A letter of Response to Reviewers

Ms. Ref. No.: egusphere-2025-3667

Title: High-resolution Emission Inventory Development and Co-emission Hotspot Identification of Air Pollutants and Greenhouse Gases in Central Plains Region, China

Reviewer #1:

The study by Li et al. developed a high-resolution emission inventory of air pollutants and major greenhouse gases in a heavily polluted province of China using a bottom-up approach. The authors compiled enterprise-level activity data and emission factors from field surveys, published literature, and statistical datasets. Following validation against existing emission inventories and an uncertainty analysis, the manuscript further examined the spatial and temporal patterns of air pollutant and greenhouse gas emissions. In addition, co-hotspot regions of both types of emissions were identified, offering useful insights for future strategies aimed at mitigating air pollution and climate change. Overall, the manuscript provides valuable information for improving the understanding of emission characteristics in China. However, several important issues need to be addressed before the manuscript can be considered for publication.

We sincerely appreciate the efforts in reviewing our manuscript and providing valuable comments. We also express our gratitude for your recognition of this research work, especially for your positive comments on the high-resolution emission inventory established by this research, the methods employed, and their practical significance. Below are the point-to-point responses to all the comments (The comments are marked in black color and our responses are marked in dark blue color). The changes that have been made according to these responses were marked in yellow color in the highlighted copy of revised version. Note that the following line numbers are shown in the corrected version.

Specific Comments:

1. I suggest that the authors carefully check and revise the manuscript for grammatical accuracy and appropriate word usage to improve its overall clarity and readability.

Response: Thanks for the constructive suggestion.
We have conducted a systematic review and revision of the grammar, vocabulary and expressions of the entire text. We have also carried out further verification using professional grammar-checking tools to enhance the overall quality.

2. Lines 85-86: “identify high-emission grids using hotspot analysis methods”: Please provide more details on the analysis methodology. Additionally, it is recommended that the authors restructure this paragraph into several sentences summarizing the main objectives of the study, highlighting the logical relationships among them, and clarifying the significance of the research.

Response: Thanks for the comment.
We have restructured the paragraphs into several sentences that summarize the main

objectives of our study. In addition, more further details have been provided about the analytical methodology used to identify high-emission grids through hotspot analysis. The specific revisions are as follows:

This study compiles an integrated emission inventory of air pollutants and greenhouse gases for Henan Province, providing a data foundation for coordinated atmospheric and climate governance. Emissions of nine air pollutants (SO_2, NO_X, CO, PM_{10}, $PM_{2.5}$, VOCs, NH_3, BC, OC) and three greenhouse gases (CO_2, CH_4, N_2O) were calculated under a unified source framework, followed by analysis of emission characteristics of different pollutants and cities. The reliability of emission inventory was evaluated through inter-study comparison and uncertainty analysis. We then allocated emissions to a monthly scale and a 3 km × 3 km grid to enable high spatiotemporal resolution analysis. To identify priority areas for emission control, a hotspot analysis was conducted: grids were ranked by emission level (top 20% assigned to four hotspot categories), and a bidirectional matching rule was applied to locate grids with concurrently high emissions of CO_2 and key air pollutants (such as SO_2, NO_X, $PM_{2.5}$, VOCs, BC and OC), thereby pinpointing key regions for coordinated mitigation. (See Lines 81-91 in the revised version)

3. More detailed information on the anthropogenic emission sources should be provided. For example, in addition to coal combustion, what other contributors are included in the energy structure? What agricultural products play a dominant role in this province compared with others, and what is their spatial distribution? For the seven emission categories, brief introductions of the major sub-sectors should be provided.

Response: Thanks for the comment and suggestions.

First, in Section 2.1, we provide more information about the energy structure and main agricultural products of the study region and their spatial distribution patterns. The specific revisions are as follows:

The total proportion of non-coal energy consumption is 37.3%, which includes a share of 15.7% for oil, 15.1% for primary electricity and other energy sources, and 6.5% for natural gas (HBS, 2023). (See Lines 109-111 in the revised version)

In terms of agriculture, in 2022, the total grain output of the province reached 67.9 million tons, with wheat and corn being the dominant crops, accounting for 56.1% and 33.5% (HBS, 2023). These crops are primarily distributed in regions such as Zhumadian, Nanyang, and Xinyang. Regarding animal husbandry, pig farming has shown remarkable scale, with an end-of-year stock of 42.6 million heads, ranking second nationwide. (HBS, 2023) Notably, Nanyang and Zhumadian have some of the highest livestock numbers in the province. (See Lines 113-117 in the revised version)

Second, for the introduction of major sub-sectors in Section 2.2, we briefly described the main sub-sectors of each category. The specific revisions are as follows:

The source of electricity and thermal power, combined thermal power generation, combined heat and power， heat production and supply, as well as biomass power generation, among others. The industrial source is further categorized into industrial combustion, non-ferrous metal, industrial solvent, cement, among others. Mobile source and oil storage and transportation include on-road mobile, railway, river transport, gas station, gasoline and diesel transport, among others. Dust emission sources consist of road dust and construction dust. The residential source cover corporate catering, residential combustion, straw household, civil solvent use, among others. Agricultural source pertains to livestock breeding, fertilizer use, among others. Waste treatment sources include wastewater treatment sewage treatment and garbage disposal. (See Lines 132-141 in the revised version)

References:

Henan Bureau of Statistics (HBS): Henan Statistical Yearbook, https://oss.henan.gov.cn/sbgt-wztipt/attachment/hntjj/hntj/lib/tjnj/2023nj/zk/indexch.htm, (last access: 10 April 2025), 2023.

4. Methodology for emission calculation: Although the authors have provided the data sources for activity levels and emission factors, additional methodological details should be clarified in the main text:
(1) For the activity data, field surveys constitute a major source for many emission sectors, as shown in Table S1. Therefore, it is recommended to describe how these surveys were conducted. What were the sample sizes for different source sectors? What measures were taken to ensure that the survey data are representative and reliable?
(2) The values and data sources of parameters used in the equations for calculating emission rates of specific species should be provided. In addition, the emission factors of methane (from mobile sources, oil storage and transportation, and waste treatment), nitrous oxide (from mobile sources, oil storage and transportation, agricultural sources, and waste treatment), and air pollutants from mobile sources should also be reported.
(3) For the spatial allocation, the authors used Google Maps rather than Chinese mapping services such as Baidu Maps and Amap. Could the authors discuss the potential impacts of using different map sources on the spatial distribution of emissions?

Response: Thanks for the constructive suggestion.

(1) For the activity data, as presented in Appendix S1, the field research in our study refers to investigations organized by management authorities. We relied on pollution source statistical data from the Henan Provincial Department of Ecology and Environment, supplemented by official statistical materials such as the "China Energy Statistical Yearbook," "China Transportation Statistical Yearbook," and " Henan Statistical Yearbook." A verification process was conducted to compare macro-level data from relevant emission sectors with field research findings, addressing any discrepancies identified. To avoid ambiguity in expression, we have made appropriate revisions in Appendix S1.

(2) We provided the parameter values and data sources used in the equation for calculating the emission rate of a specific species (See Table S9 in Supplementary Material). For emission factors, we also supplement the data of methane (from mobile sources and oil storage and transportation, and waste treatment), nitrous oxide (from mobile sources and oil storage and transportation, agricultural sources, and waste

treatment), and air pollutants from mobile sources. (See Table S5, Table S6, Table S8 and Table S10 in Supplement Materials)

(3) Regarding the use of Google Maps in spatial allocation, our primary objective is to address the issue of misalignment or displacement of geographic coordinates for certain enterprises within the survey data (for instance, some coordinates may be located outside the boundaries of Henan Province). Google Maps enables us to more accurately locate and correct these enterprises' latitude and longitude information. In this study, there is no fundamental difference between using Google Maps and domestic mapping services such as Baidu Maps in achieving the essential goal of spatial positioning. To eliminate any potential misunderstandings, we have included parentheses around "Google Maps" in the relevant supplementary tables.

5. Interpretation of the results: The authors should ensure that the results are described accurately in the main text. For example:
(1) In lines 242-243, Table 1 shows that the contribution of residential sources to SO_2 is higher than that of the electricity and thermal power sector, which differs from the statement made by the authors.
(2) In Section 3.1, it should be clarified whether the percentages represent the contribution of a specific source to the total emissions of a given species, or the contribution of a sub-sector within a source category, to avoid potential confusion.
(3) In lines 279-281, the total contribution of firewood and crop residue combustion to $PM_{2.5}$ is 6.77%, which differs from the 5.6% value reported in the text.
(4) In lines 350-351, industrial sources contribute the most to VOC emissions and have a similar contribution to CO_2 as mobile sources. Therefore, it is inaccurate to state that motor vehicles are the dominant emission source in Zhengzhou.
(5) Lind 379: According to Figure 3, Xuchang should belong to the central region of Henan Province.

Response: Thanks for the comment and valuable suggestions.
We have carefully checked and corrected the data, expressions and conclusions in the main text to ensure that all the descriptions in the text precisely correspond to the data in corresponding table and figure. The detailed modifications are as follows:

(1) Regarding the source contribution of SO_2 emissions, we have carefully verified the data in Table 1 and have revised the relevant descriptions accordingly. The specific revisions are as follows:

The high emission levels in the industrial sector are primarily attributed to its substantial coal consumption, which constituting 38.3% of the province's total, coupled with relatively lagging desulfurization control technologies in certain kilns. Among non-industrial sources, residential activity contributes 6.45% of emissions, slightly exceeding that of the electricity and thermal power sector at 5.0%. This discrepancy mainly stems from the significant progress in ultra-low emission retrofits within the power industry since the 13th Five-Year Plan period, whereas pollution control measures for residential solid fuel use, particularly the co-combustion of coal and biomass in households, remain comparatively underdeveloped. (See Lines 264-271 in the revised version)

(2) Clear instructions on the interpretation of percentages have been added at the beginning of Section 3.1. The specific revisions are as follows:
All emission contribution percentages reported in this section, unless otherwise specified, refer to the contribution of the emission source to the total emissions of the species. (See Lines 259-261 in the revised version)

(3) We have rechecked the original data and charts. The total contribution of firewood and crop residue combustion to $PM_{2.5}$ has been changed to "6.8%". (See Line 304 in the revised version)

(4) Indeed, the statement was inaccurate. We have deleted the inaccurate expression "mainly from motor vehicle emissions". (See Lines375-376 in the revised version)

(5) According to the geographical partition illustrated in Figure 3, Xuchang City indeed belongs to the central region of Henan Province. We have uniformly revised all the relevant statements in the original manuscript and systematically verified the regional classifications throughout the text to ensure accuracy. During our review of data across regions, we discovered that Eastern Henan (Kaifeng, Shangqiu) and Southern Henan (Zhumadian, Xinyang, Zhoukou) exhibit significant similarities in their emission structures; specifically, both regions are characterized by a predominant agricultural ammonia emission profile coupled with relatively low carbon emission intensity. To enhance the coherence of our paper, we have integrated the previously separately - discussed aspects regarding Eastern and Southern Henan into a unified analysis. The specific revisions are as follows:
In the southern (Zhumadian, Xinyang, Zhoukou) and eastern (Kaifeng, Shangqiu) regions of Henan Province, the most notable feature is that the NH_3 emissions from agricultural activities are relatively large, but the total amount and intensity of carbon emissions are relatively low. For instance, the proportion of the primary industry in the gdp of Zhumadian in the southern region is 17.6%, which is the highest in the province. A large livestock population, including 6.7 million pigs and 53.6 million poultry, generates 76.8 kt of NH_3 emissions (ZBS, 2024), of which agricultural sources account for 96.5%. On the contrary, the total regional CO_2 emissions were only 163.3 million tons, with an emission intensity of 50000 kt/10^8yuan GDP, which was significantly lower than that of northern industrial cities. The NH_3 emissions of Shangqiu in the eastern region were 666 kt, ranking fourth in the province and far exceeding the provincial average. In contrast, the regional CO_2 emissions were lower than the provincial average. Together, they form an "agricultural-dominated" emission model that is completely different from that of the industrial cities in the north. (See Lines 393-404 in the revised version)

References:

Zhengzhou Bureau of Statistics (ZBS): Zhengzhou Statistical Yearbook, https://tjj.zhengzhou.gov.cn/u/cms/tjj/statistical_2023/index.htm, (last access: 10 March 2025), 2024.

6. Line 337: The term “source apportionment” typically refers to analyses conducted using specific methods, such as receptor models or regional models with source apportionment techniques. Since the results here are based on emission inventories, a more accurate description would be “source contribution analysis” or “emission contribution analysis”.

Response: Yes, it is correct. Thanks for the constructive suggestion.
We have corrected the expression "source apportionment" to the more accurate "source contribution analysis" in whole paper.

7. Evaluation of emission inventories: (1) It is recommended to indicate the version of the MEIC inventory, as well as the spatial coverage and resolution for all emission inventories, in Table 2. (2) Lines 401-403: Could the authors elaborate on the differences between CHRED and this study, specifically clarifying what is meant by direct and indirect CO_2 emissions? (3) The authors claim that the estimated VOC emissions are lower than those in the MEIC inventory, attributing this to the use of localized emission factors for mobile sources and oil storage and transportation. However, since the dominant contributors to VOC emissions are industrial and residential sources rather than mobile sources, this explanation may not fully account for the observed differences. (4) It should be noted that the reference years of the current study and the other inventories differ, which may partly explain the discrepancies in emission estimates. (5) Please provide more detailed information on the Monte Carlo simulations, including the number of simulations performed for each species from each source, the software and its version.

Response: Thanks for the constructive suggestion.
Regarding the opinions on the comparison of emission results and the uncertainty analysis, we have meticulously reviewed and refined each item, and improved the relevant content and expressions.

(1) In Table 2, we have added the version of MEIC inventory, as well as the spatial coverage and resolution for all emission inventories. Please see the new Table 2.

(2) Regarding the difference between CHRED and this study, this study presents a merged emission inventory that classifies both atmospheric pollutants and greenhouse gases based on a unified source classification. For CO_2, we only calculate the direct emissions within the province resulting from the combustion of fossil fuels and industrial production processes (Scope 1). In contrast, the main greenhouse gas emission inventory of the CHRED database covers both Scope 1 and Scope 2 (indirect) emissions, the latter of which originate from the use of purchased electricity and heat (whose physical release occurs outside the province). This clarification has been added in the revised manuscript. The specific modifications are as follows:
The CHRED database accounted for direct emissions (such as the combustion of fossil fuels and industrial processes within the province) and indirect emissions (such as the consumption of purchased electricity), while this study focuses on the direct contributions at the production end and only accounted for direct emissions. (See Lines 419-422 in the revised version)

(3) Indeed, the estimated VOC emissions are lower than those reported in the MEIC inventory. This discrepancy cannot be attributed solely to the incorporation of localized emission factors for mobile sources and oil storage and transportation. Additionally, due to the reduction in the levels of industrial activities such as industrial painting and surface spraying, as well as the decrease in the activity levels of activities like dining and household solvents in the residential sector (due to the reduction in building area in 2022 and the impact of the epidemic lockdown in the second half of the year, intermittent interruptions occurred in activities such as dining services and dry cleaning), these combined factors have led to a reduction in VOC emissions. We have incorporated the addition into the text. (See Lines 435–441 in the revised version.)

(4) Yes, this study differs from other emission statistics reports in terms of the reference year used. This is indeed an important reason for the differences in the estimated emission figures. This consideration has been incorporated into the revised text. (See Lines 409–410 in the revised version.)

(5) In this study, the "Monte Carlo simulation" does not rely on external specialized software but instead employs a transparent computational algorithm based on probabilistic formulas. For each uncertain parameter (e.g., emission factor), we calculate its mean and standard deviation (sd) in Excel, and then generate 10,000 random samples following a normal distribution using the formula "=NORMINV (RAND (), mean, sd)". These 10,000 sets of sampled parameter values are subsequently input individually into the established emission calculation model, yielding 10,000 corresponding emission estimates. Statistical analysis is performed on these results to compute their mathematical expectation (mean) and 95% confidence interval. Finally, the 95% confidence intervals and means for each major source category are incorporated into the formulas $[a, b]=\left[\frac{D\text{-}E}{E}-\frac{T\text{-}E}{E}\right]$(where a and b represent the lower and upper bounds of the uncertainty range, respectively; D and T denote the lower and upper limits of the 95% confidence interval, respectively; and E is the mathematical expectation), thereby deriving the overall uncertainty range for the final emission inventory. Meanwhile, we have made corresponding additions in the main text. Specifically, as follows:

This study employs the Monte Carlo simulation method to assess the uncertainty of the emission inventory. With respect to the probability distribution characteristics of emission factors, a 10000-iteration random sampling simulation is conducted to calculate the 95% confidence intervals for pollutant emissions from each source category. (See Lines 460-462 in the revised version)

Table 2. Comparisons with other study.

Region	Year	GDP/Billion CNY	Pollutant/kt										Resolution	Reference
			SO_2	NO_X	CO	PM_{10}	$PM_{2.5}$	VOCs	NH_3	CO_2	CH_4	N_2O		
Henan	2022	62106	408.7	1336.2	4647.3	901.1	440.0	759.3	672.7	540462.0	12462.0	42.9	3km	This study
Henan	2020	54691	232.7	938.0	5784.6	342.8	265.2	1181.7	789.0	427679.2			0.25°	MEIC
Henan	2020	62106								572790.0			10km	CHRED
Henan	2020	54691								483738.0			No grids	CEADs
Guangdong	2020	88521	181.0	985.0			191.0	976.0	390.0				3km	Li et al., 2023
Shanxi	2017	55216	131.0	381.7		279.5	737.7	206.1					3km	Bo et al., 2023

Annotation: notation: MEIC1.4 V2.0, Spatial Coverage: China (Multi-resolution Emission Inventory for China, developed by Tsinghua University, http://meicmodel.org.cn/?page_id=560); CHRED, Spatial Coverage: China (China High-Resolution Emission Database, Developed by the Department of Earth System Science, Tsinghua University, College of Environmental Sciences and Engineering, Peking University; Institute of Atmospheric Environment, Chinese Research Academy of Environmental Sciences; https://www.ieimodel.org); CEADs, Spatial Coverage: China (China Emission Accounts and Datasets, Developed by Institute of Climate Change and Sustainable Development, Tsinghua University, https://www.ceads.net/data/province/)

8. Lines 458-459 and 468-470: The meteorological factors mentioned only affect the concentrations of air pollutants and are unrelated to the results presented in Figure 5, which are based on the temporal allocation of annual emissions for each species.

Response: Thanks for the constructive suggestion.
Yes, the monthly variations in pollutant emissions are essentially related to data such as activity levels. We have removed the descriptions related to meteorological factors. (See Lines 482-483 and 490-491 in the revised version)

9. It is suggested that the authors include a figure showing the population distribution in the supplementary materials.

Response: Thanks for the constructive suggestion.
A figure showing the population distribution has been added to the supplementary information (Supplementary Figure S1) as suggested.

10. Lines 507-508: What are the co-commitment types being compared against?

Response: Thanks for the constructive suggestion.
The co-commitment types (i.e., the first four categories of emission hotspots) are being compared against all other non-hotspot grid cells. At the same time, combined with the comments in Q11, we removed this sentence in the revised manuscript.

11. It is difficult to distinguish differences in the co-hotspot distributions, particularly for NO_X, $PM_{2.5}$, and VOCs, due to overlapping emission sources. In fact, the co-hotspot areas can be inferred directly from the spatial patterns of air pollutants and CO_2 shown in Figure 6. Are there any seasonal variations in the co-hotspot areas? This information could be valuable, as the dominant air pollutants vary seasonally.

Response: Thanks for the constructive suggestion.
In the revised version, we attempted to identify the seasonal co-hotspots. We selected January (winter), April (spring), July (summer), and November (autumn) as representative months to identify the co-hotspot distribution on a seasonal scale. Some interesting phenomena were discovered. At the same time, during this process, we also optimized the hotspot identification method, eliminating the grids with zero emissions and those outside the border of Henan Province. This can help alleviate the problem of difficult differentiation of the differences in common hotspot distribution due to the stacking of emission sources. We have updated Figures 7, Figure 8 and Figure S2 and revised the corresponding description in the main body as follow:

To identify priority areas for the coordinated control of CO_2 and air pollutants, grid cells with high co-emissions of both pollutants were identified at a spatial resolution of 3 km×3 km and defined as "co-hotspots" (Figure7 and 8). To analyze temporal variations in emissions, representative months—April (spring), July (summer), November (autumn), and January (winter)—were selected to identify seasonal co-hotspots, with the results shown in Figure S2.
The results indicate that SO_2 emissions show the highest spatial clustering, with Category 1 co-hotspots accounting for 72.2% of SO_2 and 57.1% of CO_2 emissions, yet

comprising only 467 locations. These are mainly concentrated in the central-northern industrial zone of Henan, centered on Zhengzhou, Luoyang, and Pingdingshan. Their contribution to CO_2 emissions peaks in winter (10%), reflecting increased industrial heating demand. NO_X emissions are also highly concentrated, with Category 1 co-hotspots covering 57.8% of NO_X and 78.8% of CO_2 emissions across 843 locations. They cluster in industrial zones and extend linearly along transport corridors, indicating both industrial and mobile sources. Seasonal contributions to CO_2 are elevated in summer (6%) and winter (10%). For $PM_{2.5}$, Category 1 co-hotspots contribute 42.2% of emissions and 77.6% of CO_2, with 721 locations largely overlapping SO_2 hotspots, highlighting industrial dominance. Seasonal counts rise from 146 in spring to about 300 in autumn and winter, with winter contributing 10% of emissions, linked to intensified industrial and area-source activities. VOCs emissions are more dispersed, with Category 1 co-hotspots contributing only 27.1%, while non-hotspot areas account for 67.1%. Spatially, they occur in industrial parks and urban areas, and show stronger coupling with CO_2 in winter within key industrial zones. BC emissions from Category 1 co-hotspots make up 34.2% of BC and 58.2% of associated CO_2, totaling 426 locations aligned with industrial and transport zones. Co-hotspots peak in number during spring (891) but contribute little to CO_2 (4%), indicating widespread low-intensity burning. In winter, they are more concentrated and contribute 8% of CO_2, reflecting centralized heating and industrial coal use. OC emissions are the most dispersed, with Category 1 co-hotspots contributing only 17.1%—the lowest among pollutants—and non-hotspot areas 81.2%, suggesting predominance of distributed area sources with limited link to concentrated energy use. (See Lines 526-551 in the revised version)

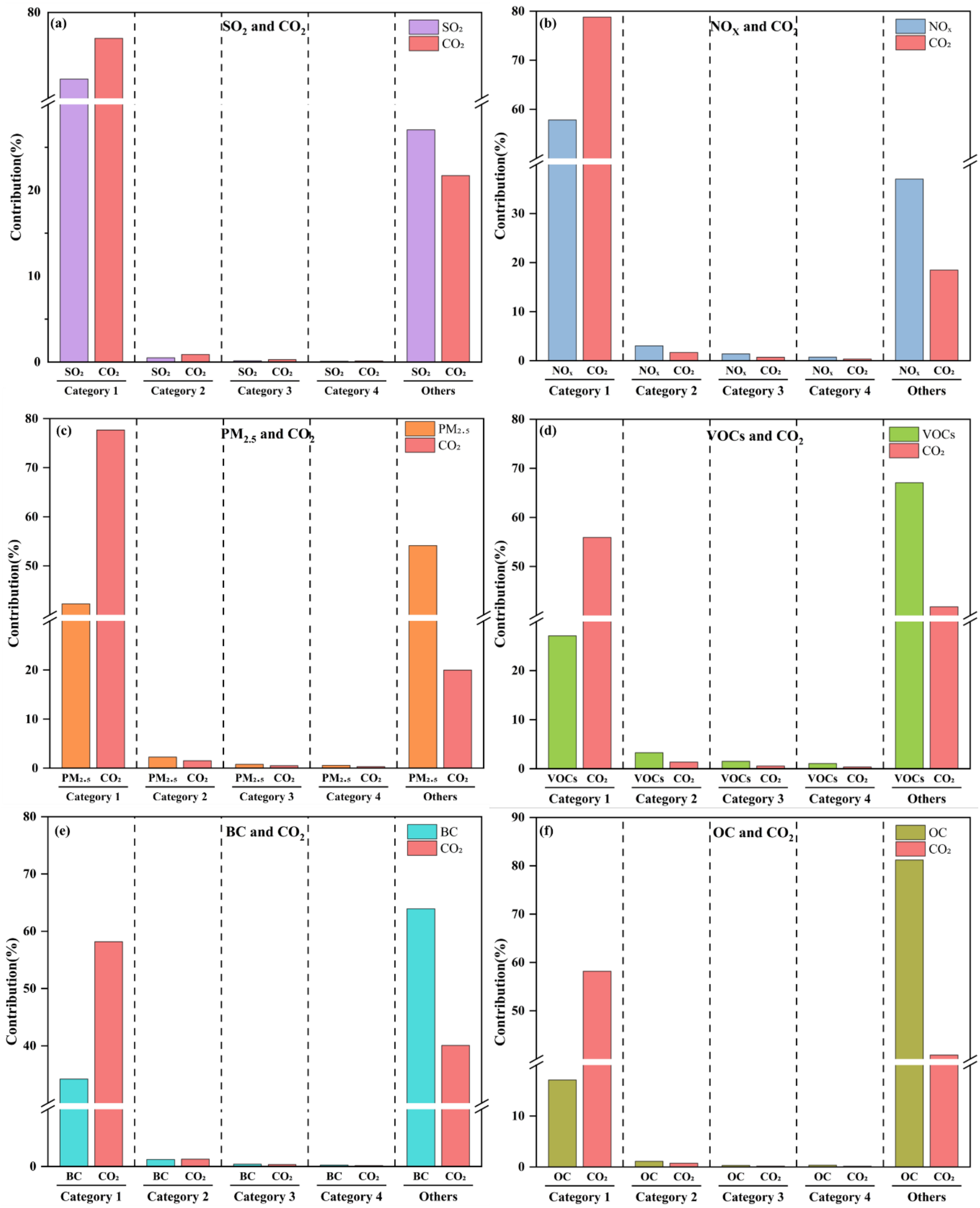

Figure 7. Emission contributions of five categories to different emission co-hotspot. (a) SO_2 and CO_2, (b) NO_X and CO_2, (c) $PM_{2.5}$ and CO_2, (d) VOCs and CO_2, (e) BC and CO_2, (f) OC and CO_2. Categories represent the grad of grid based on descending order of emissions: Category 1 (top 5% grids), Category 2 (6-10%), Category 3 (11-15%), Category 4 (16-20%), and Others (bottom 80%).

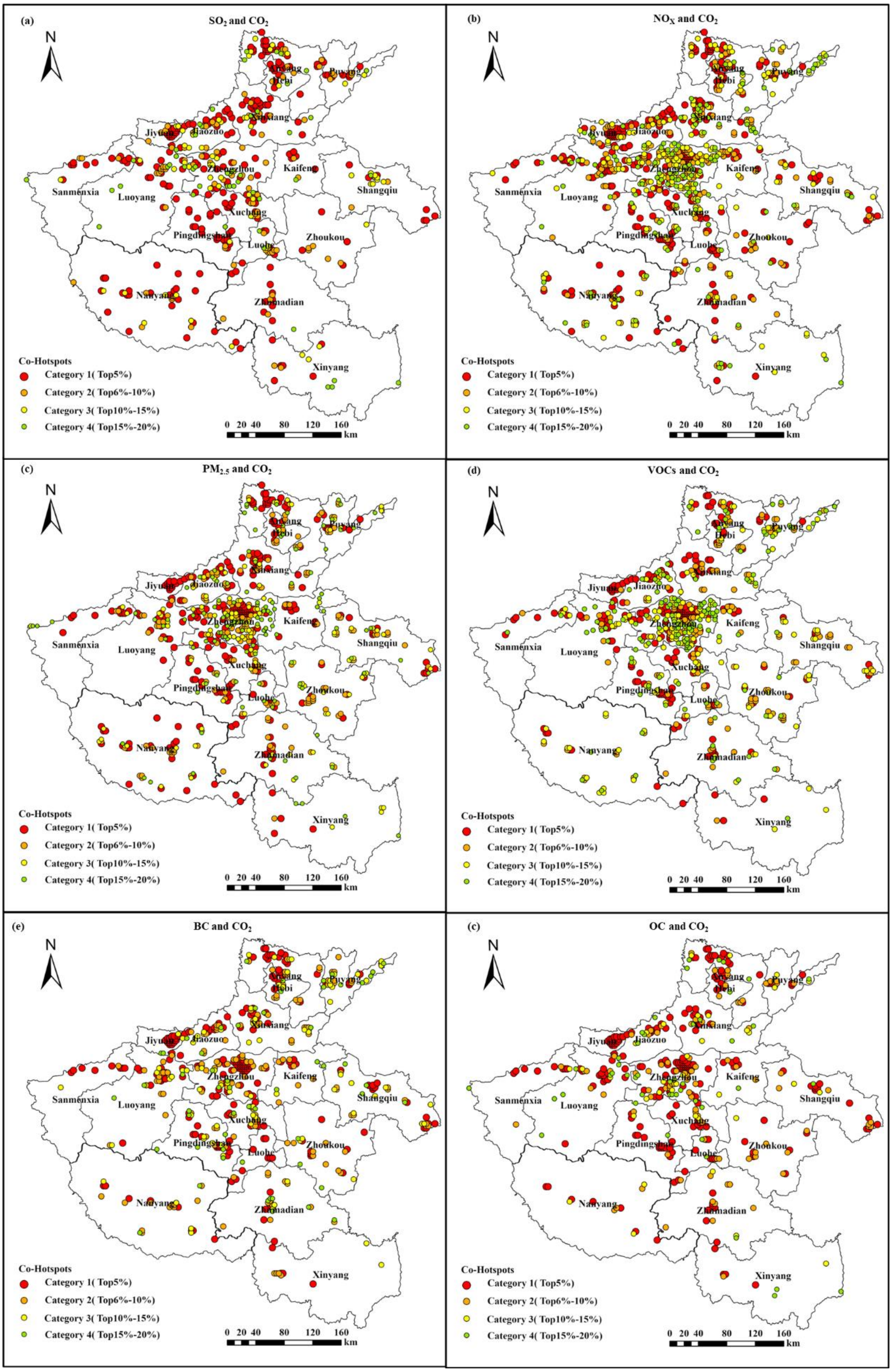

Figure 8. Spatial maps of the co-hotspots of (a) SO_2 and CO_2, (b) NOx and CO_2, (c) $PM_{2.5}$ and CO_2, (d) VOCs and CO_2, (e) BC and CO_2, and (f) OC and CO_2.

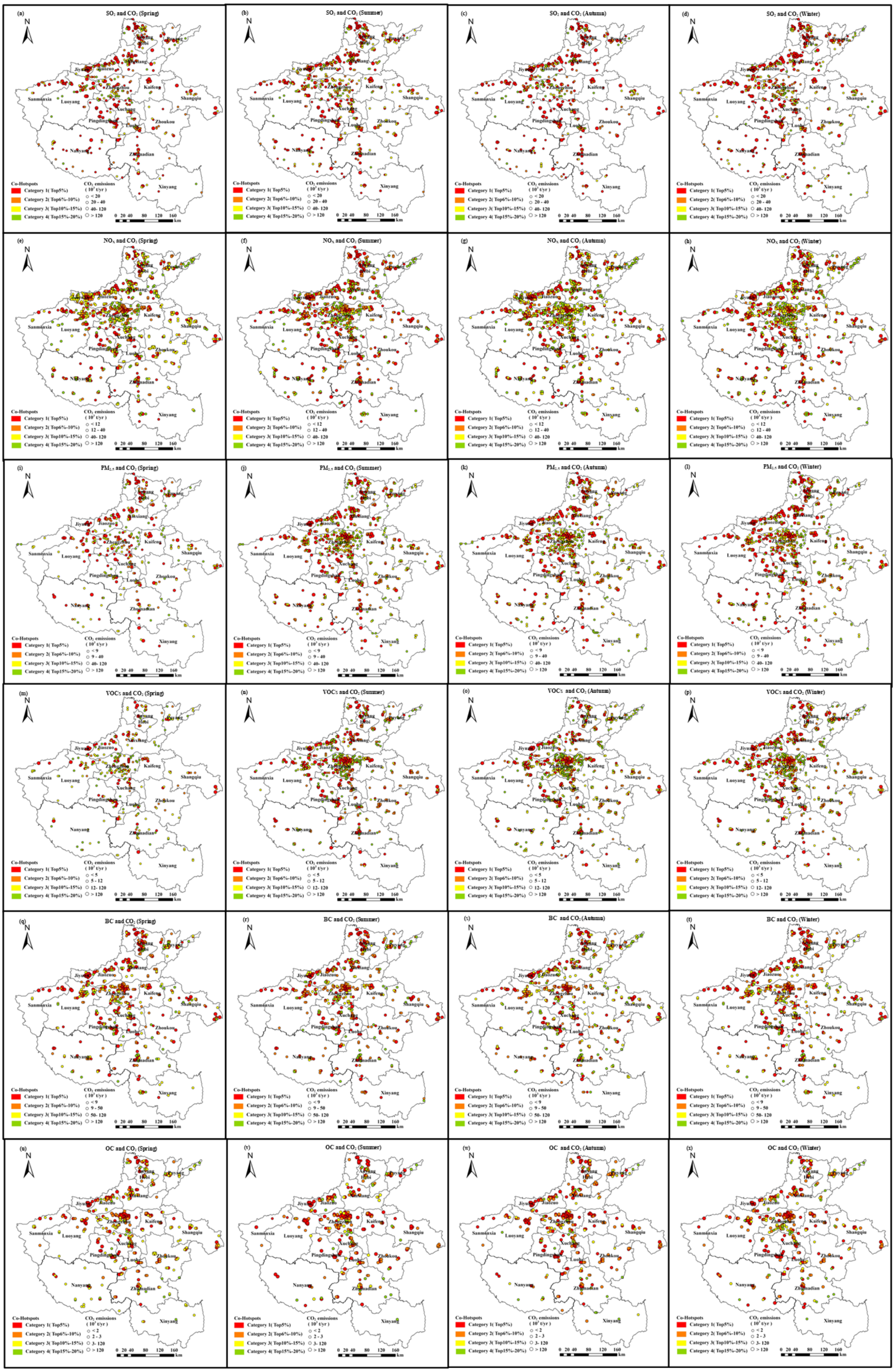

Figure S2. Seasonal spatial maps of the co-hotspots of (a-d) SO_2 and CO_2, (e-h) NO_X

and CO_2, (i-l) $PM_{2.5}$ and CO_2, (m-p) VOCs and CO_2, (q-t) BC and CO_2, and (u-x) OC and CO_2 from spring to winter.

(12) It is suggested to merge Sections 4 and 5 into a single section that summarizes the methodologies and key findings of the current study, compares them with other relevant research, and provides insights for future work.

Response: Thanks for the constructive comment.
In the revised version, Section 4 and Section 5 of the original version had been merged into one single Section 4.
In addition, we also refined the description of comparisons with other relevant studies as "This study, in comparison with the emission data from inventories such as MEIC and CEADs, as well as those from provinces like Guangdong and Shaanxi, reveals that the use of localized activity data and emission factors—alongside distinct energy structures and intensive agricultural and industrial activities—results in reasonable discrepancies in the estimation of CO_2 and various pollutants". (See Lines 581-584 in the revised version)

Technical Corrections:
1.Line 37: "they are fundamentally both gaseous ……" This is not correct. Particulate matter is also an important type of pollutant emitted from anthropogenic sources.

Response: Thanks for the constructive suggestion.
We have revised the sentence as "they are fundamentally both emitted in gaseous form highly overlapping emission sources, including ……" .

2. Lines 51-55, 71-74, 396-399, and 459-463: These sentences should be rephrased for clarity and accuracy.

Response: Thanks for the constructive comment.
The new sentences have been rephrased.
Furthermore, a series of emission inventories in China have been constructed at the regional (Xu et al., 2020; Zhao et al., 2024; Wu et al., 2022), provincial (Zhou et al., 2020; Jiang et al., 2020a), and urban (Liu et al., 2018) levels. Meanwhile, significant progress has also been made in building inventories for key industries (Gao et al., 2019; Hua et al., 2016; Jiang et al., 2020b). (See Lines 51-55 in the revised version)

Henan Province is a key polluted region with high concentrations of both $PM_{2.5}$ and O_3. In 2023, for instance, the annual $PM_{2.5}$ concentration exceeded the national standard II by 28%, and the O_3 concentration exceeded the standard II by 12%. Furthermore, the share of days with good air quality was lower than the national average of 85.5% (MEEPRC, 2024a). (See Lines 72-75 in the revised version)

This difference primarily stems from variations in activity data source and spatial resolution. While all studies employ the emission factor approach, our estimates are derived from individual enterprise-level accounting of 7390 designated size industrial enterprises, incorporating detailed terminal energy consumption data from power and heating sectors, using one-to-one enterprise-level accounting rather than aggregated statistical data. (See Lines 412-416 in the revised version)

NH_3, CH_4 and N_2O, which have higher emissions in the summer months, especially June and July, and lowest emissions in the winter months (January and December). Primarily caused by elevated NH_3, CH_4 and N_2O releases during summer months due to intensive agricultural practices, such as the application of nitrogen fertilizers and animal excreta management. In winter, when agricultural activities are reduced, emissions of NH_3 and N_2O are significantly lower. (See Lines 485-488 in the revised version)

References:

Gao, C., Gao, W., Song, K., Na, H., Tian, F., and Zhang, S.: Spatial and temporal dynamics of air-pollutant emission inventory of steel industry in China: A bottom-up approach, Resources Conservation and Recycling, 143, 184-200, 10.1016/j.resconrec.2018.12.032, 2019.

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Zhao, H., He, W., Cheng, J., Liu, Y., Zheng, Y., Tian, H., He, K., Lei, Y., and Zhang, Q.: Heterogeneities in regional air pollutant emission mitigation across China during 2012 – 2020, Earths Future, 12, e2023EF004139, 10.1029/2023EF004139, 2024.

Zhou, Z., Tan, Q., Deng, Y., Song, D., Wu, K., Zhou, X., Huang, F., Zeng, W., and Lu,

C.: Compilation of emission inventory and source profile database for volatile organic compounds: A case study for Sichuan, China, Atmos Pollut Res, 11, 105-116, 10.1016/j.apr.2019.09.020, 2020.

3.Lines 78-80: The content is repeated in the following paragraph. It is recommended to remove this sentence.

Response: Yes, it has been removed.

4.Line 89: The term "research site" is not appropriate for describing a region.

Response: It has been changed to "research region" as suggested

5.Lines 100-101: It is recommended to replace "is dominated by" with "is characterized by". Additionally, the phrase "a split of" is not appropriate in this context and should be removed.

Response: Thanks for the constructive suggestion.
We have replaced "is dominated by" with "is characterized by" and removed "a split of" as suggested.

6.Line 116: "By refer to…" => "By referring to"
7. Line 118-119: "seven emission source of" => "seven emission sources of"

Response: Thanks for the constructive suggestion.
We have completed the revisions to address the grammatical issues in Technical Corrections 6 and 7.

8. Lines 130-132: It is suggested to merge these sentences with the previous paragraph.

Response: The sentences "Emission factors were chosen using pertinent literature and local assessments, emphasizing……" have been merged with the previous paragraph.

9. Please ensure the consistent use of dash symbols and the correct formatting of subscripts for terminology throughout the manuscript.

Response: We have double-checked the entire text. The dash symbols and subscript formatting have been made consistent throughout the manuscript.

10. In Table 1, a "–" symbol in the total PM_{10} emissions should be removed.

Response: Yes, the "–" symbol has been removed.

11. Line 250: "account for" => "accounting for"
12. Lines 258-259: "Same to SO_2" => "Similar to SO_2"; "NOx emission" => "NOx emissions"
13. Line 262: "industrial source" => "industrial sources"

Response: Thanks for the constructive suggestion. All these minor issues have been corrected.

14. Lines 263-265 and lines 284-286: Are those results from the current study, or are they reported in other published literature?

Response: Thanks for the constructive comment.

Lines 263–265 and 284–286: The reported percentages (93.1% and 14.0%) correspond to the empirical findings of our study. The citations have been strategically relocated to clarify that these figures are either corroborated by, or situated within the context of, prior literature. We have revised the relevant sentences to explicitly differentiate between our original results and the supporting or comparative evidence drawn from cited studies. (See Lines 288-291 in the revised version)

15. Line 270: ", and residentail." => ", and residential sources."
16. Line 272-273: "showing that" => "indicating"

Response: Thanks for the constructive suggestion. All these minor issues have been corrected.

17. Line 274: "in this region": Which region is being referred to here?

Response: Thanks for the constructive comment.

We have revised the phrase "in this region" to "in Henan Province" to make the geographic scope explicit.

18. Lines 317-318: The conclusion stating a "heavy dependence on coal-based energy and energy-intensive industries" cannot be inferred solely from the contributions of these two source categories. It is recommended to remove this statement, as the subsequent analysis already addresses this point.

Response: Yes, the duplicated sentence has been removed.

19. Title of Section 3.2: Consider revising it to "Emission characteristics at the city level".

Response: Thanks for the constructive suggestion. The title of Section 3.2 had been revised as 'Emission characteristics at the city level'.

20. Line 385: "of" should be removed.

21. Line 440: "75%,)" the comma should be removed.

Response: Yes. They have been removed.

22. It is recommended to revise the title of Section 3.3.1 to "Comparison with other emission inventories".

Response: Thanks for the constructive suggestion.

We have revised the text accordingly.

23. It is recommended to display all panels of Figure 6 on a single page.

Response: Thanks for the constructive suggestion. We have revised Figure 6 to fit on a single page.

24. Line 481: "Sulfur" should be removed.

25. Line 486: "which is influenced not only influenced by": The second "influenced" should be removed.

Response: Yes. The above text has been deleted.

Reviewer #2:

We sincerely appreciate the efforts in reviewing our manuscript and providing valuable suggestions. We are particularly grateful for the recognition of the important of our research region. We have carefully considered all comments and revised the manuscript accordingly. Below are the point-to-point responses to all the comments (The comments are marked in black color and the responses are marked in dark blue color). The changes that have been made according to these responses were marked in yellow color in the highlighted copy of revised version. Note that the following line numbers are shown in the revised version.

Specific details:

1. BC is mentioned in Figure 2, but I do not see any maps of its emissions such as in Figures 4-8. What happened?

Response: Thanks for the constructive comment.
In the previous draft, due to the consideration of the number of complete maps, the BC layout map was initially not included in the main map series. Based on your suggestion, we have added the relevant BC diagram to Figure 4-8 to ensure the completeness of the discussion. The specific revisions are as follows:

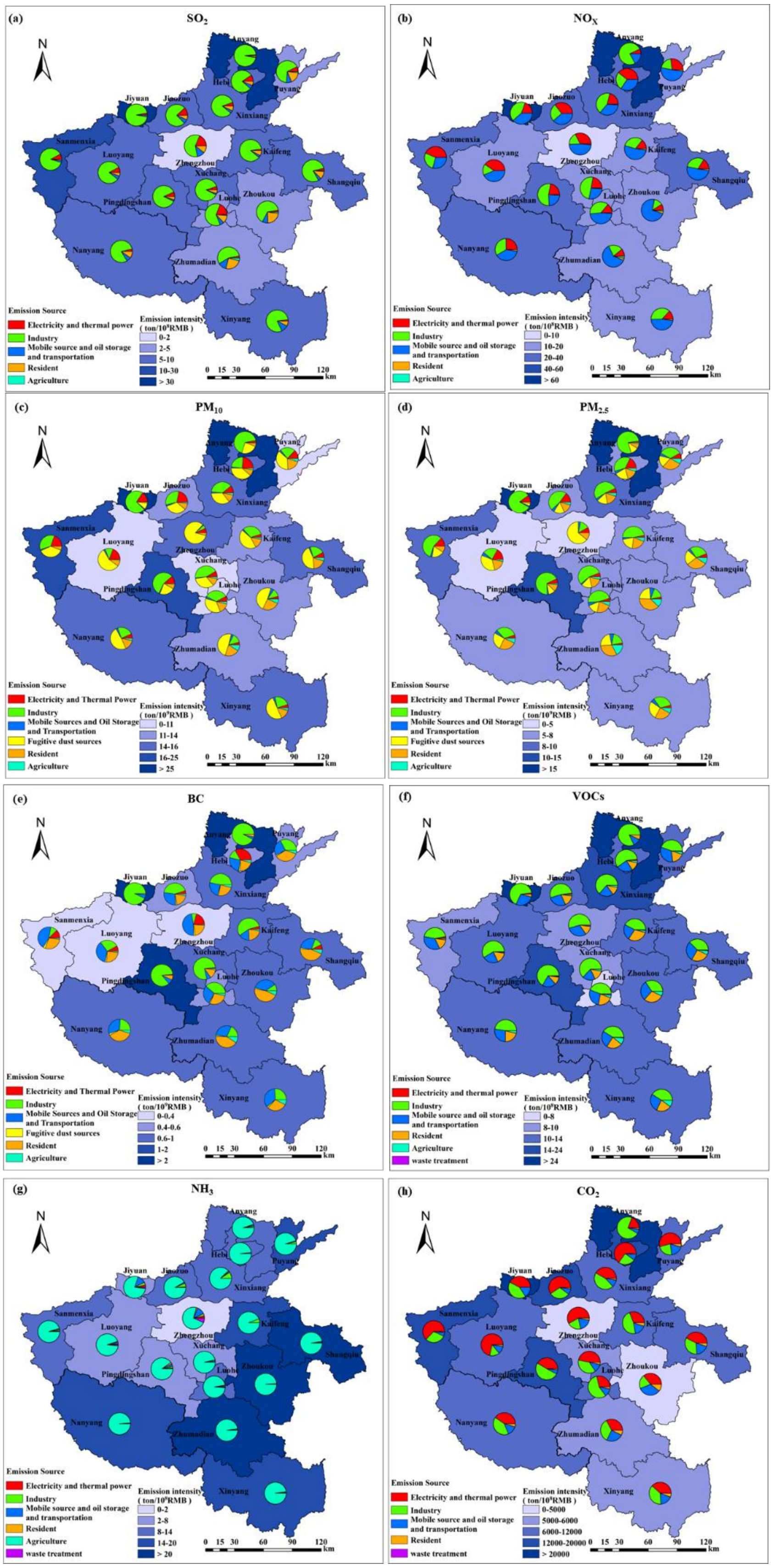


Figure 4. Maps of emission intensity per unit of GDP (ton/10^8 RMB) for (a) SO_2, (b) NOx, (c) $PM_{2.5}$, (d) VOCs, (e) NH_3, and (f) CO_2 across cities. Pie charts in maps indicate the emission source contributions of each city.

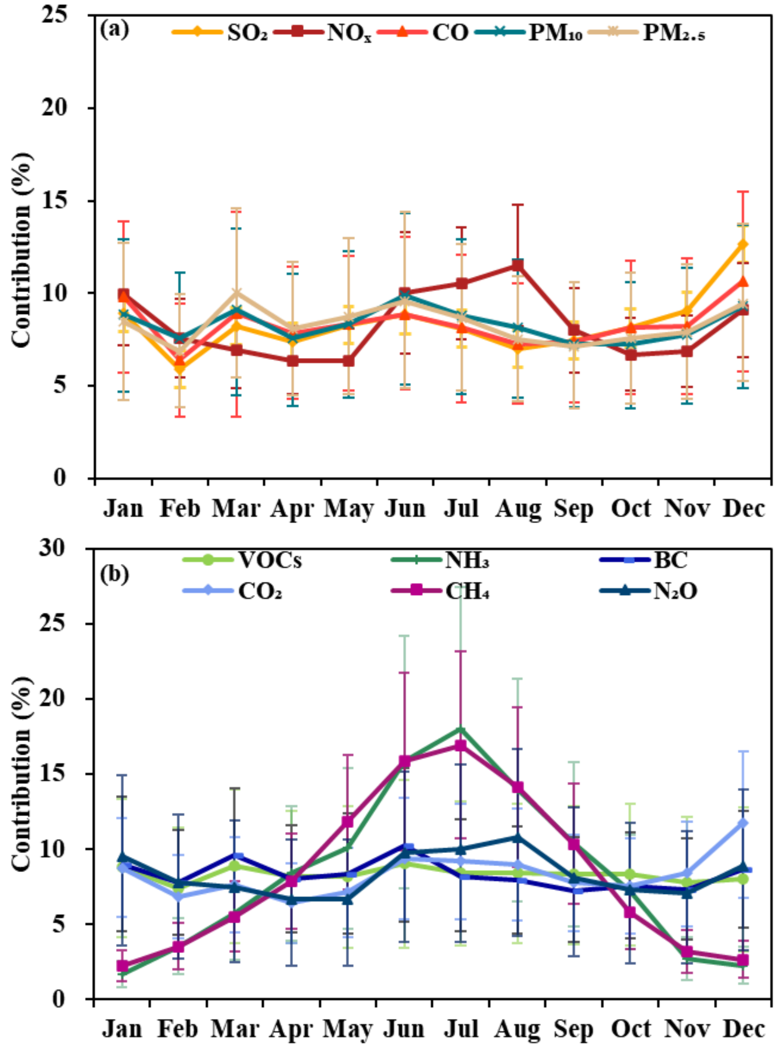


Figure 5. Monthly variation of air pollutants and GHG emissions in 2022 with uncertainty boundary.

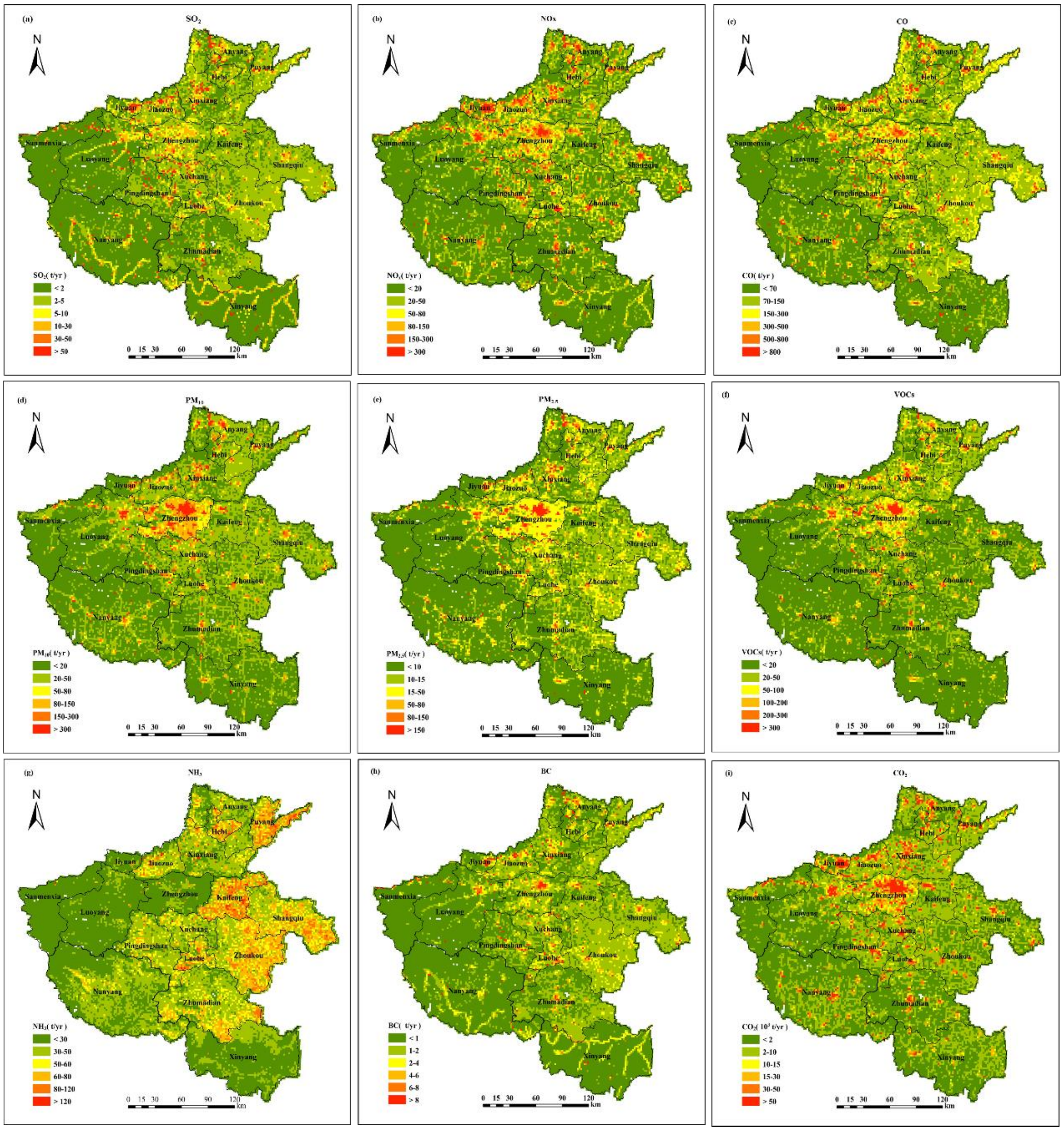


Figure 6. 3 km × 3 km gridded spatial distribution of air pollutants and GHG emissions.

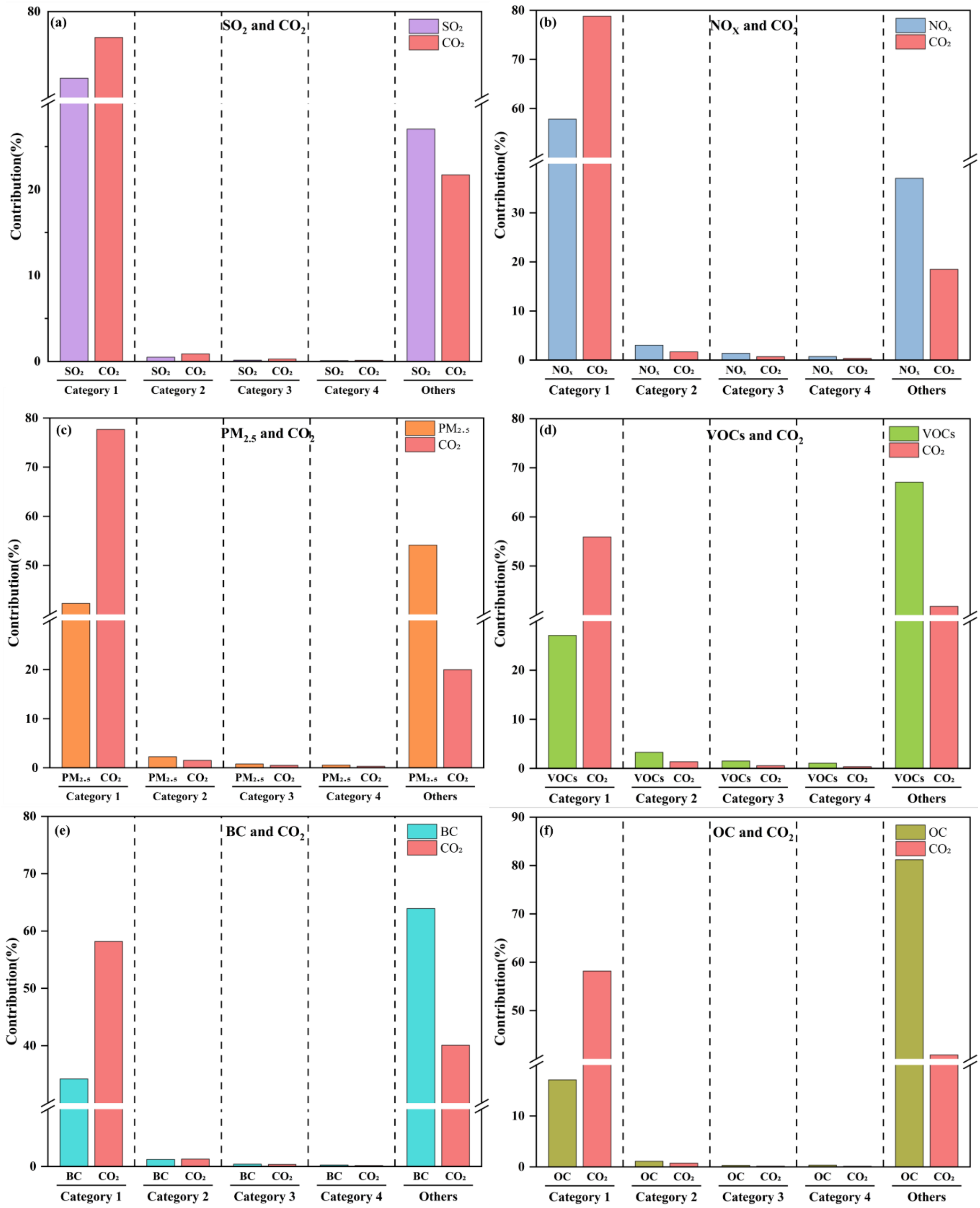


Figure 7. Emission contributions of five categories to different emission co-hotspot. (a) SO_2 and CO_2, (b) NO_X and CO_2, (c) $PM_{2.5}$ and CO_2, (d) VOCs and CO_2, (e) BC and CO_2, (f) OC and CO_2. Categories represent the grad of grid based on descending order of emission: Category 1 (top 5% grids), Category 2 (6-10%), Category 3 (11-15%), Category 4 (16-20%), and Others (bottom 80%).

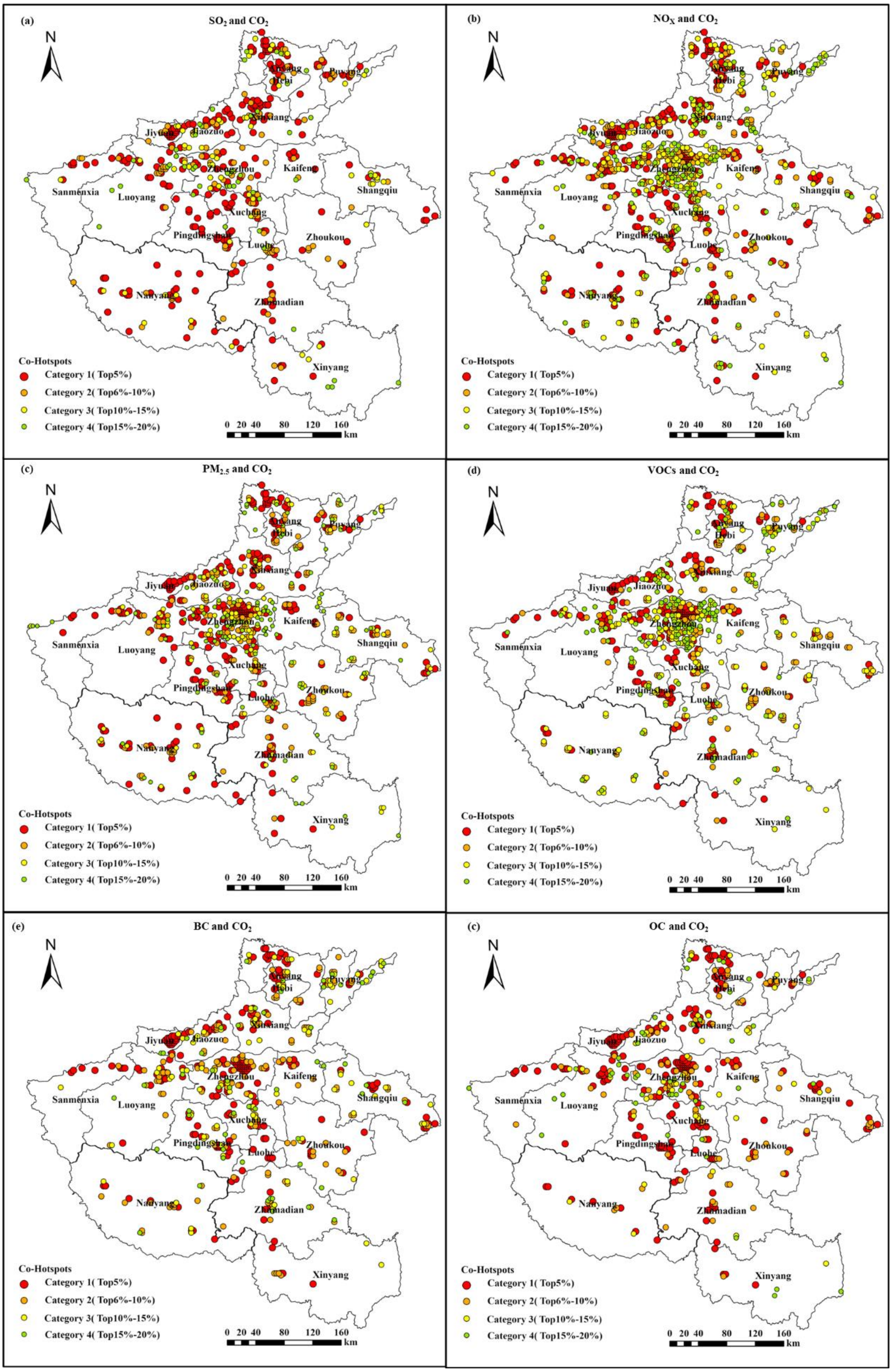


Figure 8. Spatial maps of the co-hotspots of (a) SO_2 and CO_2, (b) NOx and CO_2, (c) $PM_{2.5}$ and CO_2, (d) VOCs and CO_2, (e) BC and CO_2, and (f) OC and CO_2.

2. The equation used in 2.3.1 is not sufficiently robust to predict BC within a reasonable amount of uncertainty, since BC mass is a very small fraction of PM2.5 yet has a substantial impact on the climate system. One such issue has been moving beyond mass-based approaches to include number and particle size-based approaches. Please see work published more than 1 decade ago raising this issue such as https://doi.org/10.1029/2007JD009756 and https://doi.org/10.1002/2013JD019912, as well as more modern work at high spatial resolution which offers solutions to this issue such as https://doi.org/10.1038/s41467-022-35147-y and https://doi.org/10.1021/acs.estlett.5c00340

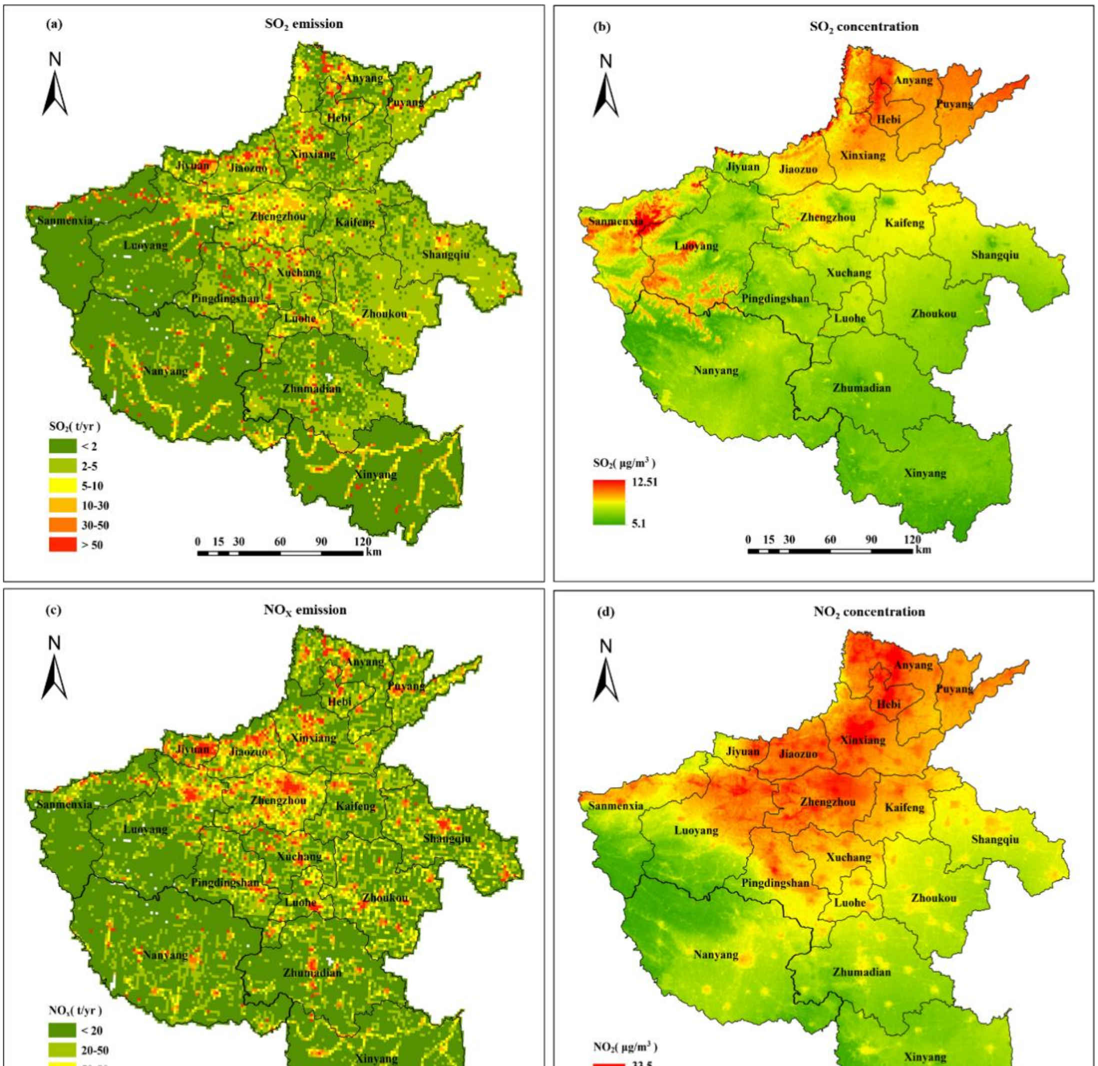
Response: Thanks for the constructive suggestion.
Thanks for the important information of relevant key literatures. We fully agree that to evaluate the climate effect and environmental concentration of BC in the atmosphere, it is necessary to go beyond the single emission factor method and consider its secondary transport process and microscopic physical properties in the atmosphere. The recommended literature (such as Chen et al., 2022; Liu et al., 2025) and the "top-down" inversion based on satellite remote sensing present in these articles have pointed out a valuable direction for our future work.
However, we would like to take this opportunity to clarify that the core objective in our study is to develop locally integrated air pollutants and co-emitted greenhouse gases emission information first based on unified source classification. There is no consideration of secondary generation at present. Given the multiple number of pollutant species involved and the corresponding requirement for distinct remote-sensing datasets, it is challenged to address all species within a single study by using emission factors and remote sensing retrieval methods both. It is hoped that the first version of integrated emissions can be provided to support subsequent air quality modeling simulations and the most urgent assessment of emission reduction potential. Therefore, based on our understanding of the articles recommended by the reviewer and the comment, we cited these two articles both in the introduction section, where we describe the "advances in high spatiotemporal resolution," and in the conclusion and outlook section, where we discuss the "need to integrate bottom-up and top-down approaches in future work."
The following revisions have been made:
In particular, the development of air pollutant emission inventories has been rapid advancements in spatial resolution and temporal distribution (Chen et al., 2024; Chen et al., 2022; Liu et al., 2025), and some studies have successfully constructed gridded data with a precision of $0.1° \times 0.1°$ or even higher (Zheng et al., 2021). (See Lines 55-58 in the revised version)
Furthermore, in order to achieve precise quantification of the synergy effect, future research needs to promote the combination of "bottom-up" inventories with "top-down" models and observation methods (such as Chen et al., 2022; Liu et al., 2025), in order to go beyond the limitations of traditional emission accounting. (See Lines 604-607 in the revised version)

References:

Chen, C., Dubovik, O., Schuster, G. L., Chin, M., Henze, D. K., Lapyonok, T., Li, Z. Q., Derimian, Y., and Zhang, Y.: Multi-angular polarimetric remote sensing to pinpoint global aerosol absorption and direct radiative forcing, Nat. Commun., 13, 10.1038/s41467-022-35147-y, 2022.
Chen, T., Xiong, Y. X., Zhao, W. D., Lin, B., He, Z. H., Tao, F. Y., and Hu, X.: Construction of Heavy-Duty Diesel Vehicle Atmospheric Pollutant Emission Inventory Based on Onboard Diagnosis Data, Atmosphere-basel, 15, 10.3390/atmos15121473, 2024.
Liu, J., Cohen, J. B., Feng, Y., Wang, S., and Qin, K.: OMI-Derived Mass- and Number-Conserved Estimation of Black Carbon Emissions, Environ. Sci. Technol. Lett., 12, 731-738, 10.1021/acs.estlett.5c00340, 2025.
Zheng, B., Cheng, J., Geng, G., Wang, X., Li, M., Shi, Q., Qi, J., Lei, Y., Zhang, Q., and He, K.: Mapping anthropogenic emissions in China at 1 km spatial resolution and its application in air quality modeling, Sci Bull, 66, 612-620, 10.1016/j.scib.2020.12.008, 2021.

3. It seems your spatial distributions of sources relating to new industrial growth (Jiaozuo, Sanmenxia, Anyang), new urban growth in in Tier 3 urban areas (i.e., Luoyang, Shangqiu), and biomass burning (many places) do not necessarily match with on-the ground observations of air pollution and multiple recently published top-down studies. Please make such comparisons in general.

Response: Thanks for the comment and this suggestion is highly valuable.
Ground-based observational data on pollutant concentrations integrate the net effects of a suite of atmospheric processes — including transport, dispersion, chemical transformation, and deposition — whereas the emission inventory developed in this study characterizes the spatial distribution of pollutants at their source locations. Consequently, these two datasets provide complementary spatial validation for one another. To further assess the reliability of the spatial distribution in our emission inventory, we conducted a spatial comparison between our 3 km × 3 km gridded emission data and the China High-resolution Air Pollutants (CHAP) concentration dataset for Henan Province in 2022, which features a 1 km × 1 km spatial resolution (Figure S3). The results indicate that, although localized atmospheric processes introduce some regional discrepancies, the high-emission zones identified by our inventory exhibit strong spatial agreement with the high-concentration areas in the CHAP dataset at the regional scale. This consistency is particularly pronounced in key areas such as major industrial zones and urban agglomerations (for additional details, see Supplementary Text 1 and Figure S3).


(a)
SO2 emission
N
Anyang
Puyang
Hebi
Xinxiang
Jiyuan
Jiaozuo
Sanmenxia
Zhengzhou
Kaifeng
Luoyang
Shangqiu
Xuchang
Pingdingshan
Luohe
Zhoukou
Nanyang
Zhumadian
Xinyang
SO2(t/yr)
< 2
2-5
5-10
10-30
30-50
> 50
0 15 30 60 90 120
km
(b)
SO2 concentration
SO2(μg/m3)
12.51
5.1
(c)
NOX emission
NOx(t/yr)
< 20
20-50
50-80
80-150
150-300
> 300
(d)
NO2 concentration
NO2(μg/m3)
33.5
12
(e)
CO emission
CO(t/yr)
< 70
70-150
150-300
300-500
500-800
> 800
(f)
CO concentration
CO(mg/m3)
1.03
0.48

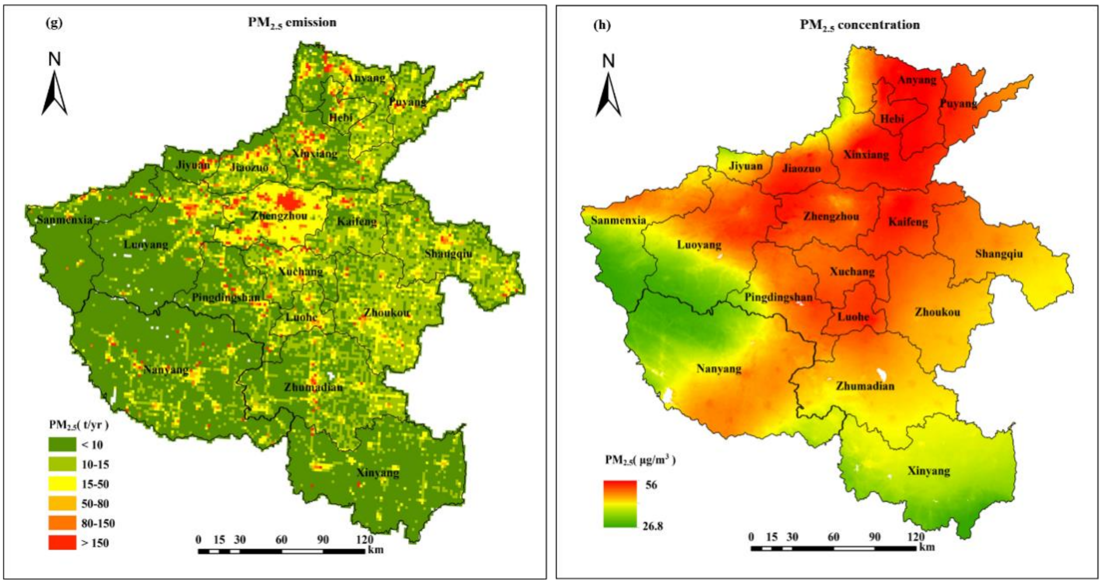


Figure S3. Side-by-side comparison of spatial distributions between 3 km × 3 km gridded emissions inventory (a, c, e, and g) and CHAP-derived (b, d, f, and h) concentrations for SO_2, NO_X, CO, and $PM_{2.5}$ in Henan Province, 2022

4. When making comparisons, it would be best to demonstrate skill of your work on a grid-by-grid and day-by-day basis between your 3km x 3km and similar 5km x 5km gridded datasets. This comparison would ideally be done at high temporal frequency, so that any uncertainties and/or biases can be clearly identified. If there are underlying reasons for these differences, they can be explored in more detail. Adding in this step is critical to demonstrate that the paper is sufficiently innovative and that the results are reasonable.

Response: Thanks for the constructive comment.
We are sorry that the original manuscript provided a 3 km × 3 km grid distribution map, but failed to clearly reflect the changes in spatial patterns over time. Based on the provided suggestions, let's explore this point, which is highly beneficial for the innovative improvement of the entire article. At the same time, in combination with the suggestion of another reviewer, we try to analyze the differences in time changes from the perspective of spatial co-hotspots. We analyzed the spatial co-hotspots for 12 months and selected four representative months - April (spring), July (summer), November (autumn), and January (winter) - to represent the co-hotspots during the seasonal changes. At the same time, during this process, we also optimized the hotspot identification method, eliminating the grids with zero emissions and those located outside the boundary of Henan Province. Some interesting phenomena were indeed discovered. The explanations for these phenomena can be found in the revised manuscript, from lines 526 to 551. Once again, we thank the reviewers for their suggestions. As shown in the figure below:

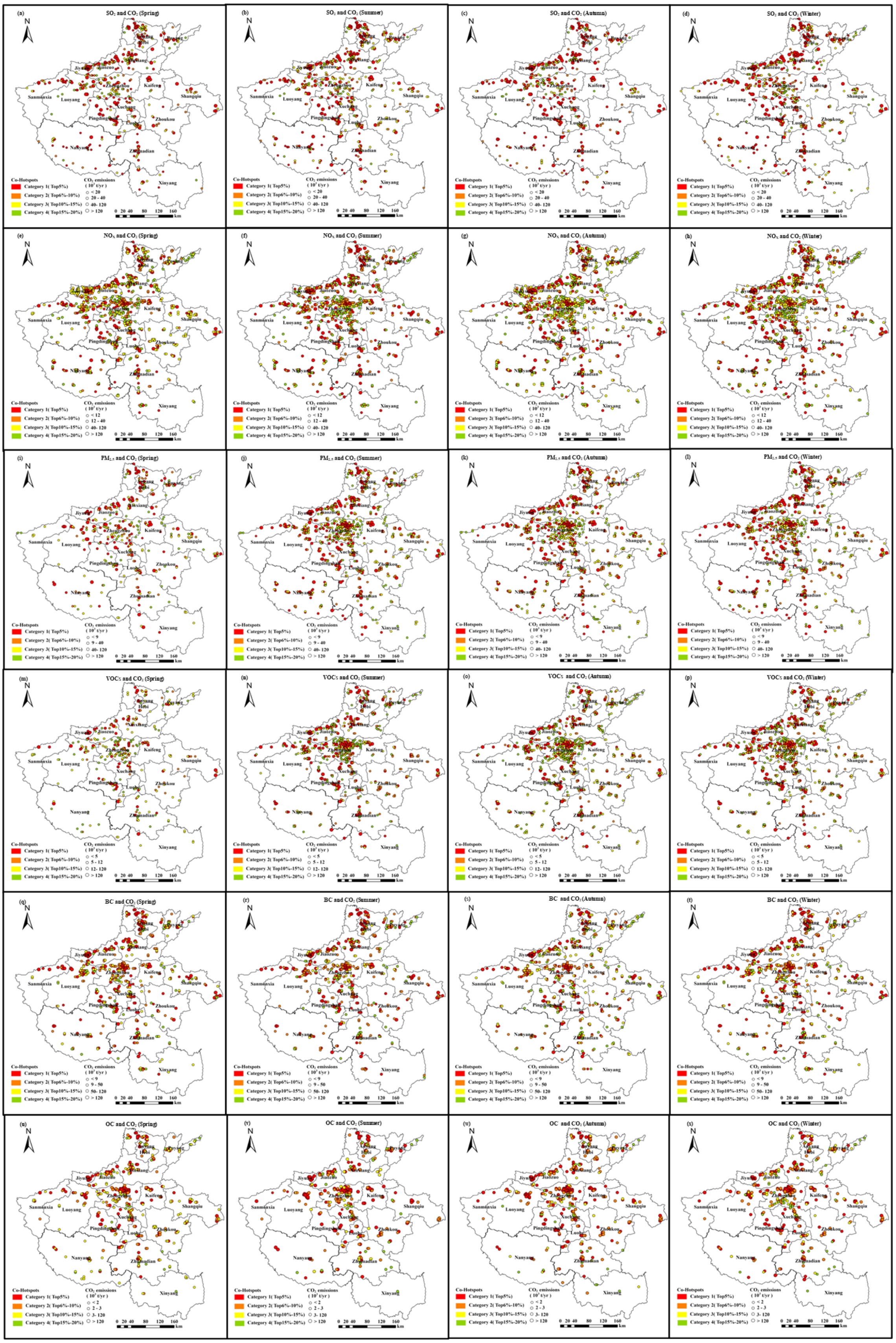


Figure S2. Seasonal spatial maps of the co-hotspots of (a-d) SO_2 and CO_2, (e-h) NO_X and CO_2, (i-l) $PM_{2.5}$ and CO_2, (m-p) VOCs and CO_2, (q-t) BC and CO_2, and (u-x) OC and CO_2 from spring to winter.