Comparative Study of the Impact of Biomass and Clean Fuels Burning as Cooking Energy on Rural Indoor Air Quality in Heilongjiang, China

Yue Zhang, Yu Zhang and Zhicheng Wang*
Energy and Environmental Research Institute of Heilongjiang Province, Heilongjiang Academy of Sciences, Harbin, Heilongjiang, 150090, China

*Corresponding author's e-mail: hnh_wzc@126.com

Abstract. In rural areas of the northeast China, the majority of residents use traditional Chinese Kang-stoves for heating and cooking by burning biomass fuels. However, this method is associated with serious indoor air pollution problems. The purpose of this study is to evaluate indoor air quality impacts from biomass combustion and indoor air quality improvements derived from using clean fuel. A comparative study was conducted in three rural households of Heilongjiang to determine indoor air pollution extent during burning biomass (rice straw, cornstalk and corncob) and liquefied petroleum gas (LPG) fuels for cooking. The results indicated that the concentration of CO from the combustion of four kinds of fuels was almost below the guidelines prescribed in Chinese Indoor Air Quality Standard. On the contrary, PM2.5 concentrations exceeded the limiting value regardless of which fuel was used, and the lowest PM2.5 concentration values, which were obtained during burning LPG, were about 2.4-7.5 times of 24-h mean PM2.5 concentration limit. The mean values of formaldehyde concentrations during using four types of cooking energy were totally higher than the standard limit, and the lowest formaldehyde concentrations were detected when corncob was used as cooking energy. Compared with LPG fuel, biomass fuels combustion in traditional Kang-stove with chimney increased the concentrations of TVOC and BTEX. To conclude, the pollutants generated from using LPG are relatively less harmful. Among the three biomass fuels, the minimal pollution derives from the combustion of corncob.

1. Introduction
In low and middle income rural areas, indoor air pollution is largely generated by household fuel combustion. For billions of people, cooking a meal or heating a room can turn the household into a dangerous place. Exposure to the smoke produced by burning polluting fuels kills 4.3 million people each year. Over three billion people use polluting fuels and devices, such as wood, coal and straw in simple stoves for their daily cooking [1]. Indoor air pollution is one of the leading causes of disease and premature death in the developing world. In 2016, household air pollution was responsible for 3.8 million deaths, 7.7% of the global mortality, and 50% of pneumonia deaths in children under 5 are due to household air pollution [2].

People in rural household of freezing Heilongjiang in the northeast China use coal and biomass fuels, such as straw, cornstalk and corncob for heating and cooking in cold winter. Due to the cold weather, doors and windows are rarely opened during the cooking process, and the smoke ventilator is rarely used. Consequently, indoor air pollution could be much more severe due to poor ventilation conditions. Exposure to the fumes produced by cooking leads to uncomfortable symptoms such as
eyes sting and throat discomfort. Researcher has revealed that fumes from biomass burning for cooking can contain volatile organic compounds (VOCs), aldehyde, H₂S and other hazardous pollutants due to incomplete combustion of carbonaceous components in the food materials [3]. Many studies have investigated the amounts and chemical compositions of PM emitted during food cooking processes. Relatively less research focusing on gaseous emissions especially formaldehyde and VOCs from cooking activities has been done. Huang W. et al. analyzed the mass and chemical composition of PM samples in rural Yunnan China, including water-soluble organic carbon, black carbon (BC) and molecular markers. The results indicated that black carbon, n-alkanes and levoglucosan dominated the most abundant fractions of the total species [4]. Huang Y. et al. measured inhalation exposure of PM in Shanxi China by using personal samplers. The mass fractions of PM₂.₅ in TSP were 90%, 72%, 65% and 68% in the kitchen, bedroom, outdoor air and personal inhalation exposure [5]. Sidhu et al. measured the real-time temporal variation of PM₂.₅ and CO concentrations in various types of rural household kitchens. The results revealed that people who use solid biomass fuel are four times more exposed to harmful pollutants than those who use clean fuel [6]. Kelp et al. monitored real-time indoor CO, PM₂.₅ and BC concentrations in houses using either traditional open fire stoves or carbon-finance-approved cookstoves. Implementation of the intervention cookstove decreased concentration of CO and PM₂.₅ but increased BC concentration [7].

The objective of this work is to: (1) assess the impact of indoor cooking activities on rural indoor air quality in Heilongjiang Province; (2) quantify the concentrations of CO, PM₂.₅, formaldehyde and BTEX (benzene, toluene, ethylbenzene and xylene) from the combustion of different cooking fuels (both biomass fuels and clean fuel); (3) identify correlations between the formaldehyde and BTEX concentration variations and fuel types.

### 2. Material and methods

#### 2.1 Study area and household selection

![Fig. 1. Examples of the kitchens and stoves in three households.](image)

Table 1. Introduction of the three households.

| Household | Location | Cooking fuel       | With or without ventilation |
|-----------|----------|--------------------|-----------------------------|
| A         | Wuchang  | Rice straw and LPG | Without                     |
| B         | Zhaodong | Corncob            | Without                     |
| C         | Zhaozhou | Cornstalk          | With                        |

The cooking emission tests were conducted in February, May and September 2018 in three villages located at Wuchang, Zhaodong and Zhaozhou in Heilongjiang separately. Since the use of cooking energy in each village is roughly the same, three representative households were selected as the research object in each village (Fig. 1), which were selected according to the fuel type they use. All of the three households adopt traditional biomass cooking-Kang stoves, while household A also adopts liquefied petroleum gas (LPG) stove occasionally (Fig. 1 square line and Table 1). The kitchens are separated from the bedroom area but connected with the bedrooms through Kang path.
2.2 Indoor air pollution monitoring

Emissions sampling was conducted during lunch time and the residents were asked to cook the same dishes and operate the stoves as they did in daily lives. The sampling process lasted for 30 min starting from fire ignition to flameout. The height of the equipment was set at 1.2 m from the floor, and kept a distance of approximately 1 m from the stoves. Gaseous CO was measured using a handheld electrochemical CO sensor (WASP-XM-E-CO, Guorui Instrument, China, accuracy: ±2%). The PM$_{2.5}$ concentration was quantified by a portable automatic dust sampler (DustTrak II 8530, TSI Incorporated, USA, measurement range: 0.001-150 mg/m$^3$; accuracy: ±0.001 mg/m$^3$). Formaldehyde was detected by a portable detector with formaldehyde sensor (PGM-6208, RAE Systems Inc., USA, accuracy: ±5%). All the above equipment was zero-checked before each sampling process. The pollutant concentrations were recorded simultaneously at an interval of 1 min.

2.3 Sampling and Analysis of VOCs

An atmospheric sampling pump (QC-2BI, Loobo Co., Ltd, China) was applied to take VOCs samples. The pump was calibrated before each sampling period using a soap film flow meter (ZM-103B, China). The gaseous samples were concentrated in stainless steel desorption tubes filled with Tenax TA adsorbent (Markes) by pump at the flow rate of 0.2 L/min, and the sampling duration was 30 min. After returning to the laboratory, VOCs were desorbed by a thermal desorption instrument (TD-100, Markes Ltd, England) and then analyzed by gas chromatography (GC) with a flame ionisation detector (FID) (GC 2010-Plus, Shimadzu Corporation, Japan). The VOCs-Tenax TA tubes were thermally desorbed at 280°C with pure nitrogen at 10 psi for 8 min. The flow path temperature was set at 180°C. VOCs were injected into the column (split ratio 31) by fast heating of the trap to 300°C using nitrogen as carrier gas (10 psi). Inertcap-1 capillary column (30 m, 0.25 mm i.d., 0.25 μm) was used for the separation of VOCs. The GC oven temperature program was set as: 50°C kept for 7 min, from 50°C to 190°C at 10°C/min, and 190°C kept for 4 min. The GC-FID system was calibrated with liquid standard solutions contained eight VOCs at concentrations ranging from 20-3000 ng/mL in advance. The R$^2$ values of the eight VOCs calibration curves were 0.9990-0.9995.

The concentrations of TVOC were the sum of identified VOCs concentrations, which were quantified by each calibration curve, while the unidentified VOCs concentrations were calculated with the response factor of toluene.

3. Results and discussion

3.1 Indoor CO concentrations

![Figure 2. CO concentration during lunch in different fuel type households.](image)

CO is formed by the incomplete combustion of wood, coal or biomass fuel. In recent years, a number of studies have focused on the CO contamination from wood or coal burning stoves, and demonstrated that CO can be a major pollutant from cooking smoke in rural household [8-11]. Considering the most adverse effects, the concentrations of CO were statistical calculated as medium values, maximum
values and the arithmetic mean values from median to maximum in this study. The highest CO concentration was detected in the household using LPG as cooking energy. CO emitted from combustion of rice straw and cornstalk were significantly higher than that from combustion of corncob (Fig. 2). This result is different from the conclusion of Indian researchers. Sidhu’s results revealed that the level of CO values ranged from 0.44 ppm to 2.42 ppm in LPG and biogas using households. The highest CO concentration was observed in kitchens using cow-dung cakes followed by agricultural residue > firewood > biogas > LPG [6]. Chakraborty observed that a higher level CO was released from coal cake, dry leaf and cow dung followed by wood, straw and least LPG [10]. The difference in results is mainly due to the release of CO is directly related to the structure of stoves and the burning rate. By comparing the pictures of Indian and Chinese stoves, it is found that Indian stoves are placed separately in the kitchens for cooking only, while the Chinese traditional stoves are connected to the Kang path for cooking and heating, then can be exhausted to outside through the chimneys.

3.2 Indoor PM$_{2.5}$ concentrations

In addition to the combustion of solid fuels such as biomass, wood and coal, the cooking operational processes such as frying, stir-frying, grilling and roasting also contribute to particulate matters and are affected by ingredients, recipes and procedures, fuel types, temperature and ventilation equipment [12]. The residents were asked to cook the same recipes using the same ingredients, and the ventilation equipment in household C was turned off in this comparative test. The lowest PM$_{2.5}$ concentration was observed in the kitchen using LPG as cooking energy, and the maximum was 430 $\mu$g/m$^3$, the mean
from median to maximum was 180 μg/m$^3$ (Fig. 3). That values were about 2.4-7.5 times of 24-h mean PM$_{2.5}$ concentration limit published in the Ambient Air Quality Standard of China \[13\]. This type of PM$_{2.5}$ concentration was mainly produced by cooking course rather than the combustion of fuel. The highest PM$_{2.5}$ concentration was detected when rice straw was used as fuel for cooking and followed by cronstalk > corncob. During lunch cooking period, results in Fig.4 shows that variation of PM$_{2.5}$ concentration in household using LPG stove was stable and much lower than those from using traditional biomass Kang-stoves. After the fire is extinguished, PM$_{2.5}$ concentrations in kitchens with three kinds of biomass cooking energy remained high level, and the values were about 6.9-249 times of PM$_{2.5}$ concentration limit (75 μg/m$^3$).

3.3 Indoor formaldehyde concentrations

Table 2. The median, maximum and med-max mean of HCHO concentrations emitted from cooking activities using different fuels (unit: mg/m$^3$).

| Fuel type    | Med  | Max  | Med-Max mean |
|--------------|------|------|--------------|
| Rice straw   | 0.472| 0.568| 0.511        |
| Cornstalk    | 0.720| 0.884| 0.784        |
| Corncob      | 0.092| 0.304| 0.149        |
| LPG          | 1.028| 1.988| 1.453        |

Aldehydes can not only be produced from the combustion of fuels, but can also be produced through chemical reactions during food cooking processes. The concentrations of formaldehyde (HCHO), as a dominant aldehyde in indoor environment, emitted from cooking activities using different fuels were monitored in this study. Table 2 listed the median, maximum and med-max mean concentrations of HCHO in this measurement. Maximum HCHO concentrations of the three selected households using four types of cooking energy varied from 0.304 to 1.988 mg/m$^3$, which were much higher than the upper limit of 0.1 mg/m$^3$ stipulated in Chinese Indoor Air Quality Standard \[14\]. It is worth noting that higher level of HCHO were generated during using LPG as cooking energy, and the variation of concentration was sharply (Fig. 5). However, it appeared that the fluctuation of HCHO concentrations, when biomass fuels were used, varied gently. Lowest HCHO concentrations were detected in household B where the corncob was used for cooking energy. In summary, regardless of which fuel was used, the concentration of HCHO during cooking activities exceeded the limit level. Compared with previous research results concluded by Kabir, Khalequzzaman and Fan \[15\-17\], it can be seen that formaldehyde level generated from cooking activities in rural households using rice straw, cornstalk and LPG as fuel energy is higher than those using charcoal, wood and natural gas.
3.4 Indoor BTEX and TVOC concentrations

VOC species mainly including benzene, toluene, ethylbenzene and xylene (BTEX), which are aromatic compounds, were detected in this work. The average concentrations are given in Table 3. Compared to LPG combustion, biomass combustion in traditional Kang-stoves with chimney increased the concentrations of TVOC by 4.3-5.7 times, and increased the concentrations of BTEX by 7.5-18.5 times. The rice straw fuel combustion showed highest levels of benzene, toluene and BTEX, while the cornstalk fuel combustion showed highest levels of ethylbenzene and xylene. In general, the highest emission of TVOC was obtained from the combustion of cornstalk in the Kang-stove. Sinha et al. assessed the exposure of cooks to benzene and toluene from dung and wood biomass fuels combustion in rural homes. Results revealed that the level of benzene exposure was 0.114 mg/m³ and 0.045 mg/m³ using dung fuel and wood fuel, respectively [18]. However, the mean values for toluene concentrations using dung fuel and wood fuel were lower than the results obtained in this work. Wang et al. reported that wood combustion caused the most serious indoor air pollution among coal, wood and biogas fuels, with the TVOC concentrations of 0.467±0.338 mg/m³ [19]. This result is close to the TVOC concentration when burning rice straw and corn cob in this study. Simultaneously, TVOC concentration for coal combustion in metal stove was reported and the value was 0.695 mg/m³, which is close to the TVOC concentration when burning cornstalk in this study.

Table 3. The BTEX and TVOC concentrations emitted from cooking activities using different fuels a.

| Fuel type | Benzene | Toluene | Ethylbenzene | Xylene | BTEX | TVOC |
|-----------|---------|---------|--------------|--------|------|------|
| Rice straw | 0.065±0.021 | 0.201±0.053 | 0.003±0.001 | 0.010±0.000 | 0.278±0.031 | 0.542±0.015 |
| Cornstalk | 0.033±0.004 | - | 0.021±0.003 | 0.080±0.008 | 0.134±0.016 | 0.701±0.339 |
| Corncob | - | 0.110±0.062 | - | 0.003±0.002 | 0.113±0.062 | 0.526±0.246 |
| LPG | - | 0.012±0.008 | 0.007±0.003 | 0.018±0.010 | 0.015±0.011 | 0.122±0.060 |

a Data are presented as mean value ± standard deviation (unit: mg/m³).

4. Conclusions

The present study revealed real-time temporal variations of CO, PM2.5, formaldehyde and BTEX concentrations in 30 min for four types of fuels (rice straw, corn stalk, corncob and LPG) in rural household settings in the northeast China, Heilongjiang. Three households use traditional fixed Kang-cookstoves with chimneys for cooking and one of which also uses LPG stove as an alternative energy source during non-heating seasons. The results showed that concentrations of CO from the combustion of four kinds of fuels were almost below the guideline prescribed in Chinese Indoor Air Quality Standard (GB/T 18883-2002). PM2.5 concentrations after the fire was extinguished in kitchens with three kinds of biomass cooking fuels remained higher level, while the PM2.5 concentration was the lowest and fluctuated less when LPG was used as cooking fuel. It is worth mentioning that PM2.5 concentrations exceeded the standard regardless of which fuel was used.

The findings also indicated that HCHO concentrations of the three selected households using four types of cooking energy were totally higher than the upper limit of 0.1 mg/m³ stipulated in Chinese Indoor Air Quality Standard. Lowest HCHO concentrations were detected when corn cob was used for cooking energy. Compared with LPG fuel, biomass fuels combustion in tradition Kang-stove with chimney increased the concentrations of TVOC and BTEX. To sum up, the pollutants generated from using LPG are relatively less harmful. Among the three biomass fuels, the minimal pollution derives from the combustion of corncob. Hence, this work highlights the significance of using clean fuel and emphasizes the necessity of improving the coverage of clean fuel for cooking purposes in rural areas.

Acknowledgments

This research was financially supported by the Basic Application Technology Research Project of Heilongjiang Institute China (ZNBZ2017NY01). The authors would like to thank the editor and anonymous reviewers for their valuable comments and detailed suggestions to improve the manuscript.
References

[1] World Health Organization (2016). Household air pollution – the world’s leading environmental health risk. http://www.who.int/airpollution/household/about/en/.

[2] World Health Organization (2016). Global Health Observatory (GHO) data – Mortality from household air pollution. http://www.who.int/gho/phe/indoor_air_pollution/burden/en/.

[3] Kim, K.H., Pandey, S.K., Kabir, E., Susaya, J., Brown, R.J.C. (2011): The modern paradox of unregulated cooking activities and indoor air quality. Journal of Hazardous Materials 195: 1-10.

[4] Huang, W., Baumgartner, J., Zhang, Y.X., Wang, Y.Q., Schauer, J.J. (2015): Source apportionment of air pollution exposures of rural Chinese women cooking with biomass fuels. Atmospheric Environment 104: 79-87.

[5] Huang, Y., Du, W., Chen, Y.C., Shen, G.F., Su, S., et al. (2017): Household air pollution and personal inhalation exposure to particles (TSP/PM$_{2.5}$/PM$_{1.0}$/PM$_{0.25}$) in rural Shanxi, North China. Environmental Pollution 231: 635-643.

[6] Sidhu, M.K., Ravindra, K., Mor, S., John, S. (2017): Household air pollution from various types of rural kitchens and its exposure assessment. Science of the Total Environment 586: 419-429.

[7] Kelp, M.M., Grieshop, A.P., Reynolds, C.C.O., Baumgartner, J., Jain, G., et al. (2018): Real-time indoor measurement of health and climate-relevant air pollution concentrations during a carbon-finance-approved cookstove intervention in rural India. Development Engineering 3: 125-132.

[8] Huboyo, H., Tohno, S., Lestari, P., Mizohata, A., Okumura, M., et al. (2013): Comparison between *Jatropha curcas* seed stove and woodstove: Performance and effect on indoor air quality. Energy for Sustainable Development 17: 337-346.

[9] Alnes, L.W.H., Mestl, H.E.S., Berger, J., Zhang, H.F., Wang, S.X., et al. (2014): Indoor PM and CO concentrations in rural Guizhou, China. Energy for Sustainable Development 21:51-59.

[10] Chakraborty, D., Mondal, N.K., Datta, J.K. (2014): Indoor pollution from solid biomass fuel and rural health damage: A micro-environmental study in rural area of Burdwan, West Bengal. International Journal of Sustainable Built Environment 3: 262-271.

[11] Hankey, S., Sullivan, K., Kinnick, A., Koskey, A., Grande, K., et al. (2015): Using objective measures of stove use and indoor air quality to evaluate a cookstove intervention in rural Uganda. Energy for Sustainable Development 25: 67-74.

[12] Zhang, Q.F., Gangupomu, R.H., Ramirez, D., Zhu, Y.F. (2010): Measurement of ultrafine particles and other air pollutants emitted by cooking activities. International Journal of Environmental Research and Public Health 7, 1744-1759.

[13] Ministry of Environmental Protection of the People’s Republic of China, GB 3095-2012: Ambient Air Quality Standards, Ministry of Environmental Protection of the People’s Republic of China, Beijing, China, 2012 (in Chinese)

[14] Ministry of Health of the People's Republic of China, GB 18883-2002: Indoor Air Quality Standard, Ministry of Health of the People's Republic of China, Beijing, China, 2002 (in Chinese)

[15] Kabir, E., Kim, K.H., Ahn, J.W., Hong, O.F., Sohn, J.R. (2010): Barbecue charcoal combustion as a potential source of aromatic volatile organic compounds and carbonyls. Journal of Hazardous Materials 174: 492-499.

[16] Khalequzzaman, M., Kamijima, M., Sakai, K., Hoque, B.A., Nakajima, T. (2010): Indoor air pollution and the health of children in biomass- and fossil-fuel users of Bangladesh: situation in two different seasons. Environ Health Prev Med 15: 236-243.

[17] Fan, G.T., Xie, J.C., Yoshino, H., Yanagi, U., Hasegawa, K., et al. (2018): Indoor environmental conditions in urban and rural homes with older people during heating season: A case in cold region, China. Energy & Buildings 167: 334-346.
[18] Sinha, S.N., Kulkarni, P.K., Shah, S.H., Desai, N.M., Patel, G.M., et al. (2006): Environmental monitoring of benzene and toluene produced in indoor air due to combustion of solid biomass fuels. Science of the Total Environment 357: 280-287.

[19] Wang, S.X., Wei, W., L, D., Aunan, K., Hao, J.M. (2010): Air pollutants in rural homes in Guizhou, China – Concentrations, speciation, and size distribution. Atmospheric Environment 44: 4575-4581.