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Contributions of internal emissions to peaks and incremental indoor $PM_{2.5}$ in rural coal use households☆

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ABSTRACT

Indoor air quality is critically important to the human as people spend most time indoors. Indoor $PM_{2.5}$ is related to the outdoor levels, but more directly influenced by internal sources. Severe household air pollution from solid fuel use has been recognized as one major risk for human health especailly in rural area, however, the issue is significantly overlooked in most national air quality controls and intervention policies. Here, by using low-cost sensors, indoor $PM_{2.5}$ in rural homes burning coals was monitored for ~4 months and analyzed for its temporal dynamics, distributions, relationship with outdoor $PM_{2.5}$, and quantitative contributions of internal sources. A bimodal distribution of indoor $PM_{2.5}$ was identified and the bimodal characteristic was more significant at the finer time resolution. The bimodal distribution maxima were corresponding to the emissions from strong internal sources and the influence of outdoor $PM_{2.5}$, respectively. Indoor $PM_{2.5}$ was found to be correlated with the outdoor $PM_{2.5}$, even though indoor coal combustion for heating was thought to be predominant source of indoor $PM_{2.5}$. The indoor-outdoor relationship differed significantly between the heating and non-heating seasons. Impacts of typical indoor sources like cooking, heating associated with coal use, and smoking were quantitatively analyzed based on the highly time-resolved $PM_{2.5}$. Estimated contribution of outdoor $PM_{2.5}$ to the indoor $PM_{2.5}$ was ~48% during the non-heating period, but decreased to about 32% during the heating period. The contribution of indoor heating burning coals comprised up to 47% of the indoor $PM_{2.5}$ during the heating period, while the other indoor sources contributed to ~20%. The study, based on a relatively long-term timely resolved $PM_{2.5}$ data from a large number of rural households, provided informative results on temporal dynamics of indoor $PM_{2.5}$ and quantitative contributions of internal sources, promoting scientific understanding on sources and impacts of household air pollution.

1. Introduction

Solid fuels like coal and biomass fuel, are still burned in inefficient stoves in many households, especially in rural areas to meet the daily cooking and/or heating demands (Lim et al., 2012). The inefficient burning of solid fuels in stoves produced a variety of harmful particles and gases, such as $PM_{2.5}$, black carbon and organic carbon (Matawle et al., 2017; Shen et al., 2019; Shen et al., 2014; Smith et al., 2009). Residential sector is one important sector contributing to ambient air pollution and premature deaths globally (McDuffie et al., 2021), and in countries like India (Guo et al., 2018) and China (Shen et al., 2019). Besides unignorable contributions to ambient air pollution, combustions of solid fuels affect indoor air quality directly and more significantly in circumstances when the air exchange was low (Li et al., 2017). As most people usually spend long time indoors (Duan, 2013), indoor air quality is more closely related to the human health and working efficiency (Kosonen and Tan, 2004).

Indoor $PM_{2.5}$ concentration has strong spatiotemporal variabilities (Qi et al., 2019; Shen et al., 2021) and even in one house, there could be large vertical differences and dynamic changes in indoor $PM_{2.5}$ (Micallef et al., 1998; Qiu et al., 2019). The intra-variability is also significant as, for example, peaks of indoor PM could be as high as several hundreds of μg/m3 due to burning biomass fuel (Salje et al., 2014). High fluctuations in indoor $PM_{2.5}$ can result in different exposure levels and consequently different degrees of the adverse health outcomes (Liao et al., 1999). However, conventional filter-based offline measurement methods can

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hardly capture detailed intra-daily variations because of its low temporal resolution (Wittig et al., 2004).

In addition to the spatiotemporal dynamics, it is also interesting to understand the sources and relative contributions of each to indoor $PM_{2.5}$, which is important in pollution mitigation and clean intervention (Xie et al., 2017; Bari et al., 2015). Generally, sources of indoor $PM_{2.5}$ include the infiltration of outdoor particle, which are from different primary sources and also secondarily formed through atmospheric chemical reactions (Hassanvand et al., 2014), and internal sources like fuel combustions and cooking oil fumes (Li et al., 2018). Quantitative estimates of source contributions to indoor air pollution are crucial but very limited so far. Traditional source apportionment approaches, like Positive Matrix Factorization (PMF) method (Bennett et al., 2019; Larson et al., 2004; Suryawanshi et al., 2016) and principal component analysis and absolute principal component score (PCA-APCS) (Ali et al., 2017), in outdoor air source apportionment were adopted to quantify sources of indoor $PM_{2.5}$. Besides these typical approaches, some studies estimated the contribution of various sources by establishing multiple regression models (Lung et al., 2020), which usually require large amounts of data and cannot separate sources of synergy. Recently, Lu et al. (2020) and Shen et al., (2020a, 2020b) developed statistical methods to evaluate contribution of outdoor infiltrations and several typical internal sources in urban households.

The present study aimed at assessing household $PM_{2.5}$ in rural households relying on coals for daily cooking and wintertime heating, largely focusing on dynamics over a relatively long winter time, indoor-outdoor relationship, and contributions of outdoor and typical internal sources. There are very few studies on the source quantification of rural indoor air pollution. We explored the characteristics of indoor and outdoor $PM_{2.5}$ concentration, including its distribution and spatiotemporal variations by using low-cost sensors. As mentioned above, traditional indoor $PM_{2.5}$ measurement using active pumps and gravimetric measurements of particle-loaded filters have high difficulties in field work as it interrupts residents' daily lives significantly (Gao et al., 2020), and it usually lacks of sufficient temporal resolutions to capture dynamics in indoor pollution. Low-cost sensors have obvious advantages in collecting $PM_{2.5}$ data with high temporal and spatial resolutions (Malings et al., 2020). It also becomes available, by using sensors, to collect large sets of data from a large sample size of households studied. A regional model to predict indoor $PM_{2.5}$ was developed, and external data were obtained to verify the transferability of the model.

2. Methods

2.1. Study area and survey

The present field measurement was carried out in a rural village in Hebei Province, north China. We randomly selected 70 households from the village. With the consent of the residents, we placed the sensors in the kitchen or living room and asked them to fill in the questionnaire. Information on housing structure, fuel and stove types for cooking and heating, family size, smoking behavior, coal consumption amount, kitchen ventilation condition, etc., was collected from the questionnaire and summarized in Table S1. In this village, coals were burned for space heating in winter. Under the clean heating campaign in the past several years, coals used in the studied village are anthracite briquettes, as seen in Fig. S1. The coal stoves were also used for cooking, and exhaust fans were equipped in the kitchen of some families. Most houses have one kitchen, one living room and 1–3 bedrooms, which are connected to each other via interior doors.

2.2. $PM_{2.5}$ measurements and validation

Low-cost sensors are receiving grown interests and used in more and more studies (Patel et al., 2017; Wang et al., 2020b). Low-cost sensor had the advantages of cheap, continuous and real-time, so it was very suitable for our research. In the present study, the monitoring device includes a laser scattering sensor for measuring $PM_{2.5}$ every 2 minutes, a module for measuring temperature and relatively humidity, and a SD card for storing measurement dataset. The sensors were placed about 1.5 meter high, 1–2 meter away from the stove, more than 0.5 meter away from the wall, and keep a certain distance from the doors/windows. Indoor monitoring was conducted in 70 randomly selected rural households. The outdoor $PM_{2.5}$ concentration was collected from the nearest local environmental monitoring station, which was about 10 km away from the study site.

Based on the data from a standard $PM_{2.5}$ instrument (Model 5030 Synchronized Hybrid Ambient Realtime Particulate Monitor, SHARP, the United States), a calibration factor, for each sensor, was determined to convert the sensor analog signal output to a concentration equivalent to standard $PM_{2.5}$ instrument. Generally, a good linear relationship between the sensor and the standard instrument was revealed and the slopes were used in sensor calibration.

2.3. Identification of indoor $PM_{2.5}$ peaks

Indoor activities such as cooking and smoking often led to a sharp rise in indoor $PM_{2.5}$ concentrations, resulting in significant peaks. Therefore, for these activities, we used *peak extraction* method to quantify the contribution based on high temporal resolution data.

To quantitatively identify peaks associated with these activities, the contribution of outdoor infiltration was firstly deducted from the total concentration to obtain the time series of indoor-originated $PM_{2.5}$ contribution. The time series were processed by moving average firstly to reduce the noise. Then, this study encoded to find peaks inside a signal based on peak properties. When seeking peaks, the 1-D array was used to find all local maxima by simple comparison of neighboring values, and peaks that met the demand were selected by specifying conditions for a peak's properties such as height, width, prominence, *etc*. The set threshold, the time of peaks appearance, and the distance between adjacent peaks were also used for peak filtering. The flow chart of peaks identification is shown in Fig. S2. Some peaks were extracted manually and compared with the results extracted by the program. Through examination, the peaks extracted by the program were reliable.

2.4. Data statistical analysis

All calculations were made in Python, Version 3.8.7 embedded in Visual Studio (2019), Version 16.8. The libraries of Python mainly used for data processing and data analysis included pandas (Version 1.2.1), numpy (Version 1.19.5) and scipy (Version 1.6.0). Statsmodel (Version 0.12.1) and some machine learning libraries such as sklearn (Version 0.0) were used to develop the model. In order to avoid normal transformation, nonparametric method was used in this study.

3. Result and discussion

3.1. Distributions of indoor and outdoor $PM_{2.5}$

Fig. 1 shows the distributions of indoor and outdoor $PM_{2.5}$ at different time resolutions. Outdoor $PM_{2.5}$ concentrations generally followed log-normal distributions, either the hourly concentration or the daily average. The peak concentration was around 80–90 $\mu g/m^3$. During the whole study period, there were 35% days in which the outdoor $PM_{2.5}$ exceeded the Chinese national standard of 75 $\mu g/m^3$ indicating severe ambient air pollution in winter, especially during the heating period.

The distribution and contamination status for indoor $PM_{2.5}$ were very different from those in ambient air. The paired data statistical test results indicated that the indoor concentrations were significantly higher than the outdoor level ($p < 0.05$). Data statistic summary results of indoor $PM_{2.5}$ at different resolutions are listed in Table S2. The distributions of indoor $PM_{2.5}$ appeared to be bimodal, which was more

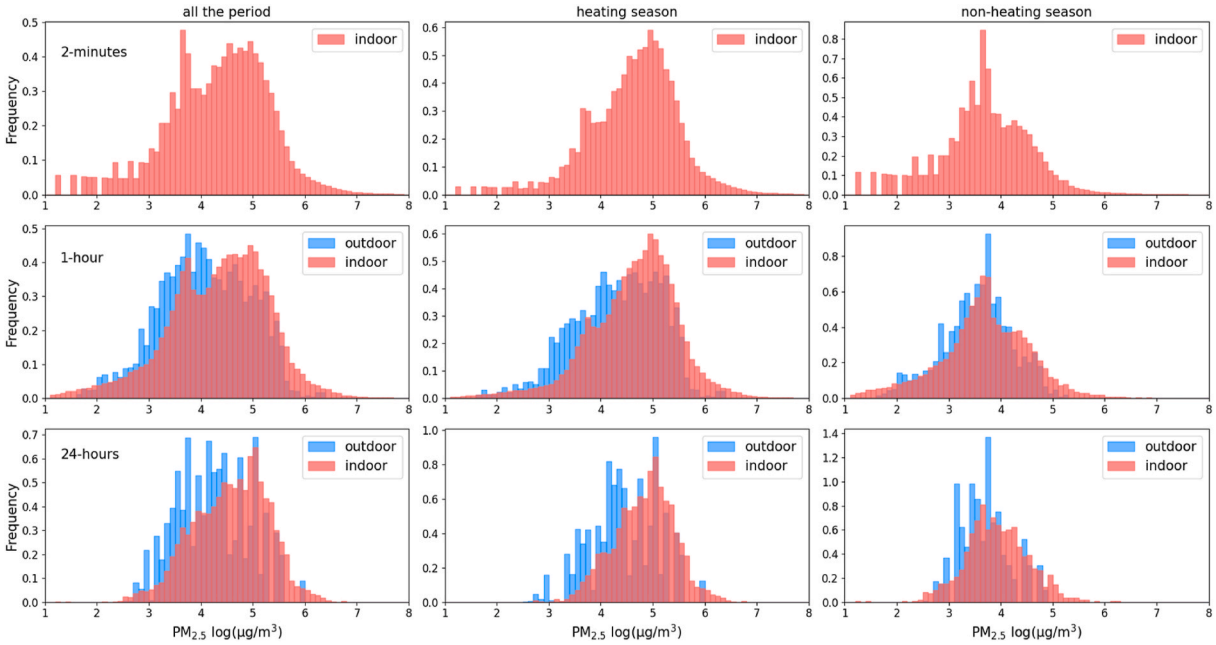

Fig. 1. Distributions of indoor and outdoor $PM_{2.5}$ concentrations (log-transformed) for the studied rural households during the study period, and for those during the heating and non-heating periods separately.

obvious at the finer time resolution. For the 2-min concentration, the peak values were at ~40 and 135 μg/m^3, respectively, and for the hourly average concentration, the two peak values were ~50 and 150 μg/m^3. The observed bimodal distribution was believed to be associated with the impacts of both outdoor air pollution and internal source emissions in indoor air quality (Mohammed et al., 2016). The left peak with relatively smaller concentrations was attributed to the influence of outdoor pollution through air exchange, while the right peak with relatively high concentrations was thought to be associated with the interference of strong internal sources such as cooking and fuel lighting or re-loading, in which high emission plumes leading to indoor $PM_{2.5}$ peaks with relatively high concentrations (Li et al., 2015).

When data from the heating and non-heating periods were analyzed separately, the right peak concentration during the heating period was obviously higher than that during the non-heating season, confirming the direct impacts of internal sources on the right peak with relatively high $PM_{2.5}$ levels. In calculating the daily average indoor concentrations, those high values would be evened since they lasted in relatively short time intervals, and the daily average concentration was more likely a unimodal distribution. The 95% quantile value of the daily mean was 277 μg/m^3, while at the 2-min resolution, the 95% quantile value was 304 μg/m^3. In a previous study in rural Sichuan, south China, where biomass fuels are widely used as cooking fuels, the daily indoor $PM_{2.5}$ distribution was also found to be unimodal and slightly right-skewed, which was similar to the present study result (Qi et al., 2019). In a study in urban Beijing households found that indoor $PM_{2.5}$ distribution could be also bimodal even without strong internal sources, which was explained by distinct pollution levels of outdoor air pollution during the clean and serious haze days (Han et al., 2015).

The average $PM_{2.5}$ concentration in the kitchen during the whole study period was 118 ± 137 μg/m^3 that was slightly higher than the 112 ± 142 μg/m^3 in the living room, and the median values in these two indoor microenvironments were 85 and 75 μg/m^3, respectively. Relatively high concentrations in the kitchen were associated with indoor activities like cooking (Lu et al., 2020). Some studies reported much larger differences in $PM_{2.5}$ concentrations between the kitchen and living room (Shen et al., 2021; Wu et al., 2015). The relative difference could be to a number of factors such as the connection of kitchen with others, temperature, etc., Even in the living room without combustion sources of indoor stoves present, the pollution was still very serious. In the rural households in this study, the living room was connected to the kitchen through an inner door, which was also a typical layout in northern rural homes. Thus, air qualities in these rooms were often closely affected by one another, resulting in pollution in the living room and relatively small differences in the average concentrations. The influence of kitchen pollution on other indoor microenvironments like bedroom and living rooms has been recognized for many air pollutants such as $PM_{2.5}$, CO, and volatile organic compounds (Kim et al., 2018; Shen et al., 2020a). The $PM_{2.5}$ distributions in the kitchen and living room were similar, as seen in Fig. S3.

3.2. Relationship between indoor and outdoor $PM_{2.5}$

I/O value is the ratio of indoor and outdoor concentration. It is an important index of relationship of indoor and outdoor $PM_{2.5}$. The larger the value is, the smaller the influence of outdoor $PM_{2.5}$ on indoor concentration is (Kalimeri et al., 2019). The I/O ratio ranged from 0.83 to 1.89, and not normally distributed. As the distribuions of indoor and outdoor concentration could be distinct largely, their ratio is not necessarily distributed normally. The average was 1.95 and the median value was 1.24, suggesting significant impacts of internal sources on indoor air quality, that is more significant as seen from higher I/O ratios during the heating period compared with that in the non-heating period, which were on average 2.08 and 1.65, respectively. High variabilities in the I/O ratio are due to distinct I/O values across the studied households in which indoor emissions and air exchange ratios were different. The I/O ratio showed a clear diurnal variation as seen in Fig. 2. The daytime ratio values were generally higher than the ratios in the night. Much higher values were observed during the cooking time, indicating sudden but significant influences of the indoor activities. Due to human activities such as cooking, intra-day variabilities in indoor $PM_{2.5}$ concentration were much higher than that in outdoor concentration, which could be also shown from the diurnal pattern of I/O value (Fig. 2).

Ambient $PM_{2.5}$ concentration was closely related to indoor $PM_{2.5}$, even in the studied rural households with strong an internal source of coal heating. The relationship between indoor and outdoor $PM_{2.5}$ concentration can be characterized by the following equation:

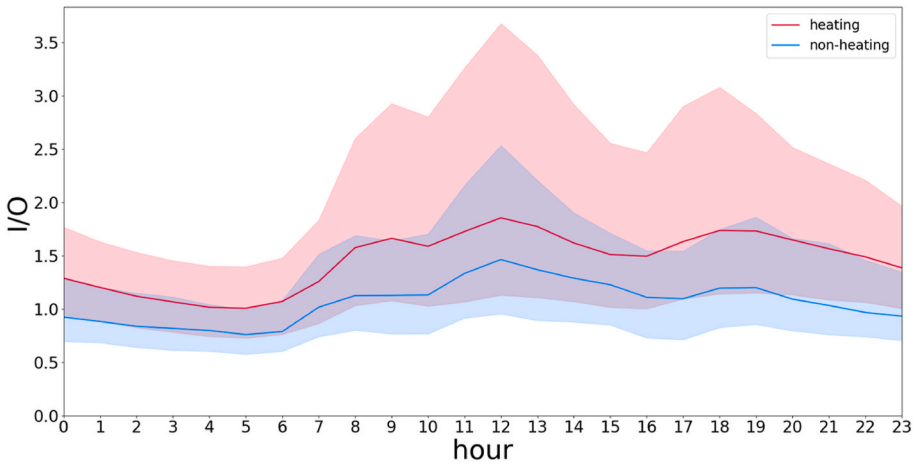

Fig. 2. Diurnal characteristics of I/O ratio from the studied rural households in winter. The shaded areas represent the inter-quartile range of the I/O ratio.

$$C_{indoor} = 0.838 \times C_{outdoor} + 46.409 \quad R^2 = 0.72$$

where C_{indoor} and $C_{outdoor}$ are the indoor and outdoor air $PM_{2.5}$ concentration, respectively. This suggested that nearly 72% of the variation in indoor $PM_{2.5}$ can be explained by the ambient $PM_{2.5}$.

As mentioned above, the influences of internal sources vary greatly between the heating and non-heating seasons. During the heating season, the solid fuel used for heating produced significant amounts of particles (Wang et al., 2020a), leading to much higher contributions of internal sources. As seen in Fig. 3A, the simple prediction of indoor $PM_{2.5}$ from ambient $PM_{2.5}$ solely always underestimated the indoor concentration in heating season but overestimated in non-heating season, which was thought to be associated with the distinct internal sources between these two periods. Therefore, one additional variable-"heating" was introduced and new model was as follows:

$$C_{indoor} = 0.685 \times C_{outdoor} - 42.458 \times heating + 61.445 \quad R^2 = 0.84$$

The *heating* can be 1 or 0, 1 for non-heating season and 0 for heating season. The new model of daily average indoor $PM_{2.5}$ concentration had relatively higher prediction accuracy, with R^2 and ten-fold cross validation R^2 values of 0.84 and 0.79, respectively. The fitting results were shown in Fig. 4. As shown in Fig. 3B, the simplified regression model could well explain the variations of the daily average indoor concentration, whether heating or non-heating season.

Cross validation had been widely used in the prediction of indoor $PM_{2.5}$ concentrations (Chen et al., 2019; Li et al., 2021; Mu et al., 2020). Cross validation could only examine the ability of the model to predict the sample data but couldn't completely test the established model. To test the generalization ability of the model, external validation was necessary. This study collected indoor $PM_{2.5}$ concentration data of households using coal fuel for heating and cooking at other times. The collected data was used to test the model and the results are shown in Fig. S4. The external validation normalized mean error (NME) and NRMSE of the model were 30.6% and 32.8%, respectively. The normalized maen bias (NMB) was 20.8%, which indicated that the model still overestimated the indoor $PM_{2.5}$ concentrations. If detailed information on the amounts of solid fuel used for cooking and heating were available and introduced in the prediction, it is expected to improve the model considerably. Other approaches like a random forest model, linear mixed-effect regression model may be considered (Li et al., 2021; Tong et al., 2020) and interesting to compare results among these approaches in future studies.

3.3. Typical internal sources of $PM_{2.5}$

Cooking oil fume emission is one important internal source of indoor

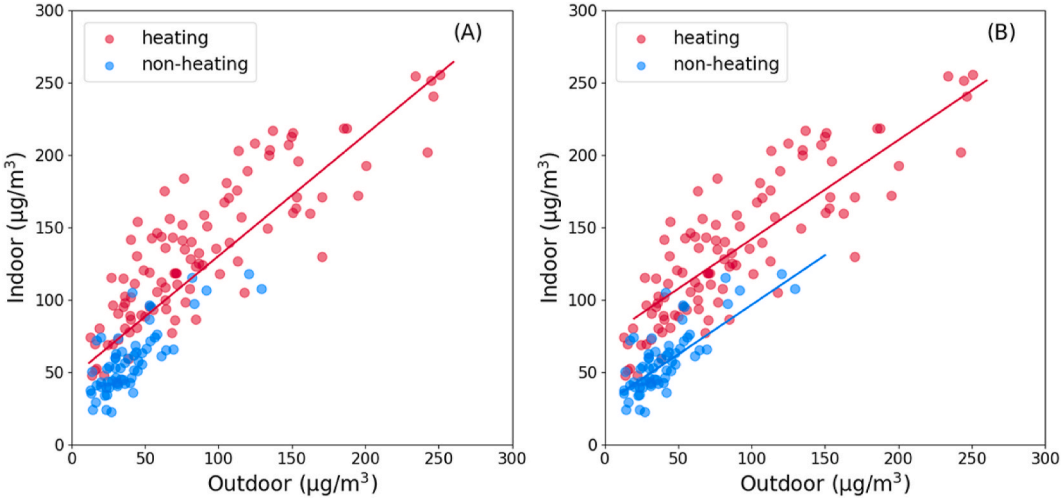

Fig. 3. Graphic expression of the model. The scatter represents the $PM_{2.5}$ concentration of indoor and outdoor air. The line in Figure A represents the simple linear regression model. And the line in Figure B represents the improved model, the lines with different colors correspond to the different seasons. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

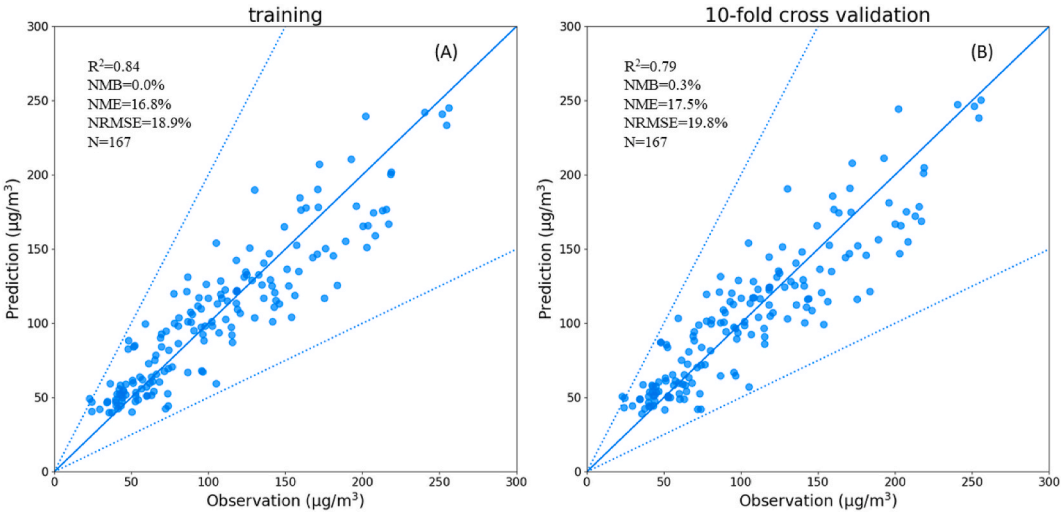

Fig. 4. (A)Performance of simplified model in predicting daily mean indoor $PM_{2.5}$ concentration. (B) Ten-fold cross-validation of the random forest model.

$PM_{2.5}$ (Li et al., 2019). Due to the close connectivity between rooms, the increase of $PM_{2.5}$ concentration caused by the cooking affects other indoor rooms (Patel et al., 2017), highlighting that the exposure caused by household air pollution is not limited to the kitchen and the person cooking (Patel et al., 2017). In this study, peaks of $PM_{2.5}$ concentrations caused by the cooking were identified during the cooking period (usually the morning -approximately 7:00–8:00, at noon 11:00–12:30 and in the evening 18:00–19:00). Fig. S5 shows a typical example of the cooking peak recognition. The average duration of a cooking peak was 47 min, ranging from 19 to 57 min. The peak was far higher than the baseline and the prominence was as high as 359 ± 352 μg/m^3, indicating significant influence of cooking on indoor air quality. The cooking peak's prominence for the lunch and dinner preparation was 365 ± 352 and 362 ± 376 μg/m^3, respectively, and it was slightly lower during the breakfast time which was about 349 ± 345 μg/m^3.

The distributions of extracted peaks were similar as shown in Fig. 5. The prominence fluctuated greatly with the coefficient of variation of 98%. This can be partly explained by the different cooking ways (Li et al., 2016), beside the method uncertainty in peak extraction. In the present relatively long-time field monitoring, it is hard to collect information on detailed cooking ways in each day for each household, thus it is unfortunately unable to investigate quantitative relationship between the incremental concentration and cooking ways here. This is an interesting topic to be further studied later. For a typically three meals per day, the average incremental contribution of each meal to daily indoor $PM_{2.5}$ concentration was about 4.0 μg/m^3 (Table 1). In a previous study on urban households using natural gas for cooking, the contribution of each meal was only 0.55 μg/m^3 (Lu et al., 2020). The difference can be explained mainly by the different cooking fuels, but also different cooking ways. The use of kitchen exhaust ventilation

Table 1
Contribution of different individual activities to indoor $PM_{2.5}$ daily mean. The "Cooking" means a weighted average of three meals.

Activity	Daily mean	25%	50%	75%	95%
Cooking*, μg/m^3/meal	4.01 ± 9.75	0.76	1.57	3.79	13.47
Breakfast	3.83 ± 9.22	0.79	1.52	3.43	13.07
Lunch	3.79 ± 7.85	0.78	1.67	3.86	12.41
Dinner	4.28 ± 11.17	0.73	1.52	3.93	14.4
Smoking, μg/m^3/cigarette	0.37 ± 0.42	0.10	0.19	0.48	1.28

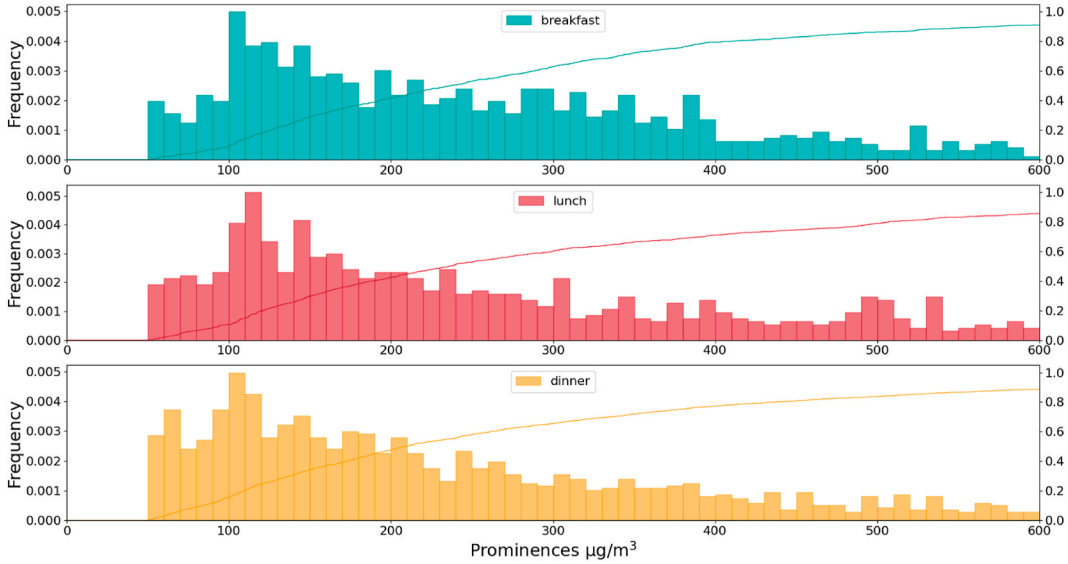

Fig. 5. Distribution and cumulative probability density of cooking peaks prominence in the morning, noon and evening. Bar chart and curves represent the frequency distribution and cumulative probability density, respectively.

systems can effectively improve the indoor air quality, especially in the kitchen (Parrott et al., 2003). Exhaust ventilation system is gradually popularized in rural China. In this study, the cooking peak's prominence of the families using exhaust ventilation systems was significantly lower than those from the households without exhauster, with the average value of 289 and 373 μg/m^3, respectively (Fig. S6).

Negative effects of smoking on indoor air quality are often reported (Canha et al., 2019; Ni et al., 2020). It was reported that e-cigarettes may produce less particles than traditional cigarettes (Ruprecht et al., 2014). Almost all cigarettes people smoke in rural China are traditional cigarettes. In the study area, some residents smoked indoor and smoking peaks were identified from these homes based on the highly temporal resolved data. The indoor $PM_{2.5}$ concentration in homes with active smokers was significantly higher than that of the non-smoking families. Smoking often occurs randomly during the day and the impact on the $PM_{2.5}$ concentration was the sharp spike. The peaks of smoking lasted for about 17 min and caused an increment concentration of 54 μg/m^3 on average. The average contribution of smoking to daily $PM_{2.5}$ mean was about 0.37 μg/m^3 with the inter-quartile range of 0.10–0.48 μg/m^3. The contribution of smoking in a previous study was estimated at 0.32 μg/m^3 (Lu et al., 2020), which was close to the result here.

High indoor concentration especially that during the heating period, was closely associated with the indoor combustions of solid fuels like coal in this study. Different from the impacts of cooking oil fumes and smoking which usually resulted in several peaks lasting for a relatively short time, the influence of fuel combustion for heating affects the indoor air quality during the whole day, leading to the overall increase of indoor $PM_{2.5}$ levels through fugitive emissions into indoor air directly (Luo et al., 2021; Shen et al., 2020b). By comparing the concentration in indoor concentrations between the heating and non-heating seasons after excluding impacts of outdoor contribution, smoking and cooking oil fume emissions, it was estimated that the combustion of coal for space heating contributed to 64 μg/m^3.

Other internal sources are possible, for example, cleaning and movement of people (Abt et al., 2000; Braniš et al., 2005). Random fluctuations in daytime $PM_{2.5}$ were very likely associated with these activities (Mousavi and Wu, 2021), although it was hard to exactly identify the contributions. Activities like burning incenses, candles, and mosquito coils, and using some electronic devices like printers can affect indoor air quality as well (Destaillats et al., 2008; Stabile et al., 2012), but these less likely happen during the present study period. The contribution of other internal sources, as a total, contributed to 17 μg/m^3.

The average contributions of cooking emission, outdoor infiltration, heating source and others to indoor $PM_{2.5}$ concentrations are shown in Fig. 6. During the heating period, the contribution of space heating was about 47%, ranging from 43% to 52%, which was the largest internal source, while during the non-heating period, outdoor air was one main source contributing to 48% (42%–53% as range) the indoor PM2.5, and the emissions from burning of solid fuels for cooking accounted for 23% on average. Variations in the estimated contributions are believed to be associated with distinct circumstances from different homes and changes in the intensities of internal and outdoor sources. In a previous study in one urban household using gas as cooking fuels, using the similar analysis approach it was estimated that outdoor contributed to about 36% of the indoor $PM_{2.5}$, varying extensively across indoor room and time (Shen et al., 2021). By adopting the typical Chemical Mass Balance source apportionment method with chemical compositions of filter-based $PM_{2.5}$ analyzed (Lai et al., 2020), estimated that indoor fuel combustion sources (i.e. biomass burning in the studied area from rural Sichuan, south China) contributed to 27%–84% of the summertime indoor $PM_{2.5}$ in homes using biomass, and in homes using LPG the biomass burning also contributed to the indoor $PM_{2.5}$ with the contribution of about 8%, while the outdoor sources comprised to about 10–20% of the indoor $PM_{2.5}$. The further analysis of samples covering a longer period showed that wood burning, food cooking, identified outdoor sources (i.e. vehicles, secondary aerosols, dust) and the others contributed to 54%, 28%, 10% and 7.6% of the kitchen $PM_{2.5}$ in the summer, and 41%, 20%, 14%, and 25% in the winter (Lai et al., 2019). Contributions of different source vary extensively among study areas, seasons, and source apportionment methods (Zhang et al., 2013), but it is clearly that in homes using solid fuels, particularly in winter, internal combustion source contributed larger than other indoor sources like cooking oil fumes, and the non-negligible outdoor contribution was probably up to 20–30%.

4. Conclusions

The present wintertime study from rural households burning coals showed that the indoor and outdoor $PM_{2.5}$ followed a bimodal distribution, and a log-normal distribution, respectively. The bimodal distribution in indoor $PM_{2.5}$ was more obvious at the finer time resolution. Indoor $PM_{2.5}$ was significantly higher than the outdoor level. The average I/O value was 1.95 over the whole study period, indicating significant impacts of internal sources on indoor $PM_{2.5}$. Indoor $PM_{2.5}$ dynamics was closely related to change in ambient $PM_{2.5}$ and contributions of indoor sources. Internal emissions like cooking and smoking led to high peaks in indoor $PM_{2.5}$ concentrations, with the peak prominence of 350 and 161 μg/m^3, respectively. Coal combustions for heating increased indoor $PM_{2.5}$ by about 66 μg/m^3. Relative contributions of internal sources to indoor $PM_{2.5}$ concentration was larger than that of external sources, especially during the heating period. The study estimated that the contribution of internal sources was ~70%, and fuel burning for heating, that was the most significant internal source, contributed ~47%. During the non-heating season, the contribution of internal sources was over 50% and the cooking emissions using solid fuels accounted for 23%. Other internal sources, such as cleaning, smoking and movement of people, contributed 11% and 29%, respectively, during the heating and non-heating periods. Based on the measured data, a model with well transferability and predictability for predicting indoor daily concentration was developed.

Household air pollution from solid fuels contributes largely to the overall air pollution exposure and human health. Unfortunately, this

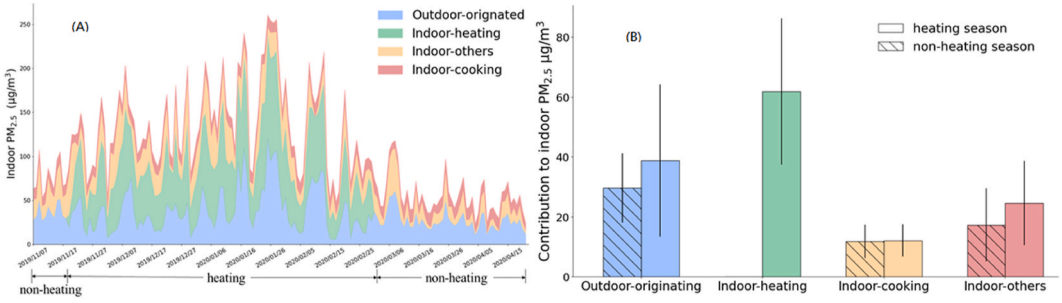

Fig. 6. Contributions of different sources to indoor $PM_{2.5}$ for each day during the study period (A) and the overall average during the heating and non-heating periods (B). Data in the panel B are means with standard deviations.

problem is sometimes underappreciated. High pollution levels and significant contributions of internal sources call for effective controls on this issue. Understanding of its temporal and spatial characteristics is critical in evaluating health impacts of indoor air pollution and actions on pollution mitigation. By using low-cost sensors, it becomes possible to collect large sets of data on indoor air quality without or with less interruption on the residents' lives, but the quality of data from sensors should be paid more attention in measuring, analysis and interpretation. The present study estimated contributions of different internal and outdoor sources to the indoor $PM_{2.5}$, which quantitatively showed high contributions of internal sources in rural households burning solid fuels, while the contribution of outdoor air was also not negligible. Note that this is different from traditional source apportionment approaches like CMB and PMF. Spatiotemporal dynamics of household $PM_{2.5}$, as well as other pollutants, and source contributions vary in different circumstances and more future studies, e.g. from different regions and seasons, are needed.

Author contribution

Yatai Men: Formal analysis, Data curation, Writing-original draft **Jianpen Li:** Formal analysis, Investigation, Data curation, Writing-original draft **Xinlei Liu:** Investigation, Data curation **Yaojie Li:** Investigation, Data curation **Ke Jiang:** Investigation, Data curation **Zhihan Luo** Data curation, Writing-review & editing **Rui Xiong:** Investigation, Data curation **Guofeng Shen**: Conceptualization, Writing-review & editing, Funding acquisition **Hefa Cheng:** Writing-review & editing **Shu Tao:** Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2021.117753.

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