Optically measured black and particulate brown carbon emission factors from real-world residential combustion predominantly affected by fuel differences

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There are 19 pages, including 6 tables and 10 figures

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S1: Calculation and Data Analysis

1. Modified Combustion Efficiency (MCE)

The MCE values reflecting burning conditions (~1for flaming phase and 0.7-0.9 for smoldering phase) were calculated with the following equation[1, 2]:

$$MCE=\frac{CO_2}{CO_2+CO}$$

2. Absorption Ångström Exponent (AAE)

The absorption coefficient (b_{abs}, Mm^{-1}, 10^{-6} m^{-1}) is calculated based on the following equation:

$b_{abs}(Mm^{-1})=\frac{ATN*A(mm^2)}{V(m^3)}$where ATN is the optical attenuation measured, A is effective area of filter and V is the air volume sampled.

Absorption Ångström Exponent (AAE) which could indicate the presence of BrC: The AAE of pure BC is considered as 1, and an AAE over 1 suggests the existence of BrC in the emissions, higher AAE means more fraction of BrC[3, 4] .The AAE values were determined as following:

$$AAE=-\frac{ln\left(\frac{b_{abs}(370)}{b_{abs}(880)}\right)}{ln\left(\frac{370}{880}\right)}$$

3. Absorption Emission Factor (AEF)

AEF (m^2/kg) which represents the light absorption cross section (m^2) of carbonaceous component per mass (kilograms) fuel used were determined were calculated with carbon-balance method:

$$\mathrm{AEF}(m^2/\mathrm{kg})=\frac{b_{abs}(Mm^{-1})*10^{-6}*EF_{CO_2}(g/kg)}{C_{CO_2}(g/m^3)}$$

4. Mass of BC and BrC

The filter-based ATN or absorption coefficient values were often found to be higher than those from the optical closure measurement[5], and there are many algorithms or approaches purposed in literature to correct the scattering, multiple scattering, and shadowing effects[6-10], so as to obtain the Mass Absorption Efficiency (MAE), or Mass Absorption Cross-section (MAC) values.

MAE is inversely proportional to the ATN as seen from the equation:

$$MAE_{BC}=\frac{ATN}{BC_S*C*R}$$

where C and R are usually empirical parameters to correct the multiple scattering and shadowing effects, respectively.

Therefore, to calculate BC mass loading (BCs, μg/cm^2) which can be converted to the BC mass concentration in air (μg/m^3), besides the ATN values directly from the instrument, it is necessary to know the C, R and MAE_{BC} values. Literature studies showed that C values can vary from 1.3 to 6.3[11-15] depending on sample types and measurement technologies, also, considerable variations exist in R values[8, 16]. For the MAE or MAC, literature-reported MAE values also vary greatly, from 2 to 50 m^2/g[3, 17-21], due to many factors such as source types (e.g., biomass or fossil fuel combustion), particle properties, and analysis method. For instance, Bond and Bergstrom[3] had suggested that the value of MAE was 7.5±1.2 m^2/g for uncoated soot particles. Shen et al.[17] measured MAE values of fuels combusted in residential cooking stoves using EC/OC analyzer and the biases of multiple scattering and shadowing effects were corrected in the calculation, finding that MAE values of 3.1 m^2/g, 6.6 m^2/g, 9.5 m^2/g, and 7.9 m^2/g for the combustion of wood, crop residue, pellet and coal, respectively. Cheng et al.[22] reported a MAE value (after biases corrected) of 3.0-3.2 m^2/g of EC from crop straw burning.

The choice of C, R, and MAE values is critical in accurately calculating the BC (also OC which could be more difficult because of very limited information available), but this is challenged because of very highly variable parameter values. The default value of MAE from the instrument manufacturer can be treated as a specific (or equivalent) MAE value, as this value is usually from the comparison experiments in which the calculated filter-based BC mass concentration based on the specific MAE value was close to the optically measured one. Thus, dividing the ATN by the default specific (equivalent) MAE value (e.g. 16.6 m^2/g here) gives the BC concentration comparable to those directly measured from the air without filter artifacts. Garland et al.[23] studied the relationship between the OT21 measured ATN and the EC from a Sunset Analyzer, deriving an equivalent MAE values of 13.7 m^2/g and 15.4 m^2/g for the cookstove emission samples from Cambodia and India, respectively. These values were close to the default 16.6 m^2/g by taking high variations across different experiments and uncertainties in EC analysis. By using ATN from the instrument without biases correction and a specific MAE value of 16.6 m^2/g, Ahmed et al.[24] found a good linear relationship (a slope of 0.91 with R^2=0.84) between the BC from the OT21 and the EC value from the thermal-optical analyzer. From a C value of 2.14 and a MAE of 7.5 m^2/g[3], as seen from the equation above, the estimated ECs values would be close to those using the default MAE of 16.6 m^2/g and uncorrected ATN. To our knowledge there was no such equivalent MAE values for BrC yet.

Thus, in the present analysis, in the calculation of BC and BrC mass on filter, we referred to the manufacturer default MAE values, like those done in some past studies[23-26]. This uncertainty should be paid more attention when generalizing the data. Given concerns on the correction of filter loading effects and uncertainties in the default specific MAE value in the estimation of carbon mass, we calculated BC with the correction for the shadowing and multiple scattering effects, and compared the results from the default specific MAE values.

The BC mass on the filter corrected (BCs', μg/cm^2) was estimated according to the following equation:

$$BC_s{}' = \frac{ATN}{MAE_{BC} * C * R(ATN)} \times 100$$

where the empirical factor C was introduced to correct the multiple scattering effect, which usually ranged from 1.3 to 3.5 in reported past studies. Here, an empirical C value of 2.14 was adopted[3, 17, 21, 27]. The parameter, R(ATN), was applied to correct the shadowing effect. R(ATN) can be estimated based on the following equation:

$$R(ATN) = (\frac{1}{f} - 1)(\frac{ln(ATN) - ln(10)}{ln(50) - ln(10)}) + 1 = (\frac{1}{f} - 1)\frac{ln(0.1ATN)}{1.609} + 1$$

where f values of 1.103 and 1.114 were suggested by Sandradewi et al.[8], and an f value of 1.1 was used following Shen et al.[17]and Zhang et al.[15].

MAE_{BC} at 880nm was calculated with the assumption of 1.0 for BC AAE value[3, 4, 28] and 7.5 m^2/g for the MAE_{BC} at 550 nm as suggested by Bond and Bergstrom[3] for uncoated soot particles, by using the following equation:

$$MAE_{BC}(\lambda) = k * \lambda^{-AAE},$$

The BC EFs (Table S2) corrected for the multiple scatting and shadowing effects (BC' EFs) were obtained and compared with the results by using the default specific MAE values recommended by the manufacturer. As seen in the Figure S1, the EFs were positively correlated (r=0.999, p<0.001), and the difference was statistically insignificant.

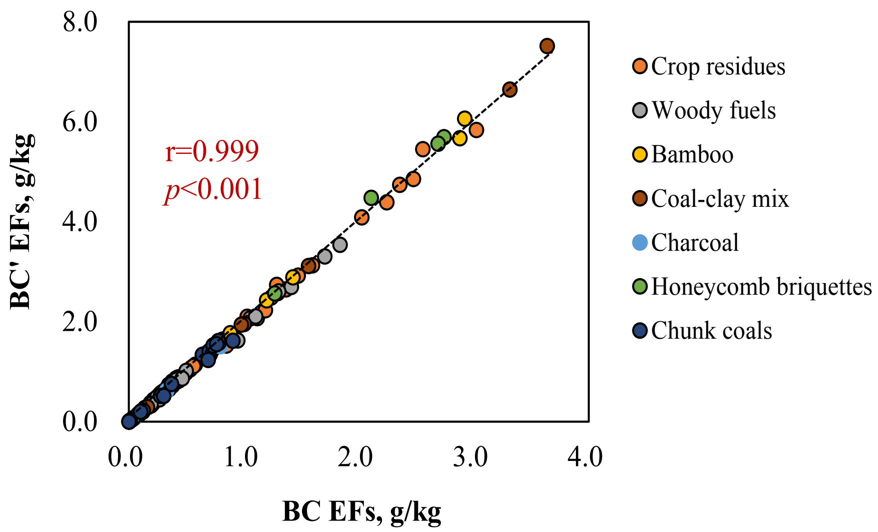

Figure S1 Comparison of BC EFs corrected for the shadowing and multiple scattering effect (BC' EF), and those calculated from the default specific MAE values recommended by the manufacturer (BC EF) for different fuels.

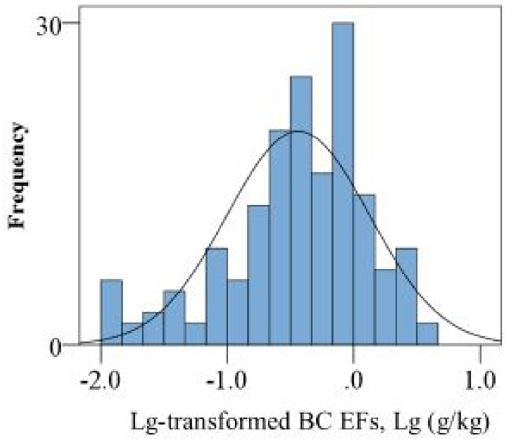

Figure S2 Distribution of measured BC EFs from residential combustion sources. Data are results from all combusted fuels.

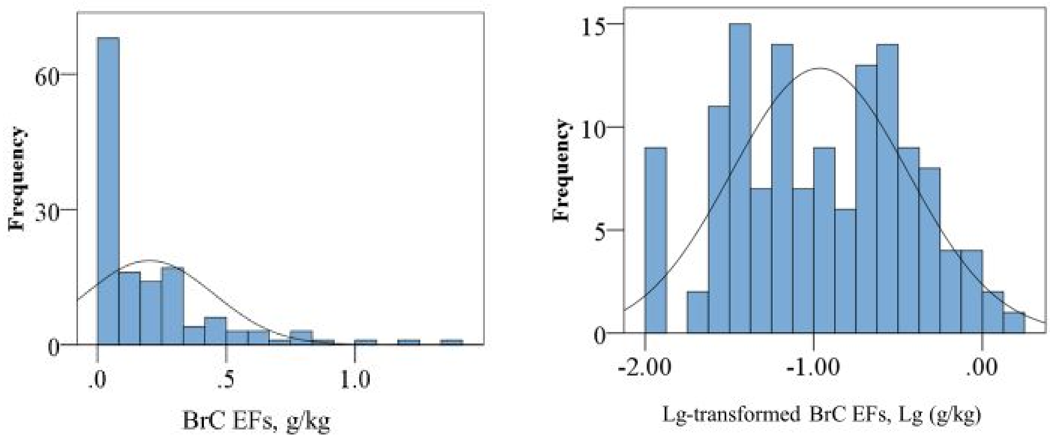

Figure S3 Distribution of measured BrC EFs from residential combustion sources. Data are results from all combusted fuels.

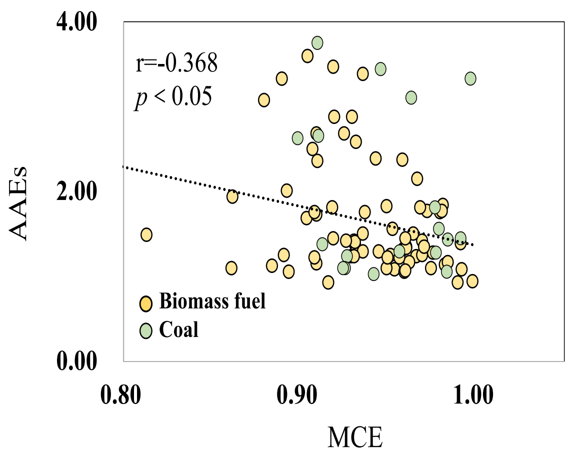

Figure S4 Correlations between AAE and MCE for solid fuels combusted in a residential stove.

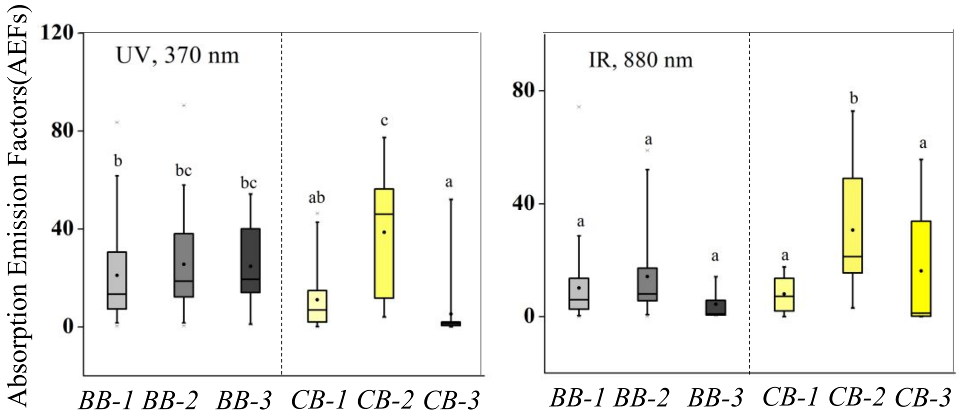

Figure S5 Absorption emission factors (AEFs) for solid fuels (CB-1 ~ 3 were chunk coals, coal-clay mix, and honeycomb briquettes, respectively. BB-1~3 were and charcoal, wood fuels, and crop residues, respectively) in residential stoves. Different letters indicate significant differences based on *p<0.05*.

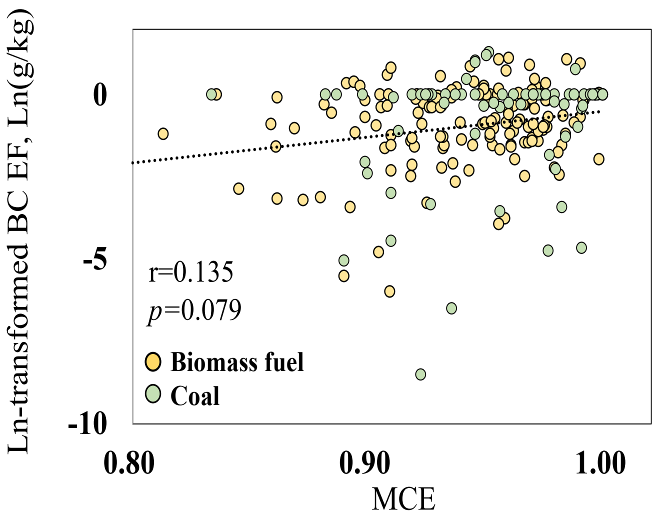

Figure S6 Correlations between BC EFs and MCE for solid fuels combusted in a residential stove.

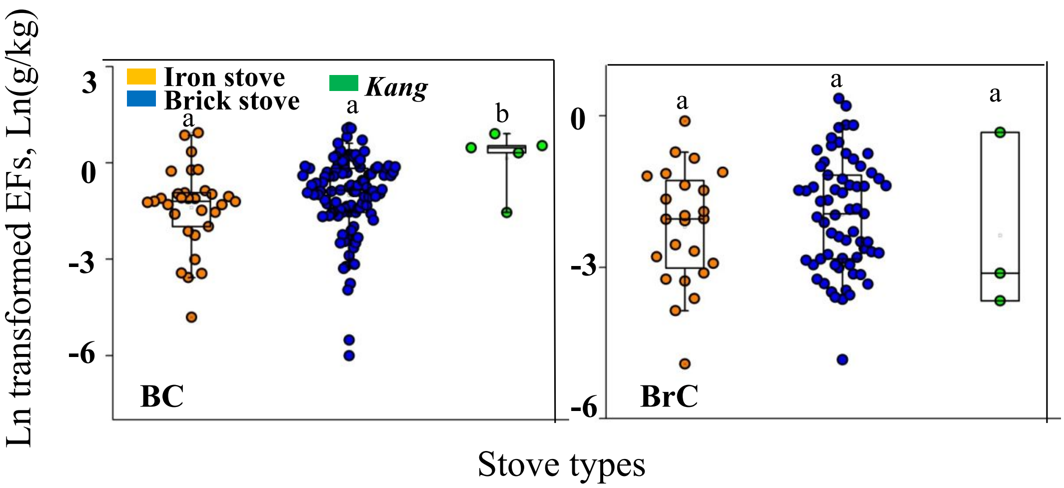

Figure S7 BC EFs and BrC EFs from burning of biomass fuels in residential stoves. Data shown are minimum, maximum, the 1st (P25), 2nd (P50) and 3rd (P75) quartile, and arithmetic means of the EFs. EFs are on the basis of dry fuel mass. Different letters indicate significant differences based on $p<0.05$.

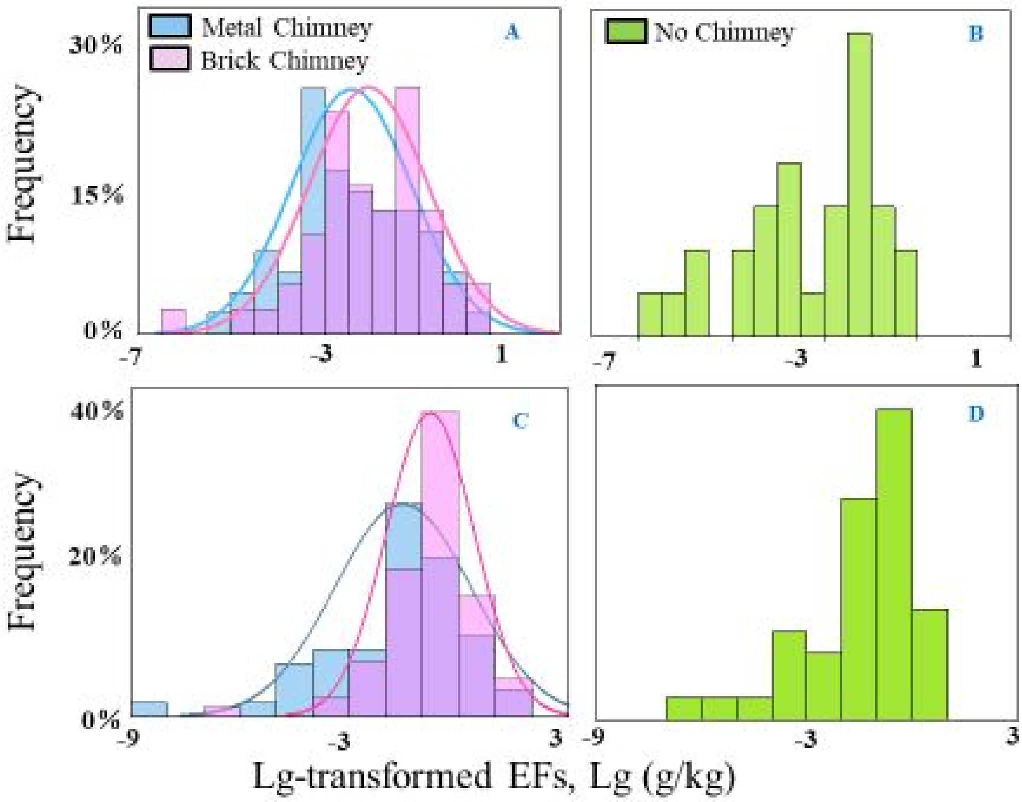

Figure S8 Frequency distribution of Ln-transformed BrC EFs (A and B) and BC EFs (C and D) from burning of solid fuels in residential stoves with/ without chimney.

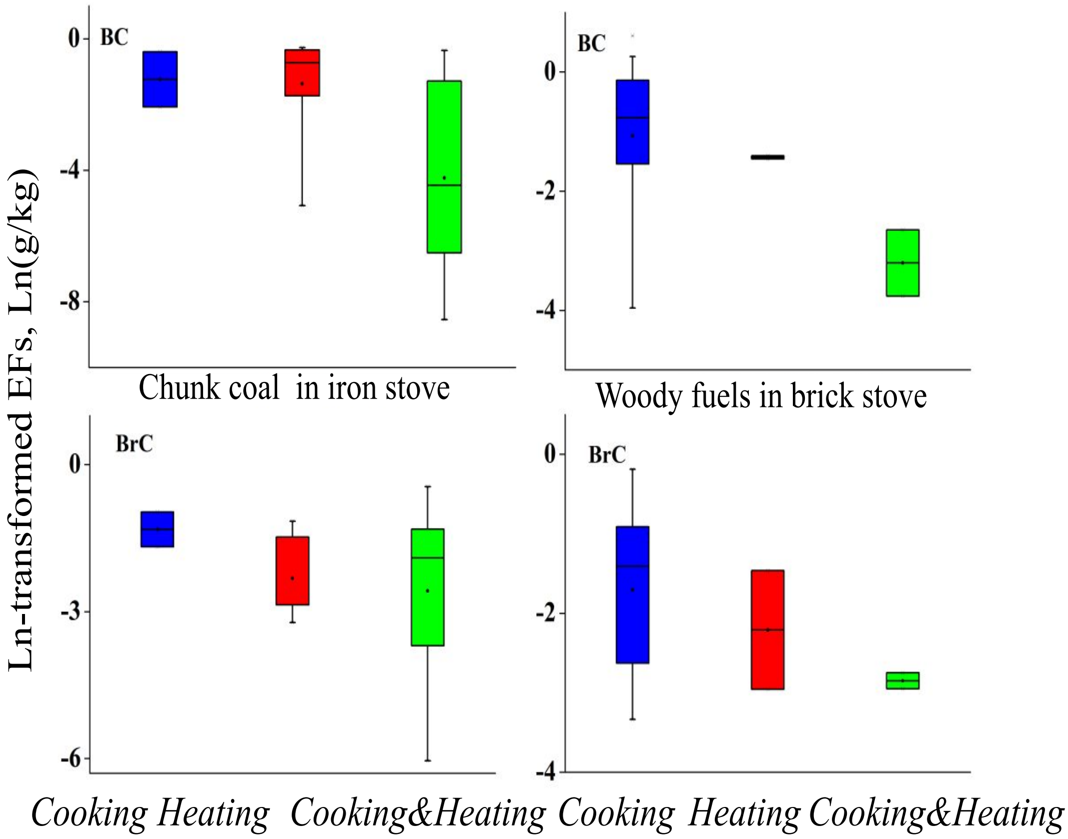

Figure S9 Ln-transformed BC and BrC EFs from burning of solid fuels in residential stoves for different daily activities (cooking or heating). Data shown are minimum, maximum, the 1st (P25), 2nd (P50) and 3rd (P75) quartile, and arithmetic means of the EFs. EFs are on the basis of dry fuel mass.

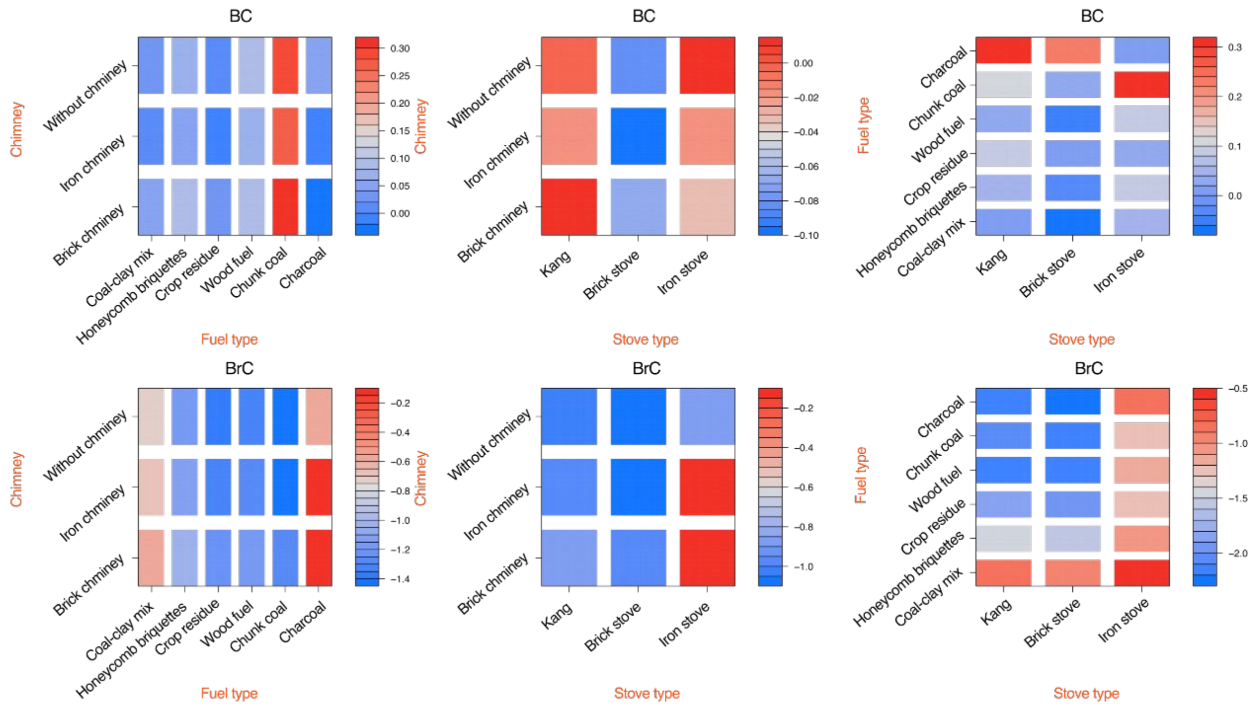

Figure S10 Interaction effects of the three influencing factors (stove type, fuel type and chimney) on EFs of BC and BrC. Stove type includes *Kang*, brick stove and iron stove. Fuel type includes coal-clay mix, honeycomb briquettes, crop residue, wood fuel, chunk coal and charcoal. The influencing factor – chimney includes brick chimney, iron chimney and without chimney.

Table S1 Testing location and fuel-stove information of the emission measurement in this study.

Area	Stove styles	Fuel styles	Chimney styles	Energy use
Henan (n=105)	brick stoves (43) iron stoves (62)	wood fuel: wood branch (17), wood log (8) crop residue: sesame straw (5), corn straw (1), soybean straw (1), O-straw (9), bamboo (1), corn cob (21) charcoal (1) Honeycomb briquette (41)	iron chimney（46） brick chimney（18） without（41）	heating (29) cooking (70) heating& cooking (6)
Hebei (n=105)	brick stove (61), iron stove (43)	wood fuel: wood logs (5) wood branch (14), crop residue: sesame straw (3), soybean straw (7), corn straw (4), O-straw (7) corn cob (20), bamboo (1) charcoal (6) chunk coal (35) coal-clay mix (3)	iron chimney (39) brick chimney (34) without (19) no record (13)	heating (29) cooking (71) heating& cooking (5)
Liaoning (n=40)	brick stove (13) iron stove (16) Kang (11)	wood fuel: wood log (7), wood branch (5) crop residue: corn straw (7), O-straw (1), corn cob (7) chunk coal (5) coal-clay mix (8)	iron chimney (25) brick chimney (10) no record (5)	heating (9) cooking (17) cooking& heating (14)
Inner Mongolia (n=18)	brick stove (7) iron stove (11)	wood fuel: wood log (8) wood branch (1) crop residue: corn cob (1) chunk coal (8)	brick chimney (16) iron chimney (2)	heating (10) cooking (4) heating& cooking (4)
Shanxi (n=41)	brick stove (5) iron stove (31) Kang (5)	wood fuel: wood log (4), wood branch (6) crop residue: corn cob (5) honeycomb briquette (5) chunk coal (19) coal-clay mix (2)	brick chimney (8) iron chimney (32) no record (1)	heating (7) cooking (8) cooking& heating (26)
Beijing (n=10)	iron stove (5) brick stove (5)	wood fuel: wood log (3), wood branch (2) honeycomb coal (2), chunk coal (3)	without chimney (9) iron chimney (1)	heating (1) cooking (4) cooking& heating (5),
Tianjin (n=6)	iron stove (6)	wood fuel: wood log (1), wood branch (1) chunk coal (4)	iron chimney (6)	heating (2) cooking (2) cooking& heating (2)

Area	Stove styles	Fuel styles	Chimney styles	Energy use
Chongqing (n=18)	brick stove (18)	wood fuel: wood log (7), wood branch (3) crop residue: corn cob (3), bamboo (5)	without chimney (2) iron chimney (16)	cooking (18)

Table S2 Fuel mass (g/kg) and fuel energy-based (g/MJ) emission factors of BC for different fuels burned in residential stoves in which BC mass on the filter was corrected for the shadowing and multiple scattering effects.

	BC' Fuel mass-based EFs	BC' Fuel energy-based EFs
Woody fuel	0.93±0.86	0.049±0.045
wood branch	0.94±0.82	0.049±0.043
wood log	0.92±0.92	0.048±0.048
Bamboo	3.4±2.0	0.19±0.11
Crop residue	1.5±1.5	0.086±0.086
corn cob	1.4±1.3	0.0801±0.075
corn straw	2.5±1.7	0.14±0.098
sesame straw	1.7±1.9	0.098±0.11
soybean straw	0.78±0.42	0.045±0.024
other straws	0.80±0.86	0.046±0.050
Charcoal	0.46±0.64	0.014±0.020
Coal-clay mix	3.3±2.7	0.18±0.15
Honeycomb briquette	2.0±2.4	0.059±0.071
Chunk coal	0.81±0.65	0.025±0.017

Table S3 The HV values of the fuels

Fuel	**HV, MJ/kg**	**Reference**
wood fuel	19.1	[29]
crop residue	17.4	[30]
bamboo	18.3	[31]
charcoal	32.0	[29]
honeycomb briquette	34.0	[32]
chunk coal	33.0	[32]
coal-clay mix	18.0	[32]

Table S4 Absorption emission factors (AEFs) for solid fuels in residential stoves.

Fuel	**Absorption emission factors (m^2/kg)**							
	880 nm				**370 nm**			
	Average± standard	Median	1st quartile	4st quartile	Average± standard	Median	1st quartile	4st quartile
wood fuel	10.2±12.1	6.01	2.68	13.6	21.1±19.4	13.4	7.34	30.6
crop residue	14.2±14.4	8.08	5.60	17.2	25.6±19.8	18.7	12.5	36.5
charcoal	4.41±5.90	0.960	0.575	5.81	24.7±19.1	19.4	15.3	35.0
coal-clay mix	30.7±23.4	21.2	18.2	40.6	38.7±26.8	46.0	12.9	55.3
honeycomb briquette	16.2±22.2	1.25	0.183	29.5	5.29±12.8	1.46	0.616	2.02
chunk coal	8.01±6.42	7.21	2.00	13.6	11.1±12.9	6.93	2.10	14.1

Table S5 Emission factors of BC and BrC from BrC from burning of different fuel-stove combinations.

Fuel-Stove	BC EFs, g/kg	BrC EFs, g/kg
honeycomb briquette-iron stove	0.97±1.2	0.12±0.27
chunk coal-iron stove	0.36±0.31	0.18±0.17
coal-clay mix-iron stove	1.6±1.3	0.086±0.089
charcoal-iron stove	0.25±0.33	0.37±0.31
wood fuel-iron stove	0.41±0.38	0.12±0.093
wood fuel-brick stove	0.49±0.43	0.25±0.24
wood fuel-Kang	0.96±1.0	0.26±0.40
crop residue-iron stove	0.61±0.82	0.15±0.13
crop residue-brick stove	0.72±0.70	0.25±0.31
crop residue-Kang	1.8±0.59	

Table S6 Partial effects of stove type, fuel type and chimney on the EFs of BC and BrC based on generalized linear model (GLM). Stove type includes Kang, brick stove and iron stove and Kang was set as the reference. Fuel type includes coal-clay mix, honeycomb briquettes, crop residue, wood fuel, chunk coal and charcoal, and coal-clay mix was set as the reference. The influencing factor – chimney includes brick chimney, iron chimney and without chimney, and brick chimney was set as the reference. Estimate: coefficient of predictor and intercept. Std. Error: standard error. Z value: the regression coefficient divided by standard errors. p-value is the statistical test of the independent variables and the significant level is set at the level of 0.05.

BC	**Estimate**	**Std. Error**	**Z value**	**p-value**
(Intercept)	2.6178	0.4149	6.31	<0.001
Kang	ref			
Brick stove	-0.8788	0.3115	-2.821	0.005388
Iron stove	-0.9616	0.3306	-2.908	0.004141
Coal-clay mix	ref			
Honeycomb briquettes	-0.5711	0.3412	-1.674	0.096082
Crop residue	-1.0211	0.2933	-3.481	<0.001
Wood fuel	-1.2312	0.3103	-3.967	0.000109
Chunk coal	-1.1864	0.3047	-3.893	0.000144
Charcoal	-1.2978	0.4016	-3.231	0.001491
Brick chimney	ref			
Iron chimney	-0.1056	0.1395	-0.757	0.450111
Without chimney	-0.1347	0.1348	-0.999	0.319221
BrC	**Estimate**	**Std. Error**	**Z value**	**p-value**
(Intercept)	0.16752	0.16554	1.012	0.3135
Kang	ref			
Brick stove	0.01475	0.13627	0.108	0.914
Iron stove	-0.06837	0.14273	-0.479	0.6328
Coal-clay mix	ref			
Honeycomb briquettes	0.04199	0.11588	0.362	0.7177
Crop residue	0.08471	0.11521	0.735	0.4635
Wood fuel	0.02414	0.11949	0.202	0.8402
Chunk coal	0.09976	0.1089	0.916	0.3613
Charcoal	0.30004	0.14071	2.132	0.0349
Brick chimney	ref			
Iron chimney	-0.03119	0.06302	-0.495	0.6215
Without chimney	-0.01591	0.06145	-0.259	0.7962

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