Response to review comments on MS# egusphere-2025-6197 by Wang et al.
(review query in black, response in blue)

Reviewer # 1

Comments:
1. Were all the flight measurement conducted on the same day? Sampling different atmospheres at different times of day for different cities in just one day could introduce variability due to changing atmospheric conditions. This may make it difficult to directly compare the measurements between cities and draw robust conclusions. Please clarify the timing of the flights and discuss how potential temporal differences were accounted for.

Yes, all the flight measurements were conducted on the same day. You are correct that this could confound the comparison between cities. The takeoff time from Beijing was 9:44 local time (Table 1), about 4 h after sunrise. The landing in Nanjing was 14:44 local time, when a fully convective boundary layer had developed. The returning flight took off from Nanjing at 18:31 local time and landed in Beijing at 22:17 local time; at these times, the stable stratification had already formed. Data during daytime and nighttime were analyzed separately to avoid direct cross-diurnal comparisons.

We noted that a high-pressure system prevailed over Eastern China during the observational day (Supplementary Figure S2). The variations in air trajectories between daytime and nighttime sampling were relatively small (Figure 9).

To further acknowledge the reviewer's point, we have added the following text to the Discussion (Section 3.1)

"We note that the profiles in Figure 5 were sampled at different times of the day. Some of the differences may have been caused by changing atmospheric conditions. The general inter-city patterns were unaffected because the temporal differences in the concentrations sampled at takeoff and at landing were smaller than the between-city differences."

2. Please define what is meant by "inversion jumps". It would be helpful to clarify how this inversion jump is identified in your observations and why it is significant. In addition, please expand on the importance of observing or measuring inversion jumps in the ABL and explain the implication of the inversion jumps observed in your results.

We now give a brief definition of inversion jumps as "differences in concentrations across the capping inversion at the top of the ABL", in the last paragraph of Introduction.

In Section 3.3, we described how these jump values were determined: "To determine the jump values, we calculated the average concentrations below 800 m for the midday observation and 500 m for the nighttime observation over Beijing, and below 1500 m for the mid-afternoon observation and 1000 m for the early evening observation over Nanjing, to represent the concentrations in ABL…"

In Section 4.4, we provide a detailed discussion on the importance of measuring the inversion jumps and on the jump values we observed.

The jump values suggest that the simple slab ABL approximation is not suitable for the urban ABL (third paragraph in Section 4.3). This point is further highlighted in Section 5: "Our results suggest that the simple one-dimensional slab model of the convective ABL may be inadequate for describing GHG budgets in the urban environments. An alternative is the advection-entrainment-diffusion model described by Lee (2023; Chapter 12)."

3. Line 182: Please expand more on how the calculations were made.

In response, we have added the following text

"First, we calculated the total emission for each city and the corresponding total area within the city administrative boundary. The emission flux (mg $m^{-2}s^{-1}$) was then calculated as the ratio of the total emission to the total area."

4. Line 196: Please clarify what is meant by "south trip" and "north trip." Does this refer to latitudinal flight direction (north-to-south vs. south-to-north) or to flights conducted over southern vs. northern cities?

We now note: "… south trip (from Beijing to Nanjing) … north trip (from Nanjing to Beijing)…"

5. Line 269, 271: The figures referenced appear to be incorrect. Please verify the correct figure citation. Additionally, please clarify what is meant by the "horizontal spikes."

The referenced figures should be Figure 6(a) and Figure 6(d). Thank you for pointing out this error.

We have added the following text to describe "horizontal spikes"

"In the profile plot in Figure 6a, the data collected in the horizontal flights were collapsed to the same altitudes, giving the appearance of small horizontal spikes (with a spike length of about 3 ppm)."

6. Line 326: Please explain how the emission fluxes of the cities were calculated.

Please refer to response to Comment 3.

7. Figure 10: It would be helpful to include labels for the city names on the maps.

Done (This figure is now labeled as Figure 9).

Minor comments:

1. Line 110: The phrase “we wish” should be replaced with more appropriate wording.
We have changed to “we aim to”.

2. Line 188: Please check spacing for “W m^{-2}in Beijing”.
Corrected.

3. Line 192-193: Please check spacing for “m s^{-1}and”.
Corrected.

4. Lines 191-194: Please revise sentence structure in this section to improve for clarity and readability.
Improved.

5. Line 226, 232: Please revise wording for “street-level concentrations.” Are the authors referring to ground-based mobile measurements?
Yes. We have changed “street-level concentrations” to “ground-based concentrations” in the manuscript.

6. Line 256: Please revise sentence structure in this section to improve for clarity and readability.
Revised.

7. Line 317: “tips” seems to be a typo.
Corrected.

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Reviewer #2

1. This paper describes airborne CH_4 and CO_2 measurements made over four Chinese cities during a single flight day in May 2023. It is well written and easy to follow. This is a nice dataset and it could form the basis for an AMT measurement report, but there are several general points that need addressing first.

We sincerely thank you for your positive evaluation of our work and for your insightful comments, which have improved the quality of our manuscript. We have carefully revised the manuscript as suggested.

2. I think the interpretation of the data above the ABL needs rethinking. As is stated in L360, the concentrations here mainly reflect wider regional emissions and long-range transport. But in plenty of other places this data is used to make statements about local emissions from the cities underneath the profiles. In particular for Hengshui there is no data in the ABL, so I don't see how any inferences can be drawn regarding local fluxes. The gradients in the free troposphere may include some contribution from local fluxes that have been transported out of the boundary layer, but they could also reflect longer-range transport. For Shangqiu the qualitative interpretation might be OK, as the flight does appear to dip into the top of the ABL briefly. But the best fit lines in Figure 8 are not representative of emissions form these cities. The current discussion surrounding this figure needs to be removed. The figure itself could either stay or go - if it stays then the discussion in section 4.2 seems far more convincing (although only in a qualitative sense - the best fit lines are not really useful here). Then I think the subsequent discussion in section 4 should only include Beijing and Nanjing.

To avoid potential confusion that might arise from original Figure 8 in the manuscript, this figure has been moved to the Supplementary Materials. The discussion in Section 4.2 has been revised to focus on Beijing and Nanjing, with original Figure 8 referenced only for a qualitative context.

3. When considering the data within the ABL for Beijing and Nanjing, until we get to section 4.2 there appears to be an explicit assumption that these are behaving as "dome" cities rather than "plume" cities (i.e. it is assumed that the storage flux dominates the advective flux). With wind data from the aircraft and/or ground observation sites and/or a weather model this assumption could easily be explored. Even if these are dome cities, it can still be difficult to make representative measurements using vertical profiles with limited horizontal extent within the city limits. Fig. 10 goes some way to addressing this, but really we need to see trajectories from the more enhanced ABL samples and the less enhanced ABL samples. Then we can see what the enhancement ratios represent (i.e. the difference between these two air histories). The wind data from the aircraft seems like a really important dataset here but there is virtually no discussion of it in the paper.

The reviewer has raised several good points. In response, we have added the following text in Section 4.2:

“In the idealized one-dimensional urban boundary layer, the CO_2 and CH_4 emitted by sources in the city are trapped in a dome above the city. Their vertical profiles and time changes are shaped by the surface emission, vertical mixing and exchanges with the free atmosphere. Horizontal advection does not play a role. In this study, the ABL deviates from this idealization because the vertical concentration profiles were also influenced by airmass advection.”

“In Nanjing, the backward trajectories in the ABL at landing and at takeoff were similar. The observed higher CO_2 concentration at the evening take-off (Fig. 5a) might have been caused by accumulation of locally-emitted CO_2 in the ABL. Interestingly, the CH_4 concentration near the surface experienced little change between these two times. It appears that at the evening takeoff, the CO_2 concentration was influenced heavily by vehicle emissions near the airport, whereas the CH_4 concentration was influenced by more distance sources such as landfills. This is a reason for why the ABL data for Nanjing in Fig. 7 exhibits two distinct clusters for the takeoff and landing.”

Since we only had measurement in one day, it would be difficult to perform accurate calculation of enhancement ratios according to trajectory history. In Section 4.2, we did point out the importance of the trajectory influence on the observed profile:

“In Beijing, the daytime air mass trajectory passed through high-emission areas located to the southwest of the city, whereas at night, few high-emission areas were present along the trajectory. This pattern was consistent with the higher CO_2 and CH_4 concentrations at the height of around 300 m in the daytime than at night (Fig. 5 a & b)”

4. For the comparison against EDGAR - I am not convinced that using the profiles later in the day negates the impact of the biosphere on CO_2. During the day there will be CO_2 uptake, which will switch to net positive emission at some point in the evening due to biospheric respiration. If these cancel out to produce net-zero biospheric impact on the aircraft profiles, it would only be by pure chance. Some sort of biospheric modelling would be needed to assess the likely impact here. That may be beyond the scope of this measurement report, but at the very least there needs to be a strong caveat placed on the EDGAR comparison stating that the impact of the biosphere is unknown and potentially significant.

We fully agree with the reviewer that the influence of the biosphere on CO_2 concentrations cannot be completely ruled out. In response to this point and Comment 5 below, we have rewritten Section 4.3:

“…Results showed that the inventory ratio was within the measurement uncertainty in Beijing and was higher by about 80% in Nanjing. Several factors may have contributed to the mismatch in Nanjing. First, vehicle and waste management are the two major CH_4 emission sources in Nanjing. In the last decade, vehicle fleets in Chinese cities have experienced two shifts in fuel use, first from gasoline to natural gas and then from gasoline and natural gas to electric (Hao et al., 2016; China's Energy Transition, 2024). These transitions were not accounted for by the EDGAR inventory. Second, using an inversion modeling with CH_4 concentration data, Hu et al. (2023) estimated that EDGAR CH_4 emission in Hangzhou is biased high by about

50% primarily due to a high bias for the waste management sector. It is possible that this source category was also biased high for Nanjing because Nanjing and Hangzhou, both located in the YRD, use similar methods to manage waste. Third, residents in Nanjing use natural gas for space heating. The inventory data are annual mean values that include contributions from space heating, whereas the aircraft observation was made in late spring when space heating was no longer needed. Finally, this study used the nighttime concentration ratio for comparison, an approach that can reduce, but not eliminate, the influence of the biosphere on CO_2. Addition of a typical respiration CO_2 flux of 0.1 mg CO_2 m^{-2} s^{-1} to the EDGAR CO_2 emission would reduce the inventory emissions ratio in Nanjing to 5.46 ppb ppm^{-1}. The relative roles of these factors cannot be inferred from one profile sampling, and a firm conclusion will require more observational data, a better estimate of biospheric respiration, and allocation of the annual inventory total to seasons and daytime and nighttime."

5. It would also be good to dig further into where thinks the CH_4 emissions are coming from. Is it really dominated by the mobile (vehicle) sector in the inventory? Perhaps it is for Chinese cities, I've never investigated, but I would have guessed most of the urban CH_4 emissions in EDGAR would be associated with heating and landfills. The fact that we are looking at data from a single flight in May also needs to be better highlighted, especially if heating emissions could be a significant source. Many emission sources are known to exhibit variability on diurnal, weekly and seasonal timescales so it is not really possible to draw inferences about EDGAR based on a single day of flying (even ignoring the issue with the biosphere).

Please refer to our response to Comment 4.

6. Another broad issue is the general lack of uncertainty values associated with the quantitative results. For example, for the jump values the standard deviations within and above the ABL could be combined to give some measure of uncertainty due to measurement variability. Considering these might then change some elements of the discussion, because in some cases the difference between values being compared is much smaller than the variability associated with them. Putting uncertainties on the enhancement ratios when comparing to EDGAR would surely result in agreement between the inventory and aircraft data for Beijing within uncertainties. The uncertainties may even be so large that there is no statistically significant difference for Nanjing either.

We have added uncertainty estimates to Tables 1 and 2. The inventory data are indeed within the measurement uncertainty for Beijing, but the mismatch is greater than the uncertainty range for Nanjing.

"Results showed that the inventory ratio was within the measurement uncertainty in Beijing and was higher by about 80% in Nanjing. Several factors may have contributed to the mismatch in Nanjing…" (Section 4.3)

Specific points:

7. I am no expert on boundary layer dynamics, but I think the first paragraph in the introduction is quite a simplification. Maybe this is OK, because this is not the focus of the paper, but I suggest that at least the language should be adapted here. For example, L33 could be "These two scalars are typically conserved if the ABL...", and

L35 should make it clear that CO_2 and CH_4 are conserved in the absence of emissions (which is obvious from the discussion in the next paragraph, but also needs stating here).

These changes have been made.

When we speak about a scalar being conserved, we refer to the situation that the scalar does not have a source or sink in free air. In this regard, CO_2 and CH_4 are conserved in the absence of chemical reactions. This point has been clarified.

8. L60 - there have that observe a change in CH_4 mole fraction between the boundary layer and free troposphere within an urban plume, although in most cases this is not the focus of the study and so the "inversion jump" values are not explicitly stated in the text. See next comment...

L74 - this presumably should say "not aware". Maybe this partly answers my previous comment in explicitly stating this is considering only studies directly over urban land. Most other urban studies have been performed downwind of the urban area, sampling the urban plume. It might be worth making this even more explicit, also at L427.

This typo has been corrected. We have expanded the background here and in Section 4.4

"Although some studies have explored the concentration ratio at different altitudes downwind of a city and have used it to infer emission origins (Li et al., 2022; Shan et al., 2022), we are not aware of published research on the CH_4:CO_2 ratio in the ABL air column directly above urban land." (Introduction)

"Our study appears to be the first to report the CO_2 and CH_4 inversion jumps in the urban ABL. Unlike previous studies targeting concentrations in urban plumes or in the urban surface layer, this study directly observed the vertical structures of CO_2 and CH_4 above cities." (Section 4.4)

9. L118 and following lines - "millions" should be "million"

Corrected.

10. Fig. 1 - the left hand panel in particular is hard to read for colour blind people. Could you change the colour scheme and run it through a colour blind simulator please? Many of the other figures are not ideal either, but this is the worst.

We have made the change to this figure and Figure 5. Thank you for this suggestion.

11. L205 - it could imply this, or it could be another temporal change. With the data here it is impossible to tell - I think this caveat needs adding here.

We have noted this point here:

"… implying a vertical CH_4 gradient of -30 ppb km^{-1} in the upper troposphere in Eastern China, although some of the difference may have been caused by temporal variations." (Section 3.1)

12. L237 - this could also be due to horizontal gradients - i.e. higher concentrations (on average) along the roads than at the airport. This possibility should be discussed here.

This point is added. Thank you.

"Another reason might be that the CO_2 concentration measured on the routes around the airport (Figure 1) was higher due to stronger traffic influence than at the airport." (Section 3.2)

13. Fig. 7 - the fit to the NJ up profile needs more discussion. There are multiple plausible causes for the structure in the data here. Looking at the profiles, one possibility is that the stable stratification leads to the local plume becoming compressed in altitude relative to the down profile, with a more methane-heavy regional plume sitting on top of it. The enhancement ratio for the lower section looks by eye to be very similar to the NJ down plume, which would support this interpretation, but there are many other possibilities too.

Please refers to our response to Comment 3.

14. L345 - also differences in the advective flux could explain the difference in concentration. This should definitely be mentioned here.

We had added this possibility to the manuscript:

"… The fourth possible reason was advection. As an air mass moves, it continuously accumulates emissions from the surface along its path, leading to higher concentrations in downwind areas. More CO_2 might have been accumulated near the surface in Beijing than in Nanjing." (Section 4.1)

15. Fig. 10 - it would be helpful to label the cities that are discussed on the text on the map.

Done.

16. L469 - It would be helpful to expand here on what this study has told us about future study design.

In response to this comment, we have added a short paragraph at the end of the paper:

"Future airborne missions should consider a spiral descent or ascent flight pattern over the target city, but unlike the patterns shown in Fig. 6, it should be extended to the lower portion of the ABL. This measurement strategy would yield information on both the ABL vertical structure and horizontal gradients in the ABL. Instead of sampling multiple cities, repeated profiles over time at the same city would allow better evaluation of the ABL dynamics and entrainment influence. Furthermore, winter season is preferred to reduce the confounding effect of biological fluxes." (Section 5)