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Quantification of emission reduction potentials of primary air pollutants from residential solid fuel combustion by adopting cleaner fuels in China

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ABSTRACT

Residential low efficient fuel burning is a major source of many air pollutants produced during incomplete combustions, and household air pollution has been identified as one of the top environmental risk factors. Here we compiled literature-reported emission factors of pollutants including carbon monoxide (CO), total suspended particles (TSPs), $PM_{2.5}$, organic carbon (OC), elemental carbon (EC) and polycyclic aromatic hydrocarbons (PAHs) for different household energy sources, and quantified the potential for emission reduction by clean fuel adoption. The burning of crop straws, firewood and coal chunks in residential stoves had high emissions per unit fuel mass but lower thermal efficiencies, resulting in high levels of pollution emissions per unit of useful energy, whereas pelletized biofuels and coal briquettes had lower pollutant emissions and higher thermal efficiencies. Briquetting coal may lead to 82%–88% CO, 74%–99% TSP, 73%–76% $PM_{2.5}$, 64%–98% OC, 92%–99% EC and 80%–83% PAH reductions compared to raw chunk coal. Biomass pelletizing technology would achieve 88%–97% CO, 73%–87% TSP, 79%–88% $PM_{2.5}$, 94%–96% OC, 91%–99% EC and 63%–96% PAH reduction compared to biomass burning. The adoption of gas fuels (*i.e.*, liquid petroleum gas, natural gas) would achieve significant pollutant reduction, nearly 96% for targeted pollutants. The reduction is related not only to fuel change, but also to the usage of high efficiency stoves.

Introduction

A variety of fuels are consumed to meet the energy demand in our society. The global total fuel consumption in 2007 was about 534 EJ, of which oil, coal, natural gas, and biomass were 154, 133, 124, and 11.4 EJ, respectively (Wang et al., 2013). Though there was only 10% (72.35 EJ) used in the residential sector for daily cooking, heating and lighting (Zhu et al., 2013), large consumption amounts of traditional solid fuels, including coal, crop straw/residues, and woody materials, resulted in significant impacts on air quality and human health (Anenberg et al., 2013; Baumgartner et al., 2014; Zhang and Smith, 2007). Household solid fuel use has been identified as an important primary source of a variety of pollutants including CO, SO_2, nitrogen oxides (NO_x), total suspended particles (TSPs), $PM_{2.5}$ (particles with aerodynamic diameter less than 2.5 μm), as well as organic and inorganic pollutants in the particles. Fine particles like $PM_{2.5}$ and $PM_{1.0}$ are of growing concern due to their deeper penetration abilities into lung areas and more adverse health outcomes. It has been estimated that residential solid fuel combustion

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produced about 33%–47% of $PM_{2.5}$, 82%–91% of primary organic carbon (OC), 46%–67% of black carbon, 10% of SO_2, and 62% of polycyclic aromatic hydrocarbons (PAHs) in China (Lei et al., 2011; Shen et al., 2013a; Wang et al., 2012; Zhao et al., 2013; Zhang et al., 2009). Globally, there are nearly 2.8 billion people relying on traditional solid fuels. In the last several decades, though the proportion of solid fuel users decreased substantially, from 62% in 1980 to 41% in 2010, the absolute population using these fuels is still very large (Bonjour et al., 2013). Recently, the first Chinese Environmental Exposure Related Human Activity Patterns Survey found that there were 43% of households using solid fuels for daily cooking, whereas for household heating, 41% of households used traditional solid fuels for heating (Duan et al., 2014). Therefore, taking the high emissions of incomplete combustion pollutants and the large population exposed to the resulting air pollution into account, household solid fuel use has been recognized as one of the top environmental risk factors, leading to over 3.5 million premature deaths in 2010 (Lim et al., 2012), and a recent updated report estimated near 4.3 million deaths in 2012 from pneumonia (12%), stroke (34%), ischaemic heart disease (26%), chronic obstructive pulmonary disease (COPD, 22%) and lung cancer (6%) (WHO, 2014).

In efforts to reduce air pollution and protect human health, it is of high priority and importance to improve our knowledge on pollutant emissions from fuel burning, which could be widely applied in the choice and evaluation of emission abatement measures, in the development of emission permits and regulations, and in intervention programs aiming to replace these so-called dirty fuels with modern and cleaner ones. Pollutant emissions often vary dramatically depending on the fuel properties, burning conditions (sometimes affected strongly by stove designs), as well as fire management behaviors (Chen et al., 2012; Shen et al., 2010; Jetter et al., 2012). Thus, local measurements are often of priority consideration in both inventory development and clean fuel intervention programs, while results borrowed from other countries may bias and increase the uncertainties of the results. It is fortunate to see that some works have been done, though still limited, during the last several years. Zhang et al. (2000) reported emission factors for CO, SO_2 and TSP for a variety of fuel-stove combinations widely used in rural China. Chen and colleagues measured emissions of carbonaceous particles for different coal types (Chen et al., 2005, 2006, 2009; Zhi et al., 2008, 2009). Shen et al. (2010, 2011, 2012a, 2012b, 2012c) reported emissions of TSP, OC, EC and PAHs from residential coal and biomass combustion in cookstoves. Li et al. (2007, 2009) and Shen et al. (2013b) also conducted field measurements to obtain more reliable emission characteristics for air pollutants from household solid fuel combustion. These studies providing valuable firsthand data filling the data gap have been widely adopted to develop emission inventories, and successfully referenced in the development of national environmental policy, regulations and emission limits.

The main objectives of this study are to carry out a comparative analysis of pollutants among different fuel types by compiling literature-reported data, and to quantitatively estimate the potential emission reductions achievable by replacing traditional dirty solid fuels with clean ones. It is expected that this mini-review study can add knowledge on pollutant emissions and possible reductions achievable for different fuel types, and solid evidence for future clean fuel and stove intervention programs.

1. Methods

As mentioned, a variety of pollutants is produced from the burning of these solid fuels. In this study, we mainly focus on CO, TSP, $PM_{2.5}$, OC, EC and PAHs. For PAHs, in addition to the summed total mass level of 15 priority PAHs (except naphthalene), the level of BeP_{eq} (Benzo[a]pyrene equivalent), calculated based on the level of individual PAHs and their corresponding toxic equivalency factors (Nisbet and LaGoy, 1992), was calculated and analyzed in this study. Measured emissions of targeted air pollutants in the literature from local measurements were collected and compiled for different fuel types commonly used in households including crop residues, firewood logs, wood branches, anthracite and bituminous coals in the forms of raw chunks and briquettes, pelletized straws, pelletized woods, natural gas and liquid petroleum gas. Most of these emission measurements were carried out after the year 2000, along with the rapid economic development in China during the last several decades. After the world-famous National Improved Stove Program during the 1980s, most cookstoves used nowadays in China are improved and modern ones. So, although it is realized that emissions vary dramatically not only within different fuel types but also among different stove designs, the current study did not cover a further fuel-stove classification. In fact, in these available studies, most cookstove designs, if mentioned, are shown as improved ones. It is noted that the difference nominally found among different fuel types, to some extent reflects emissions over the whole burning cycle affected by fuel properties, stove designs and even fire management behaviors.

Two terms of emission factor are considered. First, the fuel mass based emission factor (EF, g/kg or mg/kg) is defined as the mass of targeted pollutant emitted per unit fuel mass. This is the most frequently reported emission unit in the literature. The second unit is the per delivered energy based emission factor (EF_E, g/MJ or mg/MJ), taking thermal efficiency and fuel heating values into account, and can be calculated as $EF_E = EF / H / \eta$, where EF is the fuel mass based emission factor, H (MJ/kg) is the fuel net calorific value and η (%) is the thermal efficiency. The latter is believed to be more appropriate in data comparison due to variations in fuel properties and stove thermal efficiency.

The Emission reduction potential for pollutant i from the burning of fuel B replaced by fuel A is calculated as $R_i = 1 - EF_{E\text{-}i}(A) / EF_{E\text{-}i}(B)$, where $EF_{E\text{-}i}$ (A) and $EF_{E\text{-}i}$ (B) are delivered energy based emissions of pollutant i from the burning of fuel A and fuel B, respectively.

Statistica (StatSoft, v5.5, Tulsa, OK, USA) was used for data statistical analysis and a significance level of 0.05 was adopted. Variations in pollutant emissions are expected to result in uncertainty in the reduction estimation. The variation and uncertainty was evaluated by running a Monte Carlo simulation 100,000 times, which randomly drew input values from given probability distributions with known coefficients of variation. The results are shown as median and inter-quartile range values.

2. Results and discussion

2.1. Thermal efficiency and pollutant emissions

The thermal efficiency of traditional cookstoves in China was usually less in 10%, but after improvement, particularly the world-famous national improved cookstove program, the thermal efficiencies of most improved cookstoves are around 20%. From local emission measurements, the reported thermal efficiencies for biomass burning in household cookstoves averaged at 15.5% ± 3.9%, 17.7% ± 5.6% and 14.2% ± 0.51% in the burning of crop straw, firewood and branches, respectively. Only a few burning experiments achieved thermal efficiencies at the designed value of 20%. It is accepted that in real practice, fuel burning may not as complete as that in laboratory tests. Besides, after a period of continuous use without regular maintenance, stove thermal efficiency may decrease considerably due to flue blockage and stove degradation. Previous studies in the field have shown that pollutant emissions from old stoves may be 2–4 times those from new stoves (Roden et al., 2009; Wei et al., 2014). As shown in Fig. 1, the burning of coal briquettes in improved coal stoves and pelletized biomass fuels in modern gasifier burners have relatively higher thermal efficiencies, at about 30%-35%. In coal briquette burning, the better coal-air mixing status results in good burning conditions (Zhi et al., 2009). Pellets have been widely used in developed US and European regions, but in China are still mainly in several pilot programs. Their deployment has been strongly recommended in the Chinese renewable development plan. For pelletized fuels, usually specific modern burners should be used for these modern fuels. Emission measurements, though limited, reported that the thermal efficiency of pellet burning may as high as 35.6% ± 5.4% (Carter et al., 2014; Shen and Xue, 2014). The highest efficiencies are found in the use of liquid petroleum gas (LPG) at 42%–45% and natural gas (NG) burning, at about 50%–60% (Zhang et al., 2000). High efficiencies and low pollutant emissions for gas use are often expected, however it is important to note that there has been very scarce tests on gas burning so far. More data are expected, but it might be a challenge because of much low emissions and the influence of background levels.

As expected, the lowest pollutant emissions were found in the burning of clean gas fuels, like NG and LPG. Much high pollutant emissions are produced in the burning of raw coals, especially bituminous ones, and the burning of ordinary biomass fuels including crop straws, firewood logs and branches. The difference between anthracite and bituminous coals has been widely documented. Generally, the low volatile matter and ash contents of anthracite coals make it easier to achieve high burning efficiencies and subsequently low pollutant emissions. The difference between raw chunk and briquettes has also been mentioned previously. The change in coal form, together with slight differences in coal properties (for instance during the manufacture of these briquettes some bonding additives may be used), would cause marked differences in fuel burning conditions and pollutant emissions. Significant differences are found between firewood logs and branches/brushwood, which are explained by the different fuel-air mixing status and burning rates during woodstove combustion. The relatively fast burning and irregular sequence of branches in the stove chamber would result in insufficient air supply and less complete burning, and therefore relatively higher pollutant emissions. By compressing the materials into pellets, biomass fuel densities are increased dramatically, making these fuels convenient for storage and transport but also efficient burning. Compared to uncompressed biomass fuels, significantly lower pollutant emissions are found for pellets, which is one important consideration during their deployment.

2.2. Quantification of emission reduction by clean fuel intervention

It is gratifying, and moreover usually expected, to see a generally inverse relationship between pollutant emissions and burning thermal efficiencies (Fig. 1), though it is noted that high thermal efficiency does not necessarily mean high burning efficiency and low pollutant emissions, as documented in some previous studies (Edwards et al., 2004; Zhang et al., 2000). The burning efficiency is the ratio of energy released by combustion

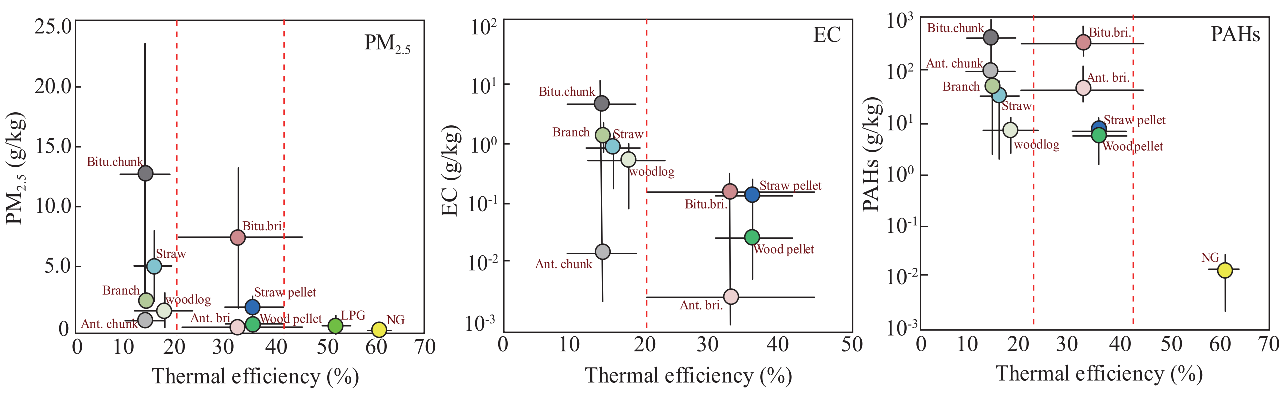

Fig. 1 – Comparison of air pollutant emission factors and burning thermal efficiencies when burning different fuel types including straw, firewood logs, branches, anthracite chunk coal (ant. chunk), bituminous chunk (bitu. chunk), anthracite briquette (ant. bri.), bituminous briquette (bitu.bri.), liquid petroleum gas (LPG) and natural gas (NG). $PM_{2.5}$, elemental carbon (EC) and polycyclic aromatic hydrocarbons (PAHs) are shown as examples of air pollutants from incomplete household fuel burning.

to the energy in fuel, while overall thermal efficiency is the ratio of cooking energy (usually tested from the water boiling tests) delivered by the fuel energy. The difference between them is the heat transfer efficiency, which is the ratio of energy delivered to cooking to the energy from combustion. The thermal efficiency directly relates to the amount of fuel consumed. Some fuel burning processes in stoves reduce fuel use (increasing thermal efficiency) by improving the heat transfer efficiency, but result in lower burning efficiency in the process, which consequently produces more pollutant emissions per unit fuel mass (Jetter et al., 2012). Low thermal efficiencies in household cookstove burning would require high amounts of fuels burned to meet the energy demand. In order to compare emissions among different fuels, per useful delivered energy based emissions, instead of fuel mass based emission factors, are calculated taking fuel net calorific values and burning thermal efficiencies into account. Low fuel mass based emissions and high thermal efficiencies, like the burning of gas, briquettes and pelletized fuels, result in much lower per energy based emissions. Fig. 2 shows per useful energy based emissions of $PM_{2.5}$, EC and PAHs, as examples, among different fuel types. Bituminous chunks appear to be the dirtiest fuels with the highest pollutant emissions, followed by uncompressed raw biomass fuels, while gas fuels are the cleanest ones, and anthracite briquettes and pellets are relatively clean fuels, producing lower pollutant emissions on a per useful energy basis.

Coal briquette burning has not only low fuel mass based pollutant emissions but also increased thermal efficiencies compared to raw chunk combustion, leading to a much greater reduction in air pollutants per unit delivered energy used. Thus for coal, briquetting technology is of wide interest in terms of emission reduction. It was estimated that 82% (71%–88% as inter-quartile range) CO, 99% (94%–107%) TSP, 76% (54%–97%) $PM_{2.5}$, 97% (95%–99%) OC, 92% (87%–96%) EC, 80% (50%–101%) PAHs and 84% (64%–109%) BaP_{eq} from anthracite chunk burning may be reduced if these anthracites were manufactured and used in briquettes. Similarly, significant reductions could be also expected for bituminous coals (Fig. 3a).

Biomass fuels are widely consumed because they are easily accessible and nearly free of charge, but inefficient burning in

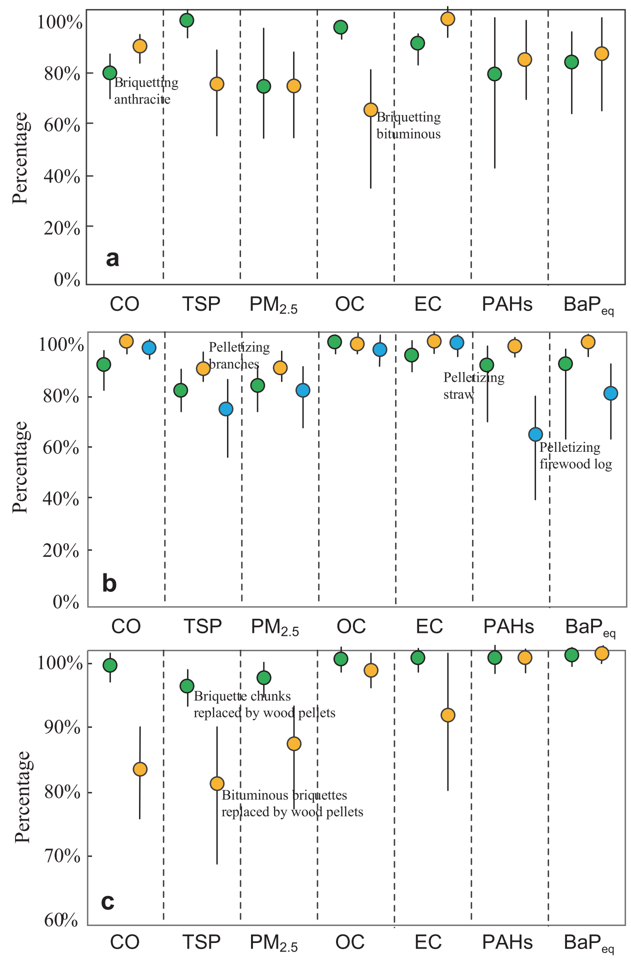

Fig. 3 – Mitigations of incomplete air pollutants from residential sector by fuel adoption with technologies of (a) coal briquetting (b) and biomass pelletizing, and (c) emission reductions by the replacement of fossil coals by modern pelletized woods. Data shown are medians and quartile ranges. OC: organic carbon; TSP: total suspended particles; BeP_{eq}: Benzo[a]pyrene equivalent.

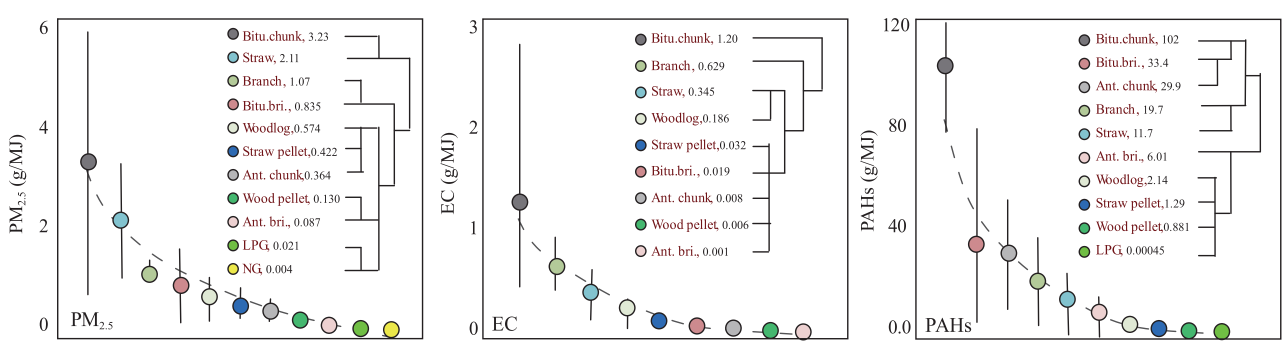

Fig. 2 – Comparison of air pollutant emissions per unit delivered useful energy among different fuel types including straw, firewood logs, branches, anthracite chunk (ant. chunk), bituminous chunk (bitu.chunk), anthracite briquette (ant. bri.), bituminous briquette (bitu. bri.), liquid petroleum gas (LPG) and natural gas (NG). $PM_{2.5}$, elemental carbon (EC) and polycyclic aromatic hydrocarbons (PAHs)are shown as examples of air pollutants from incomplete household fuel burning.

the residential sector results in extensive emissions of air pollutants, harming air quality and human health. Deployment of pelletized biomass fuels is considered as one choice. If crop straws were used in the form of pellets, there would be 88% (81%–92%) CO, 79% (71%–87%) TSP, 80% (72%–87%) $PM_{2.5}$, 96% (93%–100%) OC, 91% (85%–95%) EC, 89% (69%–94%) PAHs and 89% (60%–94%) BaP_{eq} emissions reduced compared to indoor straw burning. Similarly for woody materials, about 97% CO, 87% TSP, 88% $PM_{2.5}$, 96% OC, 99% EC, 96% PAHs and 98% BaP_{eq} emissions from branch combustions can be reduced by replacing uncompressed branches by wood pellets.

If traditional solid fuels were replaced by gas fuels, generally over 96% of most targeted pollutants could be eliminated. But the relatively high cost of gas fuels prevents large-scale adoption of gas, especially in some remote rural areas. It is of interest to look into reduction potentials from the replacement of coal fuels by pellets, since even in the form of briquettes, residential coal combustion still produced relatively high amounts of air pollutants. For instance, the EF_E for bituminous briquettes were 3.76 g/MJ (CO), 0.922 g/MJ (TSP), 0.835 g/MJ ($PM_{2.5}$), 0.524 g/MJ (OC), 0.019 g/MJ (EC), 33.4 mg/MJ (PAHs), and 4.38 mg/MJ (BaP_{eq}), whereas for wood pellets, they were 0.746 g/MJ (CO), 0.199 g/MJ (TSP), 0.130 g/MJ ($PM_{2.5}$), 0.016 g/MJ (OC), 0.006 g/MJ (EC), 0.881 mg/MJ (PAHs), and 0.015 mg/MJ (BaP_{eq}). Therefore, the replacement of bituminous briquettes by wood pellets would result in obvious emission reductions (Fig. 3c). Deployment of pelletized biomass fuel can not only result in significant reductions in air pollutants, but also benefit climate change by reducing CO_2 emissions from fossil fuel use. In fact, there has been some pellet deployment programs available in some pilot counties located in central and western China.

2.3. Implications and limitations

Nowadays, crop straws, woody materials and coals are commonly used in the residential sector, and they are large contributors to air pollutant emissions while pollutant emissions from gas burning are minor, though gas has been widely used in most urban households. The use of pellets is still only in the pilot stage. Household biomass or coal cookstoves are commonly found in the field. More and more households are equipped with more than one stove. Electric induction cookers are becoming more and more popular, but sometimes the use of these induction cookers may have difficulty meeting the requirements of residents' cooking behaviors, especially Chinese cooking habits.

As a rough estimate, annual emissions from residential crop straw, wood and coal combustion averaged 52.7 Tg CO, 3.71 Tg TSP, 3.13 Tg primary $PM_{2.5}$, 1.17 Tg OC, 0.796 Tg EC and 44.7 Gg PAHs, which comprised nearly 30%-60% of total national emissions. Among different fuel types, crop straws (59.8% CO, 57.1% TSP, 58.6% $PM_{2.5}$, 49.9% OC, 37.8% EC, and 22.8% PAHs) and bituminous chunks (16.4% CO, 25.4% TSP, 26.1% $PM_{2.5}$, 32% OC, 38% EC and 57.8% PAHs) contributed the most to the total emission from the residential sector, followed by branches (~10%–20%) and firewood logs (~5%–10%) (Fig. 4). Therefore, to reduce pollutant emissions from residential burning, the use of crop straws and bituminous chunks should be eliminated by either change-out interventions or improving burning efficiency.

If current raw chunk coals in households were replaced by briquettes, there would have a total of 9.02 Tg CO, 0.74 Tg TSP, 0.61 Tg $PM_{2.5}$, 0.25 Tg OC, 0.30 Tg EC, and 22.5 Gg PAH emissions reduced, equal to about 17.1% CO, 19.9% TSP, 19.5% $PM_{2.5}$, 21.4% OC, 37.7% EC, and 50.3% PAH emissions from the residential sector. The use of pellets instead of traditional raw biomass fuels could result in 38 Tg CO, 2.12 Tg TSP, 1.81 Tg $PM_{2.5}$, 0.72 Tg OC, 0.46 Tg EC, and 13.6 Gg PAHs reductions, equal to about 72.1% CO, 57.1% TSP, 57.8% $PM_{2.5}$, 61.5% OC, 57.8% EC and 30.4% PAH emissions in the current residential sector.

The notable reductions in pollutant emissions from the primary combustion sources would lead to considerable improvement of air quality, and consequently benefit human health and regional climate change significantly (Edwards et al., 2004; Mestl et al., 2007). It was previously estimated that there would be 166 and 65 $\mu g/m^3$ reductions in PM_{10} levels in urban and rural China, corresponding to about 0.69 and 0.63 million premature deaths avoided, respectively, if all urban households and rural areas switched to clean fuels (Mestl et al., 2007). The use of briquettes, in comparison with the use of raw chunk coal, may lead to a reduction of 53% in global warming commitment (GWC, defined as total warming committed by burning 1 kg fuel) if taking only gases like CO_2 and CH_4 into account (Edwards et al., 2004), and in terms of the climate impact of ambient particles, it was reported that a reduction of 600 Gg black carbon (BC) after the deployment of improved stoves and briquettes would be equivalent to about 1320 Mton CO_2, based on the 20-year integration period global warming potential of BC (Zhi et al., 2009).

The present study suggested that there would be an over 80% general reduction in air pollutants (89.2% CO, 77.1% TSP, 77.3% $PM_{2.5}$, 82.9% OC, 95.5% EC and 80.8% PAHs) from the deployment of two technologies (briquetting coals and pelletizing biomass), but it comes to mind that the expected decreases in the ambient concentrations of these air pollutants may differ from the reduction degree in primary emissions because of non-linear relationships. In addition, the reduction degree of ambient levels may also vary among different pollutants as their physico-chemical properties, for example deposition and degradation abilities, differ markedly. It was once reported that a 30% emission reduction in PM_{10} emission by clean fuel switching would result in 12%–15% decrease of ambient PM_{10} level (Mestl et al., 2007). A large stove change-out program in Montana found that 64% average reduction occurred in air PAHs levels, but only a 20% drop in $PM_{2.5}$ concentration (Ward et al., 2009). It is of wide interest to investigate the change in subsequent environmental loadings and impacts on human health and climate change of these pollutants after effective clean fuel intervention programs are in place, with more information available from both experimental measurements and advanced models.

One notable limitation of the comparative analysis and quantification is the scarcity of data available so far, resulting in obvious uncertainties in results and indicating the need for more emission measurements in the future. In addition, only a few pollutants are analyzed here as examples, and the combustion process produces a variety of pollutants that are not addressed in this study but which have been documented to significantly affect air quality and human health, like SO_2 and NO_x, of which residential coal combustion, especially low-quality bituminous coals, is one main contributor. Thus

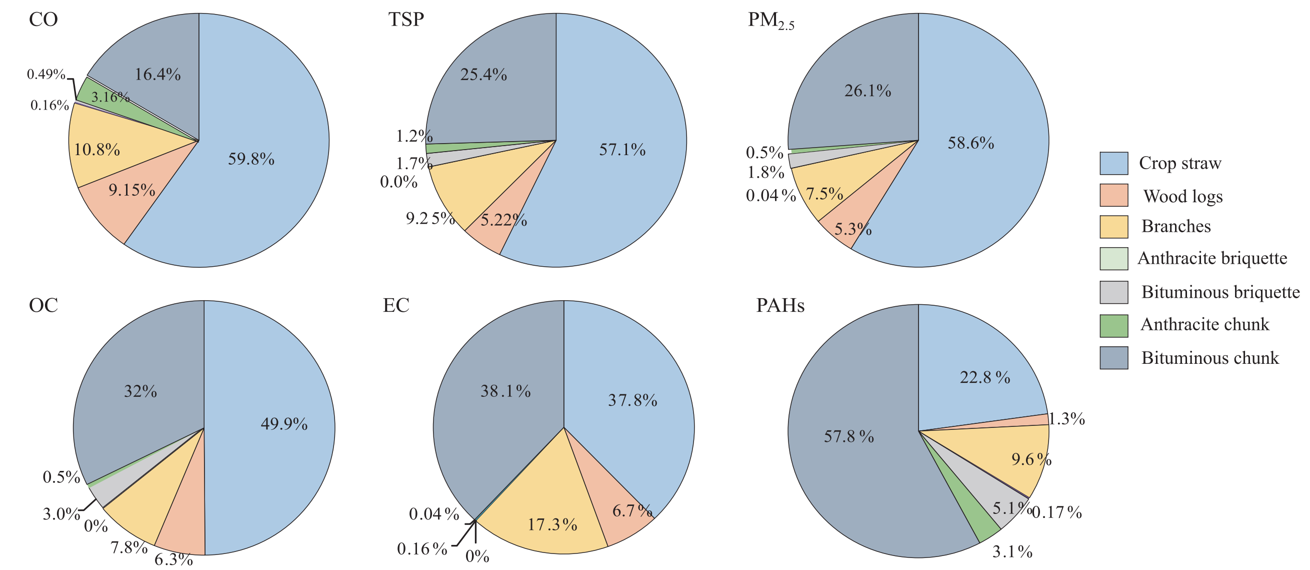

Fig. 4 – Contributions of different fuel types to the total residential emissions of CO, TSP, $PM_{2.5}$, organic carbon (OC), elemental carbon (EC) and polycyclic aromatic hydrocarbons (PAHs).

more measurements, not only laboratory tests but also field investigations, should be encouraged to provide more evidence with higher confidence levels for clean fuel intervention programs. Also, as mentioned in the Methods section, though different fuel types are considered, pollutant emissions are directly affected by stove types as well. Due to the limited information available, we did not classify pollutant emission factors according to fuel-stove combinations, but it is highly expected that this aspect will receive attention as more data become available in the future.

3. Conclusions

Based on available results from local emission measurements on residential fuel burning, which is a major source of air pollutants in China as well as other developing countries, we compared air pollutant emissions among different fuel types, and addressed reduction potentials achievable by clean fuel interventions in this study. Without doubt, gas fuels like LPG and NG are much cleaner fuels compared to solid fuels, but availability and affordable cost are usually barriers affecting a large-scale adoption, especially in some remote rural areas. For fuels such as briquettes and pellets, lower pollutant emission factors per unit fuel mass and also high stove thermal efficiencies will lead to significant reductions in air pollutant emissions, which greatly enhances our confidence in future clean fuel intervention programs. It is out of scope here to further look into the differences among different inventories, but it is highly believed and strongly recommended that more local measurements should be conducted to obtain more realistic data, so as to develop reliable emission inventories in the future. It is also necessary to identify barriers and enablers affecting clean fuel and stove adoption, which may help policy makers to issue effective and practical policies and regulations supporting the intervention.

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