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Air pollution and inhalation exposure to particulate matter of different sizes in rural households using improved stoves in central China

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ABSTRACT

Household air pollution is considered to be among the top environmental risks in China. To examine the performance of improved stoves for reduction of indoor particulate matter (PM) emission and exposure in rural households, individual inhalation exposure to size-resolved PM was investigated using personal portable samplers carried by residents using wood gasifier stoves or improved coal stoves in a rural county in Central China. Concentrations of PM with different sizes in stationary indoor and outdoor air were also monitored at paired sites. The stationary concentrations of size-resolved PM in indoor air were greater than those in outdoor air, especially finer particles $PM_{0.25}$. The daily averaged exposure concentrations of $PM_{0.25}$, $PM_{1.0}$, $PM_{2.5}$ and total suspended particle for all the surveyed residents were 74.4 ± 41.1, 159.3 ± 74.3, 176.7 ± 78.1 and 217.9 ± 78.1 μg/m^3, respectively. Even using the improved stoves, the individual exposure to indoor PM far exceeded the air quality guideline by WHO at 25 μg/m^3. Submicron particles $PM_{1.0}$ were the dominant PM fraction for personal exposure and indoor and outdoor air. Personal exposure exhibited a closer correlation with indoor PM concentrations than that for outdoor concentrations. Both inhalation exposure and indoor air PM concentrations in the rural households with gasifier firewood stoves were evidently lower than the reported results using traditional firewood stoves. However, local governments in the studied rural areas should exercise caution when widely and hastily promoting gasifier firewood stoves in place of improved coal stoves, due to the higher PM levels in indoor and outdoor air and personal inhaled exposure.

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Introduction

Solid fuels (*e.g.*, firewood, crop residues, charcoal and coal) are commonly used fuels throughout the world, especially in developing countries (Smith et al., 2004). In China, solid fuels have been widely used as the main energy source in rural households; for example, approximately 80% of the energy consumption in rural China was biomass (*e.g.*, firewood and crop residues) and nearly 10% was coal in 2003 (Zhang and Smith, 2007; Duan et al., 2014). In rural areas, solid fuel combustion was usually implemented in poorly ventilated kitchens using simple stoves and therefore led to substantial indoor emissions of various air pollutants such as particulate matter, PM (Bruce et al., 2000; Zhang and Smith, 2007; Kaplan, 2010; Leavey et al., 2015) and polycyclic aromatic hydrocarbons, PAHs (Shen et al., 2015; Chen et al., 2016). Many epidemiological studies have confirmed that continuous exposure to high concentrations of PM, particularly fine PM (such as $PM_{2.5}$ and finer particles), is closely correlated with human health risks (*e.g.*, increasing morbidity and mortality rates) of various respiratory and cardiovascular diseases (Hosgood et al., 2011; Chafe et al., 2014; Smith et al., 2014; Apte et al., 2015; Madaniyazi et al., 2015).

To diminish the health risks invoked by pollutants emitted by the indoor combustion of solid fuels, it is necessary to consume clean fuels and improve cookstoves and burning conditions. However, there is great economic pressure in rural China to rapidly and widely promote the use of some clean but relatively expensive energies, like liquefied petroleum gas (LPG) and electricity, while some common solid fuels (*e.g.*, coal and firewood) still serve as the major energy supply in households in rural China in the interim. Accordingly, adoption of improved stoves and optimization of burning conditions have been broadly promoted. For example, in Enshi County in this study, an improved coal stove (see Fig. S1 in Supplementary materials), intended to prevent fluorosis, was widely deployed. In recent years, another energy-conversation gasifier firewood stove (ZQ-JG-220, Enshi Biomass Energy Development Co., China, see Fig. S1 also), characterized by high-efficiency and low-emission through a second air supply at the top, is being strongly promoted by the local government to replace all the coal stoves. The performance of these improved stoves in pollutant emission has only been evaluated in several laboratory tests (Carter et al., 2014; Shen et al., 2012a). However, to our knowledge, the field-based measurements on the size-resolved PM emission, consequent impact on household air quality and personal inhalation exposure were quite inadequate in extensive rural areas, and thus more detailed studies are urgently required.

The objectives of the current study are to determine the exposure amounts of size-resolved PM using direct personal inhalation measurements for local residents in rural households, some of which use improved coal while the others use gasifier firewood stoves instead. In addition, daily consecutive PM with different sizes and carbonaceous fractions (elemental carbon and organic carbon, EC and OC) at different indoor and outdoor stationary sites in the rural households were also determined. The monitoring results for gasifier firewood stove were compared with improved coal stove, and also with the data on traditional firewood stove derived either from our previous studies or from the literature, so as to provide helpful advice and supporting evidence regarding the popularization of high-efficiency gasifier firewood stove in rural China.

1. Materials and methods

1.1. Description of the study area

Field sampling was performed in a mountainous area of Enshi County, Hubei Province, Central China, which has an average altitude of 1000 m and annual rainfall of 1600 mm. The local population was 4.0261 million by 2015. Due to inferior socio-economic conditions, the annual net income per capita of the rural residents in 2015 (7969 RMB) was about 30% lower than the national average of 11,422 RMB (The Bureau of Statistics of Enshi Prefecture, 2015; National Bureau of Statistics of China, 2015).

Solid fuel, including anthracite chunk and firewood, was the dominant residential energy source in the studied area. For example, anthracite chunk accounted for 74% of the residential energy consumption in 2010 (The Bureau of Statistics of Enshi Prefecture, 2010), and solid fuel combustion in households made a principal contribution to the local air pollutants. In Enshi County, a type of improved iron coal stove was utilized extensively. In recent years, another modern gasifier firewood stove was being strongly promoted by the local government to replace all the coal stoves and served as part of the national Carbon Clean Development Mechanism (CDM) project. More detailed information on solid fuels and stoves could be found in Shen et al. (2015). The houses in the local area had similar structure including a big living room and several bedrooms and storerooms. The living room usually had installed a cookstove with a chimney for daily heating and cooking, and therefore they actually served as kitchens. Accordingly, the indoor stationary sites were assigned to such living rooms.

1.2. Field sampling and pretreatment

All the personal inhalation PM samples were collected in rural households randomly selected from mountainous villages in local winter (January, 2013). With permission, one adult volunteer in each household carried a portable personal sampler. Written informed consents were apportioned individually. The subjects were required to carry the personal samplers throughout the day except sleeping, showering and using restroom, during which the samplers were placed nearby within 2 m. The size-fractionated personal exposure samples with durations of 24 hr were gathered on glass fiber filters (GFFs, 0.45 μm, BUCK, USA) using a median-volume SKC pump (Eighty Four, SKC, USA) equipped with a four-stage cascade impactor (namely, aerodynamic diameters of <0.25 μm, 0.25–1.0 μm, 1.0–2.5 μm and >2.5 μm; Sioutas Impactor, 225-370, SKC, USA) at a flow rate of 8.2 L/min. After each sampling, basic information on the selected volunteers that included age, gender, smoking behavior and cooking habits was recorded. Personal exposure measurements were implemented with 34 different residents (including 19 residents using the improved coal stove and 15 residents using the gasifier firewood stove) and each sampling lasted 24 hr (only for PM). Meanwhile, daily consecutive stationary samples (including PM and EC/OC) of indoor and outdoor ambient air were collected in pairs in the households randomly selected from the 34 households aforementioned. The stationary PM sampling equipment was exactly the same as that for the personal exposure sampling, while the 24-hr consecutive stationary EC/OC samples were collected by quartz fiber filters (QFFs, Pall QAT-UP, USA) using a low-volume pump at a flow rate of 1.5 L/min (XQC-15E, Tianyue, China).The indoor sampling was measured in the living/cooking room mentioned previously, whereas the outdoor sampling was measured in the front yard. The samplers were deployed approximately 1–2 m above the ground and ≥0.5 m from the walls. During the sampling intervals, the sampled households were similarly under poor ventilation conditions featured by most windows and doors closed regularly. Due to some sampling limitations (such as quantity of samplers and outdoors power supply), 24 available stationary samples (comprising 12 indoor samples and 12 outdoor samples) were obtained, where 12 households were randomly selected from the total of 34 households for both indoor and outdoor stationary samplings, and the corresponding sample size and cookstove types utilized in the local rural households are listed in Table S1 (Supplementary materials).

All the filters were baked at 450°C for 6 hr and equilibrated in a desiccator for 24 hr prior to weighing and sampling. After sampling, all the filters were folded and packed using aluminum foil and then stored in a refrigerator at −18°C. Finally, the filters were equilibrated in a desiccator for at least 24 hr before weighing.

1.3. Gravimetric measurements and EC/OC determinations

Gravimetric measurements were conducted using a digital balance with an accuracy of 1×10^{-5} g (XS105, Mettler Toledo, Switzerland). The EC and OC concentrations were determined using a semi-continuous Sunset EC/OC Analyzer (RT-4, Sunset Lab, USA). All of the filters were roasted in pure helium at 600°C, 840°C and 550°C for OC detection and then at 550°C, 650°C and 870°C in an O_2/He atmosphere to detect EC. The carbon results were calculated using methane at the end of each analysis cycle. The pyrolyzed OC, which was produced in helium as the temperature increased, was subtracted from the EC data according to the initial laser values (Shen et al., 2012b).

1.4. Quality assurance/quality control (QA/QC)

The flow rate of the pump was calibrated before and after each sampling interval using a primary flow calibrator (Bios. Defender 510, USA) for the SKC pump and XQC-15E pump, respectively. The blank filters in the field underwent the same pretreatment as the procedural blank for each sampling site and were then subtracted from the final results. Each filter was weighed twice before and after sampling, and the deviations were less than 5×10^{-5} g. The corresponding statistical analyses were carried out using the Statistical Program for Social Sciences (SPSS Inc., USA).

2. Results and discussion

2.1. PM concentrations and particle size distributions in indoor and outdoor air

The stationary monitoring concentrations of the indoor and outdoor size-resolved PM in rural Enshi County, including all the households using improved coal stove and gasifier firewood stove, are summarized in Table S2. The arithmetical mean of indoor $PM_{2.5}$ concentrations for all the surveyed households was up to 220.7 ± 79.2 μg/m^3, 2.9 times and 8.8 times higher than Grade II of the China national quality standard for ambient air (75 μg/m^3, Ministry of Environmental Protection of the People's Republic of China, 2012) and the air quality guideline recommended by the World Health Organization (25 μg/m^3, WHO, 2005), respectively, indicated a severe indoor air pollution even using the improved cookstoves. By contrast, the averaged outdoor $PM_{2.5}$ concentration was 100.9 ± 53.4 μg/m^3, significantly lower than that indoors ($p < 0.05$). All the indoor to outdoor (I/O) ratios for $PM_{2.5}$ were above 1.0, with the average and standard deviations of 2.5 ± 1.1, demonstrated greater emissions from indoor surroundings. Similar situations presented for $PM_{0.25}$, $PM_{1.0}$, and total suspended particle (TSP, *i.e.*, referred as the sum of PM in the four size ranges in this study) in Table S1. The PM leaked from the cookstoves increased the indoor PM concentrations, which then became concentrated due to the low-frequency of exchange between the indoor and outdoor air during the local cold winter. On the other hand, the diffusion and dilution of particles in outdoor air were usually much stronger than that in indoor air, which led to lower outdoor concentrations of PM. The local residents usually stayed indoors longer than they were in the outdoor environment during the local cold winter, and the much higher concentration of indoor PM would then cause greater potential exposures by inhalation. In urban areas, however, the concentrations of indoor PM were generally lower than those outdoors, and a few cases with elevated indoor concentrations were mainly attributed to the entrance of outdoor PM (Diapouli et al., 2011; Zhou et al., 2015), which partly indicated that the rural indoor burning of solid fuels may invoke higher concentrations of PM indoors.

Although the averaged outdoor PM concentrations were significantly lower than those indoors ($p < 0.05$), they were still higher than the reported outdoor values for most cities in China. For example, the daily mean concentration of $PM_{2.5}$ in 74 main cities in China in January 2016 was 71 μg/m^3 (Ministry of Environmental Protection of the People's Republic of China, 2016) and fell below 100.9 μg/m^3 in this study. The situation indicated the influence of the combustion of solid fuels on the local outdoor air in rural areas, consistent with the fact that the PM in rural outdoor air mainly originated from the large amount of PM emitted from the combustion of solid fuels (Zhang and Smith, 2007). A recent investigation also demonstrated that household emission was a primary and underestimated source responsible for ambient air pollution during the heating season in Northern China (Liu et al., 2016).

In view of size-resolved samples (*i.e.*, aerodynamic diameters of <0.25 μm, 0.25–1.0 μm, 1.0–2.5 μm, and >2.5 μm), the indoor and outdoor PM concentrations are listed in Table S2, and their corresponding contributions to the total PM are depicted in Fig. S2.

In the current study, fine particles were the dominant mass fraction in TSP for both rural indoor and outdoor environments. For the investigated households, the concentration ratios of $PM_{1.0}$ to TSP and $PM_{2.5}$ to TSP ranged from 56.7%–89.8%, with an average of 74.3%, and from 61.0%–93.2%, with an average of 82.1%, respectively. Former studies showed that fine particles, particularly those less than 1.0 μm, were the primary mass fraction of the total PM emitted by the combustion of solid fuels (Venkataraman et al., 2002; Chen et al., 2005; Shen et al., 2010, 2012b). For example, in reported emissions from coal combustion, fine $PM_{1.0}$ comprised up to almost 95% of the total TSP mass (Chen et al., 2005), consistent with the results in this study that fine particles with prevailing fractions in both indoor and outdoor air reflected the large influence of PM emitted from the indoor combustion of solid fuels.

Although fine PM was the majority of TSP in both indoor and outdoor air, the size distributions of PM for indoor and outdoor air were dissimilar, as depicted in Fig. 1. For indoor air, particles less than 0.25 μm occupied the largest fraction of TSP (43.5% ± 9.3%), whereas the relative contribution of 0.25–1.0 μm solid particles was predominant (46.3% ± 8.6%) in outdoor air, and <0.25 μm solid particles contributed 31.6% ± 8.0%. So the contribution of <0.25 μm indoor solid particles was significantly higher than those outdoors ($p < 0.01$). Compared with the relatively coarse particles, large amount of finer particles (<0.25 μm) emitted from solid fuel ignition in cookstoves was readily diffused into the indoor air, resulted in a greater contribution of finer particles. On the other hand, the finer particles may collapse and co-aggregate to form coarser particles during their transfer and diffusion from indoor air to outdoor air (Hinds, 1982; Richard and Spengler, 1996; Lighty et al., 2000; Shi et al., 2015), and caused the peak in the particle size distribution shifting from <0.25 μm to 0.25–1.0 μm. Another significantly higher contribution was the size greater than 2.5 μm, possibly due to resuspension of deposited ash and debris of solid fuels indoors (Zhou et al., 2015).

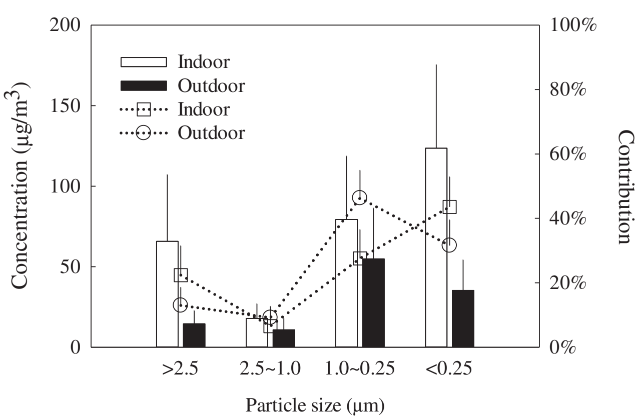

Fig. 1 – The 24-hr averaged mass concentrations of PM with different particle sizes (bar) and their contributions to TSP (line), fractionated by aerodynamic diameter at >2.5 μm, 1.0–2.5 μm, 0.25–1.0 μm, and <0.25 μm in indoor air and outdoor air.

2.2. Personal inhalation exposure concentration and particle size distribution

The arithmetical mean and standard deviation of inhalation exposure concentrations of $PM_{0.25}$, $PM_{1.0}$, $PM_{2.5}$ and TSP for all the surveyed residents using the improved coal stove and gasifier firewood stove, were simultaneously determined to be 74.4 ± 41.1, 159.3 ± 74.3, 176.7 ± 78.1 and 217.9 ± 78.1 μg/m^3, respectively. In view of $PM_{2.5}$, the exposure concentrations to local residents in rural households were apparently higher than those of the reported urban residents, with a range of 6.3 to 92.5 μg/m^3 (Williams et al., 2000; Jiang and Bell, 2008; Wheeler et al., 2011; Jahn et al., 2013; Janssen et al., 2013), which indicated more exposure to PM for the rural populations.

Our monitoring results for the exposure of all local individuals to $PM_{2.5}$ apparently exceeded the air quality guideline of 25 μg/m^3 (WHO, 2005). The finer particles ($PM_{0.25}$) ranged from 31.64 to 185.57 μg/m^3 and accounted for 33.5% ± 9.7% of TSP, also surpassed the standard. Usually, fine particles constitute the majority of detrimental components and play a governing role in the adverse effects of PM on human health (Meng et al., 2013; Chen et al., 2016). Based on an integrated exposure-response (IER) model (Burnett et al., 2014) and measured inhalation exposure concentrations of $PM_{2.5}$, more than one-third of the population-attributable fraction accounted for specific diseases (acute lower respiratory infections, chronic obstructive pulmonary diseases, lung cancer, stroke, and ischemic heart disease) could be ascribed to inhalation exposure to the local $PM_{2.5}$. The related calculation method and results could be found in Table S4, meant the local residents probably at high health risk even using the improved cookstoves. Similar to the stationary monitoring data, fine particles were the predominant part of the TSP mass for all the individual residents investigated. The concentration ratios of $PM_{1.0}$ to TSP ranged from 55.6% to 88.1%, with an average at 72.0%, and those for $PM_{2.5}$/TSP ranged from 66.3% to 94.8%, with a mean value of 80.2%.

In Fig. 2, we compared the size-resolved concentrations of PM for personal inhalation exposure and indoor and outdoor stationary monitoring. For all the particle sizes studied, the personal exposure concentrations were less than those indoors but greater than those outdoors. In this case, both indoor and outdoor air may contribute to individual exposure concentrations, and therefore it is inaccurate to establish personal exposures based on only indoor or outdoor stationary monitoring data, which only serve as rough substitutes. Portable personal sampler may be a preferable tool to quantify individual exposure. The inhalation exposure concentrations of $PM_{0.25}$, $PM_{1.0}$, $PM_{2.5}$ and TSP were more strongly correlated with the corresponding indoor concentrations than with the outdoor ones, and suggested a larger contribution from indoor air relative to that of outdoor air, as shown by the Spearman r and p values in Table S5.

Some factors like cooking and smoking behaviors, fuel/stove types and so on, were considered to influence the personal exposure extent (Balakrishnan et al., 2004; Hu et al., 2014). The exposure concentrations of the size-resolved PM for the local volunteers in different groups are summarized in Table S6. The key purpose of our study was to compare the

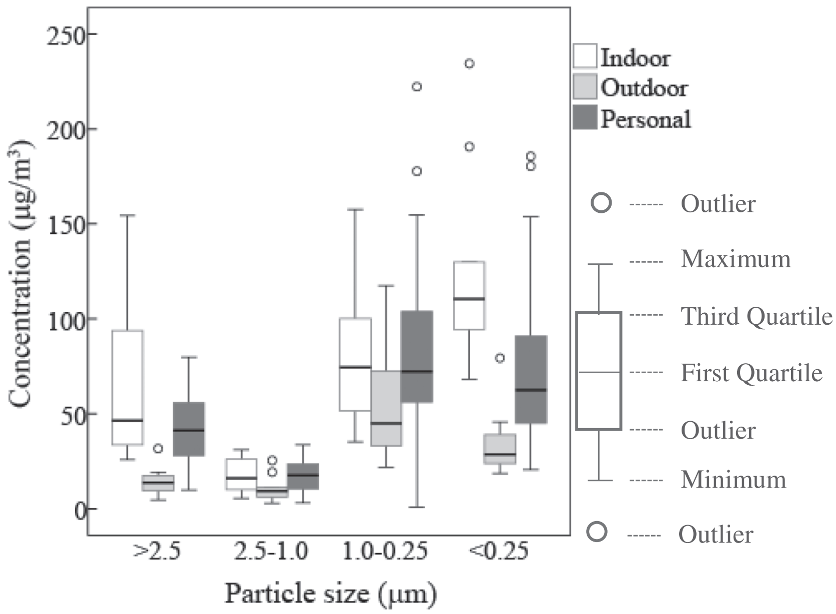

Fig. 2 – Comparison between daily averaged personal inhalation exposure and stationery concentration range of size-resolved particulate matter (open circles denote outliers), fractionated by aerodynamic diameters >2.5 μm, 1.0–2.5 μm, 0.25–1.0 μm and <0.25 μm.

performance of different fuel/stove types, so most volunteers (26 of 34 in total) in this study were non-smoking female residents mainly engaged in cooking activity. Subsequently, we did not compare their statistical significances, due to the uneven/mismatched sample size between male group and female group, between smoking group and non-smoking group, and between cooking group and non-cooking group. The corresponding differences between different households using gasifier firewood stove or improved coal stove were discussed in the following Section 2.3.2 of this study.

2.3. Comparison of indoor PM concentration and inhalation exposure level between different fuel/stove types

Different fuel/stove combinations may lead to different emitted PM amounts (Hu et al., 2014; Shen et al., 2015) and then to differences in indoor air concentrations and personal inhalation exposures. The monitoring results of gasifier firewood stove were compared with those of improved coal stove, and also with the data on traditional firewood stove obtained from our previous studies or the literature.

2.3.1. Comparison between gasifier firewood stove and traditional firewood stove

In addition to the improved coal stove, the local government is currently promoting gasifier firewood stoves currently due to their so-called high thermal efficiency and low pollutant emissions and the local energy supply characteristics in Enshi County. In this section, we compared the monitoring PM concentrations for rural households that used gasifier firewood stoves in Enshi County and in rural households that used traditional firewood stoves in other rural areas, as well as the corresponding personal exposure PM concentrations, as depicted in Fig. 3, where the value for Guizhou Province was the mean value of the data for the three types of cookstoves, *i.e.*, (557 + 533 + 337) / 3 = 476. Because various particle cutting sizes were used for PM concentrations such as $PM_{2.5}$, PM_4 and PM_{10} in previous studies, so the gathered $PM_{2.5}$ data from the cited literature were compared with the corresponding results in the present study, and the literature data for PM_4 and PM_{10} were roughly compared with our TSP results due to the absence of monitoring data on the particle sizes used in this study. Most cited data were gathered from rural households used traditional firewood stoves for heating and cooking during winter (Note: based on annual mean data in Yunnan Province). Moreover, the quoted data on Hebei Province were obtained using the same sampling, pretreatment and quantification procedures employed in this study, and due to some limitations, only the monitoring data for $PM_{2.5}$ and TSP could be offered. All the reference data are also summarized in Table S7.

The indoor stationary $PM_{2.5}$ concentration in the local rural households that used gasifier firewood stoves was 239 ± 78 μg/m^3, which was much lower than the reported range (307–1300 μg/m^3) for indoor $PM_{2.5}$ in the rural households in other areas that used traditional firewood stoves. Although we did not provide the monitoring PM_4 and PM_{10} concentrations from the current study, our corresponding result for TSP (311 ± 92 μg/m^3) was similarly much lower than the concentration ranges of PM_4 and PM_{10} (581–2400 μg/m^3) described in the literature. The individual $PM_{2.5}$ exposure concentration in the local households equipped with gasifier firewood stoves was 188 ± 80 μg/m^3, which apparently was lower than the values reported for other rural households that used traditional firewood stoves, *e.g.*, 369 ± 2 μg/m^3 in Yunnan Province (Note: based on annual average data) and 590 ± 220 μg/m^3 and 250 ± 50 μg/m^3 for cook and non-cook individuals in Hebei Province, respectively. The difference was explained that the secondary air supply in the gasifier firewood stove could enhance its air flow and combustion efficiency and subsequently diminish the PM emissions from the incomplete

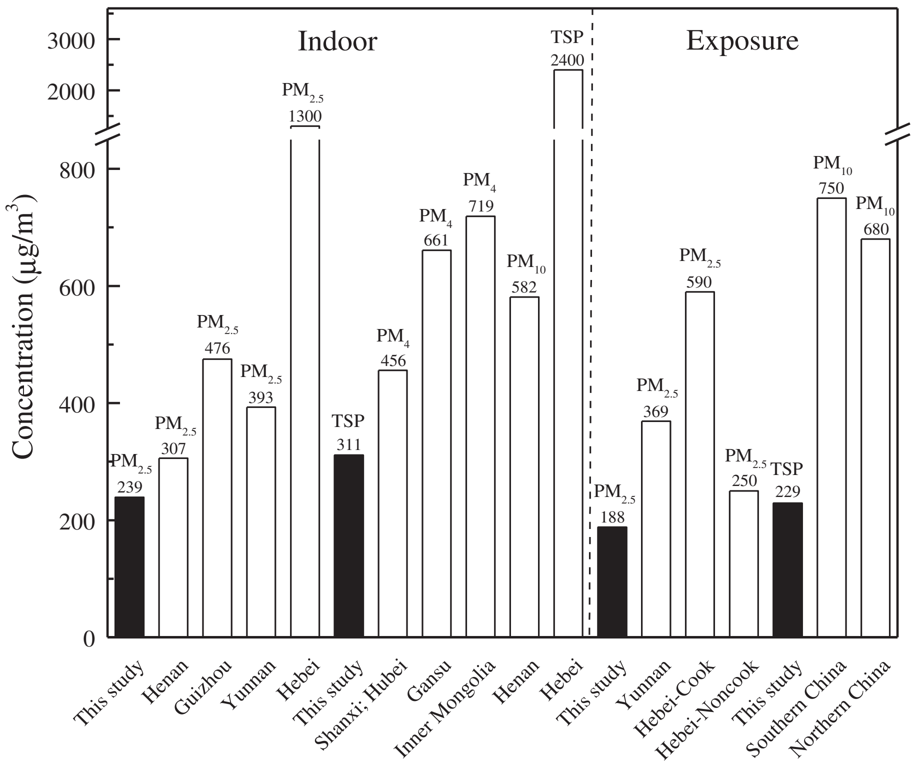

Fig. 3 – Comparison of indoor PM and inhalation exposure concentrations (μg/m^3) between traditional firewood stoves in other areas and gasifier firewood stoves in Enshi County. The data cited from the literatures in Fig. 3 are summarized in Table S7 and the corresponding references.

burning of solid fuels. In addition, the optimized inner lining and linkage between the chamber and external surface of the stove could increase the thermal efficiency and decrease the heat loss, which is also conducive to a reduction in emissions (Qiu, 2011). The obvious differences indicated that in comparison with traditional firewood stoves, the PM emission reductions using the gasifier firewood stoves was evident, which may be one of the reasons the local government is promoting that type of stove. Similar to the case of coal stoves previously mentioned, some uncertainties or influencing factors also existed such as ignition conditions and firewood properties, and further investigation therefore is required.

2.3.2. Comparison between gasifier firewood stove and improved coal stove

The daily fuel consumptions in the local households were quantified by conducting the field-based kitchen performance tests (KPTs) over an extended period of 3 days, and the related methods were described in our previous paper (Shen et al., 2015). The results of the KPT study indicated that the daily fuel consumption per capita was around 3.96 ± 1.18 kg anthracite chunk for the improved coal stove users and 4.93 ± 1.13 kg firewood for the gasifier firewood stove users, respectively. By adopting the respective conversion factors of 0.571 kgce/kg (annotation: kgce is the kg standard coal equivalent) for firewood and 0.7143 kgce/kg for raw coal (National Bureau of Statistics, 2013), the daily fuel consumptions were 2.83 ± 0.84 kgce in the local households using raw coal and 2.82 ± 0.65 kgce in the local households using firewood, respectively. Based on the measured emission factors of TSP for firewood (3.16 ± 1.5 g/kg) and coal (2.45 ± 2.62 g/kg), the mean daily emissions of TSP were 9.82 g/day/capita for the improved coal stove users and 15.4 g/day/capita for the gasifier firewood stove users. Fig. 4 in the present study shows the differences in the PM concentrations in indoor air and outdoor air and the personal exposure for rural households that used gasifier firewood and improved coal stoves. The stationary fine $PM_{1.0}$ monitoring concentrations were 220.1 ± 82.4 μg/m^3 and 100.3 ± 66.5 μg/m^3 for indoor and outdoor air, respectively, for the local rural households that used gasifier firewood stoves. The corresponding fine $PM_{1.0}$ concentrations were 185.7 ± 79.7 μg/m^3 and 82.0 ± 32.7 μg/m^3 for indoor and outdoor air, respectively, for the local rural households that used the improved coal stoves. The values for the gasifier firewood stoves were slightly higher than those for improved coal stoves. In addition, the fine $PM_{1.0}$ concentrations for personal exposure in the rural households that used gasifier firewood stoves (172.8 ± 75.0 μg/m^3, n = 15) were somewhat higher than those for the rural households that used improved coal stoves (148.7 ± 74.0 μg/m^3, n = 19); while the personal exposure concentrations were relatively approximate for the particles larger than 1.0 μm in aerodynamic diameter. To eliminate influences from factors like smoking, cooking and gender, which may interact with one another, a simple controlled analysis (Balakrishnan et al., 2004; Hu et al., 2014) was conducted to only select the exposure group of non-smoking adult females responsible for fuel burning and cooking. It was found that exposure concentrations of $PM_{0.25}$

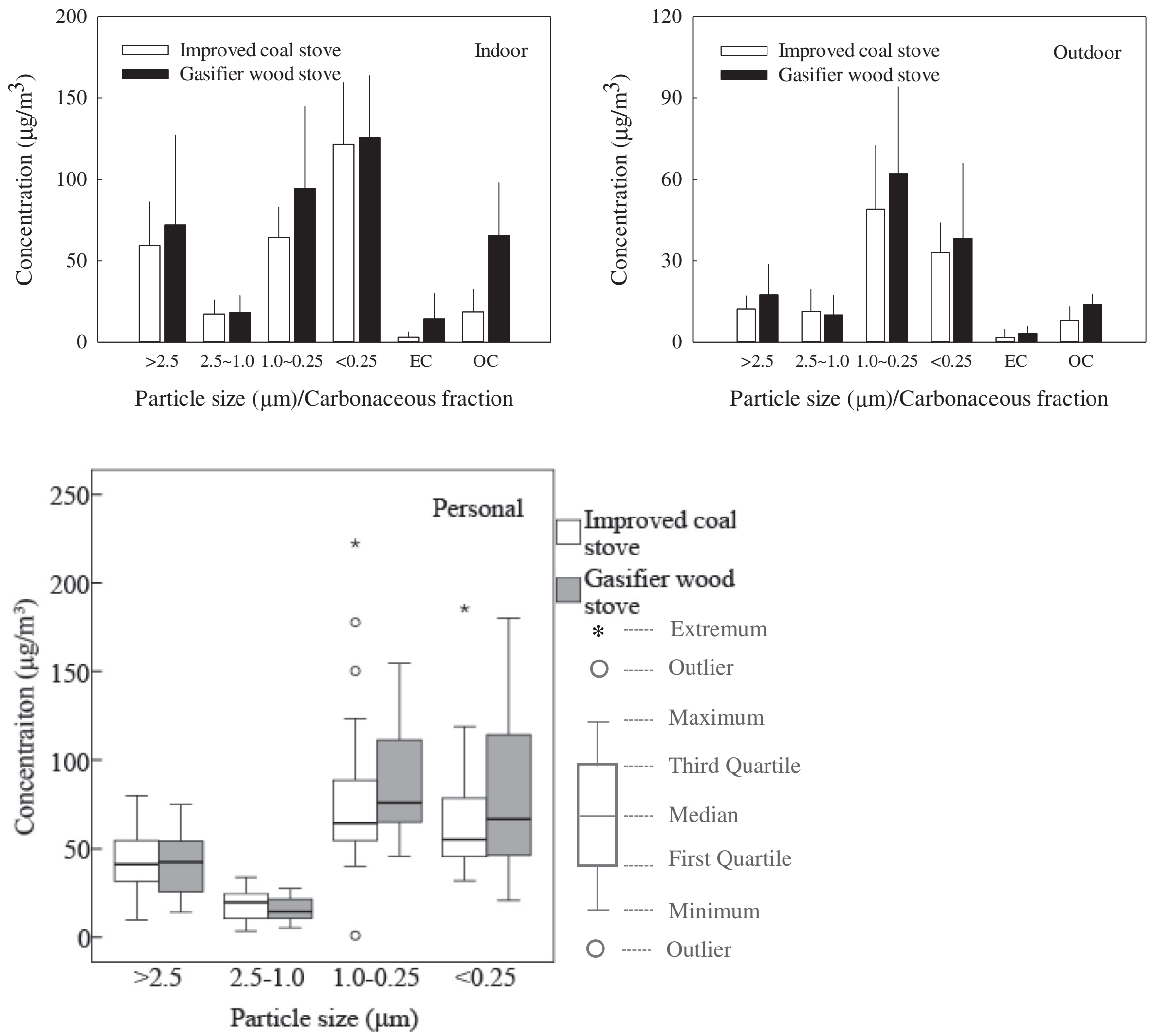

Fig. 4 – Comparison of size-resolved PM concentrations (μg/m^3) between gasifier firewood stoves and improved coal stoves in Enshi County. Upper panel: stationary monitoring concentration; Lower panel: inhalation exposure concentration.

for the households using the gasifier firewood stove (95.3 ± 45.1 μg/m^3, $n = 12$) were significantly ($p < 0.05$) higher than those using the improved coal stove (61.6 ± 38.9 μg/m^3, $n = 14$). Since the rural residents in the studied area had similar living habits, daily activity pattern and building structure, the distinction in personal inhalation exposure may, to some extent, reveal the differences in different solid fuel/cookstove types. In another work that was performed simultaneously, the emission factors (EFs) for the emitted pollutants (*e.g.*, PM and PAHs) were determined (Shen et al., 2015), where the TSP daily averaged emission amount was 15.4 g/day/capita for the local gasifier firewood stove, which was higher than that for the local improved coal stove (9.8 g/day/capita). The difference was in agreement with the stationary monitoring and personal exposure results in this study.

In addition, the total concentrations of EC and OC in both indoor and outdoor air for gasifier firewood stove (indoor: 79.9 ± 17.2 μg/m^3, and outdoor: 17.2 ± 5.9 μg/m^3) were much higher than those for improved coal stove (indoor: 21.9 ± 17.2 μg/m^3, and outdoor: 9.9 ± 7.8 μg/m^3). Similarly, the proportions of total carbonaceous fractions to TSP, *i.e.*, (EC + OC) / TSP, from both indoor and outdoor firewood combustion (indoor: 23.5 ± 14.8%, and outdoor: 18.3% ± 10.0%) were greater than those from both indoor and outdoor coal burning (indoor: 10.3% ± 9.1%, and outdoor: 8.9% ± 3.1%). A comparison experiment also indicated that EFs of EC and OC by firewood were greater than those by anthracite coal (Shen et al., 2014).

In view of the alleviation of PM emissions, use of the gasifier firewood stove as a substitute for the improved coal stove did not show the expected effects based on our monitoring data. A recently published report designated that air injection in the specially designed wood-burning stoves (similar to the secondary air supply in the gasifier firewood stoves in this study) decreased total PM emission and improved cooking performance, while increased the number concentration of total ultrafine particles ($PM_{0.1}$) during the high-power cooking (Rapp et al., 2016). In another work, it was also found that the gasifier firewood stove led to higher exposure to nitrated and oxygenated polycyclic aromatic hydrocarbons than improved coal stove (Shen et al., 2016).

Therefore, caution should be exercised in the large-scale and rapid promotion of this kind of stove in rural areas in China. As also indicated recently, the current standards adopted for evaluating healthier cookstove performance need some improvements to deliberate the total mass of pollutants emitted (Wilson, 2016). Last but not least, a small number of factors such as the different properties of solid fuels, different internal structures of cookstoves, and different combustion conditions may influence the obtained results, and thus more detailed studies with larger sample size should be implemented.

3. Conclusions

In the rural households in Enshi County, though wide adoption of improved cookstoves (improved coal stove and gasifier firewood stove), the indoor PM concentration and inhalation exposure level were much higher than the corresponding health standard. $PM_{1.0}$ fine particles predominantly contributed to indoor and outdoor PM and in the personal exposure to PM. All of the stationary monitoring concentrations of size-resolved PM in indoor air were greater than those in outdoor air, especially finer particles $PM_{0.25}$. The local individual exposure to PM with different particle sizes measured using the portable personal samplers manifested a closer correlation with indoor air than outdoor air. Due to longer exposure times to indoor PM concentrations and the resultant increased health risks *via* inhalation, local adult females should receive increased focus and concern.

The gasifier firewood stoves significantly reduced indoor PM and inhaled exposure concentrations in comparison with traditional firewood stoves. However, there was no considerable reduction in PM concentrations due to the use of gasifier firewood stoves in comparison with improved coal stoves. Accordingly, the large-scale and hasty popularization of gasifier firewood stoves as a type of clean, high-efficiency and low-emission stove by the local government to replace improved coal stoves should be met with caution. Further detailed verification studies with larger sample size should be launched in future.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.jes.2017.06.019.

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