

Dear Editor and Referees:

Thanks for your comments towards improving our manuscript. We have addressed all your comments. Our [author responses \(AR\)](#) are marked in blue and the [revisions in the manuscript are marked in red](#). It should be noted the comments raised by previous referee are marked in *italic*.

Consider the following comments from one of the reviewers:

Overall, the paper has been thoroughly revised in response to the comments; however, several comments appear to require further clarification. The main concerns regarding the revised manuscript are as follows:

AR: Thanks for your comments and we have addressed all your comments as follows.

1. Regarding the previous comment about the relationship between studies based on long-term observational data from the Beijing site and this study dealing with data observed across many regions in China, the essence of the question pertains to the differences in major conclusions between this study and such previous research. The point was not merely to indicate that long-term observational data from a single site, like Beijing, is easily accessible. Additional clarification on this matter appears necessary.

AR: Thanks for your comments and we have extended this part by comparing our results with most recent long-term observations (Line 331-337 in the revised manuscript and hereafter).

Long-term observational studies across China have widely reported decreases in BC concentrations over various periods. For example, BC concentrations in Beijing declined at an average rate of $0.19 \mu\text{g m}^{-3}$ per year between 2013 and 2022 (Xie et al., 2025). An annual decrease of $0.10 \mu\text{g m}^{-3}$ was observed in Nanjing from 2014 to 2021 (Abulimiti et al., 2025), while a trend of $-0.12 \mu\text{g m}^{-3}$ per year was recorded at Mt. Lushan from 2008 to 2022 (Liu et al., 2025). These single-site observations consistently highlight the effectiveness of air pollution control policies in reducing BC concentrations. Our study reinforces these findings from a broader spatial perspective, confirming that the observed reduction in BC levels can be attributed to the implementation of national regulatory measures, such as the Clean Air Action Plan and the Three-Year Action Plan.

2. A clear answer has not been provided regarding the previous question about the contributions of solid fuel combustion (eBC_{sf}) and liquid fossil fuel combustion (eBC_{lf}). Sufficient responses and their incorporation into the paper are essential.

AR: Thanks for your comments and we have evaluated the accuracy of BC source apportionment results, and the details can be found in AR3 below.

3. Additional review is needed regarding the author's response to the following previously asked question:

Furthermore, many studies have explained the emission sources of black carbon or elemental carbon through carbon isotope analysis, determining contributions from solid fuel and liquid fossil fuel combustion. I believe the most critical

aspect for publication of this study would be to validate the calculation results by comparing them with such previous research findings and demonstrating consistent results. For example, I suggest presenting comparisons between this study and carbon isotope analysis-based contribution assessments for specific locations and time periods in the main text.

Examining the author's response (Fig. S4), the aethalometer-based analysis results of this study show very large differences from dual carbon isotopes analysis results depending on time and location. The differences are too substantial to be explained simply by correlation coefficients (r). Therefore, the distinction between solid fuel combustion (eBC_{sf}) and liquid fossil fuel combustion (eBC_{lf}) based on aethalometer data in this study remains uncertain, and this reviewer finds it difficult to fully trust the results of this study. More detailed explanations are absolutely necessary regarding how the characteristics of aerosols from solid fuel and liquid fossil fuel combustion differ, and how and why wavelength-dependent differences in aethalometer observational data appear.

AR: Thanks for your comments and we have extended the explanation related to your concerns as follows (Line 156-167).

The Aethalometer model operates on the assumption that light-absorbing aerosols originate predominantly from two source types (Sandradewi et al., 2008), e.g., liquid fuel combustion and solid fuel combustion, which are characterized by distinct Absorption Ångström Exponent (AAE) values. Aerosols derived from solid fuel combustion (AAE_{sf}) generally exhibit higher and more variable AAE values compared to those from liquid fuel combustion (AAE_{lf}). The elevated AAE_{sf} values are attributed to the presence of significant amounts of organic carbon species, which absorb strongly in the ultraviolet and lower visible wavelength ranges (Sandradewi et al., 2008). In contrast, the lower AAE_{lf} values result from the dominant contribution BC, which absorbs light broadly across the visible spectrum. These source-specific AAE characteristics are consistent with laboratory combustion experiments, which also report higher AAE values for solid fuel emissions and lower values for liquid fuel emissions (Olson et al., 2015). Consequently, measured ambient AAE values can serve as an indicator of dominant BC sources, e.g., lower AAE values are typically associated with environments strongly influenced by liquid fuel combustion, such as road tunnels (Blanco-Alegre et al., 2020).

In addition to the above question, clear discussion of the accuracy of the methodology for distinguishing contributions of solid fuel combustion (eBC_{sf}) and liquid fossil fuel combustion (eBC_{lf}) is crucial for deriving the main results of this study, and therefore, the related content must be sufficiently explained and validated.

AR: Thanks for your comments and we have used the error propagation to discuss the accuracy of the BC source

apportionment results as follows (Line 213-223).

Due to the unknown of “real” BC source apportionment results, the accuracy of the Aethalometer model was assessed through error propagation (Martinsson et al., 2017; Zheng et al., 2020). The primary sources of uncertainty considered were the absorption coefficient and the AAE values. The uncertainty of b_{abs} was reported as 5% (Hansen, 2005) and this value was adopted for this study. For the uncertainty of AAE values, their uncertainties were defined as the difference between the values derived from the percentile method and the isotope-constrained values from Zotter et al. (2017), e.g., 0.90 for AAE_{lf} and 1.68 for AAE_{wb} . This approach was justified as the mean optimal AAE_{lf} and AAE_{sf} values obtained here (0.90 ± 0.05 and 1.70 ± 0.23 , respectively) were closely aligned with the literature values. The error propagation analysis revealed station-specific relative uncertainties in the range of 19.8%–53.8% for eBC_{sf} and 23.7%–54.0% for eBC_{lf} (**Table S3**). The reported uncertainties here were comparable to previous studies, e.g., 41% for fossil fuel combustion and 42% for wood burning estimated by Martinsson et al. (2017). Despite the relatively high estimated uncertainty, the spatial and temporal patterns of BC sources resolved from the Aethalometer model were believed reliable here.

Table S3 Relative uncertainty of the Aethalometer model in each station using the propagation of errors

Code	eBC_{sf}	eBC_{lf}	Code	eBC_{sf}	eBC_{lf}
UR	32.0%	35.3%	XA	30.7%	24.4%
TZ	53.8%	25.6%	XF	30.8%	27.5%
HM	41.8%	47.2%	WH	30.4%	35.6%
ZR	32.6%	33.0%	SX	37.7%	34.3%
YS	25.0%	27.4%	PD	35.2%	27.2%
LFS	19.8%	40.8%	LA	27.6%	54.0%
XL	29.0%	23.7%	HAZ	53.8%	33.8%
CC	21.6%	51.8%	DH	37.3%	34.7%
AS	24.3%	30.0%	LUS	23.9%	25.4%
SY	24.8%	36.5%	HJ	39.8%	29.6%
BX	25.3%	47.4%	DG	35.4%	27.3%
FS	22.7%	37.0%	NN	30.4%	45.8%
SDZ	20.7%	29.4%	PY	44.3%	45.4%
HUM	21.6%	32.6%	SZ	49.1%	48.6%
LAS	24.2%	27.8%	HZ	52.7%	31.1%
XGL	28.6%	23.9%	LS	33.2%	39.9%
ZZ	33.2%	33.5%	GL	30.5%	30.8%

5. One of the original findings claimed by the authors is the factor contributing to black carbon reduction in rural and background areas of China. As the authors argue, this is clearly distinctive from previous studies that primarily reported black carbon concentration variation characteristics in urban areas. However, overall, the main results of this study largely align with results already reported in previous studies, and the authors explain their research results in various places through citations of such previous research. It is required that this study minimize discussion of similar results compared to previous studies and provide sufficient and detailed explanations of the distinctive factors of this study. For example, regarding the conclusion of the following sentence, it is necessary to provide other observational data or emissions demonstrating reduced coal and biomass use in rural and baseline areas: The results demonstrate that the decrease in BC concentrations is mainly attributed to reduced emissions from both coal and biomass combustion in rural and baseline areas. These findings represent novel conclusions not previously reported in earlier research. (L480-482).

AR: Thanks for your comments and we have provided the emission data to support the novelty of this study, and we have also extended the discussions as follows (Line 525-529).

Evidence from BC emission data further support these findings. As shown in **Fig. S13**, BC emissions derived from the MEIC inventory (Geng et al., 2024) for biomass burning, solid fuel combustion, and liquid fuel combustion exhibited widespread reductions across most regions of eastern China between 2008 and 2020. Temporally, emissions from these sources showed consistent decreasing trends from 2008 to 2020 at rural, baseline, and urban stations. For example, BC emissions from rural residential energy consumption in northern China decreased from 0.85 Tg in 2010 to 0.55 Tg in 2020 (Zhang et al., 2023).

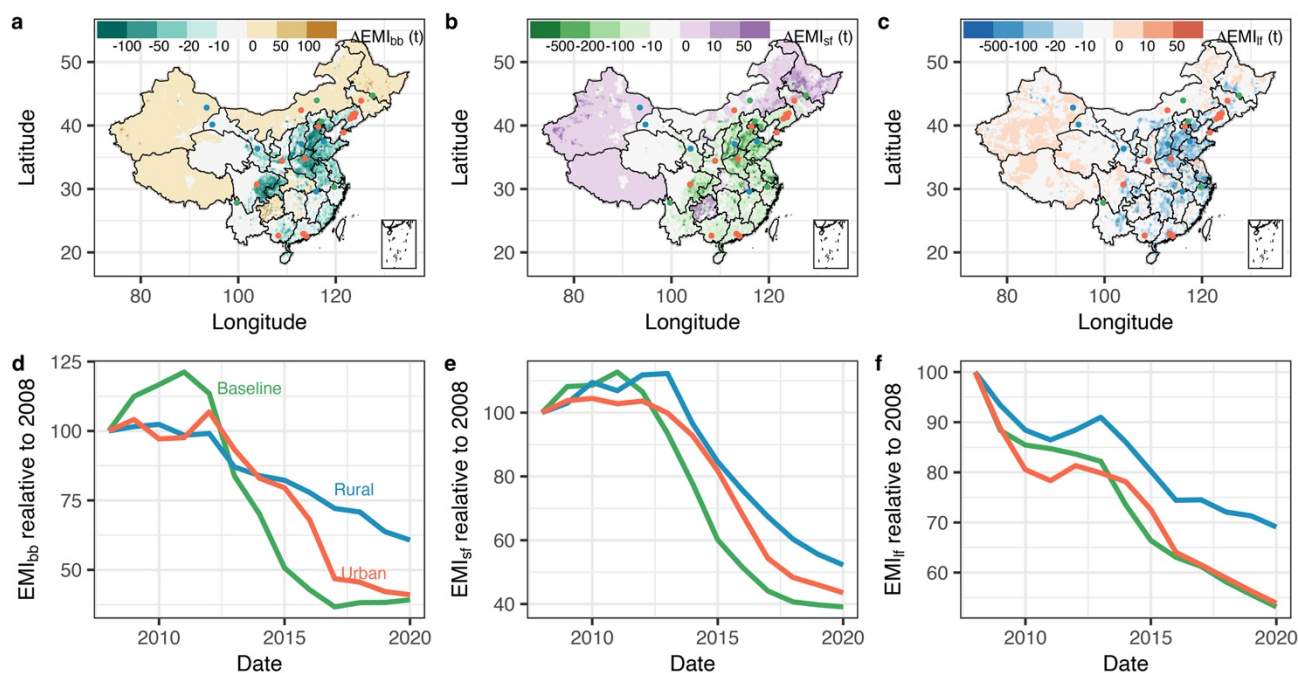


Figure S13 Changes in emission reduction of biomass burning (a), solid fuel combustion (b), and liquid fuel combustion (c) between 2008 and 2020 and their time series of BC emission amounts from different fuels compared to 2008 for different station types (d–f).

6. Additionally, it would be helpful to provide a clear definition of how "baseline area" is defined in the above sentence.

AR: Thanks for your comments. We have provided the description about the baseline station as follows (Line 98–101).

Baseline stations are established in areas remote from strong emission sources and human activities to monitor the long-range transport of BC aerosols and their natural emissions. For instance, the Lin'an (LA) baseline station is situated at an altitude of 139 m a.s.l. in Zhejiang province, positioned approximately 150 km northeast of Shanghai and 50 km west of Hangzhou city.

7. A response to this notification from the journal is required: Figures 1, 2, 7 and S1 may contain a territory that is disputed according to the United Nations. If and when the manuscript is accepted for final revised publication, you will be asked to choose one of the following options: (a) you could remove the disputed territory from the map and submit new figure files, or (b) we could add a statement that some figures contain disputed territories.

AR: Thanks for your notification and we choose **option b** that you could add a Disclaimer.