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Supplementary information

Toxic potency-adjusted control of air pollution for solid fuel combustion

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Supplementary Materials for

Toxic potency-adjusted control of air pollution for solid fuel combustion

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Supplementary Notes

1. Studied Power Plant Units

China imposed the emission standard (GB 13223-2011) in 2012 to limit PM emissions from CFPPs to 30 mg/m^3 at the standard oxygen level, whose emission limit were fully met by all CFPPs by the end of 2015[1, 2]. To further reduce PM emissions, China enforced the toughest-ever "ultralow emission" (ULE) standard for CFPPs in 2014, which limits PM to 10 mg/m^3. 72.3% of national coal-fired units had met the ULE standard in China by the end of 2017[3], while the ratio reached to approximately 90% across the whole nation in 2019[4]. There were 1510 CFPPs in northern and eastern China in 2017, accounting for 60% of total capacity across the nation[3]. There were 1510 CFPPs in northern and eastern China in 2017, accounting for 60% of total capacity across the nation[3]. Owing to severe air pollution problem, the ULE retrofits has performed most frequently in north and east regions[4]. The compliance rate with the ULE standards for CFPPs in northern and eastern regions were 70% and 93%, respectively by 2017[3]. Aiming to meet the emission standard, all the CFPPs in northern and eastern China were employed with desulfurization devices (including limestone-gypsum wet flue gas desulphurization (FGD), seawater FGD, and semi-dry FGD technologies), denitrification devices (including selective catalytic reduction, SCR; selective non-catalytic reduction, SNCR; and SCR-SNCR combined denitrification technologies), and dedust devices (including electrostatic precipitator, ESP; fabric baghouse, FAB; and wet electrostatic precipitator, WESP) to remove SO_2 to below 100 mg/m^3, NO_x to below 100 mg/m^3, and PM to below 30 mg/m^3 before retrofitting with ULE technologies, respectively[2, 3, 5]. The tested units combined several distinct configurations for ULE technologies, combining these removal systems, are representative of the CFPPs across the whole nation.

In order to investigate the emission characteristics and toxic potency of PMs emitted from CFPPs, field sampling in typical units of CFPPs was conducted in northern and eastern China from December 2017 to May 2018. The electricity generation capacities of the tested units ranged from 25 MW to 1000 MW, representing the current conditions for Chinese CFPPs under operating ULE standard. The tested units combined configurations to meet ULE standards in this study are representative of the current CFPPs across the nation. These units were equipped with pulverized coal boilers or circulating fluidized bed

boilers, the two dominant types of boiler in CFPPs. Bituminous coals, the dominant type of solid fuel in Chinese power and heating plants, were burned in these boilers during the field samplings, which were performed from December 2017 to May 2018. The tested units combined several distinct configurations for ULE technologies in this study are representative of the CFPPs across the nation. All tested units were equipped with SCR technology to reduce NO_x emissions, a dedusting system to reduce PM, and a FGD system to control SO_2. Widely employed FGD technologies, including limestone-gypsum wet FGD, seawater FGD, and semidry FGD, were installed to remove SO_2. Various dedusting configurations, including ESP, FAB, and WESP systems, were implemented for PM removal in these units. The wet and semidry FGDs were combined with WESP and FAB systems as the end-pipe control system to meet the ultralow emission standards. Information about these tested systems is detailed in Supplementary Table 7. The stack exits after the control devices, were selected as the sampling sites since the flue gas is directly discharged into the atmosphere. Each unit was operated at a stable generation load of over 75% of its capacity during field measurements (Supplementary Figure 11).

2. Household Combustion Site Description

Coal (i.e., bitumite and anthracite), straw (crop residue), and wood are categorized as major fuel types across the whole nation based on a national representative survey from 34,489 households[6, 7]. Relatively high fractions of coal were used for household heating in most provinces (i.e., North China Plain), while biomass fuels were widely used for household cooking (i.e., southwest regions) according to the nationwide survey[6]. Various residential stoves are used for household heating and cooking activities. Among these, iron stoves and brick stoves are the most widely used stoves, which can be found in almost all provinces in mainland China[6, 8]. According to a nationwide field campaign, fuel type was determined as the dominant factor resulting in variations in $PM_{2.5}$ EFs[8]. The field sampling of household solid fuel combustion was carried out in typical households in three villages in northern and southwestern China based on their commonly used fuel types (i.e., bituminous coal, anthracite coal, wood, and crop residue) and stove types (i.e., iron stoves, brick stoves, and three stone stoves), which included the main types of solid fuels and stoves across the whole nation[6, 8]

Twenty to thirty households in each village were selected to test in the current study. One village located in northwestern Tang County (114.98 N, 38.75 E, near Baoding City) in Hebei Province. Coal (i.e., bitumite and anthracite) is the most widely used fuels for household heating and cooking activities, accounting for 68% of total energy consumption in the region[9]. While a small fraction of biomass (i.e., crop residue) is also used for cooking activities. Stoves mainly divided into two types (i.e., improved coal stoves and traditional iron stoves), contributing 78% to total used residential stoves in the region[10]. The improved coal stove has a higher thermal efficiency, which was commonly used for household heating; while the traditional iron stove can be used for heating and cooking activities[10]. All stoves almost equipped with an outdoor chimney but without any air pollution control devices. In the studied village, coal has been frequently used in most of households. Each household had at least one type of the stoves equipped with an outdoor chimney. The energy consumed structure and stove types in the studied village can represent the residential combustion conditions in the rural areas of Hebei province.

The other two villages are located in Ma Yidui County (100.1108 N, 24.1164 E, near Lincang City in Yunnan Province) in southwestern China. The details of the villages were described in previous study[11]. Biomass fuels (i.e., wood and crop residue) were the most widely used fuels for household cooking activities, accounting for 89% of total energy consumption in the rural region; while a small fraction of coal briquettes (10%) was also used for household cooking activities[9]. Three types of stoves (i.e., three-stone stoves, iron stoves, and brick stoves) had been commonly used for residential cooking activities in the region[12]. All stoves equipped with an indoor or outdoor chimney except the three stone stoves. No specific control devices were employed to control pollutants emission during combustion process. In the studied villages, biomass fuels were frequently burnt for household cooking in most of households. The energy consumed structure and stove types in the studied village represent those in the rural areas of Yunnan province, southwestern China. The most common cooking activities that rural residents conducted on these stoves were heating water in kettles and frying food in pans on the flames.

3. Details of Nationwide Control Measures in Two Sectors

No explicit emission standards or end-of-pipe control technologies have been widely

implemented on residential combustion[1, 2]. Replacing coal with clean fuels (natural gas and electricity) in Beijing-Tianjin-Hebei region and their surrounding areas is the major control measure taken in the residential sector (Supplementary Table 8)[1, 13]. Although clean fuels have been used in some households, most of the rural population still depends on solid fuels for household cooking and heating activities owing to their low cost[14, 15].

China imposed the emission standard (GB 13223-2011) in 2012 to limit PM emissions from CFPPs to 30 mg/m^3 at the standard oxygen level, whose emission limit were fully met by all CFPPs by the end of 2015[1, 2]. To further reduce PM emissions, China enforced the toughest-ever ULE standard for CFPPs in 2014, which limits PM to 10 mg/m^3. More than 80% of the total capacity had installed ESP to remove PMs by 2015[5]. The existing de-dedust devices been upgraded to high-efficiency ones under ULE retrofit (Supplementary Table 8)[4]. The PM emission levels required by the ULE standard have been already lower than those in the United States (22.5 μg/m^3 for PM) and close to those in the European Union (2-18 μg/m^3) (Supplementary Table 9). By the end of 2017, 72.3% of national coal-fired units had met the ULE standard in China[3]. Although 27.7% of coal-fired units were not operated at ULE levels, they had already met the emission standards (GB 13223-2011) since 2015. The average emission factors (EF) of coal-fired units were 0.09 ± 0.03 g/kg based on national CEMS data[3], which are much lower than those observed from residential combustion.

4. Tested Solid Fuel Samples

The tested coal samples were mostly bituminous coal and were produced in Inner Mongolia, Shanxi, Shaanxi, Shandong, Henan, Ningxia, and Yunnan Provinces, the main coal-producing regions in China. Biomass samples, including wood, rice residue, corn residue, wheat residue, and bean residue were selected randomly from households in the studied regions. The properties of the tested fuel as received, including the moisture, ash, volatile matter, fixed carbon, sulfur, and lower heating value, are detailed in Supplementary Table 10.

5. Sampling Setup and Procedures

An isokinetic sampling system (Environmental Supply Company, USA) was employed to

collect $PM_{2.5}$ samples from the CFPPs in accordance with U.S. EPA Methods 17 and 201A. The system consists of nozzle/cyclone sets, a heated stainless steel sampling probe coupled with a 47 mm filter holder, an impinger box assembly with glassware sets, and a sampling console with a pump (Supplementary Figure 12). The in-stack nozzle/cyclone and the sampling probe assembly were heated and maintained at 120 °C during the whole sampling process. $PM_{2.5}$ samples were collected on quartz fibers or Teflon filters. Each sampling measurement lasted 120 min to collect enough particle samples for further analysis. A minimum of three successful measurements were conducted in parallel for each sampling site/unit combination, while field-blank filters were also collected. A leakage check for the sampling system was performed before each sampling. The sampling processes and quality assurance for industrial emissions are detailed in our previous studies[16-18]. The mass fraction of OC (3.6 ± 1.9%) contribute only minor fraction to total condensates collected in the impingers. While the concentrations of PAHs contained in condensates were below the method detection limit. The amounts of OC contained in condensates were not included in the results owing to its much lower mass concentration compared to PM collected on filters.

As it is a predominant practice for rural residents and to avoid smoke emissions from cooking activity, water heating was used as the field test for the household combustion study. For each test, a stainless steel kettle or pot filled with 5.0 kg water was placed on the top of the tested stove. The maximum temperature of the water was limited to 80 °C to prevent the generation of additional vapor from boiling water. Coal samples were ignited with pine chips until a stable flame was observed. The emissions from pine chips (less than 0.1 kg in mass) for each ignition were negligible in comparison with the solid fuel emissions in each test. The whole sampling process lasted for 2–3 h and 5–8 h from fuel ignition to fire extinction for the coal cooking and heating activities, respectively. The wood samples were cut into small pieces and weighed to 2.0 kg for each test. The biomass samples were ignited with lighters and then inserted into the stove chamber in several batches. The biomass combustion processes lasted 20 to 40 min for each test. Crop residue samples, 1.0 kg for each test, were loaded into the oven chamber at several-minute intervals after being ignited with lighters. The whole burning cycle lasted for 15 to 30 min. An exhaust hood equipped with an electric blower was installed above the chimneys of the

selected stoves in the sampled households as a simple dilution system for sampling pollutants (Supplementary Figure 16). For the household measurements, the chimney of each heating stove was replaced with a short segment of ~1.5 m in height. The fuel ignition procedure and sampling system were similar to those in the abovementioned field measurements of household cooking activity. For each fuel/stove combination, the field samplings were conducted in triplicate. Blank filters were simultaneously collected for quality assurance. The sampling and quality assurance processes for household combustion emissions are detailed in our previous studies[19-21].

6. Analysis and Quality Control

The organic carbon (OC) and elemental carbon (EC) contained in the $PM_{2.5}$ samples were determined by using a multiwavelength thermal/optical carbon analyzer (DRI Model 2015, Magee, USA). The carbon fractions were divided into OC1-OC4 and EC1-EC3 on the basis of their temperature (i.e., OC fractions at 140, 280, 480, and 580 °C, respectively, and EC fractions at 580, 740, and 840 °C, respectively). Three parallel samples were examined from each filter. The total carbon from the blank filters was below 0.2 μg C/filter. The analytical procedure has been described in detail elsewhere[22-24].

The water-soluble ions in the $PM_{2.5}$ samples were determined by ion chromatography (940 Professional IC, Metrohm, Switzerland). One-quarter of each filter was extracted with 10 mL ultrapure water (Millipore, UK) and sonicated for 40 min. C6-150/4.0 and Supp 5-150/4.0 columns (Metrohm, Switzerland) were used to analyze the cations (Li^+, Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}) and anions (F^-, Cl^-, Br^-, NO_3^-, PO_4^{3-}, and SO_4^{2-}) contained in the $PM_{2.5}$ samples, respectively. The method detection limits (MDLs) for ions ranged from 0.47 to 3.33 μg/L. Blank correction was also performed under the same conditions.

Inductively coupled plasma mass spectrometry (7500a, Thermo Scientific, USA) was employed to quantify the following trace elements contained in the $PM_{2.5}$ samples: Al, Ca, K, Mg, Na, P, S, Si, Li, Be, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Rb, Sr, Mo, Pd, Ag, Cd, Sn, Sb, Cs, Ba, Pt, Au, Ti, and Pb. The $PM_{2.5}$ samples were digested in Teflon tubes with 4 mL HNO_3 and 1 mL H_2O_2 and finally diluted to 10 mL with ultrapure water. Filter blanks and procedure blanks were also measured for each analysis. The recovery rate ranged from 83% to 118%.

One-quarter of the sampled quartz filters were extracted with dichloromethane with an accelerated solvent extractor (ASE 350, Thermo Scientific, USA), and the extracts were then evaporated, purified, and finally concentrated to 1 mL under purified nitrogen. An Agilent 7890A gas chromatograph and 5975C mass spectrometer were employed to detect the 16 PAHs (Supplementary Table 2). The operation conditions were detailed in previous studies[25, 26]. Blank samples were also analyzed following the same method. The MDL varied between 23.9 and 51.2 μg/L, while recovery rates ranged from 80% to 110%.

The A549 cell line was employed for oxidative stress (OS) and cytotoxicity (CT) analysis. The reactive oxidative species were examined with a 2',7'-dichlorofluorescein diacetate (DCFH-DA) protocol. The cells were incubated with DCFH-DA for 30 min in the dark and then exposed to PM extracts for 24 h. The fluorescence intensity was quantified with a microplate reader (Varioskan LUX, Thermo Scientific, USA) at 488/525 nm. The experimental analysis process and estimation method were detailed in our previous studies [24, 26]. The cytotoxicity was measured by a 3-(4,5-dimethylthiazol-2-yl)-2,5-diphenyltetrazolium bromide (MTT) assay. After 24 h of exposure, the absorbance was recorded at 570 nm using a microplate reader (Varioskan LUX, Thermo Scientific, USA).

7. Estimation of Emission Factors and Toxic Units

The EF of $PM_{2.5}$ emitted from CFPPs were estimated as $EF = C_{PM} \times Q/M$, where C_{PM} indicates the concentration of $PM_{2.5}$ (mg/m^3), Q represents the total flue gas flow per hour (m^3/h), and M is amount of combusted coal per hour (t/h). For household solid fuel combustion, the PM EF was determined by the direct mass weight method as $EF = m_{PM} \times R/M_{fuel}$, where m_{PM} is the PM mass sampled on a filter, R is the ratio of the total flue gas flow rate to the sampling flow rate, and M_{fuel} is the weight of solid fuels before each test. Further information on the estimation method was provided in a previous study[27]. Bituminous (B) and anthracite (A) coals account for roughly ~84% and ~16% of residential coal consumption in China based on statistical data from 2009[28]. The average EF of $PM_{2.5}$ emitted from household coal combustion was calculated as follows: $EF = (EF_B \times 84\% + EF_A \times 16\%)$, where EF_B is the average EF of bitumite samples, EF_A is the average EF of anthracite samples.

The toxic units of $PM_{2.5}$-related OS (TU_{OS}) and CT (TU_{CT}) were estimated as $\frac{1}{EC_{1.5}}$

and $\frac{1}{IC_{20}}$, respectively, where $EC_{1.5}$ is the concentration causing a 1.5-fold increase in intracellular reactive oxidative species (ROS) generation relative to that in a control while IC_{20} is the concentration causing 20% inhibition of cell viability relative to that in a control. Region-specific TUs for the residential sector, including for household coal and biomass combustion, were estimated at the provincial level (Supplementary Table 5). The average provincial TU of $PM_{2.5}$ emitted from residential combustion was calculated as follows: $TU_{OS} = \frac{1}{\sum(EC_{1.5})_j \times WF_j}$ and $TU_{CT} = \frac{1}{\sum(IC_{20})_j \times WF_j}$, where $(EC_{1.5})_j$ and $(IC_{20})_j$ are the endpoint OS and CT of sample (j), respectively; and WF_j is the weighting factor of sample (j), estimated based on the mass fraction of each individual consumed fuel type to the total amount of solid fuel consumption at the provincial level[13]. The WFs of the coal (bituminite and anthracite) and biomass (wood and crop residue) samples as well as their TUs were estimated separately. TUs for the CFPPs were adopted with same values across the nation because the toxic potency of $PM_{2.5}$ emitted from CFPPs varies slightly in different regions. The toxic equivalent values (including estimations of OS and CT) for CFPPs (TEQ_{CFPP}) were assigned to 1, and the relative toxic equivalent values for the residential sector (including household coal and biomass combustion) were estimated as $\frac{(TU_{HC})_i}{(TU_{CFPP})_i} \times TEQ_{CFPP}$ and $\frac{(TU_{HB})_i}{(TU_{CFPP})_i} \times TEQ_{CFPP}$, where $(TU_{HC})_i$, $(TU_{HB})_i$, and $(TU_{CFPP})_i$ are the TUs for the household coal combustion, household biomass combustion, and CFPPs in each geographic unit (i), respectively. The toxic potency adjusted emissions were estimated based on $PM_{2.5}$ emissions and $PM_{2.5}$ associated toxic equivalent values.

8. Model Configuration and Evaluation

In this study, we used the latest version of the Community Multiscale Air Quality model (CMAQ, version 5.3.1). Version 6 of the Carbon Bond mechanism (CB6) was used for gas-phase chemistry, and AERO6 was used as the aerosol module[29]. The meteorological fields were simulated by the Weather Research and Forecasting Model (WRF, version 3.9.1). The modeling domain and WRF modeling options were the same as those used in Zheng et al[30]. The horizontal resolution was 27 km × 27 km, and the vertical extent was divided into 14 layers.

The ABaCAS emission inventory from 2005 to 2015 has been comprehensively evaluated in previous studies[30-32]. In this study, we evaluated the emission inventory for 2017 with the aforementioned modeling system. We compared the hourly modeling concentration of $PM_{2.5}$ with the observed data from the China National Environmental Monitoring Centre (Supplementary Table 11). We used the normalized mean bias (NMB), normalized mean error (NME), mean fractional bias (MFB) and mean fractional error (MFE)[33] to quantitatively evaluate the model performance. The model underestimates the concentration slightly, but the NMB for each season is under 15% (Supplementary Table 8). In addition, the MFB and MFE values for all seasons meet the performance criteria. In general, the modeling system and the emission inventory for 2017 can be used to reproduce the pollution level in China in 2017.

9. Uncertainty Analysis

Uncertainty analysis was conducted throughout this study, from the $PM_{2.5}$ EFs to the risk index for the population-weighted TPAE. The uncertainty analysis of $PM_{2.5}$ EFs, chemical components, and $PM_{2.5}$-related toxic potency were evaluated based on the standard deviation (SD) derived from the analysis results of various parallel samples. Supplementary Table 12 shows the comparison of source apportionment results of ambient $PM_{2.5}$ from the two sectors in China with other studies. Without considering the contribution to secondary aerosols in this study, the relative contribution of power plants is lower than that from other previous studies[34, 35]. The source apportionment result for the residential combustion is also lower but with less difference. Because previous studies on the source apportionment often focused on the severe pollution periods, during which the secondary aerosol is more important contributor than the primary PMs[36-38]. To the best of our knowledge, there is no previous study focusing on the contribution of primary PMs to ambient $PM_{2.5}$ concentration. Furthermore, previous source apportionment studies usually did not conduct uncertainty analysis, given that the uncertainty relies strongly on the models themselves.

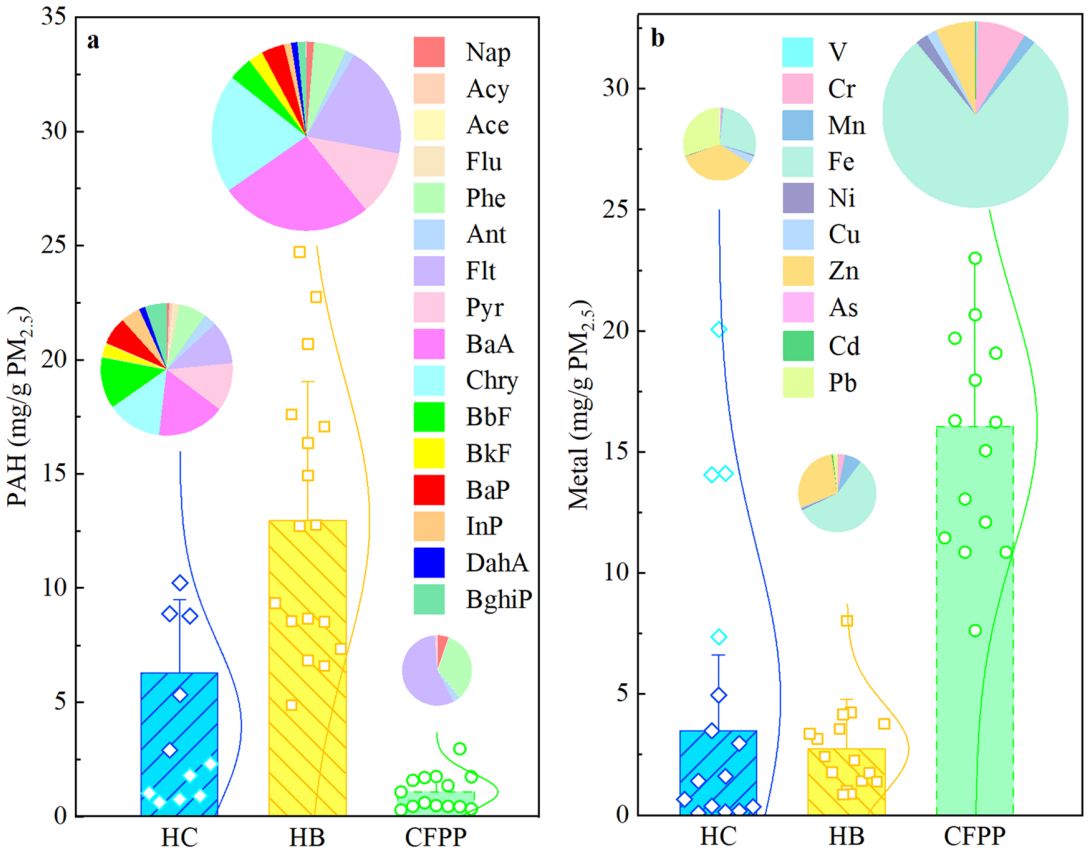
Aiming to evaluate the source apportionment in this study, we conducted two extra simulation for each scenario and year to constrain the bounds based on the uncertainties of the emission inventory and WRF-CMAQ model. The uncertainties of the $PM_{2.5}$ emissions

from CFPPs, household coal burning, and household biomass burning were assessed by 10,000 Monte Carlo runs, based on the probability distribution of the activity data, the efficiency of the control measures, and the share of each end-of-pipe control technology. The $PM_{2.5}$ emission from CFPPs, residential fuel combustion, biomass burning and the 95% confidence intervals are shown in Supplementary Table 13.

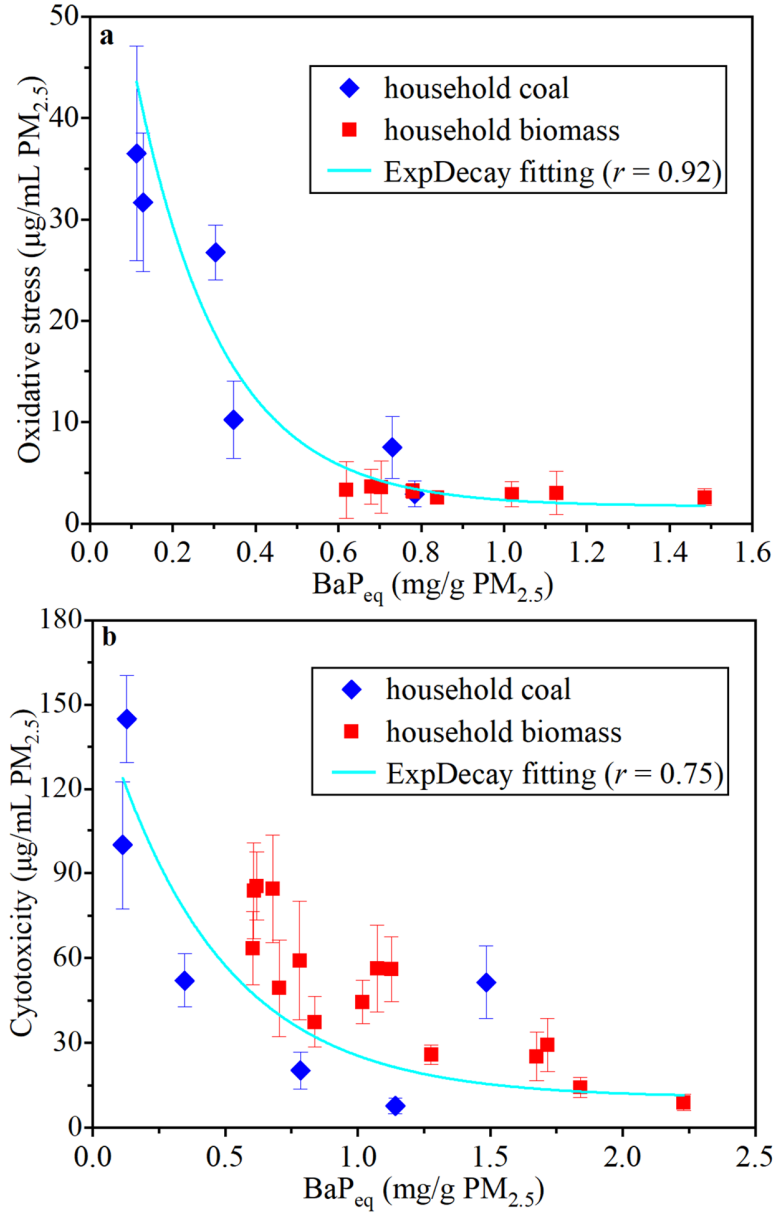
The model simulations with the upper and lower limit of the uncertainty of the emission inventory of each sector and year are run with the same model configuration, as described in METHOD to quantify the uncertainty of the source apportionment results. The uncertainty of the source apportionment in this study only includes the uncertainty of the emission inventory. Due to the near-linear "emission-concentration" response of primary PM[39], we simulated the upper and lower limits of the source apportionment using the upper and lower limits of the emission inventories (95% confidence intervals). In addition, we assume that the probability distribution of the source apportionment is proportional to that of the emission inventory.

The uncertainties of the toxic potency adjusted emissions and the toxic potency adjusted exposure to $PM_{2.5}$ are calculated by Monte Carlo method. The former includes both the uncertainties of the emission inventory and the toxicity test results, while the latter includes the uncertainties of the source apportionment results and the toxicity test results.

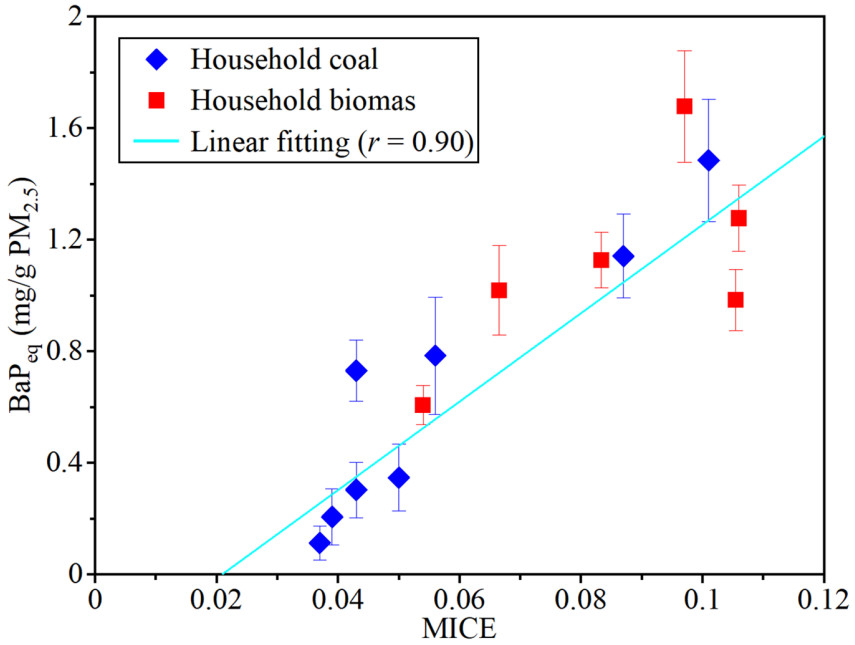
Supplementary Figures



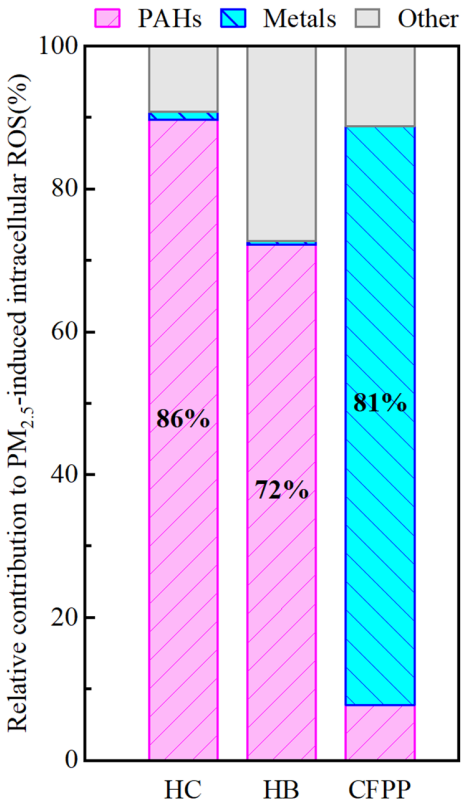
Supplementary Figure 1. Average emission factors of (a) 16 $PM_{2.5}$-bound PAHs and (b) 10 $PM_{2.5}$-bound toxic metals per unit mass of $PM_{2.5}$ emitted from the residential sector and CFPPs. The mass fractions of individual PAHs and metals are shown as pie charts at the top of each column. The areas in the pie charts in Supplementary Figure 1 a and b represent the relative EFs of PAHs and metals per unit mass of $PM_{2.5}$. The cyan and blue diamond patterns represent anthracite and bituminous coal samples, respectively. Error bars show standard deviation. Data are presented as mean values ± SD.



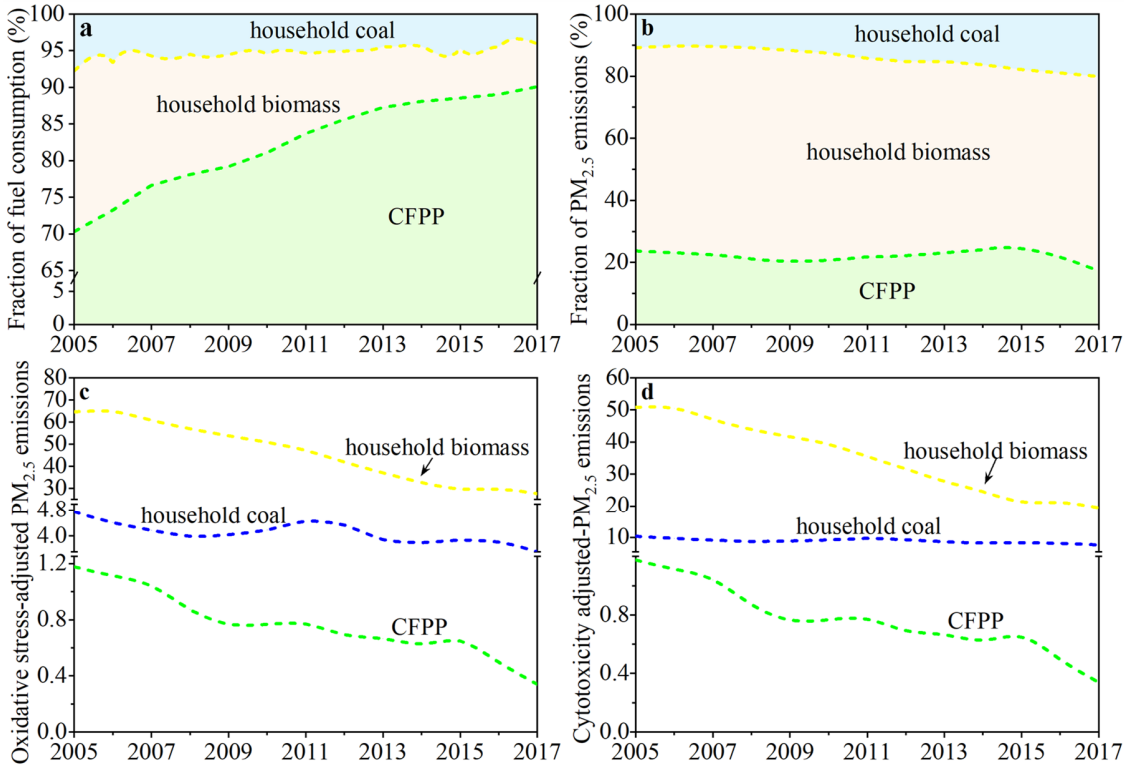
Supplementary Figure 2. (a) Oxidative stress and (b) cytotoxicity as a function of the $PM_{2.5}$-bound BaP_{eq} emitted from household solid fuel combustion. The exponential decay (ExpDecay) fitting ($y = y_0 + A_1 e^{-(x-x_0)/t_1}$) for the two cases resulted in the following equations: $EC_{1.5} = 1.71 + 35.23 \times e^{-(BaPeq - 0.15)/0.21}$ and $IC_{20} = 10.70 + 103.76 \times e^{-(BaPeq - 0.15)/0.44}$. The correlation coefficients (r) are 0.92 and 0.75, respectively. The corresponding p-values are 2.8×10^{-4} and 2.2×10^{-3}, respectively. Both measured toxicity parameters are well correlated with the $PM_{2.5}$-bound BaP_{eq}. One-sided F-test was used for data analysis.



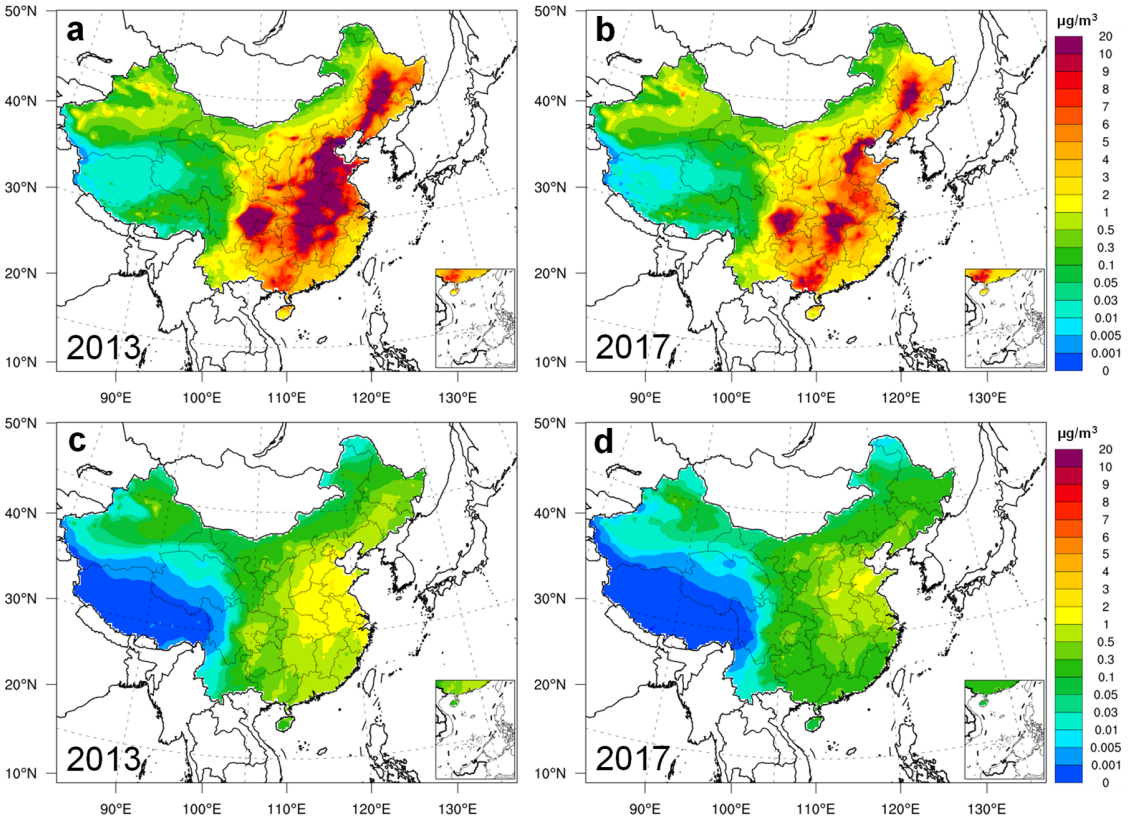
Supplementary Figure 3. $PM_{2.5}$-bound BaP_{eq} as a function of the modified incomplete combustion efficiency (MICE) of various household stoves. The linear fitting yields the following equation: $BaP_{eq} = 16.04\ MICE - 0.33$. The correlation coefficient (r) is 0.90 with a p-value of 1.3×10^{-5}. One-sided F-test was used for data analysis.



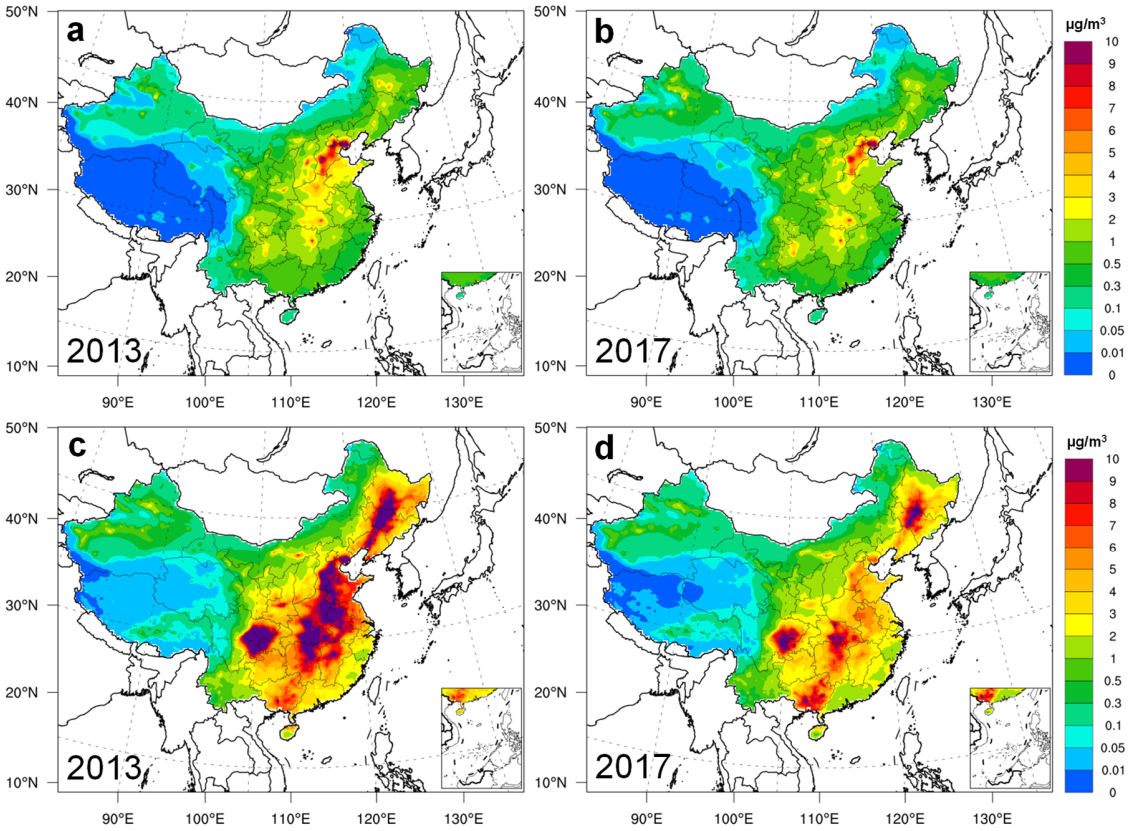
Supplementary Figure 4. Relative contributions of 16 PAHs and 10 toxic metals to $PM_{2.5}$-induced intracellular ROS for household coal burning (HC), household biomass burning (HB), and CFPPs. We cite the REP values for individual chemicals determined in a previous study since the endpoint of selected individual chemicals (Pb) for A549 cells (3.6 μg/mL) is similar to that (3.3 μg/mL) for BEAS-2B cells[40].



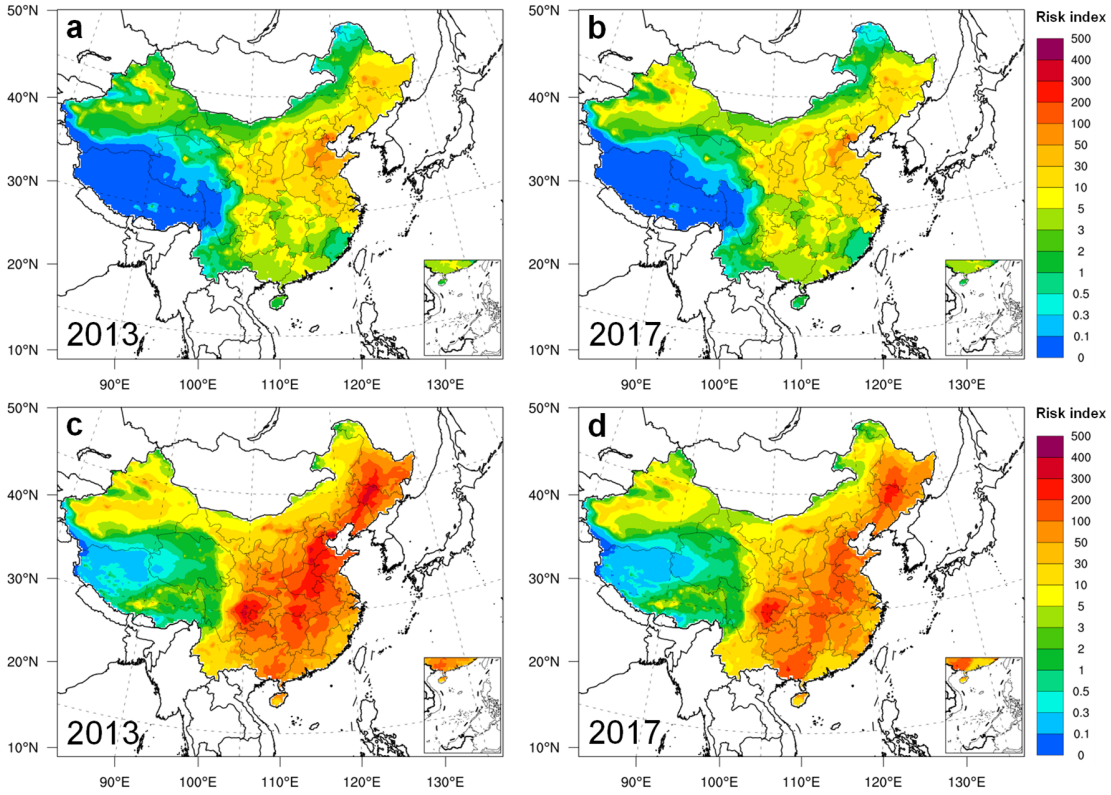
Supplementary Figure 5. Annual variation in (a) solid fuel combustion consumption (million-ton-coal equivalents (Mtce)), (b) $PM_{2.5}$ emissions, and (c and d) $PM_{2.5}$-related toxicity-adjusted emissions for the residential sector and CFPPs from 2005 to 2017 in mainland China.



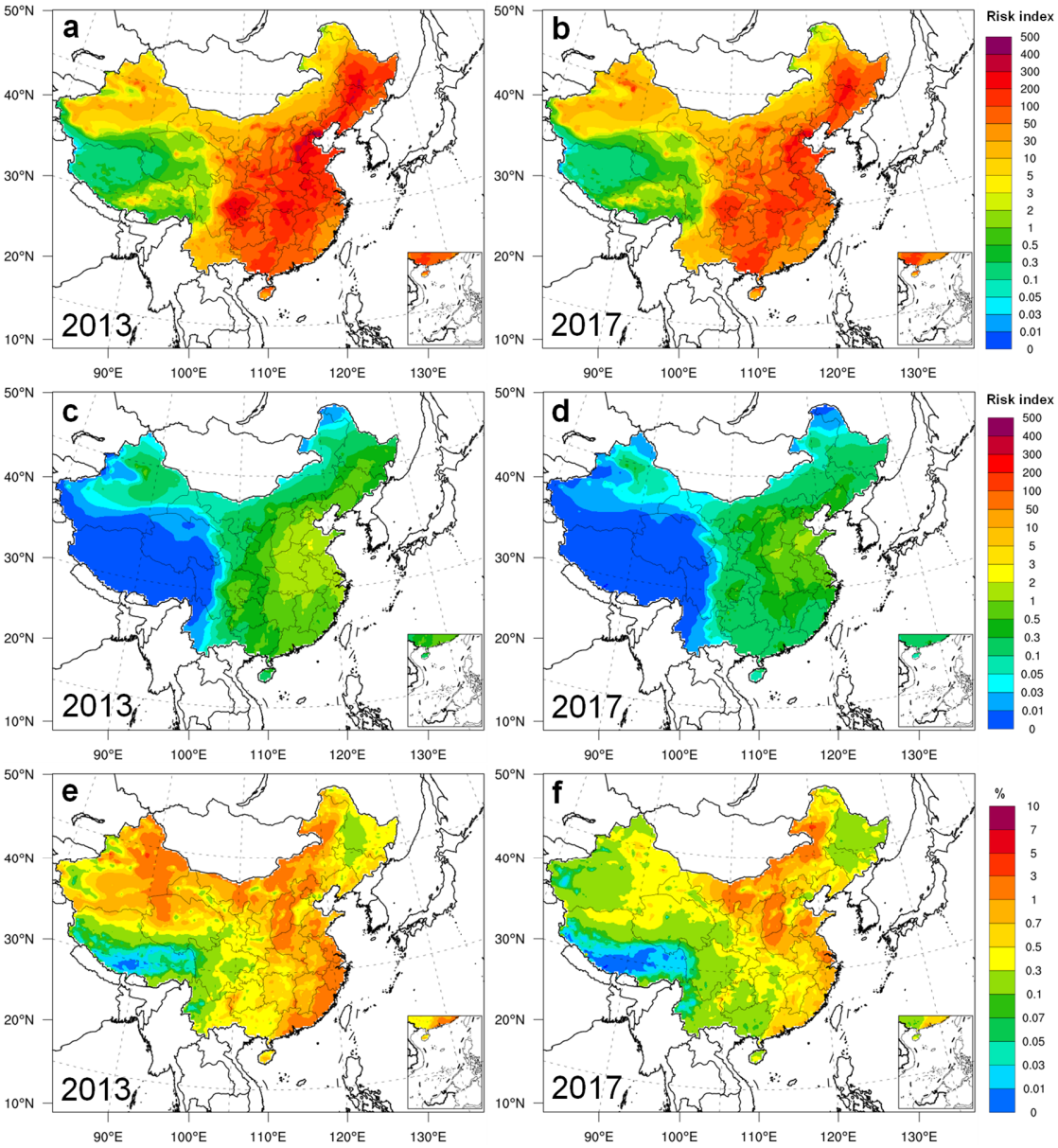
Supplementary Figure 6. Spatial distribution of the concentration of ambient $PM_{2.5}$ contributed from (a and b) the residential sector and (c and d) CFPPs in 2013 and 2017.



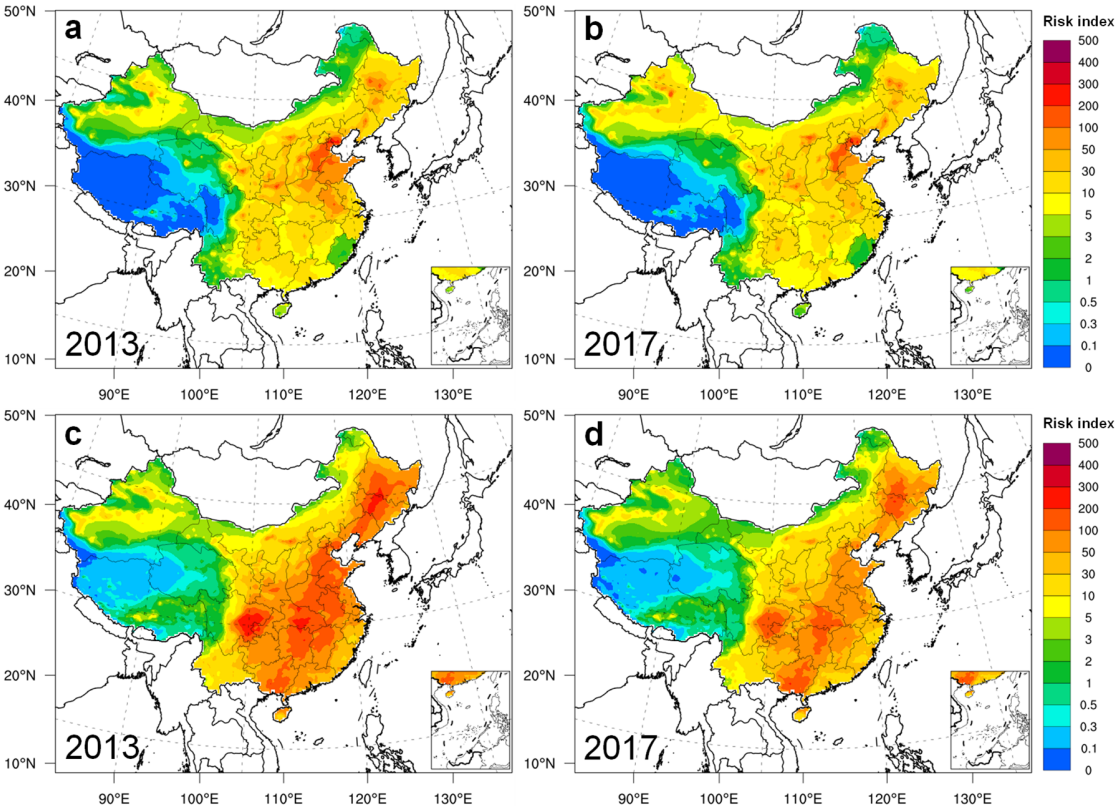
Supplementary Figure 7. Spatial distribution of the concentration of ambient $PM_{2.5}$ originating from the residential sector in 2013 and 2017, including household (a and b) coal and (c and d) biomass combustion.



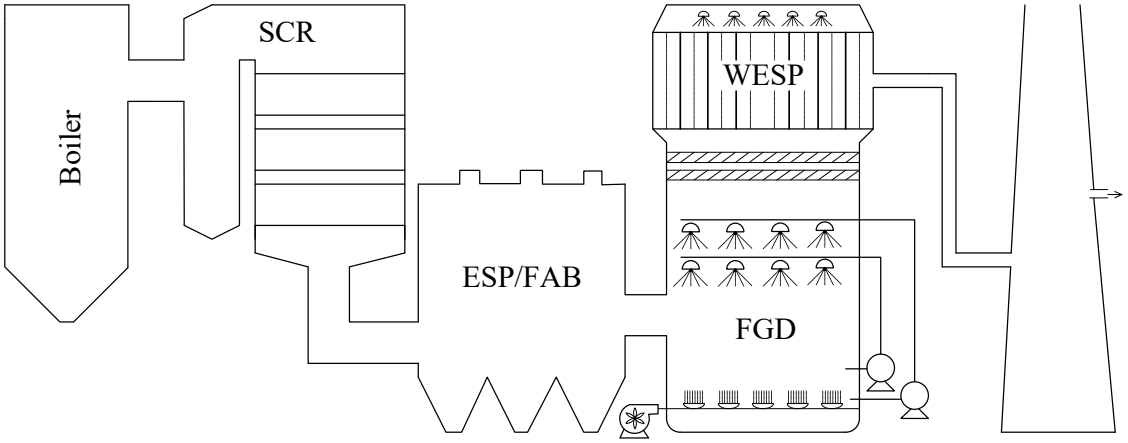
Supplementary Figure 8. Spatial distribution of primary $PM_{2.5}$-related oxidative stress-adjusted exposure to $PM_{2.5}$ ($TPAE_{OS}$) for the residential sector. Spatial distribution of the risk index for $TPAE_{OS}$ attributable to household (a and b) coal and (c and d) biomass combustion in 2013 and 2017.



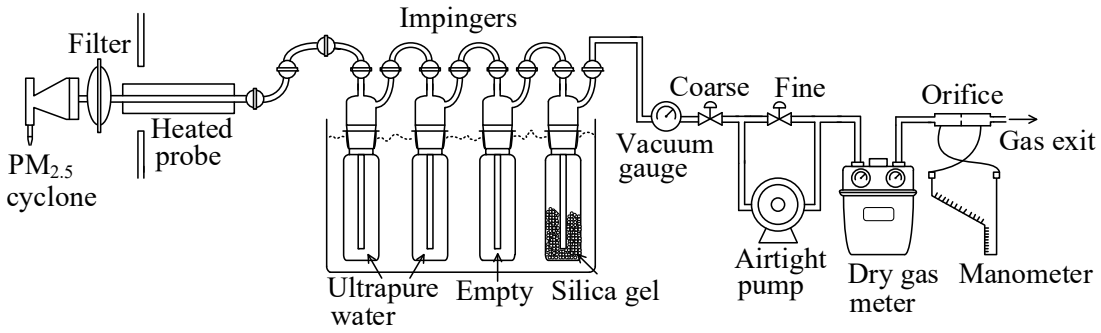
Supplementary Figure 9. Spatial distribution of primary $PM_{2.5}$-related cytotoxicity-adjusted exposure to $PM_{2.5}$ ($TPAE_{CT}$) for the residential sector and CFPPs. Spatial distribution of the risk index for $TPAE_{CT}$ caused by (a and b) the residential sector and (c and d) CFPPs in 2013 and 2017. (e and f) Relative contribution of CFPP-emitted $PM_{2.5}$ to the overall $TPAE_{CT}$ across China in 2013 and 2017.



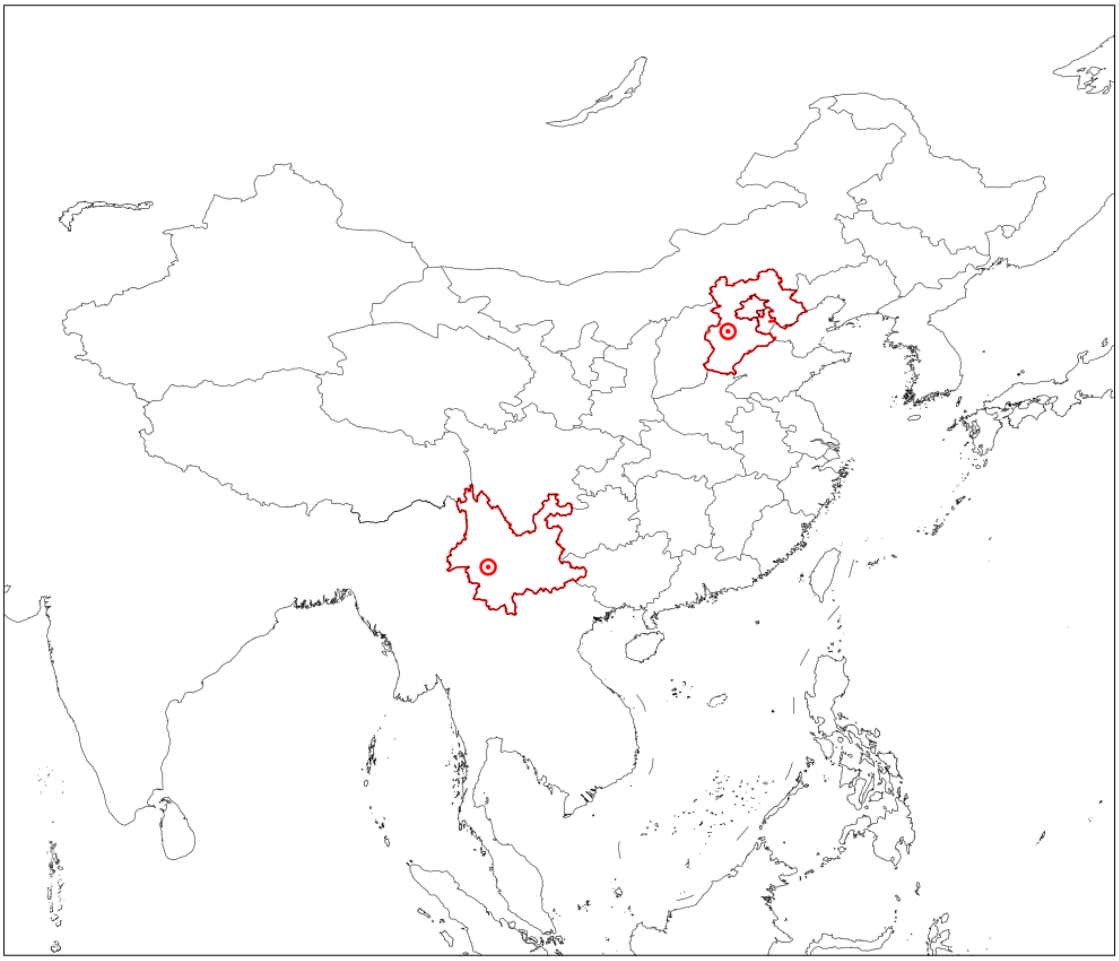
Supplementary Figure 10. Spatial distribution of the $TPAE_{CT}$ for the residential sector. Spatial distribution of the risk index for $TPAE_{CT}$ attributable to household (a and b) coal and (c and d) biomass combustion in 2013 and 2017.



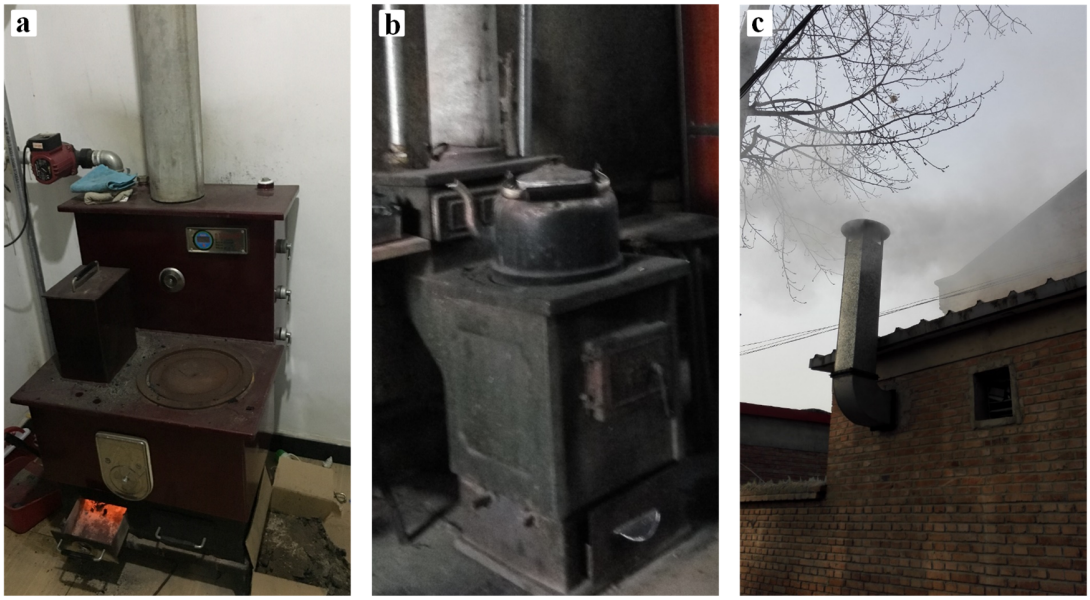
Supplementary Figure 11. Schematic diagram of air pollution control devices (including SCR = selective noncatalytic reduction system, ESP = electrostatic precipitator, FAB = fabric baghouse, FGD = flue gas desulfurization system, and WESP = wet electrostatic precipitator) and the PM sampling site for typical coal-fired power and heating plants. The sampling site is labeled with an arrow at the stack, after which no further control device is installed.



Supplementary Figure 12. Schematic diagram of the isokinetic sampling system for sampling $PM_{2.5}$ from CFPP stacks.



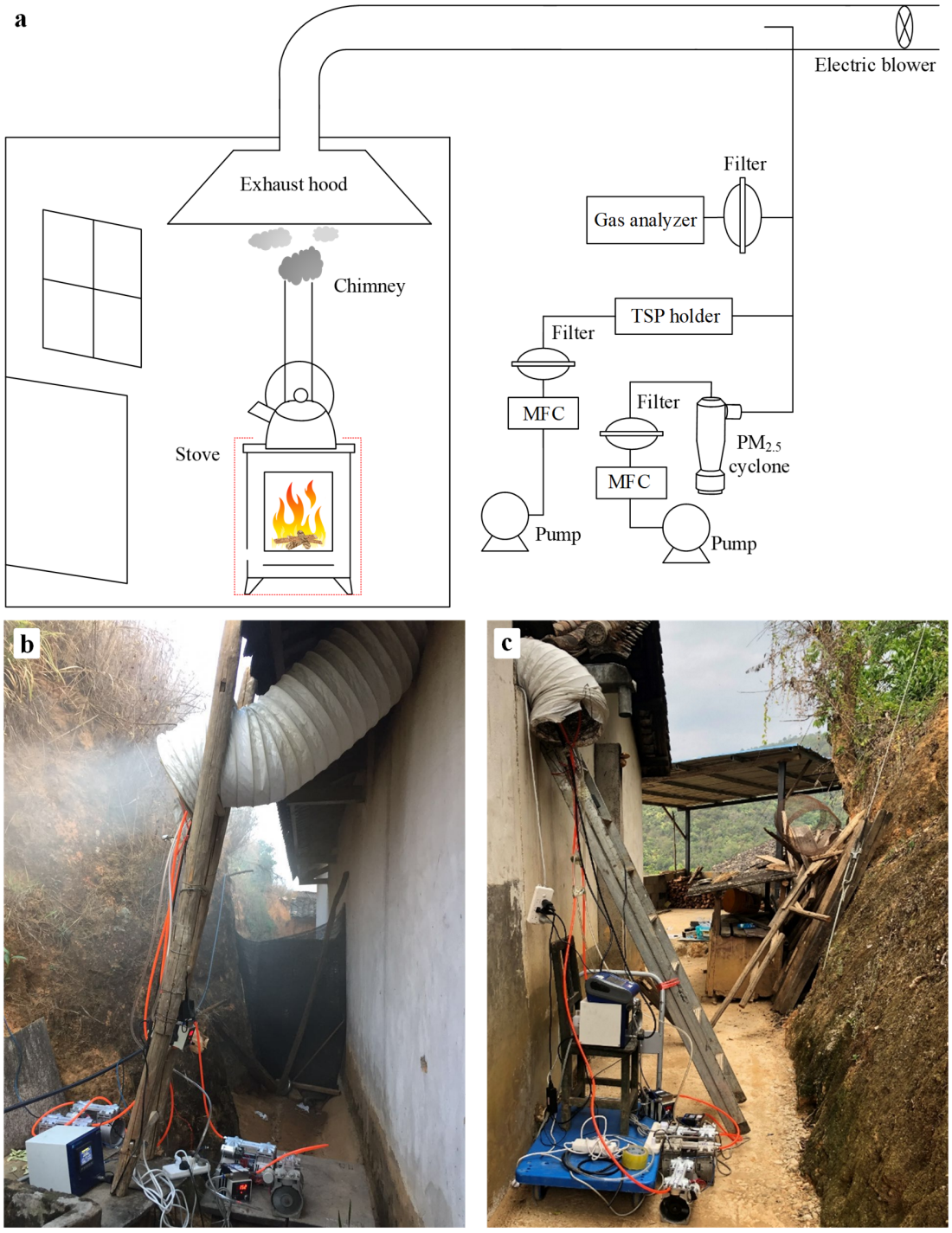
Supplementary Figure 13. Locations of the studied residential regions. The residential sampling sites located in Tang County (114.98 N, 38.75 E) near Baoding City in Hebei Province and Ma Yidui County (100.1108 N, 24.1164 E) near Lincang City in southwestern Yunnan Province, China.



Supplementary Figure 14. Photographs of (a) the improved household heating stove, (b) a traditional iron stove, and (c) exhaust gas emitted from the outdoor chimney of a typical household stove during coal combustion.



Supplementary Figure 15. Photographs of household cooking stoves, including (a) the three-stone stove, (b) the brick stove, (c) the new iron stove, and (d) the iron stove that had been used for three years.



Supplementary Figure 16. (a) Sampling system for emissions from household solid fuel combustion and (b and c) photographs of the sampling systems installed at two typical households.

Supplementary Tables

Supplementary Table 1. Comparison of PM chemical component of OC, EC, PAHs, and elements observed from residential combustion and CFPPs.

Sources	Stove type	Fuel type	OC (%)	EC (%)	PAH (%)	Elements (%)	Reference
Residential	brick/steel stove	bitumite	40.3 ± 11.7	37.3 ± 14.2			Zhang et al., 2008[41]
Industrial	industrial boiler	bitumite	1.88 ± 0.03	0.38 ± 0.01			
Residential	cooking stove	Corn residue	42.3	17.9			Cao et al., 2008[42]
	cooking stove	Cotton residue	40.4	18.1			
Residential	cooking stove	biomass	40.4 ± 12.3	21.7 ± 19.7			Li et al., 2009[43]
Residential	coal stove	bitumite			1.09 ± 0.27		Shen et al., 2010[44]
Residential	brick stove	Peanut residue			0.75		Shen et al., 2011[45]
		Soybean residue			0.83 ± 0.08		
		Cotton residue			0.95 ± 0.11		
		Rice residue			1.41 ± 0.31		
		Wheat residue			1.04 ± 0.31		
		Corn residue			0.85 ± 0.27		
Residential	brick stove	wood			1.11 ± 0.42		Shen et al., 2012[46]
Residential	cooking stove	coal				1.4	Liu et al., 2016[47]
Residential	cooking stove	wood	51.8 ± 13.8	21.9 ± 11.9			Jayarathne et al., 2018[48]
Residential	cooking stove	coal				3.00 ± 0.68	Dai et al., 2019[49]
Residential	cooking stove	biomass	42.3 ± 4.4	31.0 ± 5.3	1.30 ± 0.60	3.47 ± 2.71	This study
	heating stove	coal	56.7 ± 17.8	15.6 ± 12.7	0.63 ± 0.32	4.01 ± 1.40	
CFPP	industrial boiler	bitumite				16.59	Chen et al., 2015[50]
	industrial boiler	bitumite				20.19	
	industrial boiler	bitumite	3.37	1.06			
CFPP	industrial boiler	bitumite	5.62 ± 3.10	0.03 ± 0.07	0.11 ± 0.08	15.1 ± 9.5	This study

Supplementary Table 2. Sixteen U.S. EPA priority PAHs, their abbreviations, and their toxic equivalency factors (TEFs) obtained from previous studies[51, 52].

Substance name	Abbreviation	Quantitative ion	TEF	Number of rings
Naphthalene	Nap	128	0.001	2
Acenaphthylene	Acp	152	0.001	3
Acenaphthene	Ace	154	0.001	3
Fluorene	Flu	166	0.001	3
Phenanthrene	Phe	178	0.001	3
Anthracene	Ant	178	0.01	3
Fluoranthene	Flt	202	0.001	4
Pyrene	Pyr	202	0.001	4
Benzo[a]anthracene	BaA	228	0.1	4
Chrysene	Chry	228	0.01	4
Benzo[b]fluoranthene	BbF	252	0.1	5
Benzo[k]fluoranthene	BkF	252	0.1	5
Benzo[a]pyrene	BaP	252	1.0	5
Dibenzo[a,h]anthracene	DahA	276	1.0	5
Indeno[1,2,3-cd]pyrene	InP	278	0.1	6
Benzo[ghi]perylene	BghiP	276	0.01	6

Supplementary Table 3. Ten selected toxic metals, their abbreviations, and their TEFs estimated based on carcinogenic risk parameters (inhalation unit risk) obtained the U.S. EPA website[53].

Substance name	Abbreviation	IUR	TEF
vanadium	V	--	--
chromium	Cr	8.40×10^{-2}	1
manganese	Mn	--	--
iron	Fe	--	--
nickel	Ni	2.60×10^{-4}	3.10×10^{-3}
copper	Cu	--	--
zinc	Zn	--	--
arsenic	As	4.30×10^{-3}	5.12×10^{-2}
cadmium	Cd	1.80×10^{-3}	2.14×10^{-2}
lead	Pb	1.20×10^{-5}	1.43×10^{-4}

Supplementary Table 4. The population-weighted exposure to $PM_{2.5}$ ($\mu g/m^3$) originating from the residential sector (including household coal and biomass combustion) and CFPPs at the provincial level in 2013 and 2017.

Region	2013			2017		
	Residential sector		CFPP	Residential sector		CFPP
	HC	HB		HC	HB	
China	1.9 (1.1, 4.3)	6.2 (0.6, 15.2)	0.9 (0.6, 1.2)	1.5 (0.6, 3.3)	3.8 (0.3, 6.7)	0.4 (0.3, 0.6)
Beijing	5.9 (3.3, 13.1)	4.8 (0.4, 12.3)	1.2 (0.9, 1.6)	4.1 (2.1, 9.0)	2.5 (0.1, 4.4)	0.4 (0.3, 0.6)
Tianjin	4.5 (2.5, 10.0)	6.0 (0.5, 14.2)	1.3 (0.9, 1.7)	3.8 (1.6, 8.1)	3.2 (0.0, 5.2)	0.7 (0.5, 0.9)
Hebei	6.0 (3.4, 13.4)	8.7 (0.8, 22.0)	1.2 (0.8, 1.6)	5.2 (2.5, 11.3)	4.3 (0.2, 7.5)	0.6 (0.5, 0.8)
Shanxi	3.1 (1.7, 6.8)	3.5 (0.3, 8.2)	1.0 (0.7, 1.3)	2.0 (0.9, 4.2)	1.8 (0.0, 3.0)	0.6 (0.4, 0.8)
Inner Mongolia	0.7 (0.4, 1.5)	2.3 (0.2, 6.2)	0.4 (0.3, 0.5)	0.8 (0.4, 1.8)	1.6 (0.1, 3.0)	0.3 (0.2, 0.4)
Liaoning	1.8 (1.0, 3.9)	6.7 (0.7, 17.9)	0.8 (0.5, 1.0)	1.9 (0.9, 4.2)	3.6 (0.2, 6.6)	0.5 (0.3, 0.6)
Jilin	1.4 (0.8, 3.0)	9.1 (0.9, 24.9)	0.6 (0.4, 0.8)	1.2 (0.6, 2.5)	7.0 (0.6, 13.1)	0.3 (0.2, 0.4)
Heilongjiang	1.8 (1.0, 3.9)	7.2 (0.7, 19.3)	0.4 (0.3, 0.6)	1.3 (0.7, 2.9)	5.1 (0.4, 9.4)	0.2 (0.1, 0.3)
Shanghai	1.0 (0.6, 2.2)	2.6 (0.3, 5.4)	1.2 (0.9, 1.6)	0.6 (0.0, 1.1)	1.7 (0.0, 2.6)	0.6 (0.4, 0.7)
Jiangsu	1.0 (0.6, 2.3)	6.2 (0.6, 14.5)	1.4 (1.0, 1.8)	0.8 (0.1, 1.5)	3.6 (0.1, 6.1)	0.7 (0.5, 0.9)
Zhejiang	0.7 (0.4, 1.5)	3.0 (0.3, 6.2)	1.2 (0.8, 1.5)	0.6 (0.0, 1.1)	2.2 (0.1, 3.5)	0.5 (0.3, 0.6)
Anhui	2.1 (1.2, 4.8)	9.5 (1.0, 22.8)	1.5 (1.0, 1.9)	1.4 (0.4, 2.9)	5.1 (0.4, 8.9)	0.6 (0.4, 0.8)
Fujian	0.5 (0.3, 1.1)	2.4 (0.2, 5.1)	0.8 (0.6, 1.0)	0.4 (0.0, 0.6)	2.3 (0.2, 3.9)	0.3 (0.2, 0.4)
Jiangxi	1.2 (0.7, 2.7)	6.8 (0.7, 15.5)	0.9 (0.6, 1.2)	1.2 (0.3, 2.4)	4.0 (0.3, 6.8)	0.4 (0.2, 0.5)
Shandong	2.5 (1.4, 5.5)	7.7 (0.8, 18.8)	1.6 (1.1, 2.1)	1.5 (0.4, 3.1)	4.0 (0.1, 6.8)	1.0 (0.7, 1.2)
Henan	3.1 (1.7, 6.9)	8.7 (0.8, 20.8)	1.6 (1.1, 2.1)	1.9 (0.7, 4.0)	3.8 (0.1, 6.4)	0.8 (0.5, 1.0)
Hubei	3.1 (1.7, 6.9)	10.5 (1.1, 25.8)	1.1 (0.8, 1.5)	3.0 (1.3, 6.4)	7.0 (0.5, 12.3)	0.5 (0.4, 0.7)
Hunan	2.4 (1.3, 5.2)	8.2 (0.8, 19.8)	0.8 (0.6, 1.0)	2.3 (1.0, 4.8)	6.0 (0.5, 10.6)	0.3 (0.2, 0.4)
Guangdong	0.6 (0.4, 1.4)	3.1 (0.3, 6.8)	0.8 (0.5, 1.0)	0.6 (0.2, 1.3)	1.8 (0.1, 3.1)	0.3 (0.2, 0.3)
Guangxi	0.6 (0.4, 1.3)	6.0 (0.6, 14.2)	0.5 (0.3, 0.6)	0.5 (0.2, 1.1)	7.1 (0.8, 13.2)	0.2 (0.1, 0.2)
Hainan	0.2 (0.1, 0.5)	4.0 (0.4, 8.8)	0.3 (0.2, 0.5)	0.2 (0.1, 0.4)	2.9 (0.3, 5.4)	0.1 (0.1, 0.1)
Chongqing	1.7 (1.0, 3.7)	9.8 (1.0, 24.3)	0.6 (0.4, 0.8)	1.3 (0.6, 2.7)	6.3 (0.5, 11.4)	0.3 (0.2, 0.4)
Sichuan	1.8 (1.0, 3.8)	11.2 (1.2, 27.4)	0.5 (0.4, 0.7)	1.0 (0.5, 2.2)	6.9 (0.6, 12.6)	0.2 (0.2, 0.3)
Guizhou	1.7 (1.0, 3.8)	4.3 (0.4, 10.3)	0.4 (0.3, 0.5)	2.1 (1.1, 4.7)	3.1 (0.2, 5.5)	0.2 (0.1, 0.3)
Yunnan	0.6 (0.3, 1.2)	1.7 (0.2, 3.8)	0.1 (0.1, 0.2)	0.6 (0.3, 1.3)	1.2 (0.1, 2.1)	0.1 (0.0, 0.1)
Tibet	0.0 (0.0, 0.0)	0.1 (0.0, 0.3)	0.0 (0.0, 0.0)	0.0 (0.0, 0.0)	0.1 (0.0, 0.2)	0.0 (0.0, 0.0)
Shaanxi	2.4 (1.3, 5.4)	5.6 (0.5, 14.3)	0.5 (0.4, 0.7)	1.8 (0.9, 4.0)	2.9 (0.2, 5.2)	0.4 (0.3, 0.5)
Gansu	1.1 (0.6, 2.5)	2.3 (0.2, 5.8)	0.2 (0.2, 0.3)	1.1 (0.6, 2.5)	1.2 (0.1, 2.3)	0.1 (0.1, 0.2)
Qinghai	1.0 (0.6, 2.3)	0.7 (0.1, 1.8)	0.1 (0.1, 0.1)	0.9 (0.5, 2.1)	0.4 (0.0, 0.7)	0.1 (0.0, 0.1)
Ningxia	0.9 (0.5, 1.9)	2.3 (0.2, 5.8)	0.3 (0.2, 0.4)	1.0 (0.5, 2.3)	1.2 (0.1, 2.2)	0.3 (0.2, 0.4)
Xinjiang	0.7 (0.4, 1.6)	0.8 (0.1, 2.1)	0.1 (0.1, 0.2)	1.4 (0.7, 3.1)	0.6 (0.0, 1.1)	0.1 (0.0, 0.1)

Supplementary Table 5. Toxic units of $PM_{2.5}$-related oxidative stress (OS) and cytotoxicity (CT) for the residential sector at the provincial level in 2013 and 2017. TUs for the household coal combustion were adopted with same values in 2013 and 2017 across the nation because of no available statistical data on mass fraction of bituminous and anthracite coals to total residential coal consumption in different regions in 2017.

Region	coal		biomass			
	2013/2017		2013		2017	
	OS	CT	OS	CT	OS	CT
China	0.123 ± 0.045	0.026 ± 0.016	0.292 ± 0.098	0.021 ± 0.008	0.312 ± 0.080	0.021 ± 0.007
Beijing	0.032 ± 0.005	0.008 ± 0.002	0.356 ± 0.063	0.021 ± 0.007	0.348 ± 0.029	0.021 ± 0.005
Tianjin	0.232 ± 0.145	0.040 ± 0.036	0.344 ± 0.137	0.020 ± 0.010	0.329 ± 0.138	0.020 ± 0.010
Hebei	0.127 ± 0.048	0.026 ± 0.017	0.353 ± 0.084	0.020 ± 0.007	0.351 ± 0.051	0.021 ± 0.006
Shanxi	0.091 ± 0.027	0.020 ± 0.011	0.366 ± 0.112	0.021 ± 0.010	0.347 ± 0.026	0.021 ± 0.005
Inner Mongolia	0.251 ± 0.168	0.042 ± 0.040	0.366 ± 0.138	0.022 ± 0.012	0.367 ± 0.151	0.022 ± 0.013
Liaoning	0.199 ± 0.109	0.036 ± 0.030	0.324 ± 0.120	0.023 ± 0.011	0.335 ± 0.112	0.022 ± 0.011
Jilin	0.244 ± 0.159	0.041 ± 0.038	0.338 ± 0.129	0.023 ± 0.012	0.337 ± 0.120	0.023 ± 0.012
Heilongjiang	0.255 ± 0.173	0.042 ± 0.040	0.289 ± 0.119	0.023 ± 0.011	0.298 ± 0.101	0.022 ± 0.009
Shanghai	0.123 ± 0.045	0.026 ± 0.016	0.292 ± 0.098	0.021 ± 0.008	0.312 ± 0.080	0.021 ± 0.007
Jiangsu	0.248 ± 0.164	0.041 ± 0.039	0.202 ± 0.095	0.019 ± 0.006	0.215 ± 0.097	0.019 ± 0.006
Zhejiang	0.262 ± 0.182	0.043 ± 0.042	0.251 ± 0.083	0.022 ± 0.006	0.266 ± 0.078	0.022 ± 0.006
Anhui	0.223 ± 0.134	0.039 ± 0.035	0.260 ± 0.089	0.019 ± 0.005	0.293 ± 0.072	0.020 ± 0.005
Fujian	0.032 ± 0.005	0.008 ± 0.002	0.229 ± 0.089	0.023 ± 0.006	0.337 ± 0.035	0.021 ± 0.005
Jiangxi	0.059 ± 0.013	0.014 ± 0.006	0.222 ± 0.088	0.023 ± 0.006	0.258 ± 0.079	0.022 ± 0.006
Shandong	0.241 ± 0.155	0.041 ± 0.038	0.350 ± 0.097	0.019 ± 0.007	0.352 ± 0.081	0.020 ± 0.007
Henan	0.091 ± 0.027	0.020 ± 0.010	0.329 ± 0.085	0.018 ± 0.006	0.339 ± 0.060	0.020 ± 0.006
Hubei	0.041 ± 0.007	0.010 ± 0.003	0.229 ± 0.094	0.021 ± 0.007	0.237 ± 0.094	0.022 ± 0.007
Hunan	0.041 ± 0.007	0.010 ± 0.003	0.261 ± 0.081	0.022 ± 0.006	0.259 ± 0.082	0.022 ± 0.006
Guangdong	0.123 ± 0.045	0.026 ± 0.016	0.234 ± 0.089	0.023 ± 0.007	0.205 ± 0.093	0.023 ± 0.007
Guangxi	0.102 ± 0.033	0.022 ± 0.012	0.234 ± 0.097	0.023 ± 0.008	0.294 ± 0.070	0.022 ± 0.006
Hainan	0.123 ± 0.045	0.026 ± 0.016	0.237 ± 0.089	0.023 ± 0.007	0.226 ± 0.087	0.023 ± 0.006
Chongqing	0.076 ± 0.02	0.018 ± 0.008	0.265 ± 0.098	0.022 ± 0.008	0.267 ± 0.097	0.022 ± 0.008
Sichuan	0.069 ± 0.017	0.016 ± 0.007	0.262 ± 0.098	0.022 ± 0.008	0.324 ± 0.055	0.021 ± 0.006
Guizhou	0.073 ± 0.018	0.017 ± 0.008	0.319 ± 0.062	0.022 ± 0.006	0.334 ± 0.045	0.021 ± 0.006
Yunnan	0.070 ± 0.017	0.017 ± 0.007	0.316 ± 0.072	0.022 ± 0.007	0.335 ± 0.053	0.021 ± 0.006
Tibet	0.123 ± 0.045	0.026 ± 0.016	0.292 ± 0.098	0.021 ± 0.008	0.312 ± 0.080	0.021 ± 0.007
Shaanxi	0.261 ± 0.181	0.043 ± 0.042	0.343 ± 0.079	0.020 ± 0.007	0.347 ± 0.032	0.021 ± 0.005
Gansu	0.262 ± 0.182	0.043 ± 0.042	0.358 ± 0.084	0.021 ± 0.008	0.351 ± 0.043	0.021 ± 0.006
Qinghai	0.262 ± 0.182	0.043 ± 0.042	0.344 ± 0.073	0.017 ± 0.005	0.342 ± 0.063	0.017 ± 0.004
Ningxia	0.262 ± 0.182	0.043 ± 0.042	0.321 ± 0.121	0.022 ± 0.011	0.323 ± 0.120	0.022 ± 0.011
Xinjiang	0.260 ± 0.180	0.043 ± 0.041	0.347 ± 0.101	0.019 ± 0.008	0.348 ± 0.100	0.020 ± 0.008

Supplementary Table 6. The risk index for the population-weighted $TPAE_{OS}$ and $TPAE_{CT}$ originating from residential sector and CFPPs at the provincial level in 2013 and 2017.

Region	$TPAE_{OS}$				$TPAE_{CT}$			
	2013		2017		2013		2017	
	Residential sector	CFPP×100	Residential sector	CFPP×100	Residential sector	CFPP×100	Residential sector	CFPP×100
China	148 (102, 169)	91 (84, 102)	99 (70, 110)	44 (41, 50)	135 (102, 165)	90 (84, 102)	90 (70, 111)	45 (41, 50)
Beijing	137 (27, 236)	124 (81, 171)	72 (16, 117)	43 (28, 60)	112 (38, 203)	127 (50, 208)	65 (25, 110)	44 (17, 73)
Tianjin	224 (44, 433)	130 (86, 178)	139 (28, 285)	72 (46, 99)	224 (20, 620)	127 (52, 218)	162 (10, 479)	75 (29, 122)
Hebei	277 (70, 482)	121 (79, 167)	156 (49, 257)	65 (42, 89)	248 (52, 526)	120 (48, 206)	169 (33, 389)	67 (26, 110)
Shanxi	112 (25, 194)	99 (65, 137)	58 (15, 89)	60 (39, 82)	102 (22, 205)	97 (40, 166)	59 (16, 113)	60 (24, 100)
Inner Mongolia	74 (14, 149)	36 (23, 48)	58 (11, 118)	29 (19, 40)	60 (6, 142)	37 (14, 60)	53 (4, 134)	29 (12, 50)
Liaoning	181 (34, 367)	75 (49, 103)	115 (29, 217)	49 (32, 68)	161 (21, 355)	75 (31, 125)	113 (15, 269)	52 (20, 84)
Jilin	245 (41, 510)	60 (39, 84)	190 (34, 395)	29 (19, 40)	196 (22, 451)	60 (24, 102)	153 (19, 343)	29 (12, 49)
Heilongjiang	182 (33, 370)	43 (28, 59)	133 (26, 261)	20 (13, 28)	177 (24, 405)	44 (17, 73)	126 (21, 275)	22 (8, 34)
Shanghai	64 (11, 112)	124 (81, 171)	43 (6, 70)	58 (37, 78)	61 (12, 113)	127 (50, 208)	38 (6, 66)	60 (23, 96)
Jiangsu	110 (16, 225)	140 (93, 193)	69 (10, 138)	72 (47, 99)	122 (23, 239)	142 (59, 240)	75 (13, 146)	75 (30, 121)
Zhejiang	68 (14, 121)	118 (78, 162)	55 (10, 93)	46 (30, 64)	72 (13, 144)	120 (48, 198)	57 (9, 109)	44 (19, 79)
Anhui	212 (44, 402)	148 (97, 204)	132 (30, 231)	64 (42, 88)	199 (43, 402)	150 (62, 251)	119 (24, 237)	67 (25, 109)
Fujian	40 (4, 79)	81 (53, 111)	58 (8, 92)	29 (19, 40)	43 (8, 76)	82 (32, 135)	40 (6, 69)	29 (11, 49)
Jiangxi	114 (13, 231)	90 (59, 124)	79 (13, 149)	35 (23, 48)	129 (26, 233)	90 (37, 151)	79 (20, 137)	37 (14, 58)
Shandong	237 (47, 429)	161 (106, 221)	129 (28, 224)	95 (62, 130)	183 (25, 411)	157 (65, 271)	106 (14, 237)	97 (39, 159)
Henan	225 (37, 401)	163 (106, 225)	105 (21, 174)	79 (52, 109)	165 (41, 307)	165 (68, 273)	85 (22, 151)	82 (32, 134)
Hubei	182 (21, 383)	114 (75, 157)	128 (19, 258)	51 (33, 70)	192 (42, 363)	112 (46, 194)	136 (34, 253)	52 (21, 86)
Hunan	161 (23, 308)	80 (52, 110)	119 (19, 226)	34 (22, 47)	154 (35, 280)	82 (33, 135)	117 (29, 213)	37 (14, 57)
Guangdong	57 (9, 113)	78 (51, 107)	32 (6, 63)	26 (17, 36)	64 (15, 116)	75 (33, 131)	43 (10, 80)	29 (10, 45)
Guangxi	106 (12, 229)	45 (29, 61)	155 (25, 297)	15 (9, 20)	114 (21, 223)	44 (19, 75)	125 (24, 244)	15 (6, 25)
Hainan	70 (9, 148)	34 (23, 48)	49 (6, 104)	10 (6, 14)	72 (13, 142)	37 (14, 58)	53 (10, 102)	10 (4, 17)
Chongqing	197 (28, 412)	61 (40, 85)	128 (17, 261)	33 (21, 45)	186 (36, 366)	60 (24, 103)	122 (24, 238)	29 (13, 55)
Sichuan	220 (28, 458)	51 (33, 70)	165 (25, 297)	22 (14, 31)	202 (38, 400)	52 (21, 86)	123 (26, 233)	22 (9, 37)
Guizhou	107 (22, 189)	41 (27, 57)	86 (21, 142)	19 (12, 26)	91 (26, 164)	44 (17, 69)	77 (25, 138)	22 (8, 32)
Yunnan	41 (7, 74)	12 (8, 16)	31 (7, 54)	5 (3, 7)	34 (9, 63)	15 (5, 20)	26 (7, 47)	5 (2, 9)
Tibet	3 (0, 4)	0 (0, 0)	2 (0, 4)	0 (0, 0)	2 (0, 3)	0 (0, 0)	2 (0, 3)	0 (0, 0)
Shaanxi	183 (45, 341)	55 (36, 75)	105 (27, 191)	38 (24, 52)	162 (24, 398)	52 (22, 91)	103 (15, 275)	37 (15, 63)
Gansu	80 (20, 147)	21 (14, 29)	52 (12, 105)	13 (8, 17)	71 (10, 185)	22 (8, 36)	55 (6, 166)	12 (5, 21)
Qinghai	37 (7, 82)	9 (6, 12)	28 (5, 67)	6 (4, 9)	42 (2, 142)	7 (3, 15)	35 (1, 124)	6 (2, 11)
Ningxia	70 (14, 139)	29 (19, 41)	47 (9, 96)	28 (19, 39)	65 (7, 155)	29 (12, 50)	52 (4, 149)	29 (12, 48)
Xinjiang	33 (7, 67)	14 (9, 20)	41 (6, 101)	6 (4, 9)	34 (3, 102)	15 (5, 24)	53 (2, 183)	6 (2, 11)

Supplementary Table 7. Information on the sampling sites and parameters of flue gases.

Unit	Capacity (MW)	Boiler type	Coal	APCD
P1	2×350	PCB	bituminite	SCR+ESP+S-WFGD+WESP
P2	2×680	PCB	bituminite	SCR+ESP+S-WFGD+WESP
P3	2×25	CFB	bituminite	SCR+ESP+L-WFGD+WESP
P4	2×350	CFB	bituminite	SCR+ESP+L-WFGD+WESP
P5	135	CFB	bituminite	SCR+FAB+Sd-FGD+WESP
P6	600	PCB	bituminite	SCR+ESP+L-WFGD+WESP
P7	1000	PCB	bituminite	SCR+ESP+L-WFGD+WESP

Note: APCD, air pollution control device;

PCB, pulverized coal-fired boiler;

CFB, circulating fluidized bed;

SCR, selective catalytic reduction;

ESP, electrostatic precipitator;

S-WFGD, seawater wet flue gas desulfurization;

WESP, wet electrostatic precipitator;

L-WFGD, limestone-gypsum wet flue gas desulfurization;

FAB, fabric baghouse;

Sd-FGD, semidry flue gas desulfurization.

Supplementary Table 8. Major air pollution control measures taken in coal-fired power plant and residential sector during 2013−2017.

Sector	Standards and control measures	2013	2014	2015	2016	2017
Coal-fired power plant	Emission standard	GB 13223-2011				
			Ultra-low emission standard			
	Control technology	Installed with de-sulfurization (97%), de-nitrification (87%), and de-dust devices (> 80%)				
			Retrofitting with ULE technologies			
Residential sector	Emission standard	No specific emission standards				
	Control technology	No end-of-pipe control devices				
	Energy substitution	No specific policies				Replacing coal with clean fuels in BTH and surrounding area

Supplementary Table 9. Pollutant emission standards implemented on power plant sector in different regions, including China, the European Union, and the United States.

Pollutants	Types	China (2011 standard)[1]	China (ULE standard)[3]	European Union (2017 standard)				United States (MATS)
				<100 MW	100-300 MW	>300 MW, PC boiler	>300 MW, CFB	
NO_x (mg/m^3)	New plants	100	50	100-150	50-100	50-85	65-85	117
	Existing Plants (built after 1/1/04)	100		100-270	100-180	<85-150	65-150	
	Existing Plants (built before 1/1/04)	200						
SO_2 (mg/m^3)	New plants	100	35	150-200	80-150	10-75	20-75	160 (built after 1997)
	Existing Plants (28 provinces)	200		150-360	95-200	10-130	20-180	640 (built between 1978-1996)
	(4 provinces with high sulfur coal)	400						
PM (mg/m^3)	New plants	30	10	2-5				22.5
	Existing plants			2-18	2-14	2-10	2-8	

Notes: Mercury and Air Toxics Standards (MATS);

PC boiler: pulverized coal-fired boiler;

CFB: circulating fluidized bed boiler.

Supplementary Table 10. Solid fuel quality information obtained by proximate analysis.

Index [a]	M_{ar} [b] (%)	A_d [b] (%)	V_{daf} [b] (%)	FC_d [b] (%)	$S_{t,d}$ [b] (%)	$Q_{net,\ ar}$ [b] (MJ/kg)
B1	8.6	31.54	26.25	49.54	0.68	20.02
B2	8.4	27.18	31.84	54.38	0.95	20.58
B3	9.0	43.21	44.01	32.52	1.11	13.04
B4	22.1	22.58	37.58	47.49	0.90	16.84
B5	6.75	26.47	29.93	N.A.[d]	0.62	21.49
B6	18.1	10.54	27.23	50.92	0.41	22.87
B7	18.1	10.54	27.23	50.92	0.41	22.87
B8	6.36	3.26	33.18	64.64	0.20	29.92
B9	4.12	5.51	34.60	61.79	0.20	28.31
B10	8.44	3.24	36.56	61.41	0.25	26.55
A11	0.7	7.78	8.27	84.59	0.19	26.89
A12	1.3	21.40	9.26	71.32	0.18	25.99
A13	7.6	18.49	4.28	78.02	0.5	24.26
Bio14[c]	9.3	2.93	79.80	19.6	0.02	19.33
Bio15[c]	14.2	11.06	80.43	17.41	0.15	13.42
Bio16[c]	5.9	8.69	79.42	18.79	0.07	15.55
Bio17[c]	8.6	10.89	82.23	15.84	0.24	14.90
Bio18[c]	9.3	8.85	74.22	16.93	0.10	15.04

Notes: [a] B, A, and Bio in the sample indexes indicate bituminous coal, anthracite, and biomass samples, respectively. Fuel samples B1 to B7 were burned in industrial boilers of CFPPs, while the other samples were burned in residential stoves.

[b] M_{ar} and $Q_{net,\ ar}$ represent the moisture content and net calorific value, respectively; A_d, FC_d, and $S_{t,d}$ represent the ash, fixed carbon, and total sulfur contents on a dry basis; V_{daf} represents the content of volatile matter on a dry and ash-free basis.

[c] The biomass samples were wood, rice residue, corn residue, wheat residue, and bean residue.

[d] N.A. means no available data.

Supplementary Table 11. Model performance statistics for the comparison between simulated and observed $PM_{2.5}$ concentrations in China in 2017.

Variable	Spring	Summer	Autumn	Winter	Average	Model performance criteria
Mean SIM ($\mu g/m^3$)	38.9	25.8	35.8	61.0	40.4	
Mean OBS ($\mu g/m^3$)	43.4	27.9	41.1	70.9	45.8	
NMB (%)	-10	-7	-13	-14	-12	
NME (%)	52	61	54	49	53	
MFB (%)	-19	-25	-18	-17	-20	± 60%
MFE (%)	59	68	61	55	61	± 75%

Supplementary Table 12. Comparison of source apportionment results of ambient $PM_{2.5}$ from power plants and residential combustion in China with other studies.

Method	Emission	Year	Target region	Sectoral contribution	References
WRF-CHEM	MIX	2010	China	Power plants 33.2% Residential sector 24.2%	Gao et al.[34]
WRF-CMAQ	Multiple inventories with data fusing	2013	China	Power plants 10.3% (mortality) Residential sector 21.7% (mortality)	Hu et al.[35]
CAMx-PSAT	MEIC (2010)	2014	Beijing	Coal combustion 1.3-25.9%	Zhang et al.[54]
WRF-CMAQ	ABaCAS	2015	China	Power plants 6% Residential sector 13%	Zhang et al[30]
PMF	-	2014	Beijing-Tianjin-Hebei region	Combustion (non-vehicle) 24-40%	Gao et al.[55]
PMF	-	2011	North China Plain	Biomass burning 15.8%	Yao et al.[38]
PMF	-	2015-2016	Heze city, Shandong province	Coal combustion 17.2% Biomass burning 7.0%	Liu et al.[36]
Multiple models	MEIC (2010)	2016	Beijing	Coal combustion 16% Biomass burning 9%	Liu et al.[37]
WRF-CMAQ	ABaCAS	2013	China	Power plants 1.6% Residential sector 14.1%	This study
WRF-CMAQ	ABaCAS	2017	China	Power plants 1.1% Residential sector 13.7%	This study

Note: PMF cannot separate different kinds of coal burning (power plants, industrial boilers, and residential combustion), which makes the comparison even more difficult.

Supplementary Table 13. $PM_{2.5}$ emission from coal-fired power plants, residential coal burning and residential biomass burning and the 95% confidence intervals (Unit: kt).

Source	2013	2017
Power plants	664 (465-864)	339 (237-441)
Residential coal	438 (245-977)	396 (222-883)
Residential biomass	1761 (194-3299)	1222 (135-2306)

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