a priori* seasonality in inventory (Figure S7)." on lines 441-447.

Line 479: Where are these values of SFs shown? They are not from Table S2.

We have mentioned on line 470 as "The derived monthly *posteriori* SFs for each emission source were displayed in Table 1 for Hangzhou city.".

Figures: In general, please improve the resolution of the figures. It is very difficult to impossible to read the small text, even when expanding the figures on the screen.

Done as suggested, the main reason why the resolution of all figures seems low is caused when MS in world version transformed in pdf version, and we have updated the new pdf version with high resolutions.

Figure 1: What are the divisions within Hangzhou City?

The divisions in Hangzhou city are different districts, we have added it in the caption of Figure 1.

Figure 2: Use the same color schemes on all figures and parts within figures for the same sites.

Done as suggested, we have used the same color schemes on all figures as displayed below.

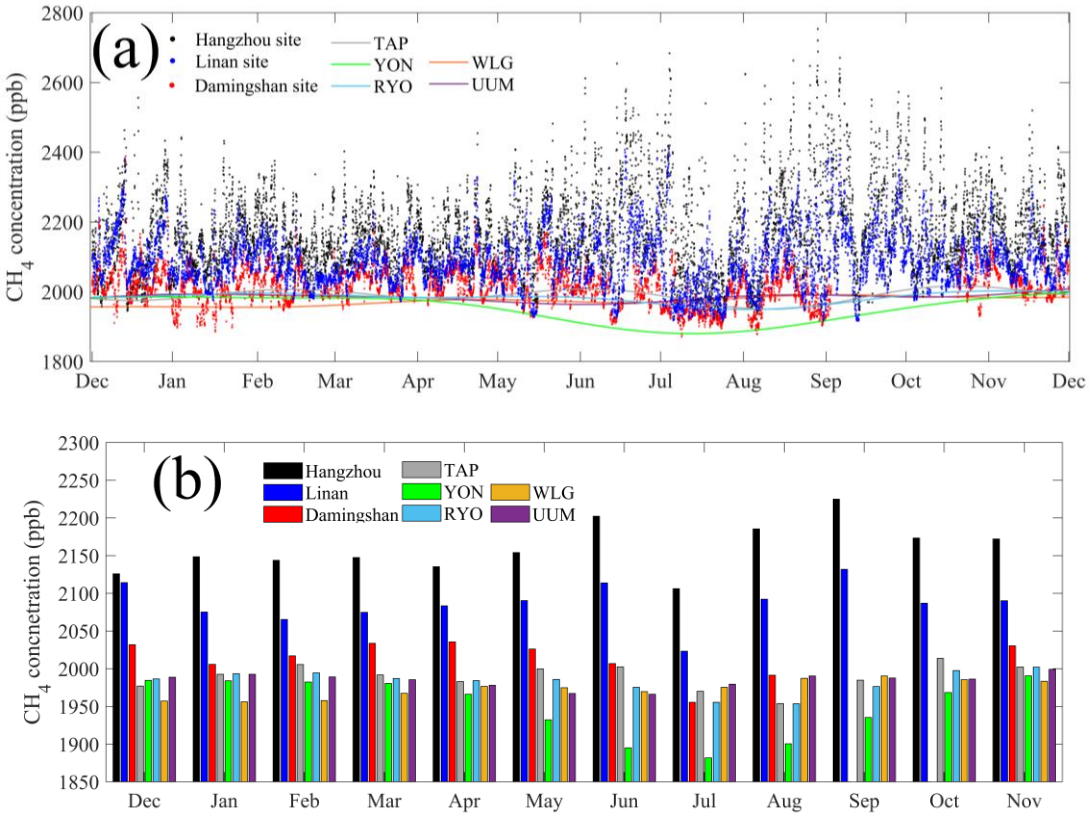


Figure 3: Replace "lg" with "log." Are the waste treatment CH4 emissions in panel (e) also from EDGAR v6.0?

Here both footprint and CH_4 emissions are expressed using log10, hence we added lg in these figures. The waste treatment CH_4 emissions in panel (e) is from EDGAR v6.0, we have added the data sources on the caption of Figure 3e on line 977.

Figure 4: Need higher resolution graphics, especially for panel (d) and (e). The note at the end of the caption may be incorrect. In panel (e), is it true that the blue color for the bar charts include all of Zhejiang, including Hangzhou? Do the blue regions in the pie charts represent Zhejiang minus Hangzhou?

Done as suggested, we increased the size and resolution of Figure 4.

Yes, in panel (e), the blue color for the bar charts include all of Zhejiang, including Hangzhou; and the blue regions in the pie charts represent the rest region of "Zhejiang minus Hangzhou". We have added more descriptions for clarification as "Note the blue color for the bar charts include all contributions from "Zhejiang", including "Hangzhou"; and the blue regions in the pie charts represent rest regions of "Zhejiang minus Hangzhou" on lines 1003-1005.

Figure 8: What region is this figure describing?

Thanks so much for pointing it out, this figure is for Hangzhou city, and we have added "for Hangzhou city" at the caption of Figure 8.