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Li, Qing, Jiang, Jingkun, Wang, Shuxiao, Rumchev, Krassi, Mead-Hunter, Ryan, [Morawska, Lidia](#), & Hao, Jiming  
(2017)

Impacts of household coal and biomass combustion on indoor and ambient air quality in China: Current status and implication.  
*Science of the Total Environment*, 576, pp. 347-361.

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<https://doi.org/10.1016/j.scitotenv.2016.10.080>



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## HIGHLIGHTS

- Contributions of CO, PM<sub>2.5</sub>, BC, and PAHs emissions from household combustion more than one-third.
- Relative contributions of PM<sub>2.5</sub> and BC in poor regions are four times higher than that in rich regions.
- Chimney can reduce indoor PM<sub>2.5</sub> level to be about 20% when burning dirty solid fuels in stoves.
- PM<sub>2.5</sub> exposure level of housewives is about 2~4 times of that of adult men in poor rural regions.
- PM<sub>2.5</sub> and BC EFs increase with solid fuel volatile matter content and up to ~100 times difference.

## 32 **ABSTRACT**

33 This review briefly introduces current status of indoor and ambient air pollution originating from  
34 household coal and biomass combustion in mainland China. Owing to low combustion efficiency,  
35 emissions of CO, PM<sub>2.5</sub>, black carbon (BC), and polycyclic aromatic hydrocarbons have significant  
36 adverse consequences for indoor and ambient air qualities, resulting in relative contributions of more  
37 than one-third in all anthropogenic emissions. Their contributions are higher in less economically  
38 developed provinces, such as Guizhou (61% PM<sub>2.5</sub>, 80% BC), than that in more developed ones, such  
39 as Shanghai (4% PM<sub>2.5</sub>, 17% BC). Chimneys can reduce ~80% indoor PM<sub>2.5</sub> level when burning dirty  
40 solid fuels, such as plant materials. Due to spending more time near stoves, housewives suffer much  
41 more (~2 times) PM<sub>2.5</sub> than the adult men, especially in winter in northern China (~4 times).  
42 Improvement of stove combustion/thermal efficiencies and solid fuel quality are the two essential  
43 methods to reduce emissions. PM<sub>2.5</sub> and BC emission factors (EFs) have been identified to increase  
44 with volatile matter content (V<sub>d</sub>) in traditional stove combustion. EFs of dirty fuels are two orders  
45 higher than that of clean ones. Switching to clean ones, such as semi-coke briquette, was identified to  
46 be a feasible path for reducing >90% PM<sub>2.5</sub> and BC emissions. Otherwise, improvement of thermal and  
47 combustion efficiencies by using under-fire technology can reduce ~50% CO<sub>2</sub>, 87% NH<sub>3</sub>, and 80%  
48 PM<sub>2.5</sub> and BC emissions regardless of V<sub>d</sub>. However, there are still some knowledge gaps, such as,  
49 inventory for temporal impact of household combustion on air quality, statistic data for deployed clean  
50 solid fuels and advanced stoves, and effect of socioeconomic development. Additionally, further  
51 technology research for reducing air pollution emissions is urgently needed, especially low cost and  
52 clean stove when burning any solid fuel. Furthermore, emission-abatement oriented policy should base  
53 on sound scientific evidence to significantly reduce emissions.

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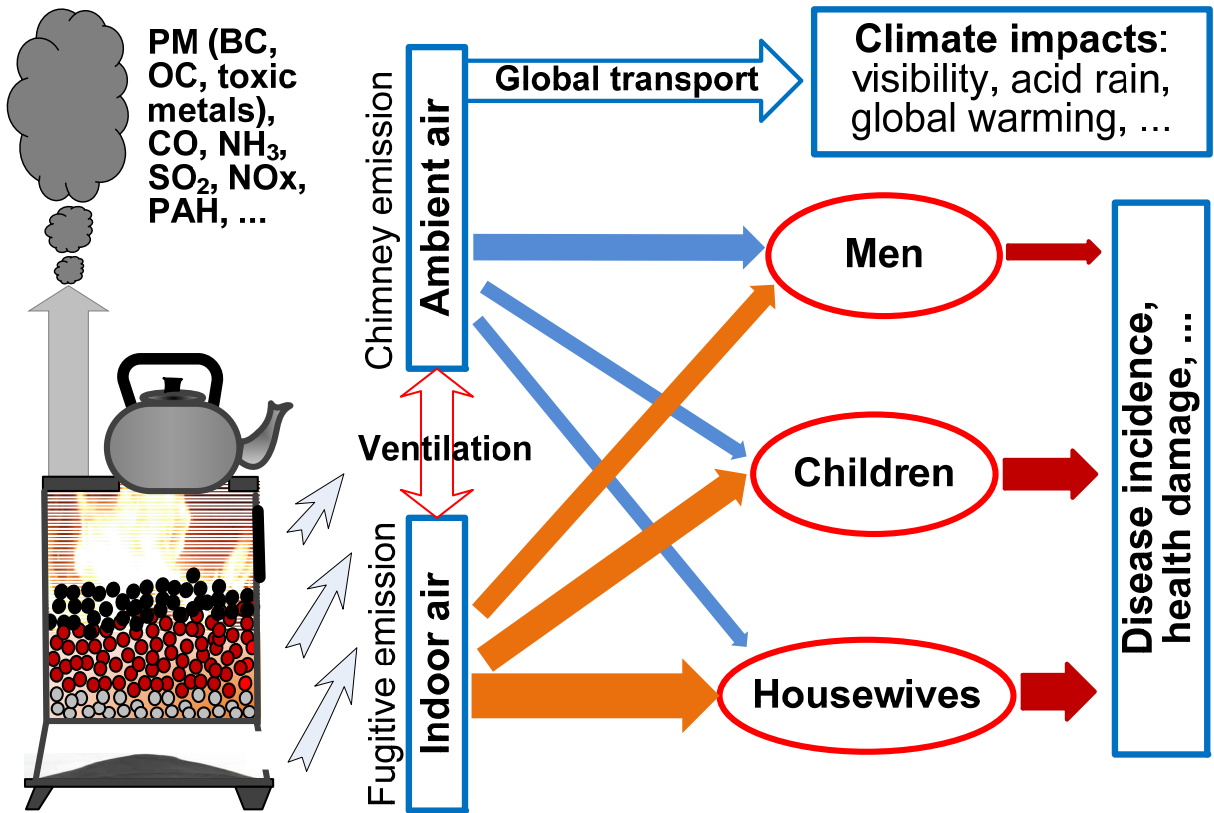
56 **Keywords:**

57 Particulate matter emissions, coal burning emissions, household stove, human exposure, air quality

58 control, pollution emissions from solid fuel combustion

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### GRAPHIC ABSTRACT



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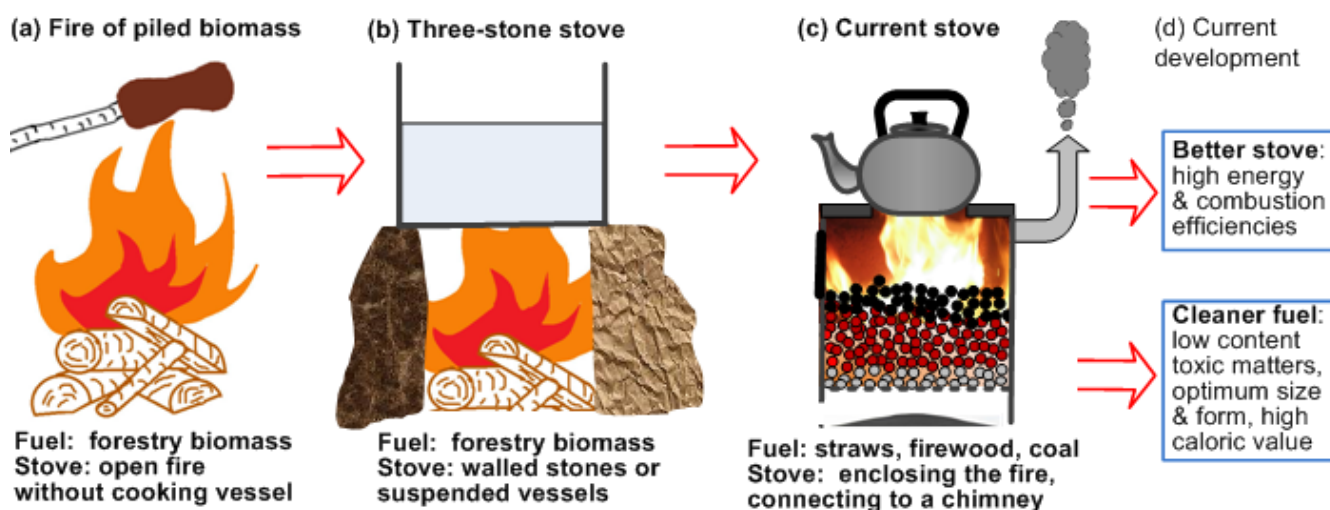
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## 81 **1. Introduction**

82 Solid fuel combustion for heating and cooking was a source of energy used by early hominids  
83 from about 1.6 million years ago (James, 1989; Roebroeks and Villa, 2011). The controlled use of  
84 biomass combustion has been commonly seen as a breakthrough adaptation in human evolution  
85 (Brown et al., 2009). The controlled fire offered human a survival advantage through cooking and  
86 heating, as well as protecting people from attacks from predatory species in China for around the last  
87 0.5 million years (James, 1989; Perez-Padilla et al., 2010). The earliest indoor air quality problems may  
88 have been caused by the use of fires inside stone-age dwellings. People brought air pollutants into the  
89 indoor living space and breathed in pollutants. Early humans may have identified that poor air quality  
90 was principally a problem in the indoor environment, resulting in early attempts to mitigate these  
91 problems through ventilation (Matson and Sherman, 2004). The Banpo villagers in China incorporated  
92 chimneys into houses to remove the products of combustion used for heating, lighting or cooking from  
93 around 4000-5000 Before Christ (Li and Jones, 2000). The living environment has been changed over  
94 the past 10,000 years, while household combustion has also evolved via improving stove structure and  
95 cooking vessels. Additionally, various solid fuels have been employed for combustion in Chinese  
96 households, mainly including biomass in the form of wood, dung and crop residues, as well as coal  
97 since approximately 220 AD. Figure 1 shows a brief illustration of the developments related to  
98 household solid fuel combustion.





99  
100 **Fig. 1.** Brief illustration of the development for household solid fuel combustion from the hominid time  
101 to the current status.

102

103 Why pollutant species are released during household solid fuel combustion? Ideally, complete  
104 combustion of fuels containing carbon, hydrogen, and oxygen should only produce  $\text{CO}_2$  and  $\text{H}_2\text{O}$ .  
105 However, in reality we know that complete combustion is impossible, as household stoves are not ideal  
106 combustors and the fuel used is in bulk form which may limit the combustion rate and the amount of  
107 oxygen available. In a substoichiometric environment, the incomplete combustion likely results in the  
108 production of carbon monoxide (CO), volatile organic compounds (VOCs), polyaromatic hydrocarbons  
109 (PAHs), and particulate matter (PM). We must also consider the formation of nitrogen oxides ( $\text{NO}_x$ ) if  
110 the combustion temperature is high enough, or there is nitrogen element present in the solid fuel (e.g.  
111 coal). In general, solid fuels contain nitrogen (N) and sulphur (S), as well as a number of mineral  
112 elements, including; Al, Si, Ca, Fe, K, Mg, Na, and P, and a number of potentially toxic trace materials  
113 (e.g., As, Pb, Hg, Mn, Cd, Co, and Cr). Table 1 lists the overall features of elemental concentrations in  
114 Chinese biomass and coals, and this list suggests that Chinese solid fuels always contain the most  
115 significant and toxic trace elements, as well as nitrogen and sulphur. Even if the ideal stove were to be  
116 used to burn them, sulfur dioxide ( $\text{SO}_2$ ) and  $\text{NO}_x$  would still be released and mineral elements  
117 including toxic ones would also be released in gaseous and/or particulate-bound states. Given the

118 incomplete nature of the combustion in household stoves, we must also consider the formation and/or  
 119 release of other nitrogenous compounds, such as NH<sub>3</sub>, and the emission of elemental/organic carbon  
 120 (EC/OC). These processes are briefly summarized in Fig. 2. Emitted PM, toxic metals, NH<sub>3</sub>, and PAHs  
 121 are harmful to the health of exposed people, while the global climate is also impacted, such as that SO<sub>2</sub>  
 122 and NO<sub>x</sub> are precursors to acid rain, NO<sub>2</sub> and reactive organic gases precursors to ozone. Thus indoor  
 123 and ambient air qualities are closely related to the household solid fuel combustion.

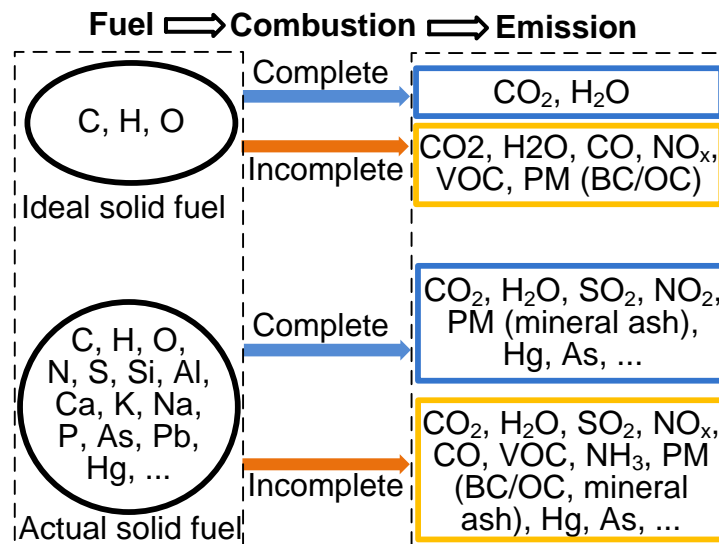
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125 **Table 1.** Characteristics of Chinese biomass (crop residue and firewood) and coals. These values were  
 126 summarized and averaged from 12 types of agricultural biomass (including rice straw, wheat straw,  
 127 corn stalk, and cotton stalk), 9 types of forestry biomass (including birch, spruce, and willow), and 3  
 128 types of coals from the previous report (Liao et al., 2004), while the listed major mineral elements were  
 129 obtained from another report (Dai et al., 2012). A<sub>d</sub>, V<sub>d</sub>, FC<sub>d</sub>, and S<sub>t,d</sub> indicate the ash, volatile matter,  
 130 fixed carbon, and total sulfur contents on a dry basis, respectively, while M and Q indicate the moisture  
 131 content and net calorific value, respectively.

Composition		Crop residue	Firewood	Coal
Proximate analysis	M (wt%)	8.3	8.3	2.8
	A <sub>d</sub> (wt%)	8.6	4.4	20.1
	V <sub>d</sub> (%)	66	71	28
	FC <sub>d</sub> (%)	17	16	49
	Q (MJ/kg)	17	19	34
Ultimate analysis	C (%)	43	48	64
	N (%)	0.94	0.41	1.13
	O (%)	43.8	41.6	10.1
	H (%)	6.65	7.32	3.97
	S (%)	0.25	0.12	0.97
elements (%)	Al	0.20	0.06	3.17
	Si	0.007	0.001	3.95
	Ca	1.20	0.95	0.88
	Fe	0.24	0.16	3.40

	K	1.17	0.38	0.16
	Mg	0.39	0.13	0.13
	Na	0.14	0.02	0.12
	P	0.14	0.06	0.04
Trace mineral elements (µg/g)	Ni	5.0	15.3	13.9
	Pb	22.6	7.8	20.9
	V	3.7	1.6	76.5
	Cd	0.4	0.4	0.2
	Co	1.6	0.8	8.5
	Cr	7.2	8.6	36.8
	Cu	27.1	33.8	27.5
	As	1.3	0.7	14.5

132



133

134 **Fig. 2.** Illustration of the pollutant emissions from complete and incomplete combustions of the

135 cleanest (ideal) and actual solid fuels.

136

137 Why the indoor air pollution from household combustion has attracted an increasing concern in

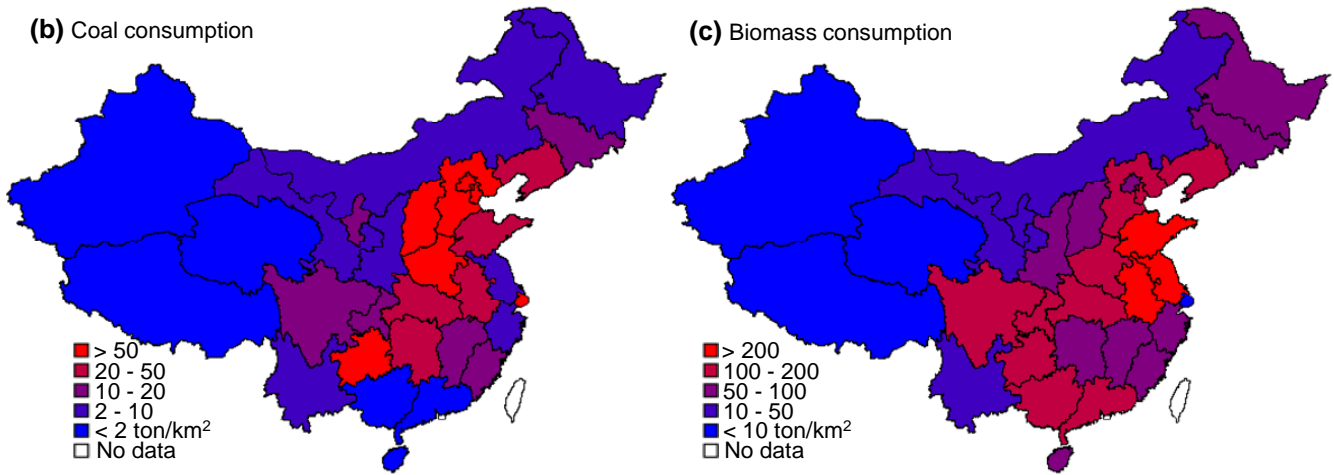
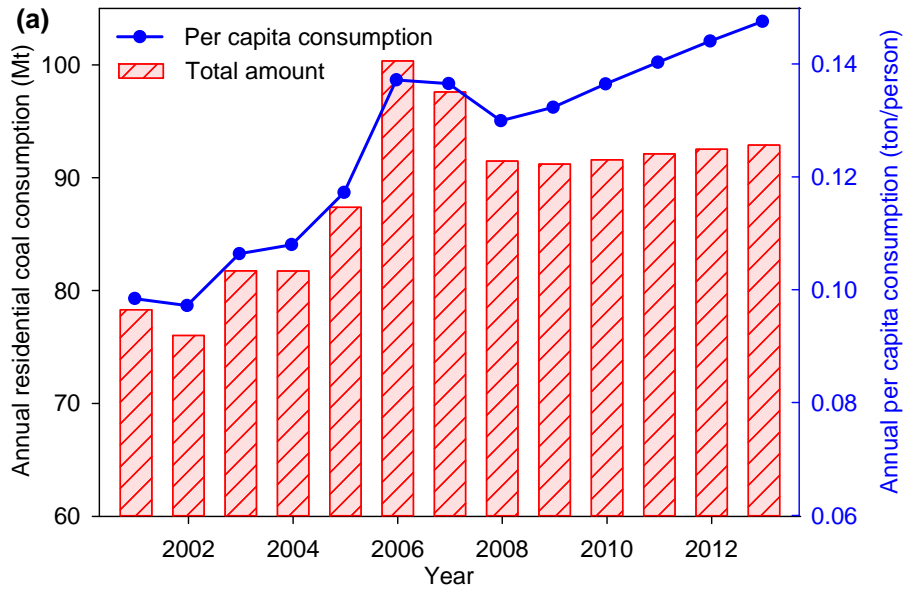
138 nowadays China? Household pollutant levels originating from the household solid fuel combustion is

139 mainly determined by the fuel quality, stove technology (including body leakage, chimney quality, and

140 combustion technology), and house ventilation (including air exchange rate with ambient air and other

141 rooms) (Peabody et al., 2005). Other conditions such as ambient temperature and geographic variables

142 should be taken into account for a detailed consideration of the potential determinants of exposure  
143 levels (Smith et al., 2012). The fuel quality and the stove technology have been gradually improving  
144 under the fast development of China's economy situation during the past several decades. Most rural  
145 buildings have been rapidly replaced by houses with reinforced concrete structure of increased  
146 airtightness and high use efficient of energy, however, some of the old sources of indoor air pollution  
147 that may not have been a problem under high ventilation rates are now creating problems (Matson and  
148 Sherman, 2004). Additionally, since per capital consumption ability increases with per capita income of  
149 rural family including rural-to-urban migrants, larger amounts of solid fuels are used. The per capital  
150 consumption of household coals, mainly used for heating in the northern part of the country, had  
151 increased from about 0.10 ton/person in 2001 to about 0.15 ton/person in 2013 in mainland China, see  
152 Fig. 3(a). Figures 3(b) and 3(c) suggest that coal and biomass are still widely used as residential solid  
153 fuels in mainland China. Residential coal has been mainly consumed in the household heating and coal  
154 supply regions, while biomass has been mainly used as residential cooking fuel in large agricultural and  
155 economic less economically developed regions.



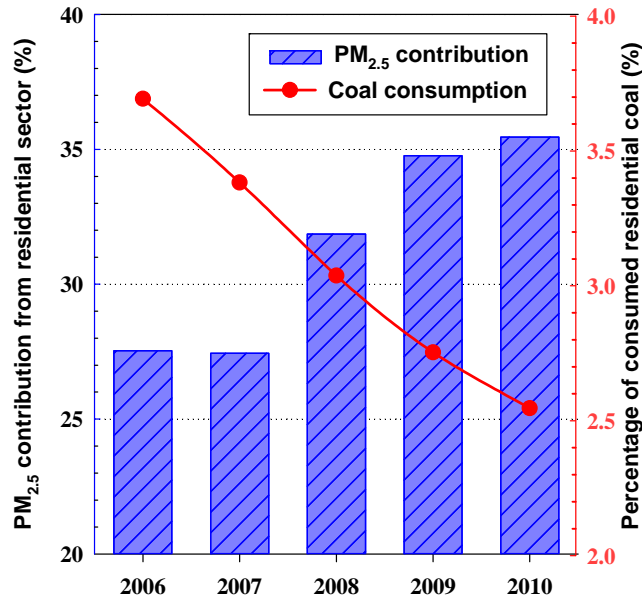
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157 **Fig. 3.** (a) Annual residential coal consumption and per capital consumption during 2001-2013 in  
 158 mainland China, and geographic distributions of residential (b) coal and (c) biomass consumption in  
 159 mainland China in 2007. The amount of residential coal consumption was directly obtained from China  
 160 energy statistical yearbook (NBSC, 2015), while per capital consumption value was the residential coal  
 161 amount divided by rural population, including people living in rural and rural-to-urban migrants.

162

163 Why household combustion has been considered to be one of major anthropogenic contributors of  
 164 atmospheric pollutants in contemporary China? As the China's population had grown from about 30  
 165 million in 221 Before Christ (China firstly unified) to 1.34 billion in 2010, the household combustion

166 of biomass and coal has significantly increased, and thus, inevitably, the potential adverse impact of the  
167 household combustion on ambient air quality. In addition, severe haze pollution episodes have been  
168 frequently occurring in China over the past decade (Guo et al., 2014; Huang et al., 2014a). Pollution  
169 emitted from the household solid fuel combustion has been attracting increasing public concern,  
170 although the percentage of fuel consumed for household use compared to that used for all other  
171 activities including industrial combustion, is very low. Comparing to industrial boilers, household  
172 stoves often feature low thermal efficiency and very incomplete combustion without air pollution  
173 control device. For instance, emission factors (EFs) of primary PM<sub>2.5</sub> (PM of aerodynamic diameter  
174 equal to or less than 2.5 μm) from household coal combustion can be ~ 100 times higher than those  
175 from power plant coal boilers (Zhang et al., 2008). Figure 4 shows that the contribution of primary  
176 PM<sub>2.5</sub> emissions from the residential coal consumption to that from all coal consumption activities in  
177 China had increased from 27.5% in 2006 to 35.5% in 2010 (Wang et al., 2014d), although the ratio of  
178 consumed coal for the residential activity had decreased from 3.7% to 2.5%. It has been estimated that  
179 in China's annual anthropogenic emissions, about 36% of primary PM<sub>2.5</sub>, 53% of EC, 62% of PAHs are  
180 from residential solid fuel combustion (Huang et al., 2014c; Shen et al., 2015; Shen et al., 2013b; Wang  
181 et al., 2012). Thus, the pollutant emission from the residential solid fuel use is a major anthropogenic  
182 source of the air pollution, while hundreds of the pollutant species are thought to increase health risk.  
183 The pollutants currently held to be the most important issue in China, including PM<sub>2.5</sub> of containing  
184 EC/OC and toxic trace components, CO, SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub>, PAHs, and VOCs.



185  
 186 **Fig. 4.** The percentage of primary PM<sub>2.5</sub> emission from residential coal consumption in that from all  
 187 coal consumption activities and the ratio of consumed coal for residential activity in mainland China  
 188 during 2006-2010. *Data source:* China energy statistical yearbook (NBSC, 2015) and emission  
 189 inventory (Wang et al., 2014d).

190  
 191 These pollutants affect both the indoor and the ambient air quality to a large extent, causing  
 192 human health damage and climate change (Anenberg et al., 2013; Bond et al., 2013; Lim et al., 2013).  
 193 Epidemiologic studies have confirmed that China's household air pollution can increase health risks,  
 194 such as, respiratory symptoms, asthma, lung function reduction, lung cancer, neural tube defects, heart  
 195 disease, elevated blood pressure, and immune system impairment. The household air pollution was also  
 196 reported as leading to over one million premature deaths in China.

197 Pollutant emissions impacting on the ambient air quality will be firstly introduced by summarizing  
 198 results from published emission inventories and source appointments. Impacts from major emitted  
 199 pollutants on the indoor air quality will be analyzed including the consideration of the fuel/stove  
 200 combustion and the household ventilation. An introduction of personal inhalation exposure level will

201 be discussed. Aiming at reducing air pollution emissions, some knowledge gaps and implications for  
202 clean household fuel and stove technologies will be presented at the end.

## 203 **2. Impacts on ambient air quality**

204 Pollution emitted from major anthropogenic emission sources, including solid fuels burned in  
205 residential stoves and industrial boilers, as well as petrol and diesel burned in vehicles, is a regionally  
206 and globally atmospheric issue (Guo et al., 2014; Huang et al., 2014a). About 14 Mt of PM<sub>2.5</sub> and 2 Mt  
207 of black carbon (BC) were annually emitted to the atmosphere in China (Bond et al., 2004; Huang et al.,  
208 2014c; Wang et al., 2012). The emitted pollutants interact to produce a mixture of hundreds of different  
209 and hazardous chemicals known as secondary pollutants via physical processes and chemical reactions  
210 in the atmosphere. This process can cause a range of health outcomes such as respiratory illnesses  
211 including lung cancer (Wang et al., 2014e), as well as global climate changes including global warming,  
212 visibility decrease, and acid rain (Bond et al., 2013; Stocker et al., 2013).

213 Since 1980s, fast urbanization development in China leads to a decrease of household solid fuel  
214 user proportion, however, there are still approximately 43% (about 600 million people) and 30% of  
215 China's households utilized them for household cooking and heating in 2012, respectively (Duan et al.,  
216 2014). Household combustion using waste (see Fig. 1b) and traditional low efficient stoves is still a  
217 common practice in rural China. Owing to incomplete combustion without air pollution control devices,  
218 EFs of many pollutants for solid fuels burned in household stoves are two orders of magnitude higher  
219 than those burned in industrial boilers (Zhang et al., 2000; Zhang et al., 2008). About 98 Mt coal, 182  
220 Mt firewood, and 340 Mt stalks in 2007 were burned in residential stoves in mainland China (NBSC,  
221 2008). Figure 5 shows a typical photo for the winter morning in the suburb of Beijing which  
222 demonstrates that the household combustion in rural China is still a serious pollutant emission source  
223 and causes negative impact on locally ambient air quality. The contribution of pollutants from these



224 residential solid fuel combustions to the local and regional air quality cannot be readily estimated  
225 owing to complex processes in the atmosphere and rare information on various pollution sources.



226  
227 **Fig. 5.** Photograph taken in rural Beijing (Huairou village) at about 8:00 am on January 19<sup>th</sup> in 2016 (a  
228 winter morning), when the sun was rising and residents commonly added solid fuels in their household  
229 stoves to warm the cold morning and cook for their breakfasts.

230  
231 Two major methods, *i.e.*, emission inventory and receptor model, have been commonly used to  
232 apportion sources of particles in China (Zhang et al., 2015). The emission inventory is based on the  
233 inventory of emission sources, while receptor models use marker species to estimate source  
234 contributions. The source appointment using the receptor model was firstly conducted by Blifford and  
235 Meeker in 1960s (Blifford and Meeker, 1967) and has been developed into several analysis methods,  
236 such as chemical mass balance and factor analysis including positive matrix factorization, principal  
237 component analysis, multi-linear engine, and unmix model (Henry and Christensen, 2010; Hu et al.,  
238 2010; Lang et al., 2013; Wang et al., 2009a; Zhang et al., 2012b). The source appointment method is  
239 now widely used for characterizing China's atmospheric PMs (Cao et al., 2005; Huang et al., 2014c;  
240 Huang et al., 2015; Wang et al., 2014a; Wang et al., 2009b; Wang et al., 2014f) and also showed that  
241 the household solid fuel combustion was a major contributor to the ambient PM level across the urban-  
242 rural spectrum (Wang et al., 2005), as well as that the relative PM and polycyclic aromatic hydrocarbon  
243 (PAH) contributions from the household solid fuel combustion are higher in heating-period than that in

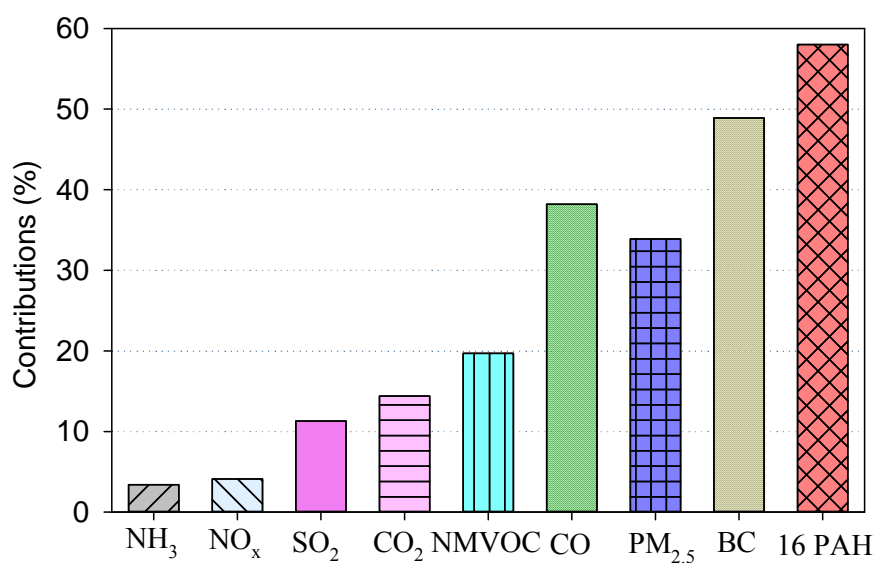
244 non-heating period in the northern China (Cao et al., 2005; Okuda et al., 2010; Tian et al., 2009; Wang  
245 et al., 2009b). However, there still lacks a systemic estimation of overall China and a detailed  
246 evaluation of pollutant emissions from the household solid fuel combustion. Thus the relative  
247 contribution of the residential solid fuel application to the ambient air pollution is introduced from the  
248 emission inventory in this section. In fact, owing to the limited information on pollutant EFs and actual  
249 distribution of fuel/stove combustions, the emission inventory is also so far not ideal on the accuracy  
250 for estimating the contribution of different sources.

251 Contributions of the household solid fuel combustion to the atmospheric pollutants will be  
252 introduced in this section. Basing on published inventories, we will firstly evaluate the contribution  
253 variety of different pollutant species. The major pollutants will be discussed in more detail on their  
254 historical contributions and geographical distributions at the province-level.

## 255 2.1. Contribution to atmospheric pollution

256 Although the ratio of household solid fuels in the total anthropogenic energy consumption is less  
257 than 15% recently in China (NBSC, 2008), relative contributions of pollutant emissions to the total  
258 anthropogenic emissions are significant. Lots of inventories report the residential emission amounts of  
259 different species, including primary anthropogenic CO<sub>2</sub>, CO, NH<sub>3</sub>, SO<sub>2</sub>, NO<sub>x</sub>, 16 PAH (Sixteen priority  
260 PAHs identified by US Environmental Protection Agency in 1976), non-methane VOC (NMVOC), PM  
261 and its contained BC (Kang et al., 2016; Lei et al., 2011; Ohara et al., 2007; Shen et al., 2013b; Streets  
262 et al., 2003; Wang et al., 2014d; Xia et al., 2016; Zhang et al., 2009a; Zhang et al., 2007; Zhao et al.,  
263 2015; Zhao et al., 2011). On the basis of the recent and available inventory data, Figure 6 shows their  
264 relative contributions to the total anthropogenic emissions in mainland China. Relative contributions of  
265 NO<sub>x</sub> in 2010 and NH<sub>3</sub> in 2012 are both less than 5%, while CO<sub>2</sub> in 2014, and SO<sub>2</sub> and NMVOC in  
266 2010 are in the range of 10% - 20%. The household solid fuel combustion contributes about 38%, 34%,  
267 49%, and 58% of total anthropogenic emissions of CO in 2014, primary PM<sub>2.5</sub> and BC in 2010, and 16

268 PAHs in 2007, respectively. BC and the dominant ratio of 16 PAHs are in the particulate-bound state.  
 269 This means that CO and PM are the most concerning pollutants. Comparing with other pollutants and  
 270 especially CO<sub>2</sub>, the high contributions of CO and PM are mainly attributed to the incomplete  
 271 combustion in the household stoves, which commonly have lower combustion efficiency than  
 272 industrial boilers. World Health Organization (WHO) also concerns on CO and PM<sub>2.5</sub> emissions and  
 273 gives their recommendation values for health protection (WHO, 2014). Hereafter, this review will  
 274 mainly focus on the emissions of CO, PM<sub>2.5</sub>, BC, and PAH during the household solid fuel combustion.



275  
 276 **Fig. 6.** Relative contributions of the residential solid fuel combustion to total anthropogenic emissions  
 277 in mainland China. Data of CO<sub>2</sub> and CO in 2014 (Xia et al., 2016), NH<sub>3</sub> in 2012 (Kang et al., 2016), 16  
 278 PAH in 2007 (Shen et al., 2013b), and the other data in 2010 (Wang et al., 2014d) were adapted from  
 279 publications.

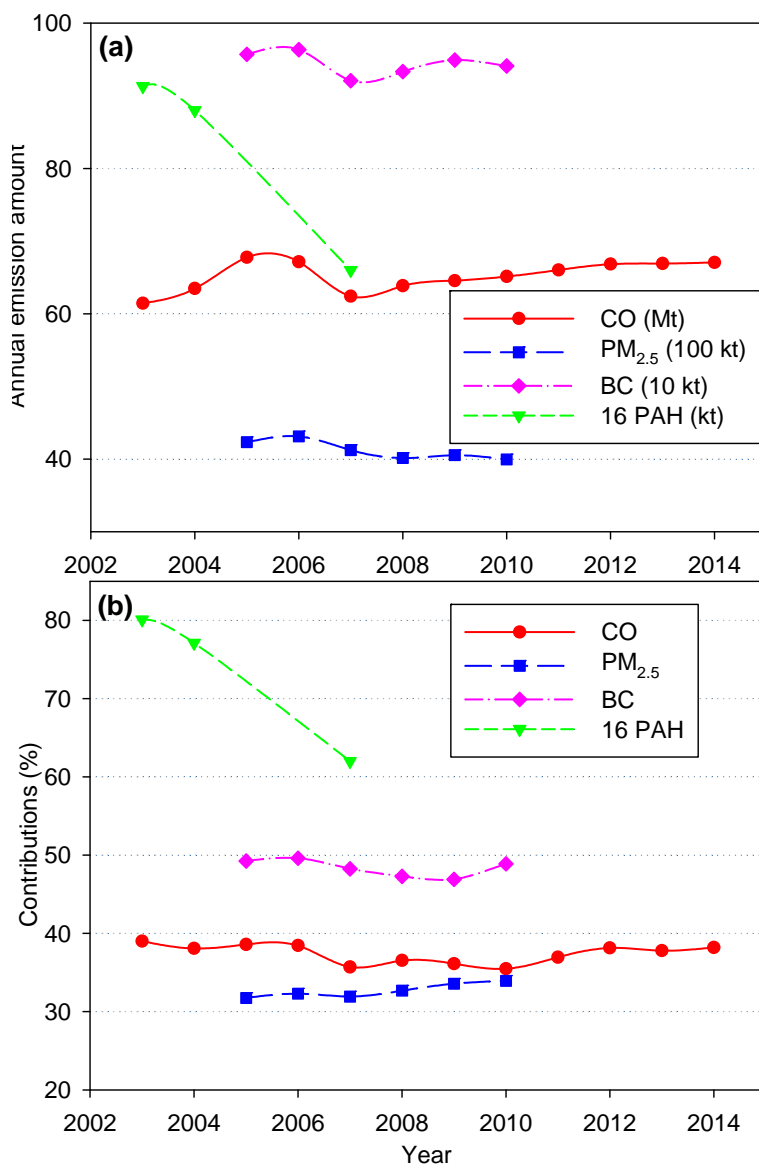
280  
 281 Figure 7 shows annual emission amounts of CO, PM<sub>2.5</sub>, and BC from residential solid fuel  
 282 combustions, and their relative contributions to the total anthropogenic emissions in mainland China.  
 283 Residential CO emission was in the range of about 60 ~ 70 Mt/year in the past decade, which suggests  
 284 that more than 100 kg/(cap·yr) in view of per-capita emissions for household solid fuel users if the used

285 amount value of residential solid fuels in the inventory was at the same accuracy for these years. The  
286 value was much higher than global average level of about 30 kg/(cap·yr) and developed countries of  
287 about 12 kg/(cap·yr). As shown in Fig. 3, the fuel per-capita consumption has continuously increased in  
288 the past decade, while the total coal consumption has also gradually increased, thus led to a continuous  
289 increase in CO emissions. However, the continuous economy growth in mainland China also results in  
290 the promotion of clean energy like natural gas and electricity, as well as cleaner household stoves. Thus,  
291 the household pollutant emissions are in race with the economy development, i.e., the increase per-  
292 capita consumption and the fast urbanization in mainland China.

293 Due to the fast urbanization and the deployment of clean fuels and stoves, the annual emission of  
294 PM<sub>2.5</sub>, BC, and 16 PAH slightly decreased in the past decade, but their relative contributions to the total  
295 anthropogenic emissions have increased. The primary PM<sub>2.5</sub> contribution in the all anthropogenic  
296 emissions had increased from 31.7% to 33.9% during 2005-2010. This is due to the lack of air pollutant  
297 devices that can reduce emissions for household stoves. However, tremendous efforts have been made  
298 to reduce pollutant emissions from industrial boilers, such as requiring coal-fired power plants to install  
299 electrostatic and bag-filtering dust precipitators for reducing PMs (~ 100% in 2012) (Li et al., 2016c;  
300 Wang et al., 2014b; Wang and Hao, 2012; Wang et al., 2010b; Xu et al., 2009; Zhao et al., 2013).

301 The BC and PAH contributions from residential solid fuel combustions in the all anthropogenic  
302 emissions were both reported to be the largest anthropogenic contributor in mainland China (Shen et al.,  
303 2013b; Wang et al., 2014c; Wang et al., 2012; Wang et al., 2014d; Xu et al., 2006; Zhang and Tao,  
304 2009). BC plays a unique and important role in the atmosphere by absorbing sunlight and emitting  
305 infrared radiation (Bond et al., 2013; IPCC, 2014). PAH has been well documented to have an adverse  
306 effect on human health and ecosystem (Gaspari et al., 2003). The significant EFs of BC and PAHs from  
307 the household solid fuel combustion have been widely investigated over the last decade in laboratory-  
308 and field-based experiments (Bond, 2002; Chen et al., 2015b; Chen et al., 2006; Chen et al., 2009; Li et

309 al., 2007; Li et al., 2009; Lu et al., 2011; Ohara et al., 2007; Shen et al., 2014; Shen et al., 2013a; Shen  
 310 et al., 2010; Wei et al., 2013; Wei et al., 2014; Zhang et al., 2009a; Zhang et al., 2008; Zhi et al., 2009).  
 311 Inventories of BC and PAH for the residential solid fuel combustions have similar trends, which are in  
 312 agreement with the results in Fig. 7 (Lei et al., 2011; Ohara et al., 2007; Streets et al., 2003; Wang et al.,  
 313 2014c; Wang et al., 2014d; Zhang et al., 2009a; Zhao et al., 2015).



314  
 315 **Fig. 7.** (a) Annual emission amounts of CO, PM<sub>2.5</sub>, BC, and 16 PAH from the residential solid fuel  
 316 combustion, and (b) their relative contributions to the total anthropogenic emissions in mainland China.

317 Data of CO (Xia et al., 2016), PM<sub>2.5</sub> and BC (Wang et al., 2014d), 16 PAH in 2003 (Xu et al., 2006),  
318 2004 (Zhang and Tao, 2009), and 2007 (Shen et al., 2013b) were adapted from different publications.

319

320 In general, the annual emission amounts of PM<sub>2.5</sub>, BC, and 16 PAH from the residential solid fuel  
321 combustion have decreased gradually due to economy development, but their relative contributions to  
322 the ambient air pollutant have increased gradually in the past decade due to the strict pollutant control  
323 for industrial boilers. Although the estimation accuracy on pollutant inventories is still not perfect, the  
324 control of pollutant emissions from the residential combustion should attract more attention.

## 325 2.2. Geographical distribution of pollutant emissions

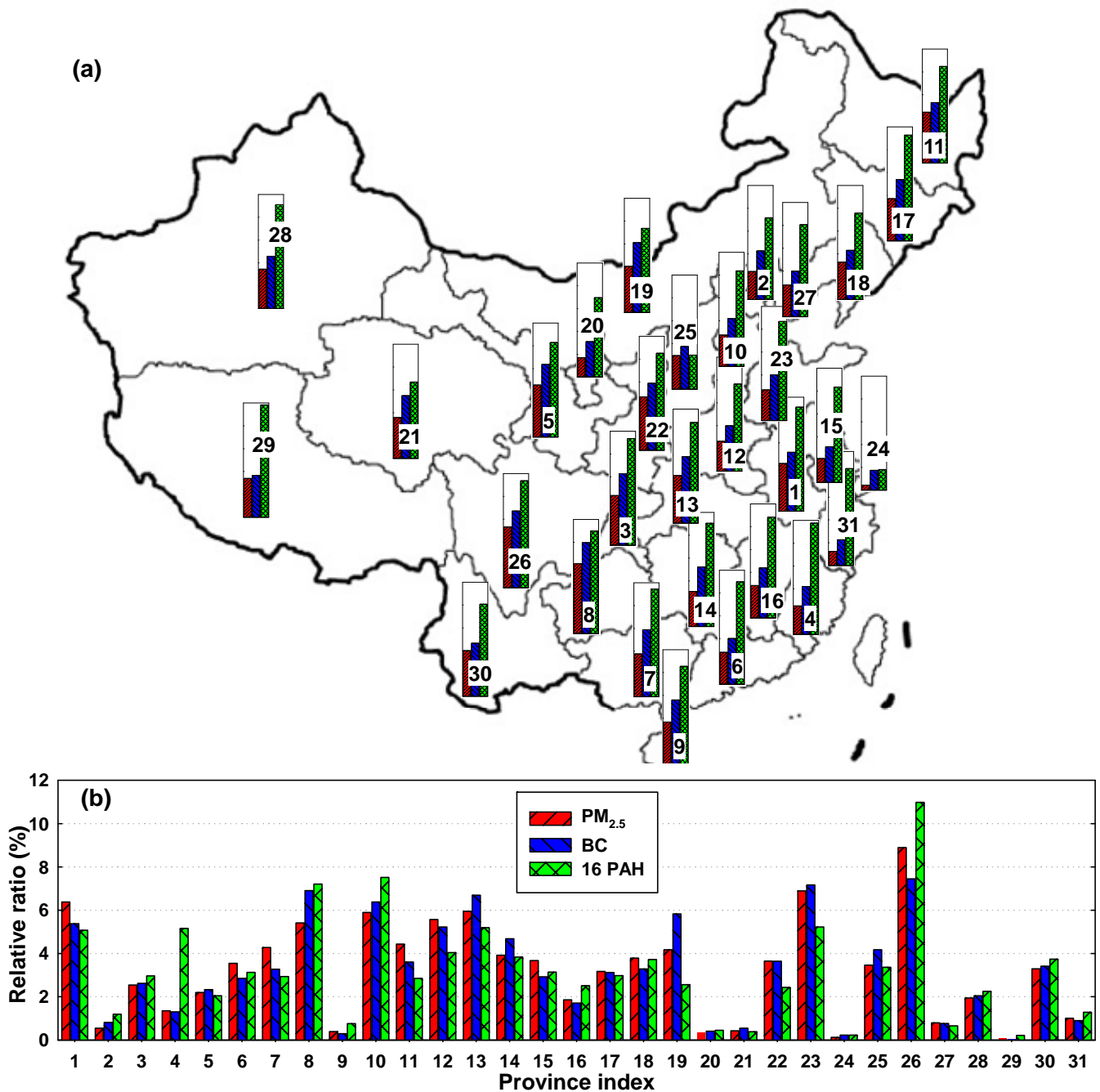
326 As discussed in the above subsection on the annual emissions, the difference of which among the  
327 historical period is mainly owing to the per-capita consumption and the urbanization. A remarkable  
328 influence is presented due to the imbalance of economic development in different regions, which leads  
329 to the difference for the ability of the per-capita consumption and the urbanization ratio. The  
330 replacement of raw solid fuels with clean energy in rural regions is difficult to achieve due to relative  
331 lower availability and higher price. The per-capita consumption amount of the household solid fuel in  
332 rural regions has increased during the economy or the urbanization development (see Fig. 3) (Duan et  
333 al., 2014). It is very difficult to distinguish the distribution of the household solid fuel in a certain  
334 province by using official statistical data (Ru et al., 2015). Here we only describe the difference among  
335 provinces in mainland China.

336 Figure 8 shows the geographical distribution of PM<sub>2.5</sub>, BC, and 16 PAH emissions from the  
337 residential solid fuel combustion at the province level. Owing to no proper data for the geographical  
338 distribution of the CO emission, it was not included in this discussion. But the CO distribution is likely  
339 to be similar as that of PM<sub>2.5</sub>. Sichuan province was the place with the highest contribution ratio among

340 all provinces, followed by Shandong, Hebei, and Guizhou provinces. In Sichuan, relative ratios of  
341 PM<sub>2.5</sub>, BC, and 16 PAH emissions are 8.9%, 7.5%, and 11.0%, respectively (Liu et al., 2007a; Wang et  
342 al., 2014d). The total relative ratios of PM<sub>2.5</sub>, BC, and 16 PAH emissions from the four provinces are  
343 27.1%, 27.9%, and 30.9%, respectively. All these provinces have with relatively low urbanization ratio  
344 and high population intensity, and thus consume relatively large amount of biomass and coals. For  
345 annual relative ratios of the household PM<sub>2.5</sub> and BC emissions, Guizhou province-the poorest province  
346 in China features the highest ratios for PM<sub>2.5</sub> and BC as 61.3% and 79.7% in 2010, respectively.  
347 Sichuan province features the second highest ratios as 53.2% and 67.2%. On the other hand, Shanghai-  
348 the richest city in mainland China features the lowest ratios for PM<sub>2.5</sub> and BC as 4.4% and 17.4% in  
349 2010, respectively, while Zhejiang province-the richest province has the second lowest ratios as 12.4%  
350 and 22.6%. In general, the regions with winter heating (northern China) and less economically  
351 developed (southwest China) suffer higher pollutant impact from household solid fuel combustion than  
352 other regions in China.

353 In a brief summary, the household solid fuel combustion has significantly negative impact on the  
354 ambient air quality, especially on the emissions of CO, PM<sub>2.5</sub>, BC, and PAH. Their emissions are  
355 mainly attributed to the low combustion efficiency of household stoves. According to the historical and  
356 geographical distributions of these species, their emission amounts and relative contributions are  
357 mainly determined by the urbanization development and the economy level.

358



359

360 **Fig. 8.** (a) Relative contributions of PM<sub>2.5</sub>, BC, and 16 PAH (in winter) from the residential solid fuel  
 361 combustion to the total anthropogenic emissions in each province, and (b) relative ratios of each  
 362 province in the total household PM<sub>2.5</sub>, BC, and 16 PAH (in winter) emissions in mainland China.  
 363 Province indexes are named as: 1 Anhui, 2 Beijing, 3 Chongqing, 4 Fujian, 5 Gansu, 6 Guangdong, 7  
 364 Guangxi, 8 Guizhou, 9 Hainan, 10 Hebei, 11 Heilongjiang, 12 Henan, 13 Hubei, 14 Hunan, 15 Jiangsu,  
 365 16 Jiangxi, 17 Jilin, 18 Liaoning, 19 Neimeng, 20 Ningxia, 21 Qinghai, 22 Shaanxi, 23 Shandong, 24



366 Shanghai, 25 Shanxi, 26 Sichuan, 27 Tianjin, 28 Xinjiang, 29 Xizang, 30 Yunnan, and 31 Zhejiang.  
367 Heights of the frame in each insert in (a) range from 0% to 100%. Data for PM<sub>2.5</sub> and BC is based on  
368 emissions in 2010 (Wang et al., 2014d), and while data for 16 PAH is based on emissions in 2003 (Liu  
369 et al., 2007a).

### 370 **3. Impacts on indoor air quality**

371 The indoor air quality is closely correlated with human health, with numerous studies linking  
372 exposure to combustion products from indoor stoves and/or heaters to a range of respiratory diseases,  
373 including lung cancer (Kim et al., 2015a; Kim et al., 2015b; Mumford et al., 1987; Smith et al., 2004).  
374 Due to the significant amount of time spent indoors (Perez-Padilla et al., 2010), the indoor air quality is  
375 a significant issue, particularly in regions which rely on the combustion of solid fuels for domestic  
376 cooking and heating applications (Smith et al., 2004). It has been found that levels of the air pollutants  
377 produced via combustion, such as PM<sub>10</sub>, PM<sub>2.5</sub>, VOCs and PAHs can vary with the fuel type and the  
378 stove type (Wang et al., 2010a; Wu et al., 2015a). Given that the direct combustion of solid fuels, such  
379 as coal and biomass, is relatively inefficient and that the rooms containing stoves/heaters are typically  
380 poorly ventilated, the potential for significant exposure is high.

381 The major determinant factors for the household air quality can be identified based on previous  
382 studies conducted in rural households in a number of Chinese provinces. Studies were conducted in  
383 Henan (Wu et al., 2015a), Guizhou (Alnes et al., 2014; Ma et al., 2015; Wang et al., 2010a; Zhang et al.,  
384 2012a; Zhang et al., 2014a), Shanxi (Chen et al., 2015a; Shen et al., 2013a), as well as a number of  
385 other studies conducted over larger areas (Mestl et al., 2007; Shen et al., 2010; Wei et al., 2014; Zhang  
386 et al., 2014b), e.g., Southwest China.

387 Despite significant interest in the indoor air quality, particularly in terms of the combustion of the  
388 solid fuel for heating and cooking applications, there is still a lack of detailed temporal data sufficient

389 to draw conclusive conclusions about exposure more broadly. This is due in part to different  
390 measurement methods used across different studies, and the variation in fuel and stoves types used.  
391 There are also, significant regional differences in terms of housing design, behavior and climate, which  
392 serve to confound these issues.

393 Key factors which influence the composition and concentration of pollutants, were the fuel type  
394 and quality, the stove design (or material), the nature of any ventilation present, and/or layout of the  
395 household (Peabody et al., 2005). Seasonal variation in levels of indoor air pollutants have also been  
396 observed, with Wu et al. (2015a) noting that the combustion of solid fuels for heating purposes was  
397 only required for part of the year.

398 In the following sections we will consider the influence of fuel and stove type, ventilation and  
399 ambient conditions on indoor air quality.

### 400 3.1. Fuel and stove type

401 A number of solid fuel sources are utilized in China, together with a variety of stove designs  
402 which often vary regionally. Coal, wood, crop residue and dried manure are all materials that have been  
403 utilized for domestic heating and cooking applications, with the choice of solid fuels based on  
404 availability. Crop residue, for example, is a common fuel source utilized in Henan Province, due to the  
405 significant amount of agriculture present (Wu et al., 2015a). The type of stove used is also a significant  
406 factor, with metal, brick or clay stoves commonplace throughout the regions studied in the literature. It  
407 is also worth noting the usage of underground coal stoves in Shaanxi, as well as the use of fire pans, or  
408 open fires (though not widespread) in Gansu (Jin et al., 2006).

409 In their study of exposure level to indoor air pollutants, Jin et al. (2006) studied a number of  
410 households in 4 Chinese provinces (Gansu, Guizhou, Inner Mongolia and Shaanxi) considering fuels  
411 used for heating and cooking, as well as the stove type and the nature of any ventilation. In all regions  
412 the fuels predominantly used for cooking were either coal or biomass (wood or crop residue). Jin et al.

413 (2006) also noted the stove types used for cooking and heating, as well as noting differences between  
414 regions, for example, the majority of coal stoves in Guizhou, as simple metal enclosures with little or  
415 no ventilation. This is in contrast to biomass cooking stoves that were found to be made of brick or clay.

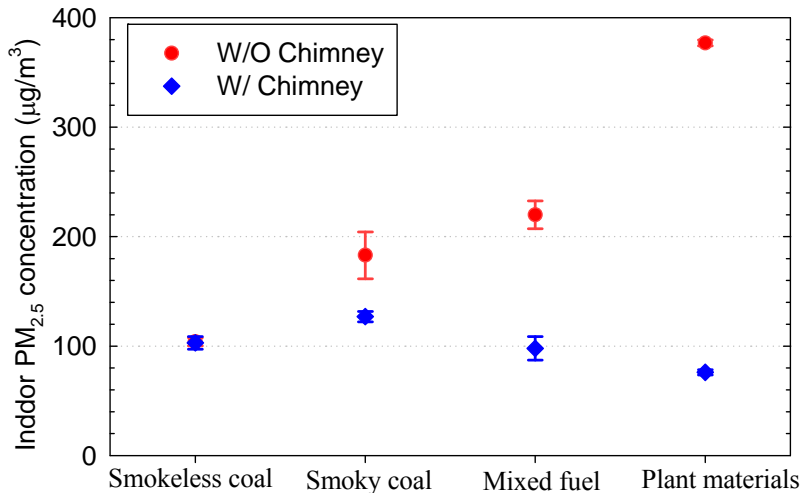
416 A number of authors have highlighted the absence of chimneys in many stoves, with Wang et al.  
417 (2010a) recording significantly higher levels of PM<sub>2.5</sub> and PM<sub>10</sub> in households featuring stoves without  
418 chimneys. Jin et al. (2006) refer to stoves as “improved” or “unimproved” based on the presence or  
419 absence of a chimney, respectively. In the studies reviewed, levels of air pollutants were typically  
420 higher in households with “unimproved” stoves, however, the fuel type still played an important factor,  
421 with PAH levels found to be generally higher in households burning coal than in households burning  
422 crop residue (Wu et al., 2015a).

423 Zhang et al. (2000) tested 28 different fuel-stove combinations, with the objective of developing a  
424 database of EFs. While this work was more focused on environmental air pollution, it does serve to  
425 illustrate the variability in emissions from a range of fuel-stove combinations, which may be commonly  
426 found in China. Importantly, this study highlighted the low thermal efficiencies of biomass and coal  
427 stoves, and the resultant high levels of CO<sub>2</sub> and incomplete combustion products per unit of delivered  
428 energy (Zhang et al., 2000). The implication of this is significant, as not only does it highlights the  
429 comparatively high levels of pollutants in relation to other fuel sources, it also indicates that a much  
430 greater mass of fuel needs to be used to achieve the same energy.

431 In their study of particle emissions from rural households in Guizhou, Zhang et al. (2012)  
432 analyzed the composition of PM<sub>2.5</sub> emissions from wood and coal burning, reporting a significant  
433 amount of the composition is carbonaceous (55% for coal and 41% for wood). They also reported  
434 ratios of OC to EC of 7.6 for coal and 10.7 for wood, which were comparable to those obtained by  
435 Wang et al. (2010a) that also considered emissions from rural households in Guizhou.

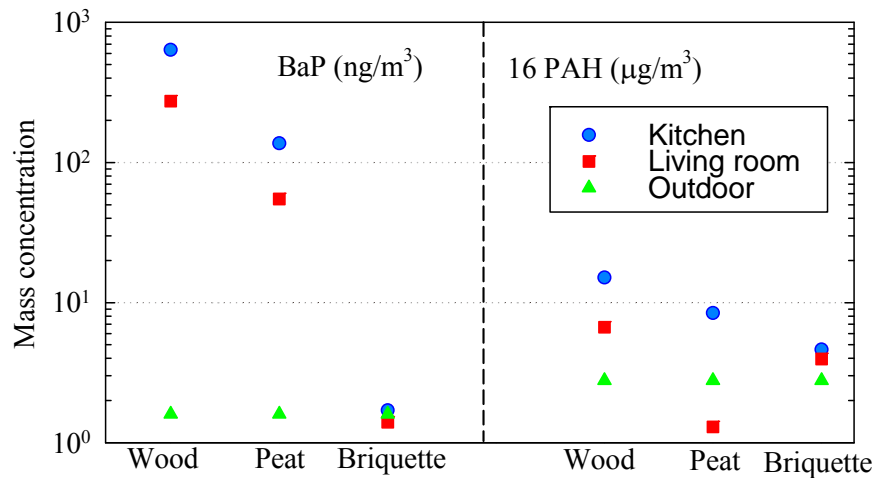
436 Figure 9 illustrates the significance of the fuel type. In general, EFs of PM, BC, and PAHs are  
437 generally in the order of smokeless coal (briquette and chunk) < smoky coal (briquette and chunk) <

438 wood < brushwood/branch < crop residue (He et al., 2005). This is supported by the results of Hu et al.  
 439 (2014) that found similar patterns were found for both measured indoor pollutant levels and personal  
 440 exposures. Jin et al. (2006) also observed higher concentrations of particulate matter in households  
 441 utilizing crop residue than those utilizing coal. The implication of a higher EF infers a higher pollutant  
 442 emission. Figure 10 indicates the relationship between indoor pollutant concentrations of PAHs and the  
 443 fuel type. However, some surveys for CO and NO<sub>x</sub> concentrations indoors showed no distinct  
 444 difference resulting from solid fuel types (Alnes et al., 2014; Edwards et al., 2007; Ma et al., 2015; Wu  
 445 et al., 2015b; Zhang et al., 2014a).



446  
 447 **Fig. 9.** Average indoor PM<sub>2.5</sub> (µg/m<sup>3</sup>) concentrations from different houses related to stoves with (W/,  
 448 vented) and without (W/O, unvented) a chimney for different solid fuel types, obtained from 24-hour  
 449 continuous measurements in main living room for each sample. The mixed fuel is combinations of  
 450 wood, plant materials and coal, where the plant materials include combinations of wood, tobacco stem,  
 451 and corncob. Data source: the field study conducted in 30 villages from Xuanwei and Fuyuan counties  
 452 in Yunnan province, in southwest of China (Hu et al., 2014).

453



454

455 **Fig. 10.** Indoor BaP and 16 PAH concentrations related to fuel type and household location. Data  
 456 source: the field study conducted in rural Shanxi province in summer (Chen et al., 2015a).

457

### 458 3.2. Household ventilation and layout

459 In the absence of a vented stove, the household ventilation is through windows and doors and is  
 460 also influence by the presence of stairwells in the room, where the stove is located (Hu et al., 2014). It  
 461 is expected that house layout will also influence pollutant levels indoors. Indeed, mixed effect  
 462 modelling conducted by Hu et al. (2014) found that the number of windows was an important factor  
 463 influencing the personal exposure to PM<sub>2.5</sub>. Additionally, the ventilation through the poor sealing of  
 464 walls and rooftops, in traditional households in rural parts of China, should also be considered.

465 In their study, Hu et al. (2014) found that households with one window in their main cooking area  
 466 typically lead to a higher PM<sub>2.5</sub> exposure than those with zero (due to the aforementioned poor sealing  
 467 of walls and rooftops in traditional homes) or two windows.

468 A number of households studied in previous work (He et al., 2005; Hu et al., 2014; Jin et al., 2006)  
 469 have featured some form of chimney or flue duct. It has been observed that significantly lower levels of

470 indoor pollutants are present when some forms of vented stoves were used (Hu et al., 2014), with  
471 corresponding reduced levels of the personal exposure also reported.

472 While the presence of a chimney will lead to reduced levels of pollutants in the room where the  
473 stove is located, this does not guarantee reduced exposure. A number of houses in Guizhou have been  
474 found to have chimneys that end in the attic (He et al., 2005; Jin et al., 2006), resulting in the highest  
475 concentrations of airborne pollutants in the home to be in the attic (He et al., 2005). The chimney  
476 ending in the attic appears to be limited to the Guizhou province, where the attic is used to dry food  
477 (He et al., 2005; Jin et al., 2006). In their study of 1741 household across 4 provinces, Jin et al. (2006)  
478 found chimneys were commonplace, however, in provinces such as Gansu and Guizhou, the chimney  
479 often did not go outside the house. In cases where the chimney did exit the house, over 90% of  
480 chimneys were below the height of the eaves (Jin et al., 2006). This raises the potential for pollutants to  
481 re-enter the house through the roof space.

482 Figure 9 shows indoor PM<sub>2.5</sub> concentrations related to household stoves, with and without  
483 chimneys, for different solid fuel types (Hu et al., 2014). According to the random-survey in rural  
484 Yunnan province, the reduced ratios for indoor PM<sub>2.5</sub> concentrations by the chimney were about 1%,  
485 31%, 55%, and 80% for smokeless coal, smoky coal, mixed fuel, and plant materials, respectively (Hu  
486 et al., 2014). The reduction ratio is much higher for fuels with higher volatile matter content, which  
487 possess higher PM<sub>2.5</sub> EFs (Li et al., 2016b). In 2010, approximately 76% of stoves used for cooking in  
488 Guizhou did not feature a chimney (Alnes et al., 2014). Combustion in these household stoves  
489 generally resulted in higher indoor pollutant levels (Alnes et al., 2014; Armendariz-Arnez et al., 2010;  
490 Hu et al., 2014). In the absence of a chimney, often the only source of household ventilation is through  
491 open door and windows. There are times of the year, however, where the doors and windows will be  
492 close, which is likely the explanation for higher winter levels of indoor pollutants reported by Wu et al.  
493 (2015b). This will be discussed further in Section 3.3.

494 The location of stoves in households has also been shown to influence the pollutant concentrations  
495 (Alnes et al., 2014; Ding et al., 2012; Downward et al., 2014; Edwards et al., 2007; Fischer and  
496 Koshland, 2007; Gao et al., 2009; He et al., 2005; Hu et al., 2014; Jin et al., 2005; Ma et al., 2015; Shao  
497 et al., 2013; Zhang et al., 2014a; Zhong, 2011; Zhong et al., 2012). Typically, the concentration of  
498 pollutants in the living room is lower than that in the kitchen (stove location), as shown in Fig. 10  
499 (Chen et al., 2015a). However, it should be noted that this relies on there being a clear demarcation  
500 between the rooms in the house and also assumes that only one stove is present. It has been noted in a  
501 number of studies that the kitchen and living room utilize the same space. Jin et al., (2006) for example  
502 found in their study that approximately 72% of households in Guizhou usually cook in the living room  
503 and that older homes in Inner Mongolia consisted of a large single room, which served as the living,  
504 cooking and sleeping space.

505 In cases where the household have multiple rooms, the levels of indoor air pollutants in bedrooms  
506 (where no heaters are present) are greatly dependent on dispersion, and heavily influenced by house  
507 layout (He et al., 2005). The exception to this is in the houses in Guizhou, with chimneys ending in the  
508 attic where, similar concentrations of pollutants were found in the bedroom (no heater) and the living  
509 room (heater), in some cases despite the presence of a door (He et al., 2005). In these cases it seems  
510 reasonable to conclude that the smoke is collecting in the attic and being distributed around the house  
511 (He et al., 2005) that is likely due to poor sealing between the rooms on the ground floor and the attic, a  
512 factor also noted by Hu et al. (2014).

513 Comparison of the studies by Hu et al (2014) and He et al. (2006), the province study indicates a  
514 large level of regional variation, in terms of house layout, stove type and fuel type. This serves to  
515 highlight the regional differences that make the results of individual studies, or region specific studies  
516 difficult to apply more broadly.

### 517 3.3. Ambient conditions

518 As well as considering the stove and fuel type and ventilation and house layout, some  
519 consideration must also be given to ambient conditions including outdoor air pollution, as well as  
520 seasonal and geographic factors, such as ambient temperature. Given that coal and biomass commonly  
521 used fuels for household heating applications, the ambient temperature is likely to be a significant  
522 influence on the indoor pollution levels and the exposure. Cold winters in the northern China typically  
523 result in significantly higher residential solid fuel (such as coal) consumption for heating than that in  
524 summers (Duan et al., 2014).

525 Households in Guizhou and Inner Mongolia, often utilize the household stoves in the living area  
526 for both cooking and heating purposes (Jin et al., 2006). In winter when in use for heating, the stoves  
527 will often be operating longer than when used for cooking alone, as may be the case in summer months.  
528 The duration of the period where household stoves are used for heating varies geographically, with Jin  
529 et al. (2006) noting that stove were used for 4-6 months in Gansu, 6-7 months in Guizhou, 6-7 months  
530 in Inner Mongolia and 5-6 months in Shaanxi.

531 Wu et al. (2015b) found higher levels of pollutants in households in Henan in winter than in  
532 autumn, with significantly higher pollutant levels (PM<sub>2.5</sub>, PM<sub>10</sub>) being recorded in living rooms in  
533 winter. A similar pattern has been observed by studies conducted in Hebei province, which reported  
534 higher concentrations of BC, OC and benzo-[a]-pyrene (BaP) in winter than in summer (Ding et al.,  
535 2012; Zhong, 2011; Zhong et al., 2012).

536 In addition to the higher pollutant levels typically observed in winter, consideration must also be  
537 given to other contributing factors. While higher pollutant levels in winter can be easily linked to  
538 higher fuel consumption (for heating), we must also consider any behavioral factors that may  
539 contribute. As previously noted, Wu et al. (2015b) observed that windows and doors which may be



540 open to provide ventilation in summer, will be closed in winter to limit heat escape. This will have  
541 addition effect of keeping more pollutants indoor and potentially increasing exposure.

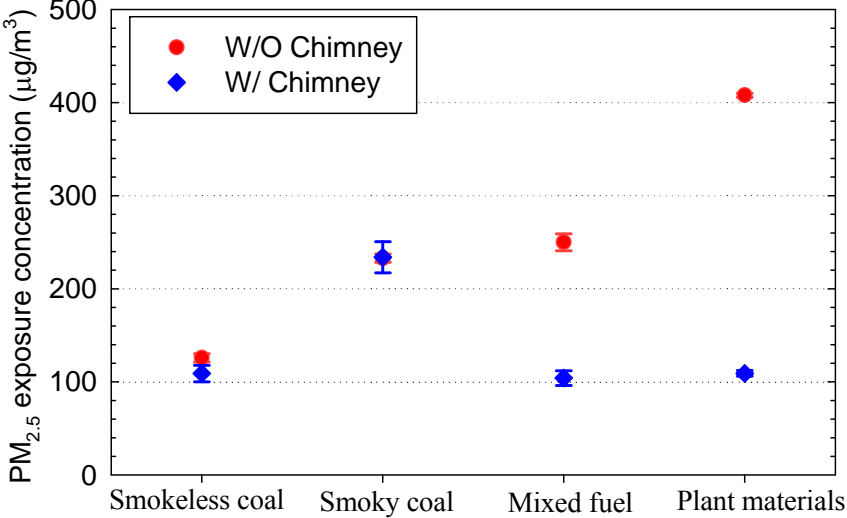
#### 542 **4. Impacts on personal inhalation exposure**

543 The household solid fuel combustion is likely to be one of the largest emission sources of indoor  
544 air pollution in rural China, as well as an important source impacting on the ambient air quality, as  
545 described in the last two sections. Poor indoor and ambient air qualities can thus induce substantial  
546 exposure to air pollutants which mainly occurs inside the house since people spend more time indoors  
547 (Brauer et al., 2012). Exposure to these air contaminants can pose a serious health hazard, particularly  
548 for women, young children, and elders who typically spend much of their time inside the household  
549 (Bruce et al., 2000; Chafe et al., 2014; Lim et al., 2013; Mestl et al., 2007; Smith et al., 2009).  
550 Household air pollution has been estimated to be one of the main health risks worldwide such as  
551 respiratory and cardiovascular risks, including cancer. In addition, a recent report showed that poor  
552 indoor air quality accounts for 3.7% of total deaths in mainland China (GBD2010, 2013). The Chinese  
553 government is one of the first in the world to define a national health based indoor air quality (IAQ)  
554 standard for residences (Edwards et al., 2007). The standard for a 1-day average has been set at 150  $\mu\text{g}$   
555  $\text{PM}_{10}/\text{m}^3$ , a level which still represents substantial health risks. Direct measurements of personal  
556 exposure to air pollutants in mainland China are extremely scarce (Baumgartner et al., 2011). In this  
557 section we introduce some results of the potential impact of fuel types, seasons, and geographic  
558 location on exposure levels to selected air pollutants among different population groups.

559 Figure 11 shows the effect of the fuel/stove combinations on the exposure levels to  $\text{PM}_{2.5}$  among  
560 housewives (Hu et al., 2014). Similar is the effect on indoor  $\text{PM}_{2.5}$  concentrations (see Fig. 9), from the  
561 combustion of solid fuels with a high volatile content which results in high  $\text{PM}_{2.5}$  exposure levels,  
562 while combustion in stoves without chimney also leads to a higher  $\text{PM}_{2.5}$  exposure level compared with

563 stoves with chimney. This result is in consistence with the WHO recommendation, i.e., to use clean  
564 solid fuel such as processed coal for the household combustion (WHO, 2014).

565

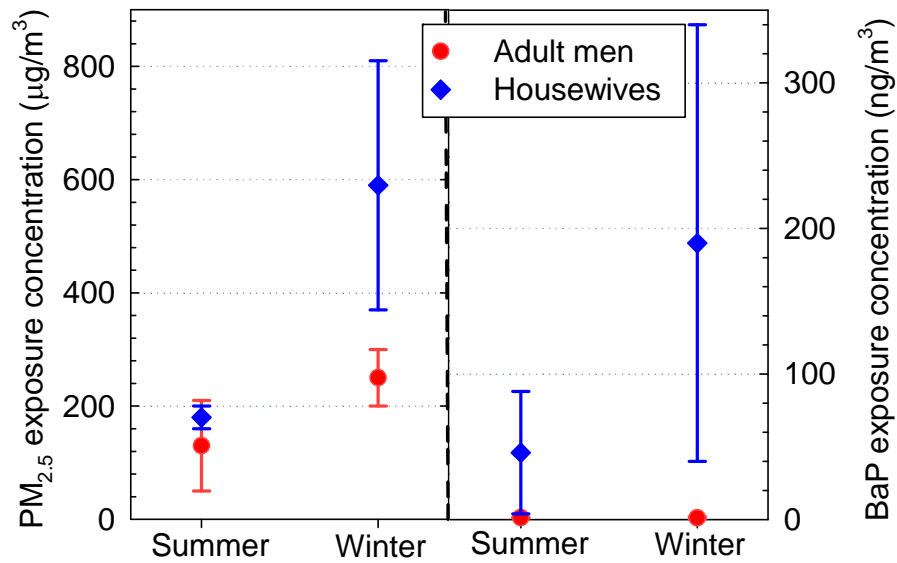


566

567 **Fig. 11.** Average concentrations of PM<sub>2.5</sub> of housewife exposure in different houses related to solid  
568 fuel type, as well as stoves installed chimney or not (see Fig. 9 caption for the explanation of mixed  
569 fuel and plant materials). The field studies were conducted in rural Yunnan province (Hu et al., 2014).

570

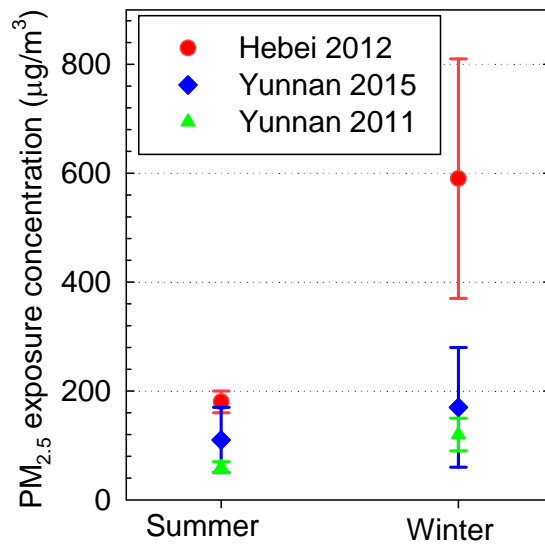
571 Other factors may also have effect on human exposure concentrations. Season can have an impact  
572 on exposure concentrations via household combustion activities, time spend inside the household, as  
573 well as the household ventilation. Figures 12 and 13 suggest that PM<sub>2.5</sub> and BaP exposure  
574 concentrations in winter are higher than in summer. In addition, the geographic location may also affect  
575 the exposure concentration. According to the reported indoor PM<sub>2.5</sub> levels from different provinces  
576 (Edwards et al., 2007; Jin et al., 2005), people in Northern China (Hebei province) are exposed to  
577 higher PM<sub>2.5</sub> concentrations than those who live in the southern China (Yunnan province)  
578 (Baumgartner et al., 2011; Huang et al., 2015; Zhong et al., 2012) (see Fig. 13).



579

580 **Fig. 12.** Exposure concentrations of PM<sub>2.5</sub> and BaP. Housewives suffer much higher personal  
 581 inhalation exposure levels than the adult men, especially in winter. Data was obtained from field  
 582 studies in rural Hebei (Ding et al., 2012; Zhong et al., 2012).

583



584

585 **Fig. 13.** Exposure concentrations of PM<sub>2.5</sub> in rural Hebei (in Northern China, winter heating region)  
 586 and Yunnan provinces (in Southern China). People in the northern China are exposed to higher PM<sub>2.5</sub>  
 587 concentrations than those who live in the southern China. Data was obtained from field studies  
 588 (Baumgartner et al., 2011; Huang et al., 2015; Zhong et al., 2012).

589 Figures 12 shows that the exposure to fine PM among housewives is higher than that among the  
590 adult men, especially in winter, which is mainly due to housewives spending more time near the stove  
591 and inside the house. In addition, housewives from the rural northern China are exposed to 2 and 4  
592 times higher PM<sub>2.5</sub> concentrations compared to those who live in the southern China during summer  
593 and winter, respectively.

594 In a brief summary, the factors affecting the personal inhalation exposure to fine PM are similar  
595 with those factor of affecting the indoor air quality, including the fuel/stove combination, household  
596 ventilation, and ambient air quality conditions. In addition, the time people spending in the house and  
597 near household stoves is another important factor. Thus the housewives have higher personal inhalation  
598 exposure level than the adult men. Children usually spend more time with their mother and also are  
599 likely to be exposed to higher indoor air pollution. The household solid fuel combustion leads to  
600 increased health risk via increasing the exposure to their emissions.

## 601 **5. Determinants of emission factors**

602 Flue gas from the household stove combustion is commonly and directly released to the  
603 atmosphere without any treatment by air pollution control device that has been widely installed  
604 for China's industrial boilers, such as for coal-fired power plants (Wang et al., 2014b; Wang  
605 and Hao, 2012; Wang et al., 2010b; Xu et al., 2009; Zhao et al., 2013). Pollutant EFs are mainly  
606 determined by two factors, i.e., the fuel quality and the combustion process, as briefly  
607 illustrated in Fig. 2. Parameters for the solid fuel quality mainly include contents of caloric  
608 value, moisture, volatile matter, ash, sulphur, chloride, toxic elements such as As, Hg, and Pb,  
609 as well as the fuel form such as briquette and pellet. No policy on biomass quality comes out in  
610 mainland China, while the government has introduced and updated policies on the residential  
611 coal quality to reduce pollutant emissions, e.g., increasing caloric value, lowering the threshold  
612 values for ash on dry basis ( $A_d$ , required to be less than 16% for Beijing-Tianjin-Hebei region),

613 and volatile matter content on dry and ash-free basis (required to be less than 10% and 20% for  
614 anthracite and bituminous briquettes, respectively, in Beijing) (BMAQTS, 2013; NDRC, 2014).

615 The combustion process is mainly controlled by the stove technology and operational  
616 technique that depends on stove structure and resident skill level. The stove technology had  
617 been spontaneously developed until 1980s for at least 4000 years, the length of written China's  
618 history, as briefly illustrated in Fig. 1. China government has undertaken many programs to  
619 develop the household stove technology since the early 1980s (Qiu et al., 1996; Sinton et al.,  
620 2004; Smith et al., 1993b; Zhang et al., 2009b). China's Ministry of agriculture stated that about  
621 85% rural households had improved biomass or coal cooking stoves in 2007 (CMA, 2008).  
622 Thermal efficiencies of these improved stoves should be not lower than 30%, while no fugitive  
623 emission from chimney is allowed (CMA, 2006). In fact, the current policy on residential solid  
624 fuels lacks of solid scientific supports from residential experiments.

625 Aiming to present the dominant determining factors on pollutant EFs, this section  
626 introduces the published results on fuel parameters and the stove technology. The fuel  
627 parameter mainly focuses on the fuel form, size, and contents of volatile matter content, ash,  
628 and sulphur. The stove technology mainly covers widely reported traditional stoves (see Fig.  
629 1c) and recent developed advanced stoves in China.

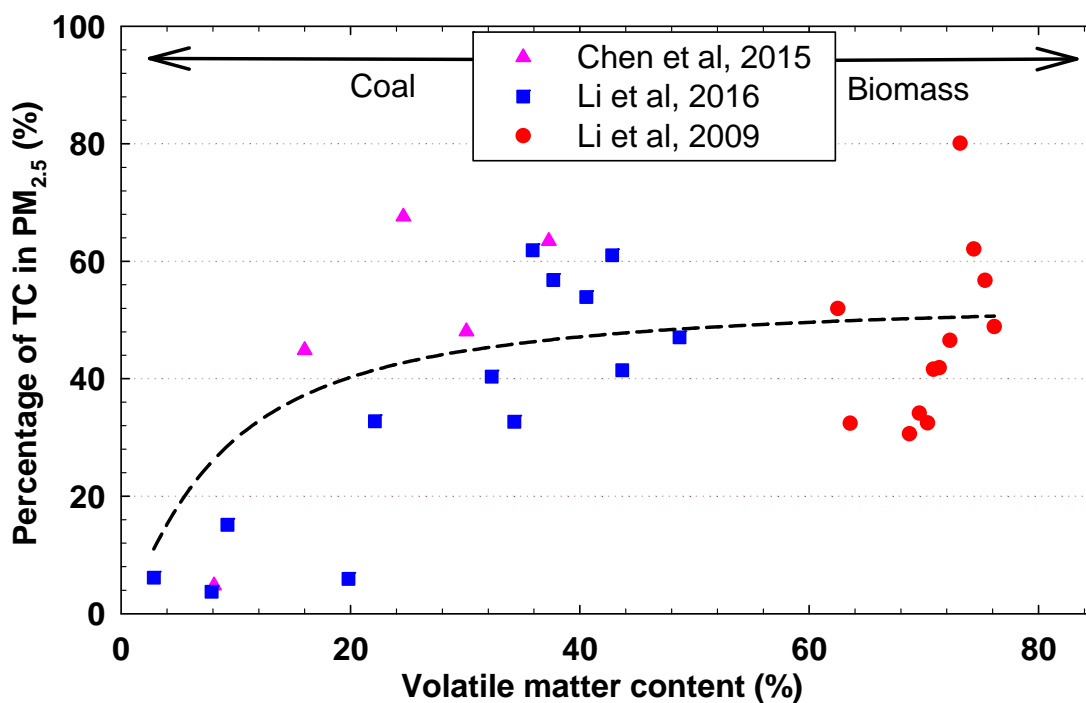
## 630 5.1. Fuel quality

631 The solid fuel quality has been intensively investigated on its effect on pollutant EFs during  
632 household combustions. Coal's volatile matter content, one of the controlled coal parameters by  
633 Chinese government (BMAQTS, 2013; 2014), was found to be the dominant factor affecting EFs of  
634 PM, OC, and BC (Shen et al., 2010). Comparative investigation of chunk and briquette coal samples  
635 showed that honeycomb-coal briquette had about 40-80% lower EFs in various stoves (Chen et al.,  
636 2015b; Ge et al., 2004; Zhi et al., 2009). Significant variation of EF exists among different qualities of

637 biomass. Biomass fuels made into pellets and briquettes were found to have much lower EFs than their  
638 raw (or unprocessed) materials (Johansson et al., 2004; Lim et al., 2012; Ravichandran and Corscadden,  
639 2014; Shen et al., 2012). Figure 14 summarizes the reported PM<sub>2.5</sub> EFs from the residential solid fuel  
640 combustion. The PM<sub>2.5</sub> EFs fast increase with an increase in the fuel's volatile matter content till about  
641 30% and then stay roughly flat for combustion in similar traditional household stoves. The PM<sub>2.5</sub> EFs  
642 of low volatile matter content fuel-- anthracite coal have been reported to an order of magnitude less  
643 than those of high volatile matter content fuels—bituminous/lignite coal and raw biomass (Chen et al.,  
644 2006; Li et al., 2016b; Li et al., 2016c; Shen et al., 2014). Measured PM<sub>2.5</sub> EFs for fuel samples with  
645 high volatile contents (lignite coals and biomass) often have large uncertainties, which are possibly  
646 related to unstable combustion during the coal ignition and pyrolysis stages.

647 Figure 15 shows the fractions of total carbonaceous species in PM<sub>2.5</sub> samples as a function of the  
648 fuel volatile matter content. Their trends are similar to the correlation between the PM<sub>2.5</sub> EF and the  
649 fuel volatile content (see Fig. 14). This indicates that the increase of the PM<sub>2.5</sub> EFs is driven by the  
650 incomplete combustions for high volatile content solid fuels, whose PMs contain high ratio of  
651 carbonaceous components. Because of the low burning efficiency in the household solid fuel  
652 combustion, generating more devolatilized matters (for higher volatile matter content fuel) in a short  
653 time period of pyrolysis stage leads to more unburned organic compounds which then serves as  
654 precursors (OC and EC) for the PM formation (Li et al., 2016b).





662  
 663 **Fig. 15.** The percentage of total carbonaceous component (TC = EC + OC) in PM<sub>2.5</sub> samples emitted  
 664 from the residential solid fuel combustions as a function of the solid fuel volatile matter content. Data  
 665 was adapted from published literatures (Chen et al., 2015b; Li et al., 2016b; Li et al., 2009). The dashed  
 666 line roughly indicates the trend of TC ratio in PM<sub>2.5</sub> with the increase of the fuel volatile matter content.  
 667

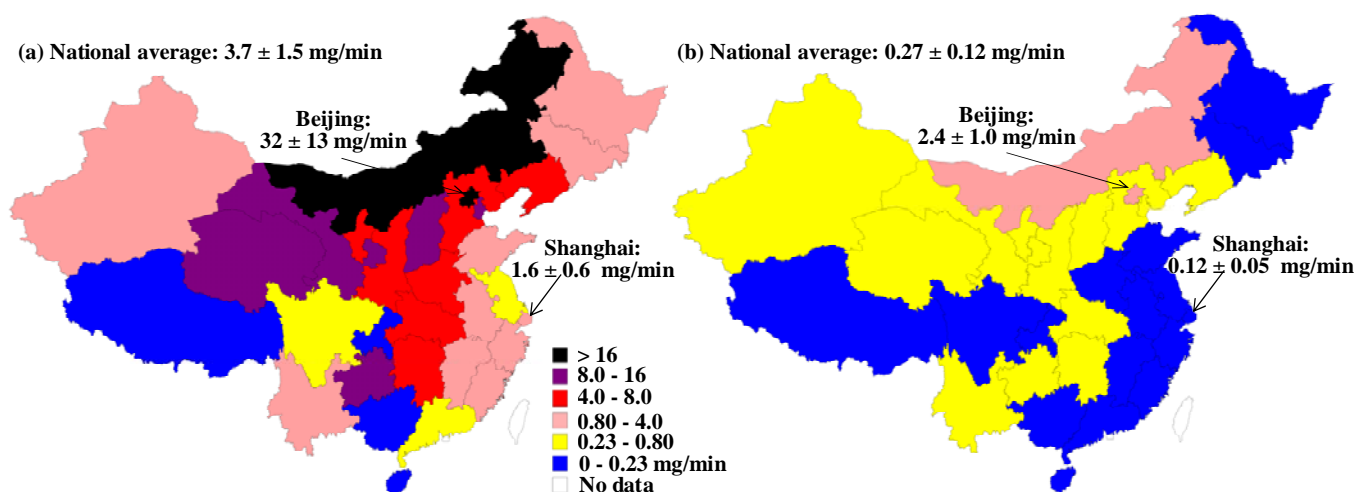
668 The fuel form is another widely accepted method to control pollutant emissions. Making raw  
 669 biomass (crop straws and wood) and coals into pellet and/or briquette forms were reported to have  
 670 relatively low pollutant EFs (Bond et al., 2002; Chen et al., 2015b; Shen, 2015; Shen et al., 2012).  
 671 However, particle-bound PAHs (the dominant form of PAHs in household flue gas) from coal  
 672 briquettes were also reported to have significantly higher EFs than those from coal chunks (Chen et al.,  
 673 2015c). The increase of clay content in the briquette was thought to serve as catalyst to crack down the  
 674 coal tar into carbon and hydrogen, and thus reduce the emission of PM precursors (Bond et al., 2002;  
 675 Landis et al., 1997). However, briquetting and pelleting technologies change fuel size and ash content  
 676 simultaneously, while solid fuel size and ash content both was identified to affect PM emissions (Li et  
 677 al., 2016b). Unfortunately, there are still no detailed report on the optimum form of solid fuels to



678 reduce pollutant emission, including size, additive addition, molding pressure and humidity, and  
679 powder diameter ranges.

680 The fuel volatile matter content has been recognized to be the dominant factor among the reported  
681 solid fuel parameters on fuel quality. Beijing-Tianjin-Hebei governments have been making new  
682 policies to reduce pollutant emissions from the residential coal consumption by replacing low rank coal  
683 (high volatile matter content) with anthracite (low volatile matter content) with the help of financial  
684 subsidy since 2012. However, anthracite coal is not well accepted due to its difficulty in ignition and  
685 high price. Semi-coke briquette, made from bituminous and lignite coals by industrial carbonization  
686 treatment, was reported to be the clean solid fuel due to low volatile matter content (Li et al., 2016c).  
687 According to estimation, switching to semi-coke briquettes can reduce averaged EFs of PM<sub>2.5</sub>, EC, and  
688 OC by about 93%, 98%, and 92%, respectively. Additionally, the semi-coke briquette has relatively  
689 lower price and higher burnout ratio in household stoves.

690 The replacement of raw coals with semi-coke briquettes was evaluated to be a feasible path to  
691 reduce pollution emissions. Figure 16 shows the hypothesized impact of this replacement on averaged  
692 PM<sub>2.5</sub> emission rates per household coal stoves at the provincial level in 2012, with referring to the  
693 WHO recommendation values (WHO, 2014). The replacement with semi-coke briquettes significantly  
694 reduces pollution emissions, especially in regions with high household coal consumption. The  
695 hypothesized analysis shows that the adoption of clean solid fuel can relieve the pollutant emissions  
696 from the household combustions in most rural China.



697  
 698 **Fig. 16** Averaged PM<sub>2.5</sub> emission rates from household coal stoves at the provincial level for  
 699 the year of 2012 in rural area of mainland China: (a) current raw coal consumption and (b) after  
 700 the hypothetical replacement with semi-coke briquettes. The colours were marked with  
 701 referring to the WHO recommendation values: 0.23 mg/min and 0.80 mg/min are for unvented  
 702 and vented cases, respectively (WHO, 2014). These estimations of emission rate are with high  
 703 uncertainties since there was no information for stove and coal distributions in China. Here we  
 704 used the number of rural households as the stove number. Averaged amount of coal  
 705 consumption in a household stove was obtained by dividing the total consumed coal amount in  
 706 rural with the total stove number in each province's rural. The averaged emission rate was then  
 707 obtained by timing averaged coal amount per stove with emission factors, obtained from the  
 708 experimental estimation (Li et al., 2016c).

709

## 710 5.2. Stove technology

711 Owing to the relatively inefficient stove technology associating with very incomplete  
 712 combustion process, solid fuels burned in household stoves have much higher PM EFs than that  
 713 in industrial boilers. Improved stoves with a high burning efficiency may have relatively low  
 714 EFs and low fuel consumption (Edwards et al., 2007; Zhang et al., 2000). According to the

715 estimation of reduction scenarios applied to a year-2010, cleaner stoves have the potential to  
716 provide better emission reduction when free fuels are used than the deployment of cleaner fuels  
717 (Winijkul and Bond, 2016).

718 The concept of “advance stove” has a different meaning at different periods. It mainly  
719 denoted the stove installed a chimney to remove emissions from indoor to outdoor without  
720 significant reduction of final emissions, and possessing a relatively higher energy efficiency  
721 than the three-stone stove before 1980s in mainland China (Smith et al., 1993a; Winijkul and  
722 Bond, 2016), see Fig. 1. However, the energy efficiency of these “advance stoves” for cooking  
723 was commonly lower than 10% (Sinton et al., 2004). After a continuous improvement since  
724 1980s (Smith et al., 1993a), the thermal efficiency of cooking stoves for the household solid  
725 fuel combustion can be improved to be about 27% - 35% by changing the fuel burning form  
726 (Carter et al., 2014; Panwar and Rathore, 2008; Shen et al., 2015). Thus the current “advance  
727 stove” requires a higher thermal efficiency, such as about 30% and 70% for cooking stoves and  
728 heating stoves, respectively, as well as significant low pollutant EFs.

729 There are several reports on the evaluation of in-use emission and the thermal efficiency of  
730 updated biomass cooking stoves—gasifier cooking stove (Carter et al., 2014; Panwar and  
731 Rathore, 2008; Shen et al., 2015) and a new heating stove of employing under-fire combustion  
732 technology (Li et al., 2016d; Li et al., 2016e). The two advanced stoves were initially designed  
733 to reduce air pollution emissions from household biomass and coal combustions, respectively.  
734 Since the biomass has much higher volatile matter content than coal, there is no difficulty for  
735 carbonization and ignition in household stoves. The gasifier cooking stove increases the  
736 combustion efficiency of devolatilized organic matter by secondary air supply. However, the  
737 gasifier cooking stove is still no successful in burning coals, which can’t be carbonized and  
738 ignited easily in the small size household stoves (Shen et al., 2015). The under-fire technology,  
739 different from the over-fire technology commonly used in household stoves, separates the

740 chamber space of solid fuel storage and combustion region. The solid fuels are carbonized and  
741 burned in the junction of the two regions, while the devolatilized organic matter was burned in  
742 the combustion region (Li et al., 2016e). The new heating stove was reported to improve the  
743 average thermal efficiency to be more than 80% for burning all tested fuel types (semi-coke,  
744 anthracite, briquette, bituminous, lignite, and biomass), as well as be effective in reducing CO<sub>2</sub>  
745 and pollutant emissions regardless of the volatile content in solid fuels. The average reductions  
746 of delivered energy-based EFs were ~87% for NH<sub>3</sub>, 50% for CO<sub>2</sub>, 79% for PM<sub>2.5</sub>, 95% for EC,  
747 85% for BaP–equivalent carcinogenic potency, and 66% for 8 selected toxic elements (Pb, Cu,  
748 Sb, Cd, As, Ag, Se, and Ni) in PM<sub>2.5</sub> (Li et al., 2016d; Li et al., 2016e). Improvement in  
749 household stove technologies has been demonstrated as a practical approach to improve the  
750 thermal efficiency and reduce the pollutant emissions and CO<sub>2</sub>. However, further laboratory and  
751 field investigations on the combustion/thermal efficiency and pollutant emissions of updated  
752 combustion technologies are still needed. Furthermore, inventory of the stove changes are  
753 needed to be characterized quantitatively, since household heating and cooking stoves used in  
754 mainland China are changing very fast recently.

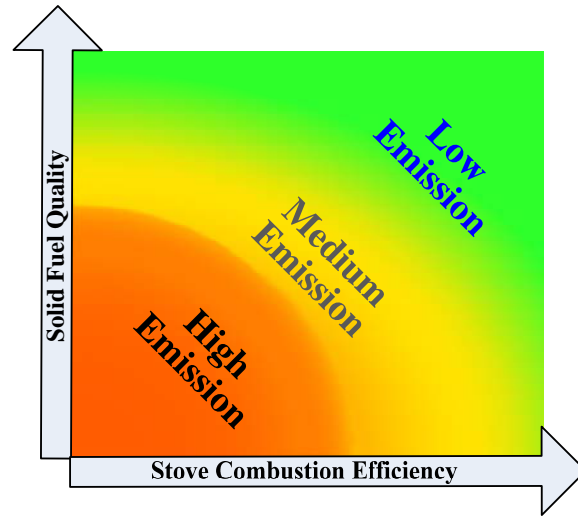
## 755 **6. Summary**

756 The current status of the indoor and ambient air pollution originating from the household coal and  
757 biomass combustion was reviewed in this study. The incomplete combustion of solid fuels in  
758 household stoves for cooking and heating activities has resulted in much higher pollutant EFs (such as  
759 PM, BC, PAH, and CO) than that of solid fuels combusted in industrial boilers, and thus caused  
760 seriously negative impacts on the air quality in China. Although annual pollutant emission has not  
761 increased significantly in the past decades due to the decreasing population of household solid fuel  
762 users (resulting from the urbanization rate and the economic development), the household relative

763 contributions of CO, PM<sub>2.5</sub>, BC, and PAH emissions have increased owing to increasingly strict  
764 pollutant control policy made to limit air pollution emissions from industrial boilers recently.  
765 Summarizing from various inventories, the household relative contributions of CO, PM<sub>2.5</sub>, BC, and  
766 PAH emissions in all anthropogenic sources are more than 30%, 30%, 45%, and 60% in mainland  
767 China, respectively. Winter heating (northern China, such as Hebei) and less economically developed  
768 regions (southwest China, such as Guizhou and Sichuan) have suffered more negative impacts from the  
769 household solid fuel combustions than other regions (such as the richest region-Shanghai and Zhejiang).  
770 The geographical distribution of the indoor air quality has similar characteristics as that of impacts on  
771 the ambient air quality. Additionally, the household stoves without installing chimneys and poor  
772 household ventilation can lead to a poor indoor air quality especially when burning high volatile  
773 content solid fuels. Closely related with the ambient and indoor air quality, the personal inhalation  
774 exposure level in winter heating and less economically developed regions is higher than that in other  
775 regions. The time people spending near the cooking stoves is another important factor. The inhalation  
776 exposure level of various people in the rural area is generally in the order of adult men < children <  
777 housewives. Thus the household solid fuel combustion has led to an increased health risk, especially  
778 for household solid fuel users.

779 Pollutant EFs of the household solid fuel combustion rely on the fuel/stove combination. PM<sub>2.5</sub>  
780 and BC EFs have been identified to increase with the solid fuel volatile matter content in traditional  
781 household stoves. Advanced stoves, such as gasifier biomass cooking stoves for biomass and “under-  
782 fire” heating stoves for all solid fuels, can reduce pollutant emissions (including PM, BC, OC, PAH,  
783 CO, NH<sub>3</sub>, and CO<sub>2</sub>) by improving the combustion and thermal efficiencies. Thus the improvement of  
784 the combustion/thermal efficiencies of household stoves and the improvement of the solid fuel quality  
785 are the two essential methods to reduce pollutant emissions from households, as summarized in Fig. 17.  
786 Clean solid fuels, such as semi-coke briquette, and improved stoves have been both proposed to reduce  
787 PM emissions in China. However, cleaner solid fuels are typically more expensive, which in turn may

788 result in residential consumers not choosing it. Thus a clean stove with low cost regardless of the fuel  
789 quality will be a good practical approach to reduce the air pollution in China.



790

791 **Fig. 17.** Schematic diagram for the control of major pollutant emissions from household stoves. Solid  
792 fuel quality mainly includes the fuel volatile matter content, fuel size, and fuel form (Li et al., 2016b).  
793 Emission level mainly includes PM, BC/OC, and VOC, while some other pollutants such as SO<sub>2</sub> and  
794 NO<sub>x</sub> are not included.

795

796 Although the severe pollution situation, the resulting negative impacts from the household coal  
797 and biomass combustion, and reduction methods have been identified and emphasized, a systemic  
798 investigation of this problem and development of effective policies to significantly reduce pollutant  
799 emissions is still critically needed in China. There are large uncertainties in evaluating the air quality  
800 and health impacts resulted from the household combustion. Three major knowledge gaps are urgently  
801 needed to be filled. (1) Inventories for the temporal impact of the household solid fuel combustion on  
802 local ambient air. Since the solid fuel consumption strongly relies on season and local fuel availability.  
803 In addition, the population migration and the urbanization strongly affect the residential energy  
804 consumption. (2) Statistic data for the clean solid fuels and clean stove technologies, as well as the  
805 fuel/stove combination. As summarized in this review, the clean solid fuels and the advanced stoves

806 can bring a significant reduction of air pollutant emissions. The deployment of the residential clean  
807 fuels and clean stoves used in mainland China changes very fast and depends on local government  
808 policy. There is no statistical data for the trend of these changes, but it is extremely important for  
809 estimating the indoor and ambient air quality in China. Thus the data should be better characterized in  
810 further studies. (3) The effect of socioeconomic development on the residential solid fuel consumption.  
811 It is well known that China's fast economic development and rapid urbanization have resulted in the  
812 per-capita consumption of energy and millions of rural-to-urban migrants, as well as the solid fuel and  
813 stove selections. However, there is still no quantitated estimation of this socioeconomic effect. Further  
814 investigation on this topic is needed to better understand the effect of China's socioeconomic  
815 development on residential pollutant emissions and associated health impacts.

816 In addition to filling these gaps, further research for practically reducing household pollution  
817 emissions is also urgently needed, especially on the topic of the high quality household stove and fuels  
818 with low cost for solving the problem in long-term. The clean stove should also be low pollutant  
819 emission when burning any solid fuel of containing any level of volatile matter. As a feasible approach  
820 to control household air pollution, costs of emission reduction by the clean stove and solid fuel should  
821 be considered with comparing to other clean energies, such as natural gas and electricity. The use of  
822 proper fuel/stove combinations will improve the air quality and benefit the human health. Furthermore,  
823 emission-abatement oriented policies should base on sound scientific evidence to reduce emissions,  
824 such as that improper contents in current residential coal standards should be corrected, the household  
825 stove standard should be updated soon, and potential policy alternatives beside subsidy for really clean  
826 fuels and stoves.

827

828        **Acknowledgements**

829    Financial supports from the National Key Basic Research and Development Program of China  
830    (2013CB228505), MEP's Special Funds for Research on Public Welfare (201409002), and National  
831    Natural Science Foundation of China (41227805, 21422703, and 21521064) are acknowledged. The  
832    authors are grateful to supports from the Australia-China Centre for Air Quality Science and  
833    Management (ACC-AQSM).

834



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