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DATA DESCRIPTOR

Optical emission inventory of carbonaceous aerosol from the residential sector in China

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The residential sector in China is a major contributor to light-absorbing carbonaceous aerosols, including black carbon and brown carbon, which have significant impacts on climate change. This study developed a province-level inventory of optical emissions of carbonaceous aerosols from the residential sector in China from 1960 to 2019. The inventory was based on activity data from the PKU-GEMS database and absorption emission factors derived from laboratory-based combustion experiments, which reduced uncertainties associated with traditional mass-based methods relying on mass absorption efficiency. The dataset provided annual light absorption at the ultraviolet region (370 nm) and the infrared region (880 nm), offering valuable insights into the spatial and temporal trends of optical emissions from residential carbonaceous aerosols. This inventory would support more accurate evaluations of radiative forcing impacts of carbonaceous aerosols.

Background & Summary

Carbonaceous aerosols, consisting of black carbon (BC) and organic carbon (OC), play a significant but still poorly understood role in influencing the Earth's climate system^{1–3}. BC is a major contributor to atmospheric warming, exerting a significant radiative effect rivaling that of CO₂². Meanwhile, emerging evidence indicated that some OC, referred to as “brown carbon” (BrC), also contributes substantially to solar radiation absorption at specific wavelengths, accounting for around 19% of the total light absorption by anthropogenic aerosols⁴.

The residential sector is a significant source of BC and BrC emissions, accounting for approximately 49% of BC emissions and 71% of BrC emissions in China, respectively^{5,6}, and playing significant roles in climate change. Notably, the residential sector is also a major source of uncertainty in emission inventories due to uncontrolled and highly variable combustion conditions^{7–9}. For example, BrC emission factors from residential solid fuel combustion vary widely, from 0.098 to 20 g/kg, reflecting substantial uncertainties and highlighting broader challenges in accurately evaluating the climate impacts of carbonaceous aerosols^{10–13}. Several mass inventories have been developed in China over the past two decades using both bottom-up and top-down methods^{14–17}. These inventories provide critical data for calculating the direct radiative forcing of carbonaceous aerosols. However, significant uncertainties remain, particularly for BrC, as its complex composition makes it difficult to separate from other aerosol components, making accurate mass quantification nearly impossible¹⁸.

Although several methods have been developed to characterize BC and BrC, there remains controversy about definitions and quantification methodologies, particularly for BrC. At present, no universally accepted “gold standard” has been established^{13,19,20}. Typically, the mass of BC and BrC is estimated with the ratio of light absorption to mass absorption efficiency (MAE). However, reported MAE values vary significantly, ranging from 0.16 to 50 m²/g, depending on factors such as fuel type, particle properties, and experimental methods^{21–25}. Additionally, MAE calculations often require corrections for multiple scattering and shadowing effects, but the inconsistency of parameters and the variability across studies further complicate the assessment^{21–24}.

Given the uncertainty therein, it is expected to directly obtain the optical absorption ability of aerosols by using absorption emission factors (AEFs), defined as the absorption cross-section released into the atmosphere per kilogram of fuel consumed (m²/kg), which serves as a measure of the emission potential of the absorption

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coefficient for BrC or BC. Unlike existing optical emission inventories specifically for BrC^{26,27}, this study did not attempt to separate the contributions of BC and BrC, avoiding the additional uncertainties associated with distinguishing between BC and BrC absorption. The separation of BrC contribution is commonly based on the assumption that the absorption Ångström exponent (AAE) for BC is fixed at 1.0, while experimental studies have shown that the AAE for BC can vary between 0.9 and 1.5^{28,29}. By combining AEFs with PKU-GEMS data, a national-scale optical emission inventory (1960–2019) was developed. This dataset reduces uncertainties in traditional methods and provides a foundation for assessing the climate impacts of residential carbonaceous aerosols in China. Furthermore, it could serve as an important basis for formulating targeted mitigation strategies in China.

Methods and Data Source

Laboratory-based combustion emission experiments. A total of 29 types of solid fuels were tested in the laboratory system (Figure S1), comprising 14 types of coal with varying geological maturities and forms, 11 types of biomass fuels (including crop residues and woody fuels), and 4 types of biomass pellet fuels produced from straw and wood. These typical fuels were selected to represent the most commonly used solid fuels across China, ensuring coverage of regional variations in fuel types and usage patterns. Fuel properties were provided in Table S1 in the *Supporting Information* (SI). Combustion experiments were conducted using three stoves, including a traditional stove commonly used in rural China, an improved stove, and a self-made three-stone stove designed to simulate open combustion emissions. Detailed images and structural specifications of the stoves are shown in Figure S2.

The combustion process was conducted following established protocols^{30–32}. Briefly, approximately 1 kg of woody fuel or 200–400 g of crop residues were used per test, lasting 20–40 minutes. For biomass pellets, the fuel was supplied through an automatic feeding system at a constant rate of 3 kg/h, with the entire combustion cycle lasting approximately 1–2 hours. For coal fuels, approximately 1.5 kg of chunk coal or two briquettes were initially used, followed by the addition of ~1 kg of test coal or one briquette as the initial combustion began to fade. Each coal combustion test lasted 4–7 hours. All combustion experiments were conducted in duplicate or triplicate to ensure reproducibility.

Real-time gaseous pollutants, including CO, CO₂, and total hydrocarbons (THC), were monitored online (Thermo Fisher Scientific Inc.). Additionally, PM_{2.5} (particulate matter with an aerodynamic diameter ≤2.5 μm) was collected on quartz fiber filters (QFFs) at a flow rate of 16.7 L/min.

Light absorption measurement. In this study, the light absorption of carbonaceous aerosols collected on QFFs was measured at wavelengths of 370 nm and 880 nm using a SootScan OT21 optical transmissometer (Magee Scientific, USA). The light attenuation (ATN) caused by particles was determined by comparing the transmission intensity of a blank filter to that of a loaded filter. To minimize potential biases from the properties of the reference filter, field blank filters were used as references during the analysis. Additionally, the OT21 instrument was calibrated before the measurements using a NIST-traceable neutral density filter.

Absorption emission factors. Emission factors, defined as the mass (grams) of the pollutant emitted per mass (kilograms) of fuel consumed, were calculated using the carbon mass-balance method¹³. This method assumes that the total carbon in the fuel is primarily released into the gaseous phase as CO₂, CO, and THC, as well as into the particulate phase as carbon. The carbon mass balance can be expressed as:

$$C_f - C_a = C_{C-CO_2} + C_{C-CO} + C_{C-THC} + C_{C-PM}$$

where C_f and C_a represent the carbon mass in the fuel and ash, respectively. C_{C-CO_2} , C_{C-CO} , C_{C-THC} , and C_{C-PM} are carbon released as CO₂, CO, THC, and PM, respectively. Here, C_{C-PM} refers to the mass of OC and elemental carbon (EC), which were measured using a DRI Model 2001 thermal/optical carbon analyzer (Atmoslytic Inc., Calabasas, CA) following the Interagency Monitoring of Protected Visual Environments (IMPROVE) program.

The Product of Incomplete Combustion (PIC) is defined as:

$$PIC = (C_{C-CO} + C_{C-THC} + C_{C-PM}) / (C_{C-CO_2})$$

The EF of carbon in CO₂ per unit mass of burnt fuel (M) can then be calculated as:

$$EF_{C-CO_2} = (C_f - C_a) / [(1 + PIC) \times M]$$

To calculate the EF of CO₂ (g/kg), a conversion factor (f_{CO_2}) is used to account for the relationship between the carbon mass in CO₂ and the total mass of CO₂:

$$EF_{CO_2} = EF_{C-CO_2} \times f_{CO_2} = (C_f - C_a) \times f_{CO_2} / [(1 + PIC) \times M]$$

EFs of other components (EF_X) can be calculated based on the EF of CO₂ and mass concentration ratios of these components to CO₂:

$$EF_X = EF_{CO_2} \times C_X / C_{CO_2}$$

where C_X and C_{CO_2} are mass concentrations (g/m³) of component X and CO₂, respectively.

In the present study, the AEF (m²/kg), which represents the light absorption cross-section (m²) of carbonaceous component per mass (kilograms) of fuel consumed, was performed using the following equation:

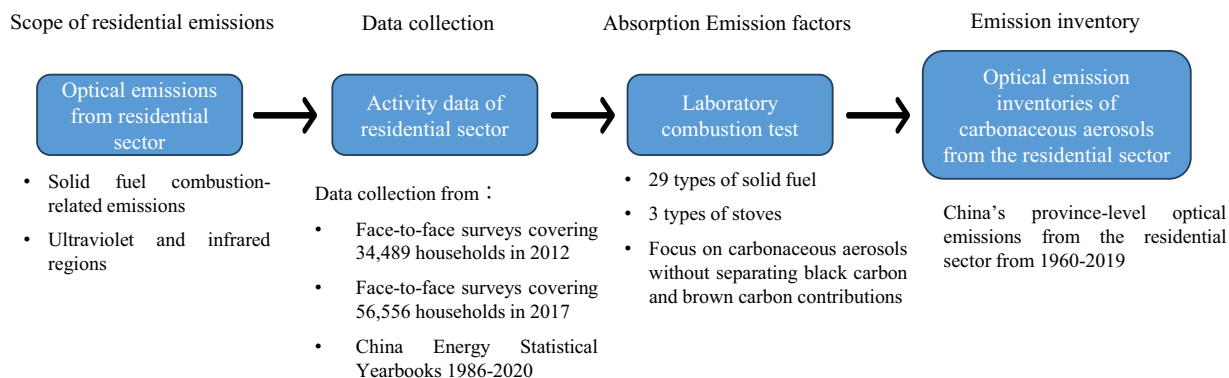


Fig. 1 Construction process of China's provincial optical emission inventory for the residential sector. AEFs were determined at specific wavelengths (370 nm in the UV region and 880 nm in the IR region). The flow chart presents a detailed, step-by-step outline of data processing and analytical methodology.

$$AEF = b_{abs} \times 10^{-6} \times EF_{CO_2}/C_{CO_2}$$

where b_{abs} (M/m), the intrinsic absorption coefficient, is influenced by various factors and requires correction using the empirical factors C and R(ATN), as expressed in the following equation:

$$b_{abs} = b_{ATN}/[C \times R(ATN)] = ATN \times A/[V \times C \times R(ATN)]$$

where ATN is the optical attenuation measured from the SootScan OT21 (for 370 nm and 880 nm), and A and V are the effective filter area (mm^2) and the volume of air sampled (m^3), respectively. The empirical factors C and R(ATN) were applied to account for multiple scattering and the shadowing effect, respectively. Further details on the calculations are provided in Section S1 of the SI.

Besides, modified combustion efficiency (MCE), which reflects combustion condition, is provided in Table S2 in the SI and was calculated using the following equation:

$$MCE = CO_2/(CO_2 + CO)$$

where the CO_2 and CO are excess molar mixing ratios of CO_2 and CO, respectively.

Estimation of optical emissions. Figure 1 shows the overall structural design of the optical emission estimation methodology employed in this study. The bottom-up inventory was developed by summing the products of individual activity levels and AEFs³³. Specifically, optical emissions from individual activities were estimated using AEFs (Table S3) combined with activity level data, as described by the following equation:

$$Optical_{K,\lambda} = \sum \sum A_{i,k} \times T_{i,j,k} \times AEF_{i,j,k,\lambda}$$

where $optical_{k,\lambda}$ represents the total optical emissions from residential sources in year k at the wavelength of λ ; $A_{i,k}$ is the activity level of the i th source in year k; $T_{i,j,k}$ denotes the proportion of the j th technological category for the i th source in year k; $AEF_{i,j,k,\lambda}$ is the absorption emission factor of the carbonaceous aerosols for the corresponding technological category of the i th source in year k at the wavelength of λ .

Data source. The dataset for fuel energy consumption in both rural and urban areas was derived from the Global Emission Monitoring System (GEMS, accessible at <https://gems.pku.edu.cn/>). For rural areas, energy consumption data were obtained through two comprehensive national surveys conducted in 2012 and 2017^{34,35}. These surveys provided detailed information on rural residential energy use, a significant source of carbonaceous aerosols, with daily biomass fuel consumption data collected from 34,489 households in 2012 and 56,556 households in 2017. Data for years not covered by the surveys were estimated through linear extrapolation, using per capita energy consumption data from the surveyed years³⁶. Residential straw consumption was categorized by crop type, including corn, wheat, rice, sugarcane, soybeans, cotton, and other crops, based on crop production data from the Food and Agriculture Organization of the United Nations³⁷ and the straw-to-grain mass ratios for different crops³⁸. Biomass pellet consumption was further classified into wood pellets and straw pellets, depending on the usage of firewood, branches, and straw. Energy consumption for unmonitored years was extrapolated from historical biomass consumption trends reported by Xu *et al.*³⁹. For urban households, coal consumption data were derived from province-level energy statistics compiled by Zhang *et al.*⁴⁰ based on the China Energy Statistical Yearbooks⁴¹. It is generally assumed that urban residents have limited or no access to biomass fuels⁴².

Data Records

The dataset is available at Figshare (<https://doi.org/10.6084/m9.figshare.28769657>)⁴³, including emission factor dataset named “Absorption Emission Factor”, which measured in the laboratory system with explanations provided in the SI, and province-level datasets of optical emissions at 370 nm (UV) and 880 nm (IR) spanning 1960 to 2019, covering 31 provinces in China, excluding Hong Kong, Macao, and Taiwan which are named “Optical Emission_UV Region_1960–2019” and “Optical Emission_IR Region_1960–2019”, respectively. Besides the total emission, those for different fuels, such as anthracite, bituminous coal, crop residues, woody fuel, and biomass pellets, are also estimated and available in the datasets.

Technical Validation

Uncertainty analysis. A Monte Carlo simulation was conducted 10,000 times ($n = 10,000$) to quantify uncertainties in the compiled inventory arising from activity data and AEFs. The calculated uncertainties are evaluated at the level of individual source types within each province. The parameters followed uniform distributions, with assumed coefficients of variation set at 10% for residential biomass fuels and 5% for coal^{44,45}. AEF were sampled from a lognormal distribution, with the mean and standard deviation derived from the combustion tests. For emission calculations, arithmetic means of AEF were used instead of geometric means to enhance accuracy⁸.

The resulting emission distributions for optical emissions were characterized using their 2.5th, 50th (median), and 97.5th percentiles, representing a 95% confidence interval. For the year 2019, the uncertainty in total residential emissions was calculated to range from -14% to $+21\%$ for the emissions at the UV region, and from -10% to $+12\%$ for the emissions at the IR region.

Comparison with existing estimates. Since the uncertainty analysis did not provide absolute truth values, we compared our results with other emission inventories for validation. However, to the best of our knowledge, there is no comprehensive optical emissions inventory specifically for carbonaceous aerosols. As a result, we conducted a basic comparison with existing inventories of the absorption cross-section of BrC (ACS_{BrC}). Based on the assumption of an AAE of 1 (detailed calculation methods are provided in the SI), we estimated ACS_{BrC} emissions, with results for specific years listed in Tables S4 and S5.

Tian *et al.*²⁶ estimated total annual ACS_{BrC} emissions in 2015 from biomass combustion and residential coal combustion to be 4194 Gm^2 (uncertainty: -33.2% to $+41.2\%$) and 615 Gm^2 (uncertainty: -39.3% to $+40.1\%$), respectively²⁶. In contrast, our results were approximately 30% lower, with total annual ACS_{BrC} emissions from residential biomass and coal combustion estimated at 3762 Gm^2 and 440 Gm^2 , respectively. It is important to note that the biomass emissions reported by Tian *et al.* (2015) included contributions from various sources, such as agricultural burning and industrial biomass use, which could partly explain the higher values reported in their study. On the other hand, our study demonstrated relatively low uncertainty levels, with the 95% confidence intervals for coal emissions and biomass emissions of $[-12\%, +14\%]$ and $[-6.9\%, +8.0\%]$, respectively, due to the higher precision and accuracy of the PKU-GEMS activity data^{46–48}. Additionally, Yan *et al.*²⁷ reported significantly higher values, suggesting that residential coal combustion emissions were 1930 Gm^2 in 2017, with total uncertainty of -57.6% to $+57.6\%$ ²⁷.

Limitations. This study developed a province-level inventory of optical emissions from carbonaceous aerosols in the residential sector in China, providing important insights for assessing their climate impacts and supporting mitigation strategies. However, several limitations need to be addressed. First, while the AEFs were derived from controlled laboratory experiments, combustion conditions in the real world and user behavior affect combustion processes, leading to differences between lab- and field-based emissions. Incorporating field measurements under diverse conditions would enhance the inventory’s accuracy and better reflect real-world scenarios by capturing the variability in fuel properties, stove technologies, and combustion practices. Second, while spatial and temporal variations in activity data (e.g., fuel consumption and stove technologies) were included, uniform AEFs were applied for each fuel type and stove combination. This approach overlooked region-specific differences in combustion characteristics, potentially reducing precision. Future studies should aim to refine EFs by integrating region-specific field data and considering the variability in combustion processes to improve the accuracy of the inventory. Additionally, uncertainties in historical activity data, particularly for earlier decades, were considerable, due to limited regional statistics and dependence on extrapolation. Improving the spatial and temporal resolution of activity data and refining emission factors specific to provinces and years would help address these uncertainties. Future work should focus on reducing these uncertainties and improving the reliability of the dataset to better evaluate residential emissions and their climate impacts.

Code availability

No custom code was used for this study.

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Author contributions

L.Z., G.S. and S.T. led the project. J.L. compiled the activity data. L.Z., Y.L., and X.L. conducted the experiment. L.Z. wrote the manuscript, and all authors provided revisions to the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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