Supplementary materials of

Carbon reduction requires attention to the contribution of natural gas use: combustion and leakage

NUMBER OF PAGES:13

NUMBER OF FIGURES:9

NUMBER OF TEXTS:4

This supplementary information contains about details about Beijing Meteorological Tower, eddy observation system and navigation observation station, daily summer variation of CO_2 flux from 2009 to 2017, the total consumption, electricity inflow and the proportion of natural gas in total energy consumption from 2013-2022, the spatial distribution of CO_2 and CH_4 flux with wind speed and direction, grid distribution of natural gas consumption in Beijing, calculation methods of flux source area and natural gas leakage rate, uncertainty analysis of flux calculation, estimation of non-natural gas sources

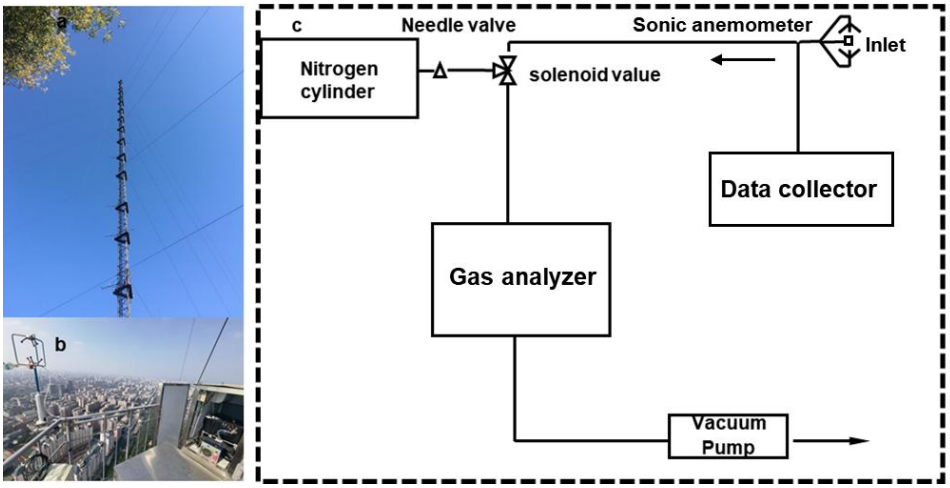


Figure. S1 Beijing Meteorological Tower(a), Eddy Observation System(b) and Schematic Diagram(c)

Text S1 Calculation methods of flux source area

Eddy covariance method belongs to single point observation, so the observation results at a certain height can only reflect the information of a specific area around the site. The area where the upwind direction of the observation site contributes to the measured flux is called the flux contribution source area. For a homogeneous underlying surface, the flux from all directions and distances around the observation point is the same, so the contribution of flux to the source area is not a major issue to consider. However, for the heterogeneous underlying surface studied in this article, as the wind direction and speed change over time, the measured flux comes from different directions and distances around the observation points, including the flux contributions of different underlying surface patches. Therefore, the analysis of spatial representativeness in flux observations is particularly important. The flux footprint model can be used to quantitatively analyze the range of flux contribution source regions. The flux footprint function estimates the position and relative importance of the flux source that affects a given height of flux observation based on observation height, atmospheric stability, and surface roughness, expressed as a set of distances between observation points and flux sources in different directions, the specific

calculation formula is as follows:

$$x_{peak} = X^*_{peak} z_m \left(\frac{\sigma_w}{u_*}\right)^{-0.8}$$

$$x_{nn\%} = X^*_{nn\%} z_m \left(\frac{\sigma_w}{u_*}\right)^{-0.8}$$

Among them, x_{peak} refers to contributing distance to maximum flux, $x_{nn\%}$ refers to contributing distance to nn% flux, X^*_{peak} and $X^*_{nn\%}$ refer to the dimensionless distance related to surface roughness, z_m refers to observation altitude,σ_w refers to the standard deviation of vertical wind speed, u_*refers to friction wind speed.

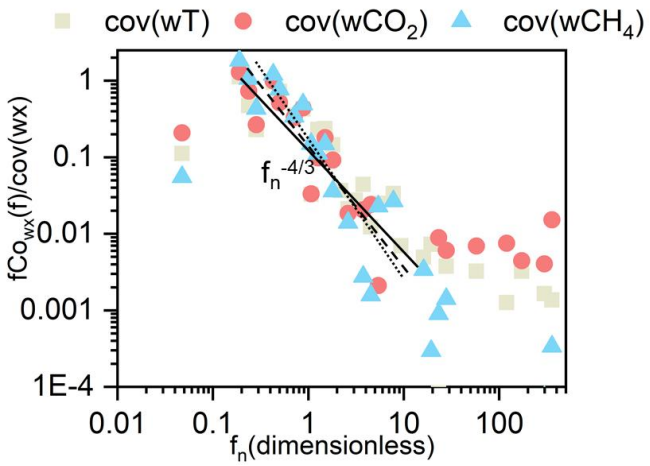


Figure. S2 Normalized cospectra of T(sonic temperature), CO_2 and CH_4 with respect to w(vertical wind component. The normalized frenquency f_n is defined as f_n =f(z-d)/U, where z,d and U denote measurement height(m), zero plane displacement(m) and wind speed(m/s), respectively. Each cospectrum is an average of half-hourly cospectra between 12:00-16:00. Regression of spectra for f_n between 0.1-10Hz marked by solid line for Co(wT) and dashed line for both Co(wCO_2) and Co(wCH_4)

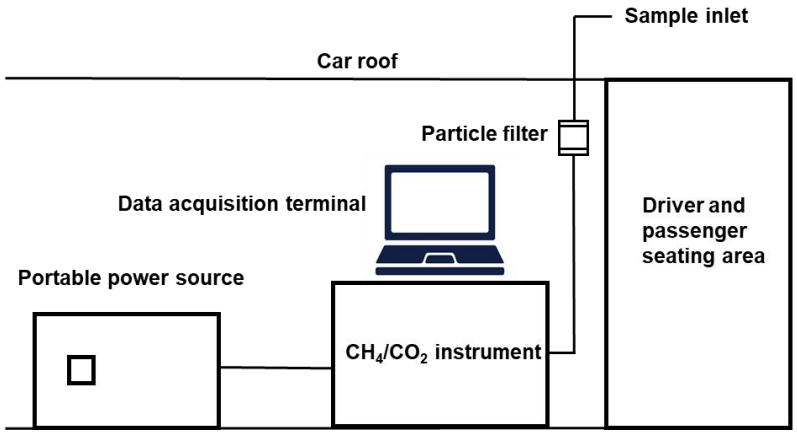


Figure. S3 Schematic diagram of navigation observation station

Text S2 The uncertainty analysis of flux calculation

Just like other complex observations, flux observations based on eddy covariance method are subject to both systematic and random errors. However, different many other observations, it cannot be directly calibrated on-site for the measured flux. In addition to systematic errors caused by instrument calibration and data processing, sampling errors are one of the largest sources of uncertainty in vortex related flux observations. Sampling errors cannot be eliminated through experimental design due to the limited number of independent samples that contribute significantly to the flux within any fixed sampling period, and there are various auto-correlations and cross-correlations in the two covariates. A longer sampling period will increase the number of independent samples, thereby reducing sampling errors, but it often leads to other problems such as lack of atmospheric stability.

To answer questions such as whether any two flux observations varies significantly from each other, whether a specific flux observation is different from zero notably, and whether the flux observation is in alignment with the model results, it is necessary to quantify the contribution of sampling errors. We adopted the scheme proposed by Finkelstein and Sims (2001)[1] to calculate the sampling error, and the specific calculation formula is as follows:

$$\sigma_F = \sqrt{\frac{1}{n}[\sum_{p=-m}^{m} \gamma_{w,w}(p)\gamma_{c,c}(p) + \sum_{p=-m}^{m} \gamma_{w,c}(p)\gamma_{c,w}(p)]}$$

Where n is the number of samples within the average period of flux, m needs to be greater than the turbulent integration time scale, p is the lag time, and $\gamma_{w,w}(p)$ and $\gamma_{c,c}(p)$ refer to the auto-covariance of w and c, respectively.

Figure.S4 shows the daily variation and random error of CO_2 and CH_4 flux during the observation period. The range of random error for CO_2 flux is 1.43-3.72 $\mu mol \cdot m^{-2} \cdot s^{-1}$, with a mean of 2.33 $\mu mol \cdot m^{-2} \cdot s^{-1}$. The range of random error for CH_4 flux is 10.92-29.54 $nmol \cdot m^{-2} \cdot s^{-1}$, with a mean of 18.43 $nmol \cdot m^{-2} \cdot s^{-1}$. Both flux and random error peak between 10:00 and 15:00, when the absolute random error is large while the relative random error is small.

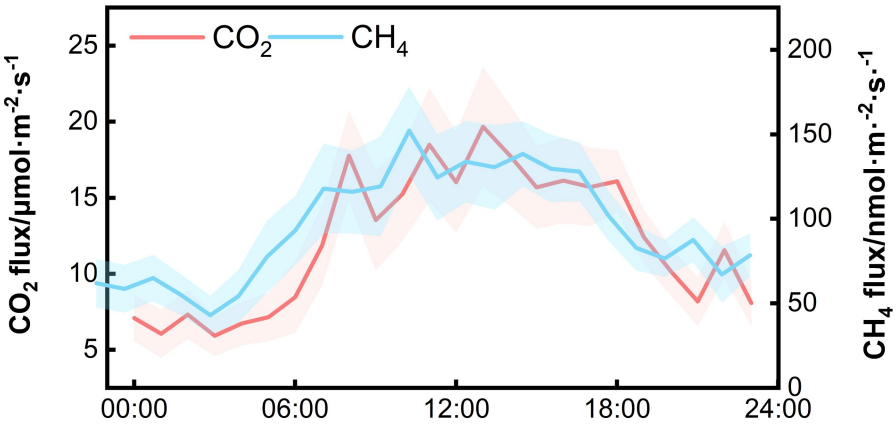


Figure.S4 The daily variation of CO_2 and CH_4 flux and their error bands

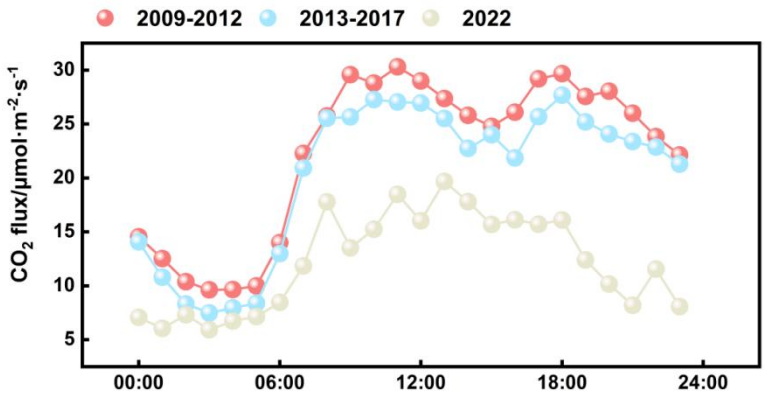


Figure.S5 Daily summer variation of CO_2 flux from 2009 to 2017 compared to this study

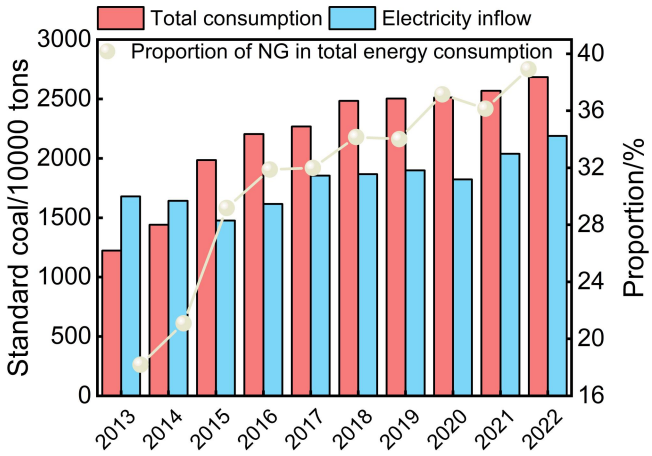


Figure. S6 The total consumption, electricity inflow and the proportion of natural gas in total energy consumption from 2013-2022

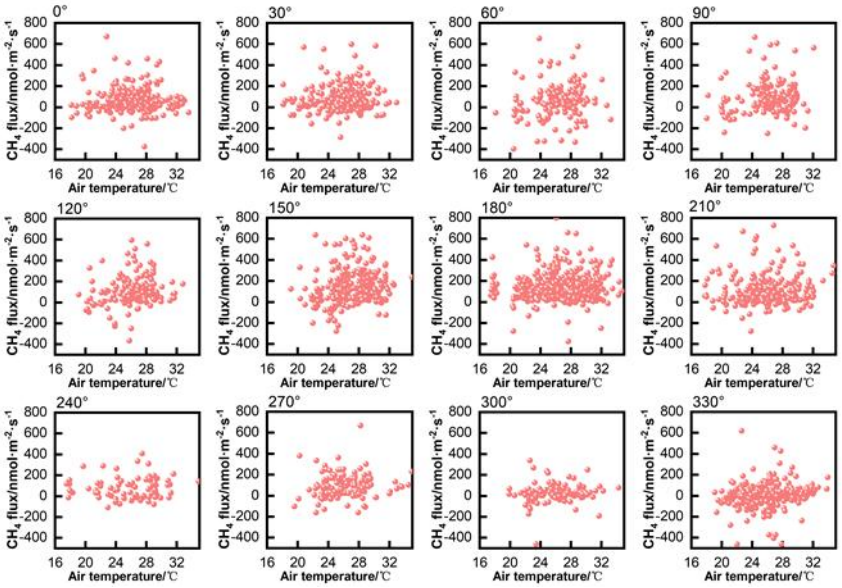


Figure. S7 Scatter plot of temperature and CH_4 flux along 12 directions

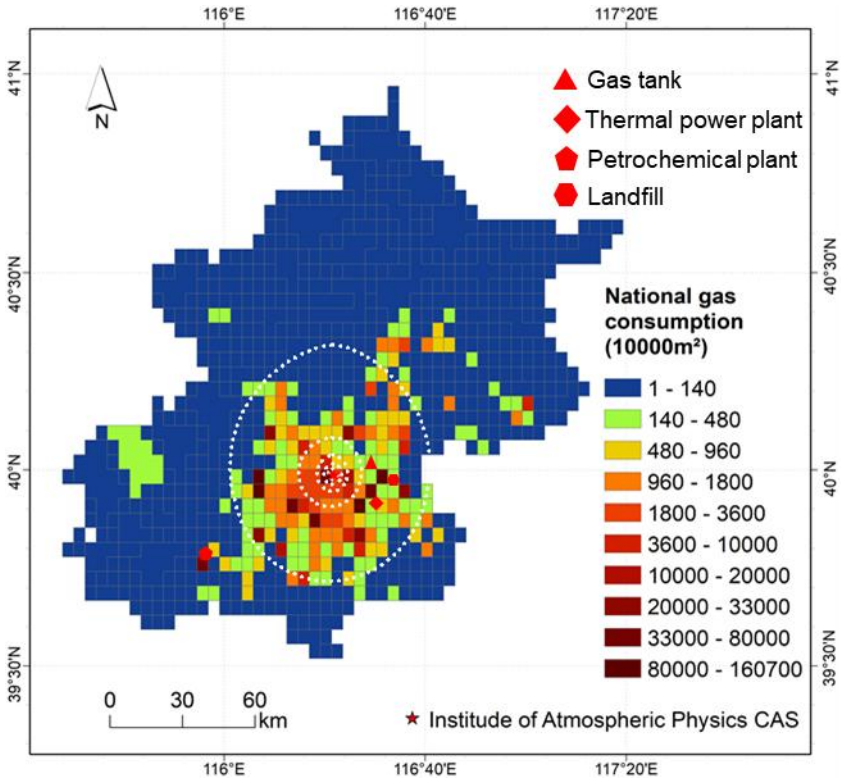


Figure.S8 Grid distribution of natural gas consumption in Beijing (the total residential consumption is gridded according to population density, the final grid result is the superposition of the enterprise and residential consumption, the white dotted line refers to the flux source area)

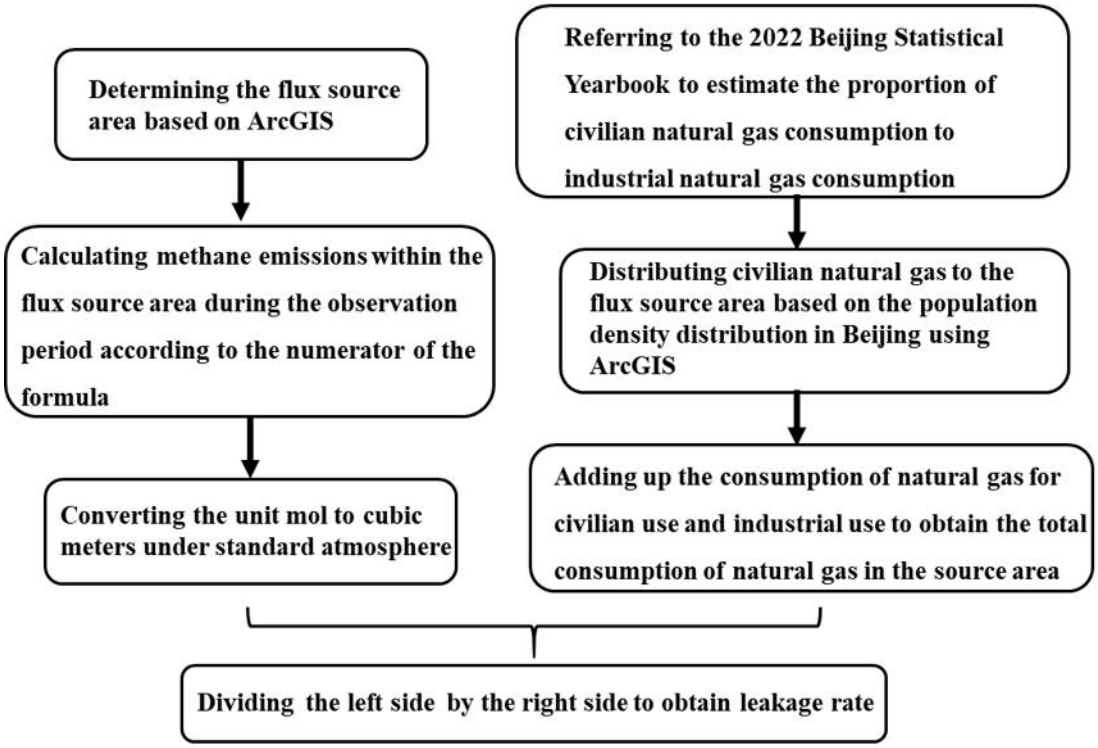
Text S3 Calculation methods of natural gas leakage rate（LR）

$$LR = \frac{Flux \times A \times F \times T}{NG_{sum}}$$

Among them, *LR* refers to methane leakage rate, *Flux* refers to CH_4 flux, *A* refers to area of flux source, *F* refers to factor converting amount of substance(mol) to volume(m^3), *T* refers to Observation duration, NG_{sum} refers to sum of natural gas consumption during observation period within the flux source area. The following diagram shows the specific calculation steps.

Considering that the natural gas consumption data of industry during the observation period was directly obtained by the Beijing Ecological Environment Center, the uncertainty of the total

natural gas consumption during the observation period mainly comes from civilian natural gas, however, the annual consumption of civilian natural gas only accounts for about 10% of industrial natural gas consumption in 2022. Therefore, we reckon that the uncertainty of natural gas leakage rate based on this method is relatively small though civilian natural gas was evenly distributed among observation durations without considering seasonal variability here.



Text S4 Estimation of non-natural gas sources

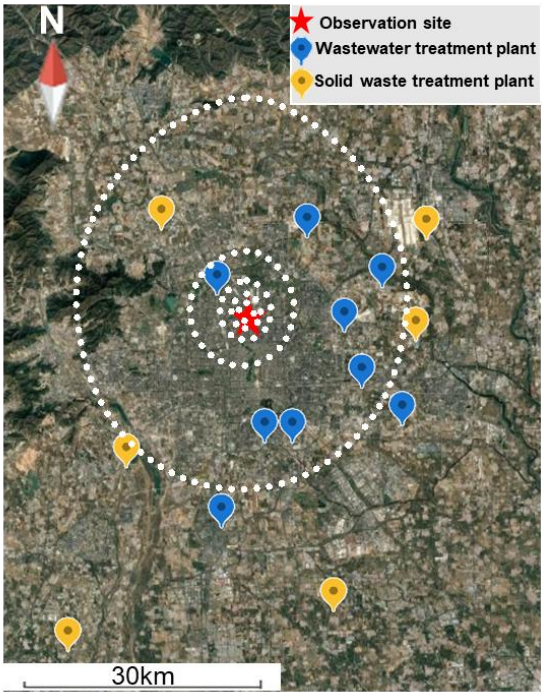


Figure.S9 Locations of large-scale sewage treatment plants and solid waste treatment plants in Beijing (the map was extracted from © Google earth: https://earth.google.com/)

The existing inventory shows that the waste treatment process mainly includes solid waste disposal, wastewater treatment (including anaerobic treatment of domestic and industrial wastewater, CH_4 emissions from sludge), which are important sources of methane in Beijing. The common methods of solid waste treatment include composting, incineration, and landfilling. Previous studies have shown that landfill treatment produces several orders of magnitude higher CH_4 than composting and incineration treatment.[2] Therefore, we mainly considered the methane emissions from landfilling treatment. Large-scale landfill sites in Beijing were selected here, it can be seen that most of them are not located within the flux source area (only one), so we assumed that methane emissions from garbage dumps within the source area could be approximately ignored (Figure.S10). However, methane emissions from wastewater treatment plants should be considered, as there distributed many sewage treatment plants within the source area (Figure.S10) with actual treatment capacities of over 40 million tons per year, which will make a certain contribution to methane emissions in the source area. In addition, urban sewage pipeline systems

also play an important role in methane emissions in cities, and CH_4 emissions from which in some areas may be much higher than CH_4 emissions from sewage treatment plants.[3,4]

Although based on current results, this contribution seemed relatively small compared to natural gas. Firstly, if biogenic sources dominated methane emissions within the source area, methane flux would show a high correlation with temperature. Hu et al.'s study indicated that a 10℃ increase in temperature will lead to a 38%-50% increase in waste based methane emissions,[5]which contradicts our observations., for no correlation was observed between methane flux and temperature in all directions (Figure.S7). Moreover, the observation experiments conducted by Yin et al. at the Guiyang wastewater treatment plants showed that the methane emission would peak at night considering that it was related to the wastewater treatment time and hydraulic retention time,[6] which was not reflected in our observation results.

The contribution of wastewater treatment plant emissions within the flux source area was roughly estimated based on the results of the literature. According to Zhao et al.'s conclusion,[7] the total amount of methane emissions from urban sewage treatment plants correlated strongly with the city's GDP, household consumption level and urban population density (correlation coefficient is as high as 0.7-0.9), In megacities like Beijing, the per capita CH_4 emissions from urban sewage treatment facilities can reach 2.9kg/year. According to the national census results(http://sedac.ciesin.columbia.edu//data/collection/gpw-v4/sets/browse), the permanent population of Beijing in 2022 is about 21.8 million, it is estimated that the population proportion in the flux source area is 64.8% of the total population in Beijing using ArcGIS 10.2 on basis of the world population grid data, with a corresponding population of approximately 14.1 million and a flux source area of approximately 2400 square kilometers. The methane flux emitted from the sewage treatment plants within the flux source area is approximately 12.35 $nmol \cdot m^{-2} \cdot s^{-1}$, which accounts for 13% of the methane flux during the observation period. As the CH_4 emissions during wastewater treatment are significantly correlated with COD concentration. we also roughly estimated its methane emissions based on emission factors. According to the estimates of Zhao et al. for 229 cities in China,[7] the CH_4 emission coefficient of urban sewage treatment plants is about

0.017-0.24gCH4/gCOD, while the average per capita COD production in Chinese cities is approximately 24kg/year.[8] the emission flux of sewage treatment plants within the flux source area is about 1.73-20.73nmol·m^{-2}·s^{-1} combining the flux source area and the population above, the flux results calculated based on the correlation between CH_4 emission and population density are within this range.

The methane emissions from urban sewer systems was estimated with the above emission factor method. The CH_4 emission coefficient of urban sewage pipelines is about 0.0532 gCH4/gCOD.[8] Based on the area of the flux source area and the population in the source area, the sewage pipeline flux is around 5 nmol·m^{-2}·s^{-1}. Therefore, the total methane flux from wastewater within the flux source area comes to 6.73-25.73 nmol·m^{-2}·s^{-1}, and whose contribution to the methane flux is the highest at 27%, making the lower limit of natural gas leakage in Beijing estimate to be 0.82%, 1.12% can be used as the upper limit of natural gas leakage ratio if all methane emissions are attributed to natural gas. Unfortunately, due to a lack of data, we are currently unable to quantify the uncertainty of the calculation results. However, it can be confirmed that natural gas is the main source of methane emissions within the flux source area.

Text S5 Formula of natural gas leakage rate

$$\ln maxexcessCH_4 = -0.988 + 0.817 \times \ln emission\ rate$$

Where the maxexcess CH_4 is the maximum CH_4 enhancement concentration (ppm)，the unit for emission rate is g/min.

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