Dear Editor and reviewers:

We are submitting our revised manuscript, entitled "**Carbon emission reduction requires attention to the contribution of natural gas use: Combustion and leakage**" to *Atmospheric Chemistry and Physics*.

We thank the Associate Editor and reviewers for the detailed and helpful comments to improve the manuscript. Responses to the individual comments are provided below. Reviewer comments are in **bold**. Author responses are in blue plain text. Modifications to the manuscript (Tracked changes) are highlighted in red, similar issues are merged into one point, the numbering of the figure in this responses letter is the same as the manuscript or the supplementary.

The submitted manuscript has been revised based on reviewers' comments.

Sincerely,

Guiqian Tang,

Professor

Institute of Atmospheric Physics, Chinese Academy of Sciences

Beijing, China

Response to Reviewer #1's comments:

The manuscript presents the results of a 73 day long observational campaign of methane (CH_4) and carbon dioxide (CO_2) fluxes made on a tall tower in Beijing. In addition, surface mobile measurements over different time periods were also made to address some specific geospatial regions within the domain covered by the flux tower. The methods to prepare and analyze the data are very standard. The findings include that the emissions of CH_4 are likely anthropogenic in nature due to their similarity in time and direction to those of CO_2. Comparisons were made with other previous campaigns in earlier years. They then draw some conclusions about the changes in CO_2 and CH_4 over time and relate these to various different policies. The authors clearly have demonstrated that their basic measurements of flux are reasonable and representative. There should be no doubt about this point, and hence the fundamental data underlying the project looks sound.

I recommend that the work undergo major revisions before it be considered further. However, due to the strong people on the team, I do believe that with a considerable amount of hard work and time, that they can raise the level of the paper to such that it will make a good ultimate contribution to ACP. I am happy to continue to work with any future revisions which are brought forward.

Response: We thank the reviewer very much for the positive comments on this work. We have revised the manuscript carefully according to your valuable feedbacks, please see the replied below, the similar issues are merged into one point. We hope that the revised version can address your concerns.

Point 1: One such issue about the data is that the individual half-hour averaged flux time series over the entire time studied is not available anywhere. However, the details in the figures of the entire-campaign averaged hour-by-hour data clearly demonstrates that the hour-to-hour and day-to-day variability are both important. They also demonstrate that there are issues likely occuring at the half-hour scale, but they cannot be analyzed or discussed based on the current figures and data provided. Therefore, analyzing the data or evaluating analysis done cannot be validates, and the potential strong impacts of these 30-minute scale variations cannot be analyzed or presented. This weakens the paper.

Lines 205, 206, and 213: I do not agree with your statement based on the data presented in Figure 1. It looks like CH_4 emissions start to rise at 5am, rise with a different rate than CO_2 emissions, stay much flatter, and start to decrease at 5pm. Thus, there is a shift between these two which is not similar. However, the point is that if the entire time series were analyzed, we could be even clearer. You have 30-minute data, so please re-do this analysis more carefully and precisely.

Response 1: Thank you very much for the comment. We improved the analysis of diurnal CO_2 and CH_4 flux variations by utilizing data at a 30-minute resolution instead of hourly data, enhancing the accuracy of our results. and there are indeed some differences between the two. The corresponding wording has also been modified in the corresponding section as follows:

The CH_4 flux began to increase gradually from 04:00 to around 08:30, and then remained stable until after 10:30, when it began to rise rapidly again, reaching its daily

peak of approximately 157.1 nmol m^{-2} s^{-1} around 11:30. After 17:30, it slowly declined. Its diurnal variation pattern showed some differences compared to CO_2 flux, which rose from 03:30 to around 08:30 similar to CH_4 flux. However, the peak for CO_2 flux occurred around 13:30, then slowly decreased, after 18:30, it decreased rapidly.

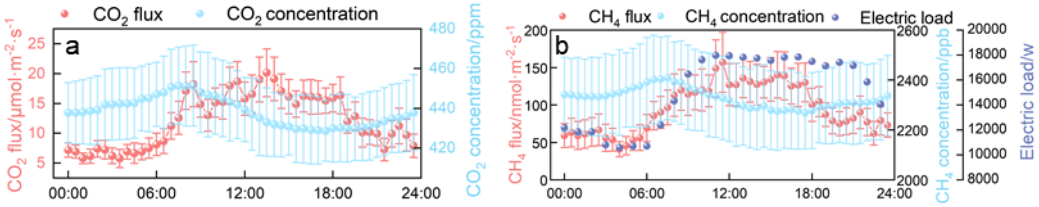


Figure 1 Daily variations in the CO_2 and CH_4 concentrations, fluxes, and electricity loads

Point 2: The use of background subtraction may lead to substantial errors. First, there are the issues of the observational uncertainty in the background value. Second, there are more modern papers demonstrating that background subtraction is not needed. Third, in those cases in which long-range transport is present, background subtraction is flawed in connection with the flux tower computational assumptions. This is because the equations underlying the flux calculation assume that the upper air is clean and that the emissions come from the local surface. Recent papers have demonstrated that there is in fact long-range transport into Beijing from upwind industrial sources in central China, and therefore any such events would need to be excluded from the data before analysis is performed. I raise this point since in analysis done both by my group as well as others, the time period studied in this work contains at least one such long-range transport event. Analysis of the 30-minute time series may help identify this event, and possibly others as well. In addition, this paper introduces the use of a 5-minute window to identify background values. However, given the size of the domain, this is not consistent. The observed wind speed will take more than 5 minutes to go from the edge of the domain to the tower location, and hence the length of the averaging period must be at least this long. This will change from day-to-day and hour-to-hour. The time likely needs to be longer, to account for any atmospheric recycling occurring within the domain.

Lines 264-265: This point is raised earlier. Since it takes more than 5 minutes for the wind to flow from the edges of the footprint to the flux tower observation point, why do you use a 5-minute window? Furthermore, why do you choose the 5th percentile? What happens if you choose the minimum value? Or the 1st percentile? Or the 10th percentile? We know what the large-scale average CH_4 measurements are from the long-term base station in Shangdiaizi, why not use their value? You need to carefully consider the error introduced by such a sweeping set of assumptions. Please quantify how a change in the calculation of the background changes the results? Please quantify how the observational uncertainty could lead to the value of the 5th percentile background value to change? How would this uncertainty propagate into the calculation of the enhancement, when it is applied at both the lower and upper ends simultaneously?

Of course, there are newer techniques such as published in ACP in 2025 this year

based on a study of CH_4 in central China which completely does away with background subtraction and enhancement calculation. You could consider this new approach as well and completely avoid the issues of enhancement and background subtraction. Or you can work hard to justify why your background subtraction is valid and how it contributes to overall uncertainties in the conclusions.

Response 2: Thank you very much for the comment. Considering the influence of meteorological conditions and regional transport, background concentrations are constantly changing. Therefore, using the minimum value is unreasonable. As the reviewer noted, using methane concentrations from the Shangdianzi station, which is very close to Beijing, as the background value is indeed a good approach. Unfortunately, data from the Shangdianzi station after 2022 is currently unavailable for download. In previous studies, the time windows used for background subtraction were primarily 5 minutes or 10 minutes, and the quantiles employed were either the 5^{th} or the 10^{th} (Pu et al., 2023, Well et al., 2018; Well et al., 2019), However, few studies have evaluated the impact of using different combination of time window and quantile on background value calculation. Yet, the choice of both the time window length and the quantile does indeed affect the final calculated background concentration. Here, using mobile measurement results near the gas storage tank in summer as an example, we evaluated the impact of different window-quantile combinations on background value calculation. This part has been added to supplementary, it can be seen in the following figure that the calculated background concentration varies only slightly when using different combinations, with a maximum difference of only about 12 ppb (after 14:45). This value is very small relative to the total methane enhancement (observed value minus background value). Nevertheless, we still evaluated the confidence level of the background concentrations calculated using different metric combinations. This method references the approach of Schiferl et al (2025). The specific procedure is: for each data point, based on the 5-min or 10-min time window, we take all CH_4 observation data within a rolling window extending 2.5(5) minutes forward and 2.5(5) minutes backward from its timestamp (totaling 5(10) minutes). Then, we perform 1000 bootstrap resamples (sampling with replacement) from these data. For each resampled dataset, we calculate a background value (either the 5^{th} or 10^{th} quantile). This yields 1000 background value estimates, forming a distribution. Finally, we determine the middle 95 % range of this distribution (the 2.5^{th} to the 97.5^{th} percentile), which represents the 95 % confidence interval (CI) for the background value at that point. Based on our calculations, the mean widths of the confidence intervals for the combinations: 5-min window/5^{th} quantile, 5-min window/10^{th} quantile, 10-min window/5^{th} quantile, and 10-min window/10^{th} quantile were 32.1 ppb, 26.9 ppb, 17.2 ppb, and 29.6 ppb, respectively. A smaller confidence interval width indicates lower variability and better reliability. Therefore, we selected the 10-min window with the 5^{th} quantile as the background value. As for the observational uncertainty, we consulted the official website of the instrument and found that its accuracy is 0.2 ppb at a10 Hz frequency, thereby having a limited impact on the calculation results of the 5^{th} percentile. We recalculated the background value and CH_4 concentration enhancement value based on the fifth percentile of the 10-minute window, the calculation results differ from the previous manuscript. We also found leakage from the petrochemical plant, and the maximum value of natural leakage was captured near the gas storage tank. Then, we used Weller's method to calculate the methane leakage rate, and the confidence interval

based on the Bootstrap method was used to estimate the uncertainty of the leakage rate. The natural gas leakage rate from the gas storage tank and power plant in winter were 7.4 ± 0.1 g/min and 0.6 ± 0.03 g/min, respectively, and the natural gas leakage rate from the gas storage tank and power plant in summer were 1.2 ± 0.04 g/min and 2.1 ± 0.07 g/min, respectively. The natural gas leakage rate near the petrochemical plant was 0.6 ± 0.04 g/min.

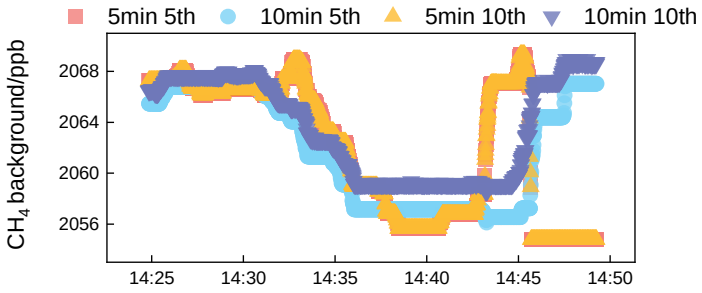


Figure.S10 The time series of the calculated background value with different methods

Point 3: Line 101: Why do you use a 219ppb standard for CH_4? Would the results change if a more reasonable 1900ppb or 2000ppb standard were used?

Response 3: Thank you very much for the comment. We apologize for the misunderstanding caused by our writing error. The standard gas concentration is actually 2190 ppb. We have modified it in the revised manuscript.

Point 4: Lines 116-118: There are many studies which apply mean and standard deviation or even more complex analysis such as EOF, SVD, etc. You need to describe in more detail things such as: how many standard deviations are used, is the data normally distributed, lognormally distributed, etc. Even using the more advanced techniques you need to demonstrate the variance explained or reduced. The current work is incomplete.

Response 4: Thank you very much for the comment. The methods by Vickers and Mahrt.,(1997) are widely applied in spikes removals during flux calculation. Considering the concentration are usually not normally distributed and avoid removing reasonable data mistakenly, a suitable method was used as follows and has been added to the Section 2.2:

Take a moving window with a width equal to 1/6 of the averaging period (typically 5 minutes). Calculate the mean and standard deviation of the time series within the window. Define outliers as any data points deviating from the mean by n times the standard deviation (initial n = 3.5). Replace identified outliers with linearly interpolated values from adjacent points. Consecutive outliers ≤ 3 are treated as a single outlier; consecutive outliers ≥ 4 are considered local trends and excluded from outlier classification. Iteratively increase n by 0.1 per cycle until no outliers are detected or 20 iterations are reached. Advance the window by half its width (step size) and repeat outlier detection/removal for the next window. Continue this process until all outliers are processed within the averaging period. If outliers exceed 1 % of total data points in any averaging period, discard that entire period.

Point 5: Lines 134-136: This raises many issues. I will outline three of them. First, can you please show us a map of the footprint area, I cannot seem to find it anywhere. Second, it takes wind more than 5 minutes to cross the urban area of Beijing, so why do you use a 5-minute averaging time period to compute background CH_4 in this case? Given the very large number of sources just upwind from your area (including oil production in nearby Hebei and Shandong), how do you work to exclude long-range transport from outside of your footprint area?

Response 5: Thank you very much for the comment. The source area map is shown as follows and has been added into the supplementary. It can be seen that the source area covers the most urban area of Beijing. It basically covers the entire Fifth Ring Road area of Beijing but does not extend to other provinces, thus excluding long-range transport from other provinces. The 5-minute time window applied in background calculation is referred as Weller (2018, 2019) and Tettenborn (2025), but there are also some uncertainties indeed, we explained it as answered in Point 2.

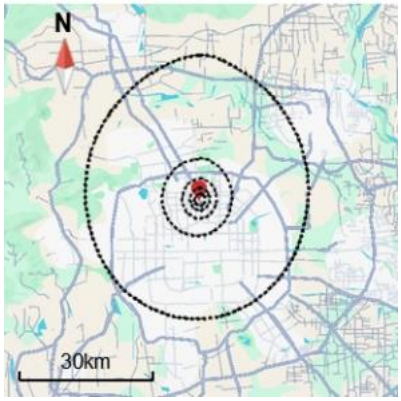


Figure. S2 The range of source area (The dashed lines represent the 10%, 30%, 50%, 70%, and 90% contribution source areas from inside to outside, respectively, the map is from Google earth: https://earth.google.com/)

Point 6: Given that the size distribution of particulate matter in Beijing (as published by others in your same institute, aerosols have a very large number in the sub-micron range) how do you filter the particles without altering the air flow? Or do you not filter these 100nm sized particles? If not filtered, how would they impact the observations?

Response 6: Thank you very much for the comment. In our experiment, a 0.45 μm sized polytetrafluoroethylene filter was employed, the use of this type of filter membrane will have a certain impact on the flow rate, so the 2 L/min mentioned in the article is the measured flow rate value after the filter membrane was installed, the setting of this value has taken many factors into account. Since the pore size of the polytetrafluoroethylene is significantly larger than 0.1 μm, particles with size smaller than 0.1 μm cannot be completely filtered. However, they do not affect the observational results because the mid infrared wavelength range is 2.5-25 microns, Consequently, 100nm sized particles do not interfere with the absorption of mid-infrared light by the target gases.

Point 7: Lines 179-182 are very hard to follow. Do you mean you are comparing a sort of summer-average from this work with summer-averaged from previous

works which also used tall towers, but towers with different heights? Even if my understanding is correct, you need to re-word this sentence.

Response 7: Thank you very much for the comment. Your understanding is correct. We apologize for the non-standard language and we have rewritten this sentence as following, also, a map is added to explain this:

The diurnal CO_2 flux ranged from 6.05 to 19.66 μmol·m^{-2}·s^{-1} with an average of 12.21 ± 1.75 μmol·m^{-2}·s^{-1}(Figure 1a), which was generally lower than the summer observations by Cheng et al., (2018) and Liu et al., (2012) at 200 m and 140 m in this tower, respectively (Table.S1).

Table.S1 Comparison with the CO_2 flux in this study with previous studies based on this tower at different heights

Period	Height(m)	Results(μmol m^{-2} s^{-1})	Reference
200-2009	140	16.2 ±4.1(8-20)	Liu et al(2012)
2013-2016	200	14.5(5-30)	Cheng et al(2018)
2009-2017	140	21.4(7.5-30.32)	Liu et al(2020)
2022	220	12.2 ±1.8(6.1-19.7)	This study

Point 8: Significant digits. Can you really trust the wind and concentration measurements as well as the analysis technique to 4 digits of precision?

Response 8: Thank you very much for the comment. According to the manuals of the 3D sonic anemometer and high-frequency instruments, the measurement precision can reach to 0.01m/s for 3D sonic anemometer(https://www.baidu.com/link?url=Pn8oHkR_lCj5A-f0PNx19CTO5PVGL9T3iq3ebvmnYX4Bsby842-gEFCKVFbPTgOYad0-P4lY3sV2GLN0JFErJvSPpI3U_W-I091Okt4q1Q25u3HYg0ZS6sboKM2S687r&wd=&eqid=f46c92c500049ff400000003682c3ef9), and around 0.75 ppm for CO_2 and 1200 ppt for CH_4 at a sampling frequency of 10 Hz (https://www.aozuo.com.cn/productinfo/1875506.html), Therefore, the flux(unit μmol·m^{-2}·s^{-1} for CO_2 and nmol·m^{-2}·s^{-1} for CH_4) calculated as their covariance can achieve 4 significant digits of precision.

Point 9: Lines 191-192: This is a clear mistake. The paper was submitted in 2024. How can you not know if the 30ug/m^3 by 2022 was achieved or not. Also, why is this written in future tense?

Response 9: Thank you very much for the comment. We apologize for our carelessness and have modified corresponding part as follows:

Beijing has increased the amount of electricity flow from other provinces in recent years (Figure S6), which has further driven a decrease in the annual average concentration of $PM_{2.5}$, dropping to 30.5 μg·m^{-3} by 2024.

Point 10: Lines 215-216: How do you explain negative CH_4 fluxes? Why are both negative and positive fluxes considered in tandem with each other? Isn't one a source and the other a sink? A maximum R of 0.82 means that it accounts for 67% of the variability, which has some amount of correlation power. However, it is not as strong as the authors make it out to be. Introducing more advanced comparisons between the emissions of CH_4 and CO_2, concentrations of CH_4 and CO_2, Temperature, incoming surface solar radiation, and other variables in

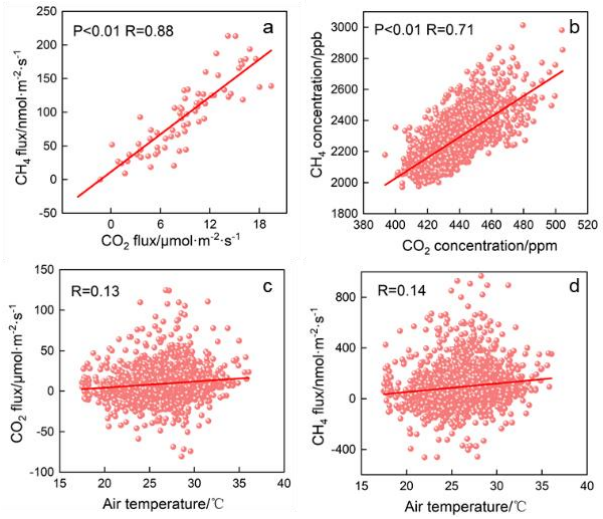
tandem will make the analysis stronger. Using EOF or even SVD to analyze the time series in tandem will also make the analysis stronger. This result looks like a reasonable start, but an insufficient analysis to support the remainder of the paper.

Response 10: Thank you very much for the comment. There are two potential causes for the observed CH_4 negative values in this study. First, the existence of a CH_4 sink within the urban environment. Based on literature reports, soil uptake is currently the only known CH_4 sink (Lee et al., 2023), which could explain a portion of the negative CH_4 fluxes. Second, the explanation lies in errors associated with the turbulent flux measurement system. This uncertainty is difficult to quantify because the sources of error are diverse, such as signal loss due to frequency attenuation in closed-path systems, the occurrence of negative values when real fluxes approach zero caused by the instrument's low signal-to-noise ratio, and the failure of the steady-state assumption underlying the eddy covariance method under conditions of weak turbulence.

Unfortunately, no study can fully quantify the causes of negative values in flux observations currently, particularly over highly heterogeneous urban surfaces, where quantifying these uncertainties becomes especially challenging. Due to weaker turbulence development at night, flux measurement uncertainty increases, and the probability of observing negative fluxes is higher. Fluxes frequently fluctuate around zero during these periods. Therefore, considering both positive and negative flux values simultaneously helps mitigate these uncertainties. This is why most studies analyzing fluxes effectively consider the net flux (the combination of positive and negative values)."

We also aimed to conduct a more in-depth analysis using methods such as Empirical Orthogonal Function (EOF) analysis or Singular Value Decomposition (SVD), as suggested by the reviewer. However, based on our attempt, EOF or SVD analysis is primarily more applicable to multi-site time series data. Given that our study relies on single-site data, applying EOF or SVD could not yield meaningful outcomes in this context.

Although the concentrations and fluxes did not perfectly align, a linear fitting was performed on the concentrations of CO_2 and CH_4 in the figure below, revealing a certain correlation between them ($R=0.71$). This indicates that both are influenced by the same emission sources, a fitting of their net fluxes at the daily scale showed a significant correlation with a relatively high correlation coefficient ($R=0.88$), which also supports this view. Also, the linear fitting between the air temperature with CO_2 flux and CH_4 flux was conducted, there is no correlation among them, implying a ignorable emission by biogenic sources like waste disposal or soil emission driven by temperature.



Point 11: Lines 246-247: I cannot access the document.

Response 11: Thank you very much for the comment. We check the webpage and find that it has been updated and the original webpage is invalid. The cur -rent webpage is as follows: https://www.gov.cn/xinwen/2019-12/30/5465088/files /e3682ce168c8427b886a43a790d66c2c.pdf

Point 12: Lines 247-249: There is a distance that the wind must travel from the source to the observation point, and what time lag would this produce between the production time and the observed time? Please demonstrate using the 30-minute flux time data and the day-by-day electricity data.

Response 12: Thank you very much for the comment. The data we obtained o n the webpage is hourly daily variation of electric load (https://www.gov.cn/xin wen/2019-12/30/5465088/files/e3682ce168c8427b886a43a790d66c2c.pdf). Unfortun ately, we are unable to acquire higher-resolution continuous time series of pow er production data. Though it is a good try to quantify the time lag between t he electricity production time and observed time, we are sorry we are not able to do this currently owing to limited data.

Point 13: Lines 255-257: This is not logical. There was a paper published in Communications Earth and Environment in 2025 which showed that CO emissions from central China are much higher in November-January due to increases in production to meet the end of the year production cycle, as well as possibly due to more small and remote emissions (possibly heating or small business energy needs in winter). This CO would then be transported to Beijing in part and chemically decay into CO_2 as it is being transported. You need to consider these findings before you make such statements.

Response 13: Thank you very much for the comment. As answered in Point 3, the source area only covers the most urban area of Beijing, but does not extend to other provinces, thus excluding long-range transport from other provinces.

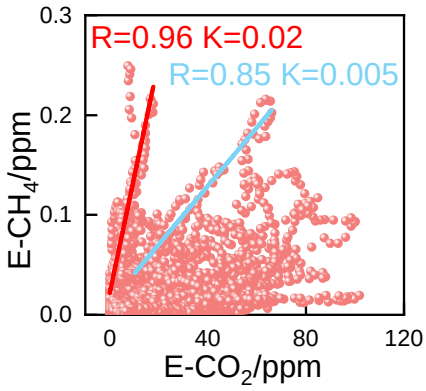
Another a essential point is that CO has a long lifespan in the atmosphere, and it takes several tens of days to decay into CO_2 (Drummond et al., 2009; Weinstock et al., 1969), so the impact of long-distance transmission of CO is relatively small.

In summer, the sink of CO_2 is the largest due to photosynthesis, and the consumption

of natural gas is at a relatively low level. Nevertheless, there is a good correlation between CO_2 and CH_4. With the arrival of the winter heating season, the consumption of natural gas will greatly increase, resulting in better homology between CO_2 and CH_4.

Point 14: Lines 281-283: This is an interesting finding. However, the scaling of the plots and the lack of data make it impossible to validate. I am happy to support the authors to improve upon this, but at the present time have insufficient data or readability to do so.

Response 14: Thank you very much for the comment. After changing the method of calculating background values, we also discovered natural gas leakage near the petrochemical plant, as shown in the following figure. The line with a slope of 0.02 was related to the gas storage equipment, and the line with a slope of 0.005 was relevant to the natural gas combustion equipment.



Point 15: Figure 5 is scaled differently on each plot. It makes comparison of the already very small data points nearly impossible.

Response 15: Thank you very much for the comment. We unify the scale on each plot as follows, it can be seen the most distinct leakage of natural gas near the gas storage tank in winter and power plant in summer, with the CH_4 enhancement of 1759 ppb and 630 ppb, respectively. Also, strong CH_4 concentration enhancemant (1375 ppb) near the waste disposal stations was captured, suggesting the non-negligible CH_4 emission from the waste disposal processes.

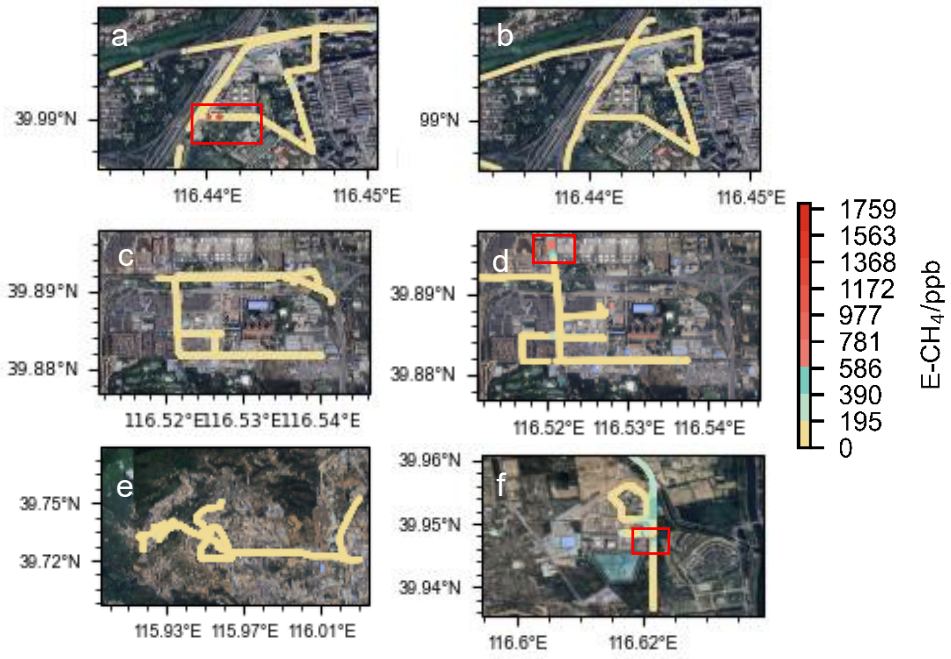


Figure 5 CH_4 enhancement concentration distribution map based on vehicle observations (a, c show storage tanks and thermal power plants in winter; b, d show storage tanks and thermal power plants in summer; e shows petrochemical plants; f shows waste disposal stations; and the red box represents high leakage value)

Point 16: Lines 21-22: Natural gas contains a very large amount of CH_4, but does not contain CO_2. You need to explain how these could be co-emitted? If you mean that more natural gas being used for combustion produces more CO_2, and that it also leads to more leaks, than this could be one such way to analyze this. But the way it is currently written raises questions about the rest of the paper since it is in the abstract.

Response 16: Thank you very much for the comment. We admit that the current writing style is indeed somewhat ambiguous so we added explanations after this sentence as bellows:

The combustion of which releases CO_2, while its leakage processes emit CH_4.

Point 17: Line 32: the second sentence requires a reference.

Response 17: Thank you very much for the comment. The reference has been added as following.

Seneviratne, S. I., Rogelj, J., Seferian, R., Wartenburger, R., Allen, M. R., Cain, M., Millar, R. J., Ebi, K. L., Ellis, N., Hoegh-Guldberg, O., Payne, A. J., Schleussner, C. F., Tschakert, P., and Warren, R. F.: The many possible climates from the Paris Agreement's aim of 1.5 degrees C warming, Nature, 558, 41-49, http://doi.org/10.1038/s41586-018-0181-4, 2018..

Point 18: Line 95: please do not use so many abbreviations.

Response 18: Thank you very much for the comment. The abbreviations have been deleted in the revised manuscript as follows:

Under the action of a vacuum pump, the air sample enters the instrument room at a flow rate of 2 lpm through a polytetrafluoroethylene sampling tube with a length of 3 m and an inner diameter of 3 mm.

Point 19: Again, in specific refer to line 135: "the flux source area covers most of the urban area of Beijing and reflects the average emission characteristics at the regional scale." Could the author add a figure to describe the space covered.

Response 19: Thank you very much for the comment. The source area has been added to Supplementary as follows: it can be seen that the flux source area covers most of the urban area of Beijing.

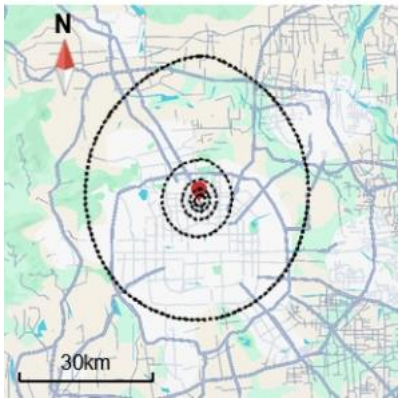


Figure. S2 The range of source area (The dashed lines represent the 10%, 30%, 50%, 70%, and 90% contribution source areas from inside to outside, respectively, the map is from Google earth: https://earth.google.com/)

Point 20: Again, in specific refer to line 110, the observation period of the article is between June 11 and September 7, 2022. Is the data from this time period really sufficient to reflect the characteristics of emissions of CH_4 and CO_2 over this region? If so, provide evidence to support this.

Response 20: Thank you very much for the comment. Due to the limitation of our observation period to summer months, we admit the measurement results cannot fully characterize the annual flux characteristics. Previous studies indicate that CO_2 and CH_4 fluxes typically exhibit seasonal variations. For instance, in Beijing, the summer net CO_2 flux is significantly smaller than winter values, as the CO_2 sink (plant photosynthesis) weakens markedly during winter while emissions increase with the heating season(Liu et al., 2012; Liu et al., 2020). Regarding CH_4 fluxes, there are currently no annual-scale measurements available for Beijing.

However, according to measurements from other developed cities(Gioli et al., 2012; Helfter et al., 2016), if natural gas consumption drives changes in regional CH_4 flux, the CH_4 flux in winter would increase due to higher gas usage during the heating season. In our observations, CH_4 flux is driven by natural gas consumption; therefore, we predict that CH_4 flux in Beijing will rise substantially during the winter heating season. Taking all the considerations above, the CO_2 and CH_4 fluxes in other seasons of Beijing would indeed differ from summer measurements, yet such differences would not alter the main arguments and conclusions of this study. A essential point is that the calculated leakage rate will not alter in different seasons, the reason is that the CH_4 flux was positive with the natural gas consumption, leading to their ratio (leakage ratio) to be seasonally constant. This part has been added to Section 4.2 as following:

If the CH_4 fluxes were solely attributable to pipeline leakage processes, CH_4 fluxes should remain relatively stable throughout the day without significant diurnal variations, given that the constant pressure in urban pipeline pressures. Yet in our observations, CH_4 fluxes exhibit pronounced diurnal patterns and their spatial distribution positively correlates with natural gas consumption. This indicates that CH_4 emissions in Beijing predominantly originate from consumption-oriented leakage processes. Consequently, as natural gas consumption surges during winter heating periods, CH_4 emissions from these processes (e.g., fugitive emissions from electrical devices) also increase. As a result, the ratio of emissions to consumption (leakage rate) remains relatively stable. Thus, the CH_4 leakage rate measured in summer demonstrates year-round representativeness.

Point 21: Again, in specific line 162: "Before the particulate matter entered the instrument, it was removed using a filter head." What are the components of this filter head? Can it filter without disturbing the airflow? I have seen a patent and a paper describing such a material in China, but it can only filter down to about 300nm particles. Would this possibly have an impact, especially since your claim is that combustion is the source of the CH_4 and the CO_2 and that there is a large amount of small BC particles also produced by such combustion? There was a paper in the Chinese-Language GuangXueXueBao journal specifically raising issue with this topic.

Response 21: Thank you very much for the comment. As ansered in Point 4, the components of this filter head is polytetrafluoroethylenea and the flow rate mentioned in the article is the measured flow rate value after the filter membrane was installed. Although 0.45 μm sized polytetrafluoroethylene filter exhibit low removal efficiency for sub-100 nm particles, it demonstrates significant retention efficiency (~90%) for particles in the 0.1-0.3 μm size range (Soo et al., 2016; Xu et al., 2018), this performance stems from the synergistic effect of physical sieving and deep-bed adsorption enabled by its asymmetric porous structure. Moreover, our low flow rate operation (2 L/min) further enhances the filtration efficiency for sub-0.3 μm particles. Crucially, any remaining sub-100 nm sized particles pose negligible interference to optical instruments using mid infrared technique (2.5-25 μm), thus introducing no substantial measurement influences.

Point 22: Lines 180-182: Please provide a map, which will make it much easier for the audience to follow.

Response 22: Thank you very much for the comment. The map has been added to the Supplementary as follows:

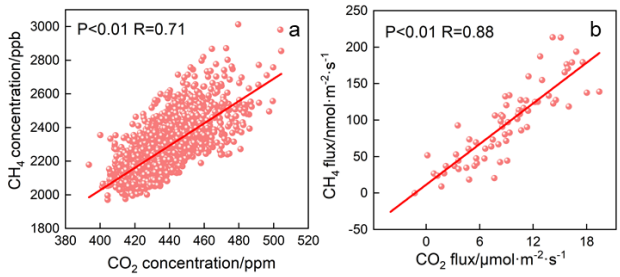
Period	Height(m)	Results(μmol m^{-2} s^{-1})	Reference
200-2009	140	16.2 ±4.1(8-20)	Liu et al(2012)
2013-2016	200	14.5(5-30)	Cheng et al(2018)
2009-2017	140	21.4(7.5-30.32)	Liu et al(2020)
2022	220	12.2 ±1.8(6.1-19.7)	This study

Point 23: Section 3.2: The results from Figure 6 do not demonstrate that the concentration enhancements are well correlated, especially given that positive and

negative enhancements are needed to be used in tandem for the analysis. How can someone conclude that the fluxes are linearly correlated? Especially so when the 30-minute data is not presented directly.

Response 23: Thank you very much for the comment. Firstly, concentrations and fluxes are fundamentally distinct. Concentrations are significantly influenced by meteorological dispersion conditions, whereas fluxes primarily reflect the source-sink characteristics of the target gas within the flux footprint area. Furthermore, mobile surveys and tall-tower observations capture different emission information. Mobile surveys predominantly detect instantaneous emission sources within a certain distance from the observation vehicle (e.g., traffic emissions). Clearly, when instruments sample plumes associated with traffic, the correlation between methane CH_4 and CO_2 concentrations weakens.

In contrast, observations at the 220-m height of the tall tower capture uniformly mixed signals from multiple emission sources, offering greater regional representativeness. A key piece of evidence is the significantly stronger correlation between CH_4 and CO_2 concentrations observed at the tower site compared to near-ground mobile surveys. This finding is consistent with the flux measurements results.



Point 24: Line 272: What are the units of E-CH_4 and E-CO_2? How are they computed?

Response 24: Thank you very much for the comment. The units of the E-CH_4 and E-CO_2 are ppb and they refer to the enhance concentration of CO_2 and CH_4, respectively. In the original manuscript, the values of E-CO_2 and E-CH_4 were obtained by subtracting the background value of this observation from the real-time observed value.

Point 25: Usually, discussion comes after the results are fully presented. However, line 234 is "DISCUSSION", while the later line 235 is "3.3 Driver of the homology between CO_2 and CH_4". Please carefully considering re-structuring or change around header titles.

Response 25: Thank you very much for the comment. We apologize for the mistake in our writing. The title of corresponding sections have been modified, the title of section Driver of the homology between CO_2 and CH_4 has been change to 4.1, the title of section Climatic effect of natural gas (NG) losses and the impact on carbon neutrality has been change to 4.2, the title of section Policy implications has been changed to 4.3.

Point 26: Why is there no summary or conclusion section?

Response 26: Thank you very much for the comment. The section Conclusion has been added to the revised manuscript as bellows:

This study utilized the eddy covariance method to measure CO_2 and CH_4 fluxes at 220-m height in urban Beijing, providing critical insights into surface-atmosphere

exchanges of greenhouse gases in the region. First, urban areas unequivocally act as net sources of both CO_2 and CH_4. The daily mean fluxes were 12.21±1.75 $\mu mol \cdot m^{-2} \cdot s^{-1}$ for CO_2 and 95.54±18.92 $nmol \cdot m^{-2} \cdot s^{-1}$ for CH_4, with daytime emissions significantly exceeding nighttime levels, highlighting the importance of anthropogenic influences.

Although diurnal variation patterns differed slightly between CO_2 and CH_4 fluxes, their strong correlation indicates shared dominant sources. Spatial distribution analysis revealed high consistency between both fluxes and natural gas consumption patterns, confirming natural gas as a common source. With Beijing's energy restructuring, natural gas has become the dominated terminal energy consumption. Its combustion releases substantial CO_2, while leakage processes emit CH_4, as validated by mobile observations detecting CH_4 fugitive emissions during production, storage and use stages. Although biogenic sources could contribute to CH_4 emissions, they account for at most 27 % of total CH_4 fluxes in the source area, ruling out the view that biological sources dominate both emissions. Attributing all CH_4 emissions to natural gas usage, the upper leakage rate of natural gas in Beijing was calculated as 1.12 % ± 0.22 %.

The CH_4 emissions from natural gas will exacerbate climate warming. Calculated flux results showed that the contribution of CH_4 to climate warming on a century and 20-year scale can reach as high as 8.37 % and 23.17 % of CO_2, respectively. On the basis of predicted energy report and calculated leakage rate, it is predicted that natural gas leakage will delay China's realization of carbon neutrality, which necessitates urgent attention to mitigate associated climate effects.

Point 27: In the supporting information section, the entire content is a long sentence, making it difficult to understand. Furthermore, I cannot find the underlying datasets and therefore it makes it very difficult for me to properly review.

Response 27: Thank you very much for the comment. We apologize for the lack of conciseness in our language and we have reorganized the language for the entire content in the Supplementary.

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