The impact of household cooking and heating with solid fuels on ambient PM$_{2.5}$ in peri-urban Beijing

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HIGHLIGHTS

- In a village near Beijing, ambient PM$_{2.5}$ and household stove use was monitored in a heating season.
- Primary PM$_{2.5}$ emissions from household space heating and cooking were estimated.
- Multivariate analysis shows 39% of local PM$_{2.5}$ associated with household emissions.
- The peri-urban area and urban Beijing experienced similar levels of ambient PM$_{2.5}$

ARTICLE INFO

ABSTRACT

Household cooking and space heating with biomass and coal have adverse impacts on both indoor and outdoor air quality and are associated with a significant health burden. Though household heating with biomass and coal is common in northern China, the contribution of space heating to ambient air pollution is not well studied. We investigated the impact of space heating on ambient air pollution in a village 40 km southwest of central Beijing during the winter heating season, from January to March 2013. Ambient PM$_{2.5}$ concentrations and meteorological conditions were measured continuously at rooftop sites in the village during two winter months in 2013. The use of coal- and biomass-burning cookstoves and space heating devices was measured over time with Stove Use Monitors (SUMs) in 33 households and was coupled with fuel consumption data from household surveys to estimate hourly household PM$_{2.5}$ emissions from cooking and space heating over the same period. We developed a multivariate linear regression model to assess the relationship between household PM$_{2.5}$ emissions and the hourly average ambient PM$_{2.5}$ concentration, and a time series autoregressive integrated moving average (ARIMA) regression model to account for autocorrelation. During the heating season, the average hourly ambient PM$_{2.5}$ concentration was 139 ± 107 μg/m^3 (mean ± SD) with strong autocorrelation in hourly concentration. The average primary PM$_{2.5}$ emission per hour from village household space heating was 0.736 ± 0.138 kg/hour. The linear multivariate regression model indicated that during the heating season — after adjusting for meteorological effects — 39% (95% CI: 26%, 54%) of hourly averaged ambient PM$_{2.5}$ was associated with household space heating emissions from the previous hour. Our study suggests that a comprehensive pollution control strategy for northern China, including Beijing, should address uncontrolled emissions from household solid fuel combustion in surrounding areas, particularly during the winter heating season.

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1. Introduction

Ambient PM$_{2.5}$ (particulate matter with an aerodynamic diameter less than 2.5 μm) pollution poses a great health risk to the Chinese population and was associated with nearly 1 million premature deaths in 2013 (GBD, 2013 Mortality and Causes of Death Collaborators, 2015). Ambient air pollution (AAP) often reaches very high concentrations during the winter season in northern China; several studies have identified unfavorable weather conditions, high coal consumption, and traffic emissions in urban areas as the main sources of air pollution (Chen et al., 2013; Wang et al., 2013; Xiao et al., 2015a; Zhang et al., 2013). Additionally, indoor combustion of household solid fuel, including biomass and coal, generates household air pollution (HAP), which is associated with a significant additional health burden worldwide and an estimated 800,000 premature deaths in China in 2013 (GBD, 2013 Mortality and Causes of Death Collaborators, 2015; Smith et al., 2014).

Household solid fuel use is also an important source of AAP, especially in northern China. Modeling studies indicate that residential coal and biomass combustion accounted for 34% of total PM$_{2.5}$ emissions in China between 1990 and 2005 (Lei et al., 2011) and that residential emissions accounted for 35–43% of the daily surface PM$_{2.5}$ concentration in the Beijing, Tianjin and Hebei region during January and February 2010 (Liu et al., 2016). In addition, other studies estimated the relationship between household emissions' contributions to AAP and subsequent health impacts in China. In 2010, Chafe et al. attributed 10% of the population-weighted ambient air PM$_{2.5}$ concentration in East Asia to cooking with solid fuels (Chafe et al., 2014). Lelieveld et al. attributed 32% of premature deaths associated with outdoor PM$_{2.5}$ pollution in China to residential energy use (Lelieveld et al., 2015). Source apportionment of PM$_{2.5}$ in Beijing indicates that coal and biomass combustion contributes 30% of PM$_{2.5}$ annually (Zhang et al., 2013). Even though most coal is used in coal-fired power plants, central heating stations, or for industrial activities (China Energy Statistical Yearbook, 2012, 2013), rural household coal use for space heating was recently found to be a larger source of overall emissions in northern China than previously expected, both due to historical underestimation of rural fuel consumption and a lack of pollution control at the household level (Zhi et al., 2015). Similarly, several studies reported that residential heating is a crucial source of combustion, which influences indoor air pollution significantly in households heated with biomass in the Tibetan Plateau (Carter et al., 2016; Xiao et al., 2015b).

Furthermore, both satellite observation and field measurement studies in northern China found that ground level PM$_{2.5}$ concentrations were higher in rural or suburban areas than in high-population-density urban areas or remote field sites, particularly during the winter heating season. This suggests strong local emissions in rural and suburban sites, potentially from coal used for space heating (Li et al., 2014; Xiao et al., 2015a; Xie et al., 2015).

Despite these findings — which suggest the contribution of household solid fuel combustion to ambient air pollution — relatively few direct ambient PM measurements are made outside of China's cities, and the contribution of household solid fuel use for space heating to local ambient air pollution remains poorly characterized. Guided by the limitations of previous studies, we measured use of solid fuel cookstove and heating devices to estimate hourly primary PM$_{2.5}$ emissions, and then to assess the relationship between emissions and hourly ambient air PM$_{2.5}$ concentration. By investigating the impact of household space heating on ambient concentrations of PM$_{2.5}$ on a relatively fine temporal scale, we provide suggestions on future air pollution control strategies in China.

2. Material and methods

2.1. Site description

This study was conducted in Er He Zhuang (EHZ) village (116°05′ E, 39°41′E), which is located in a peri-urban area, approximately 40 km southwest of central Beijing, China, in the northwest part of the North China Plain. Most peri-urban households in the North China Plain use liquefied petroleum gas (LPG) or electricity for cooking, but still rely heavily on coal and biomass for space heating in the winter. EHZ village has an approximate area of 0.25 km^2 and a population of approximately 200 households and 500 residents. Most households are one-story, uninsulated brick homes with rooms arranged around a central outdoor courtyard. A map of the study site is shown in Fig. 1.

2.2. PM$_{2.5}$ and meteorological measurements

We installed an Onset HOBO Micro Station Data Logger (Onset Computer Corp., Bourne, MA, USA) on the rooftop of a centrally located 2-story village office building (Fig. 1). Temperature (T), atmospheric pressure (P), relative humidity (RH), wind speed (WS) and wind direction (WD) were collected at 5-min intervals between January and March 2013. To facilitate analysis and represent air pollutants from urban Beijing (northeast of EHZ), we converted WS and WD into two orthogonal velocity components, i.e. southwest wind speed (SWWS) and northwest wind speed (NWWS), illustrated by Equation (2–1). A positive value of SWWS indicates the speed of wind coming from the southwest direction, whereas a negative value indicates the speed of wind coming from the northeast direction. Similarly, a positive value of NWWS indicates the speed of wind coming from the northwest direction, whereas a negative value indicates the speed of wind coming from the southeast direction.

$$SWWS = \sin\left((315 - WS) \times \frac{180}{\pi}\right) \times WS$$

$$NWWS = \cos\left((315 - WS) \times \frac{180}{\pi}\right) \times WS \qquad \text{(Equation 2 −1)}$$

Two DustTrak II Aerosol Monitors (Model 8530, TSI Corp. Shoreview, MN, USA) with PM$_{2.5}$ size-selective impactors were set up in locally constructed protective metal boxes on rooftops of single-story homes located in the northwest and southeast quadrants of the village (Fig. 1). Real-time ambient PM$_{2.5}$ concentration data were collected at 1-min intervals from January 9, 2013 to March 10, 2013 during the heating season. To calibrate the real-time measurements to the local EHZ village aerosol, we performed two and three 24-h gravimetric calibrations for each of the two DustTrak Monitors using the DustTraks' built-in gravimetric sampler and collected PM$_{2.5}$ on 37 mm PTFE filters. A field blank - a 37 mm filter placed in a closed, capped cassette — was collocated in the same protective box as each DustTrak during each gravimetric sampling period. After gravimetric sampling was completed, filters were placed in sealed, airtight petri dishes in Ziploc bags and stored at −20 °C. The sealed and bagged petri dishes were transported back to the USA on ice in an insulated container. All filters were weighted in triplicate before and after sampling on a seven-digit microgram balance with accuracy of 1 μg at Kirk R. Smith's laboratory in a temperature and humidity controlled room at the University of California, Berkeley.
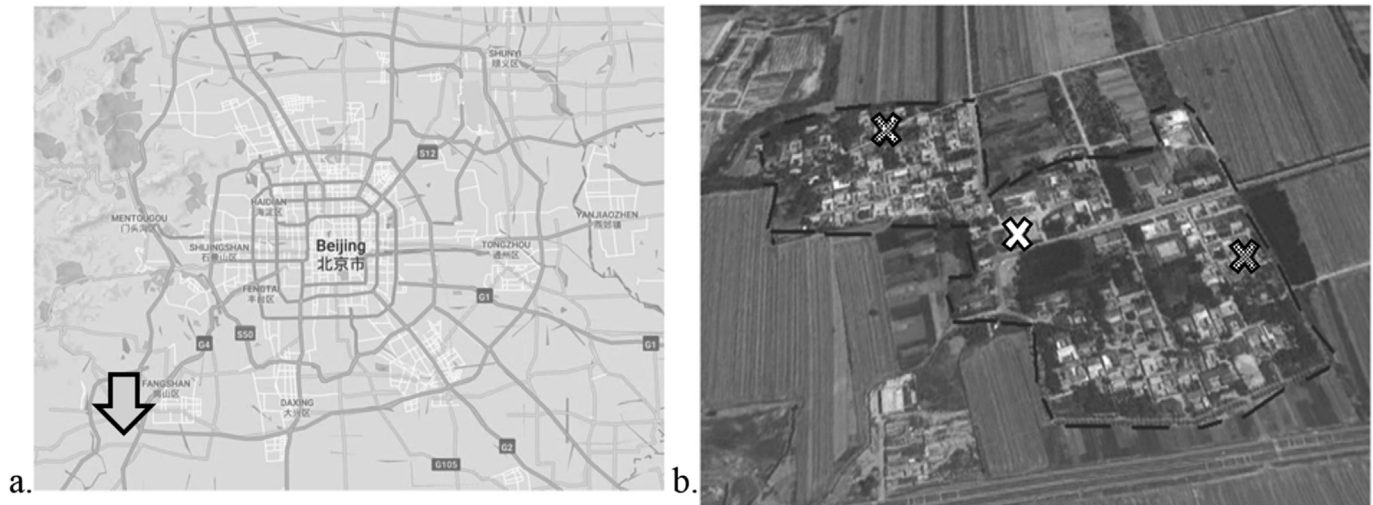
Fig. 1. Map of Study Site. a) The arrow shows the location of EHZ village outside the 6th ring road in southwest Beijing. b) The dotted and blank Xs indicate the rooftop monitoring locations of the two DustTrak aerosol monitors and the meteorological station, respectively, within EHZ village (shown with dashed line).

2.3. Household surveys and stove use monitoring

In June 2012, we randomly selected 36 households (one-sixth of all EHZ household) and asked them to participate in our study. We administered two household surveys and set up continuous stove use monitoring. Of the 36 households, 33 households agreed to participate and provided written consent. The household surveys were used to collect information on demographics, socio-economic indicators, energy use, fuel types used and fuel purchasing history, and the number of cooking stoves, water boilers, furnaces, floor heating systems and heated platform beds (called *kang*) in each household. In each household, we identified all cookstoves and heating devices and monitored the surface temperature of each appliance continuously using commercially available iButton SUMs (Maxim IC, USA) described previously (Ruiz-Mercado et al., 2013, 2012). We employed 61 SUMs on 61 solid-fuel-using appliances identified in the 33 participating households. Cookstoves were monitored between July 2012 and March 2013, while heating devices were monitored between December 2012 and March 2013. Pictures of each type of monitored device and stove are shown in Fig. 1S in the supplemental information (SI) material part 1. A coal boiler in the main village office was also monitored during the winter heating season. The characteristics and distribution of monitored devices and SUMs placement by device type are listed in Table 1.

Before implementing long-term SUMs monitoring, we conducted pilot tests on each type of cookstove and heating device in a subset of households to determine the optimal sampling interval and placement of SUMs to capture typical temperature changes when devices were in use. A SUMs sampling rate of 5-min was used because it was frequent enough to capture multiple temperature readings during short cooking events, yet long enough to allow continuous sampling for approximately 28 days (the maximum period between household visits). Every two to four weeks, SUMs data from each household were downloaded using the OneWire Viewer software program (Maxim Integrated, San Jose, CA), household members were asked if they had stopped using or added new cookstoves or heating devices, and SUMs monitors were removed or added accordingly.

The human subject protocol for this study was approved by (1) the University of California, Berkeley's Committee for the Protection of Human Subjects, (2) Tsinghua University, and (3) the local Er He Zhuang government.

2.4. Household PM$_{2.5}$ emissions estimates

We estimated hourly PM$_{2.5}$ emissions per hour (EPH) from household solid fuel combustion in EHZ village by combining SUMs measurements, household energy survey results and previously published primary PM$_{2.5}$ emission factors (EFs) for solid fuel combustion in rural China (Fig. 2). First, we characterized the use of cookstoves and heating devices based on SUMs data indicating

Table 1
Characteristics and Distribution of the Solid Fuel Combustion Stoves/Devices in the Sampled Households and SUMS Monitors (n = 33 households).

| Device type	Purpose	Typical device location	SUMS placement	Number of devices monitored	Number of households with device (percentage)
Biomass Portable Bucket Stove	Cooking	Outdoor, portable	Near stove base on brick	2	2 (6%)
Biomass Fixed-location Stove	Cooking	Outdoor, enclosed structure	Top corner of surface	2	2 (6%)
Biomass Floor Heating	Space heating	Indoor, combustion chamber outdoor	On floor	2	2 (6%)
Biomass *Kang*	Space heating	Indoor	On platform surface	7	7 (21%)
Biomass Water Boiler	Cooking	Outdoor, portable	On spout	5	5 (15%)
Coal Honeycomb Briquette Stove	Mostly Space Heating	Indoor, portable or fixed location	On side or base of stove	14	13 (39%)
Coal Furnace with Radiator	Space heating	Indoor, furnace in separate room	On pipe of furnace or radiator	29	27 (82%)
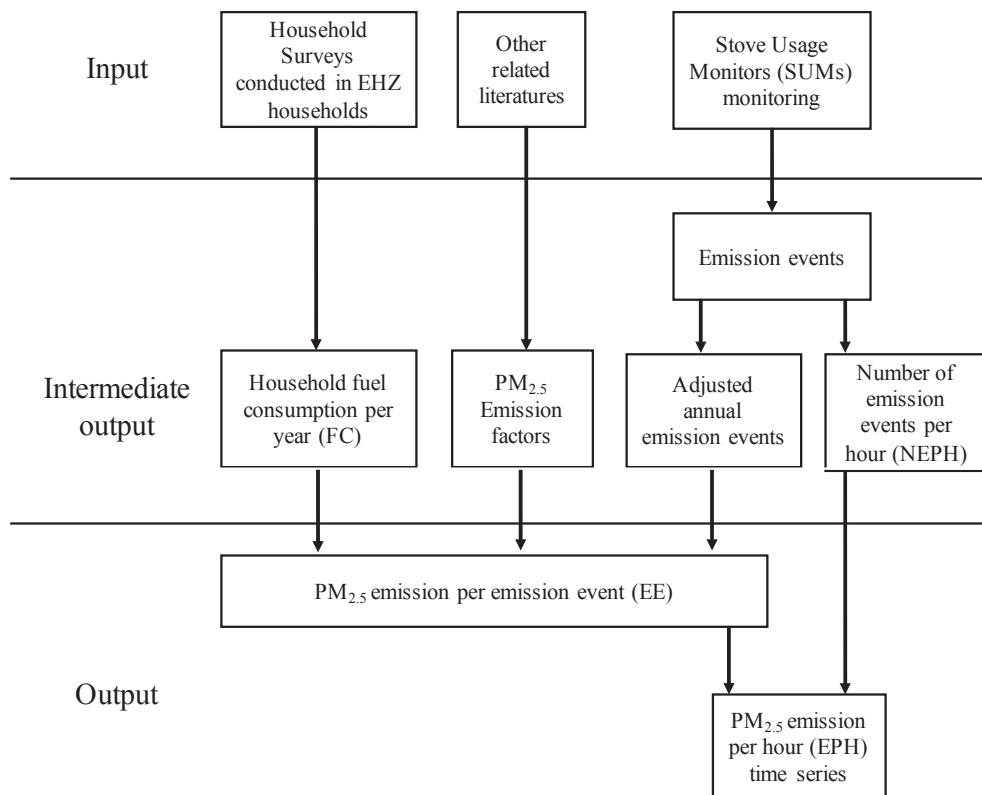
Fig. 2. Flowchart showing inputs used to estimate PM$_{2.5}$ emission per hour (EPH).

whether heat was generated in a given device. Due to the heterogeneity of the stoves and heating devices, custom usage event estimating algorithms were developed for each type of biomass and coal combustion cookstove and heating device and implemented in R software version (3.2.0). The algorithms to estimate emission events, marked by peaks in monitored temperature data, generally consisted of two components: the lagged difference between eight consecutive SUMs temperature data points and a threshold related to both the daily SUMs temperature range and rate of temperature change during solid fuel use events. The development of algorithms was based on a randomized visual check of results for each type of cookstove and heating device, and comparison to ambient temperature data, followed by additional testing on another set of samples for each type of cookstove and heating device. Detailed information on this methodology is included in the SI. The methodology for SUMs data handling and algorithm development can be found elsewhere (Mukhopadhyay et al., 2012; Pillarisetti et al., 2014).

Because we were unable to monitor stoves and heating devices over the course of an entire year, we adjusted the monitored SUMs emission events to estimate the number of annual SUMs emission events. To derive the number of space heating emission events, we applied the ratio between the monitoring time for each space heating device and the average length of the heating season in Beijing (Duan et al., 2014). To derive the number of cookstove emission events, we applied the ratio between monitoring time for each cookstove and one year.

Household survey results were used to determine annual household fuel consumption (FC) for both loose/chunk coal and honeycomb briquette coal, based on reported coal quantity purchased, used, and remaining at the end of the heating season. Because biomass fuels used in EHZ — wood branches or agricultural residue — were collected rather than purchased, the survey did not

collect information about the mass of biomass fuel purchased and consumed by EHZ households. Thus, we used results from similar surveys conducted in rural Beijing in 2009 (Zhang and Koji, 2012) to estimate total biomass FC per household. We then proportionally allocated the total biomass FC to cooking and space heating according to the ratio of SUMs emission events estimates for each category.

EFs of PM$_{2.5}$ from coal and biomass combustion in our study were chosen from field measurements and testing in China (Chen et al., 2015; Shen et al., 2013; Zhang et al., 2000; Zhi et al., 2008). Due to the several-fold difference in EFs of PM$_{2.5}$ from briquette versus loose or chunk coal, we calculated an adjusted coal EF weighted by the ratio of briquette to chunk coal consumption among EHZ households. The EFs for biomass were calculated by averaging reported values from published studies conducted in China.

We then incorporated estimated EFs, annual fuel consumption (FC), and the number of adjusted 5-min emission events per year to calculate PM$_{2.5}$ emissions per event (EE) by Equations (2–2):

$$EE\left(\frac{g}{event}\right) = \frac{EF\left(\frac{g}{kg}\right) \times FC\left(\frac{kg}{year}\right)}{Adjusted\ Emission\ event\left(\frac{events}{year}\right)}$$

(Equation 2 −2)

We used Equations (2–2) to estimate PM$_{2.5}$ emissions for each 5 min SUMs event for each device-fuel combination separately (e.g., biomass water boiler, coal furnace for space heating). We then multiplied EE by the number of SUMs emission events per hour (NEPH) and the sampling ratio (SR) of households in EHZ village and summed the calculated emissions per hour (EPH) for each device-fuel combination to estimate total household emissions per

hour for EHZ village (Equation (2–3)).

$$EPH\left(\frac{kg}{hour}\right) = \sum_{i=1}^{3} EE_i\left(\frac{g}{event}\right) \times NEPH_i\left(\frac{event}{hour}\right) \times SR \times \frac{1\ kg}{1000\ g}$$

(Equation 2 −3)

2.5. Data analysis and modeling

Gravimetrically-corrected data from the two DustTrak monitors were averaged to estimate ambient $PM_{2.5}$ concentrations near ground level in EHZ at 1 h intervals. The derivation of a custom DustTrak calibration factor used to correct real-time DustTrak data is shown in Table S1. Meteorological variables (T, P, RH, SWWS, NWWS) and household $PM_{2.5}$ EPH were then averaged at 1 h intervals during the sampling period of the heating season (January 9th to March 10th, 2013). Even though the SUMs and meteorological datasets cover a longer period of time, we only have complete ambient $PM_{2.5}$ concentration data from January 9th to March 10th, 2013; thus our analysis was limited to this period. The relationship between EPH and hourly ambient $PM_{2.5}$ concentrations was assessed by a multivariate linear regression model, and a time-series regression model was fitted among the hourly $PM_{2.5}$ concentrations, meteorological variables, and $PM_{2.5}$ EPH due to auto-correlation of the these variables (Cryer and Chan, 2008; Gocheva-Ilieva et al., 2013; Liu, 2009, 2007; Reisen et al., 2014).

First, we log transformed the hourly $PM_{2.5}$ concentrations, since these data showed a log-normal distribution. Then, we fit a multivariate linear regression model among log-transformed hourly $PM_{2.5}$ concentrations with lagged household EPH and meteorological variables. Third, to account for strong autocorrelation of the log-transformed ambient $PM_{2.5}$ concentration series, we applied a time-series autoregressive integrated moving average (ARIMA) regression model on the log-transformed hourly $PM_{2.5}$ mass concentration ($\ln(Y_t)$) series with household $PM_{2.5}$ EPH and meteorological variables (m_t, including T_t, P_t, RH_t, $SWWS_t$ and $NWWS_t$). The details of the time series ARIMA analysis are illustrated in the SI materials part 2. Multivariate regression and time-series ARIMA regression modeling were performed using R software version (3.2.0) and the "astsa" package (version 1.3) at a significance level of 0.05.

3. Results

3.1. Survey results, fuel use, SUMs events and $PM_{2.5}$ emission estimation

Table 2 summarizes the results of the household fuel use survey by cooking and space heating task. In nearly all of the 32 households surveyed, improved fuels (electricity and LPG) were the primary fuels for the majority of cooking tasks year-round, though many households continued to use both biomass and coal as supplemental fuels for certain cooking tasks, particularly heating water and cooking animal food. Coal and biomass were used as space-heating fuels, with coal as the primary space-heating fuel in 88% of surveyed households.

Fig. 2S shows illustrative SUMs temperature profiles and corresponding emission events for each type of biomass and coal cookstove and heating device for a week. Table 3 lists the aggregated results of SUMs monitoring emission events for 33 households by fuel and device type and adjusted emission events for each fuel-device combination.

Our surveys showed that all of the household coal was purchased from local vendors in EHZ. Most of the coal used in EHZ households was loose or chunk coal (2 930 ± 1 680 kg/household/year) burned in furnaces for space heating; a smaller amount of honeycomb briquette coal (400 ± 520 kg/household/year) was also used. Our survey also noted that biomass fuels used by EHZ households mostly consisted of wood branches or agricultural residues collected from the fields. Estimated total biomass FC per household was 190 kg/household/year, based on a similar survey conducted in rural Beijing in 2009 (Zhang and Koji, 2012). After proportionally allocating the total biomass FC to cooking and space heating according to the ratio of SUMs emission events for each category, we estimated that on average, 87 kg/household-year of biomass was used for cooking and 103 kg/household-year of biomass were used for space heating. The EFs of $PM_{2.5}$ from coal and biomass chosen in this study were 6.33 g/kg and 4.48 g/kg, respectively (Table 4).

Combining adjusted emission events, FC and EFs, we estimated primary $PM_{2.5}$ emissions per 5-min event (EE) of 0.37 g/event, 0.33 g/event and 1.00 g/event for biomass cooking, biomass space heating and coal space heating, respectively. During the monitored heating season, nearly all household emissions were from space heating, over 90% of which were from coal furnaces connected to a radiator. Fig. 3a shows the time series plot of total combined $PM_{2.5}$ emissions per hour (EPH) from biomass and coal use in EHZ village households during the winter heating season from January 9th to March 10th, 2013.

The average $PM_{2.5}$ EPH from biomass and coal in EHZ during the heating season was 0.736 ± 0.381 kg/hour (mean ± standard deviation). The EPH time series usually peaked at night and reached its lowest point in the afternoon. There was an overall trend of EPH decreasing after mid-January and a notable drop between February 10th and February 19th, which was probably due to the Chinese New Year, during which some people left the village and hence the space heating devices were not used.

Table 2
Reported Primary Fuel Used by Households for Cooking and Space Heating Tasks (n = 32 households).

		Honeycomb Briquette	Loose or Chunk Coal	Biomass	Clean Fuel[a]	None
Summer	Stir-fry	0	0	1	31	0
	Rice	0	0	0	32	0
	Bread	0	0	1	29	2
	Animal Food	1	0	4	10	17
	Water Boiling	1	0	7	24	0
Winter	Stir-fry	1	0	0	31	0
	Rice	0	0	0	32	0
	Bread	0	0	1	29	2
	Animal Food	1	1	3	10	17
	Water Boiling	1	3	3	24	0
	Space Heating	5	23	3	1	0

[a] Including electricity, LPG.

Table 3
Stove Use Monitors (SUMs) Monitoring Results and SUMs 5-min Emission Events by Cookstove and Heating Device Type (n = 33 households).

Fuel and device	Number of households with device	Number of emission events (thousands)	Total 5-min points measured (thousands)	Main usage	Adjusted emission events for a year (thousands)
Biomass Water Boiler	5	2	133	Cooking	22.9
Biomass Fixed Stove	2	7	71	Cooking	7.9
Biomass Bucket Stove	2	3	94	Cooking	20.7
Biomass Floor Heating System	2	15	56	Heating	6.7
Biomass *Kang*	7	14	182	Heating	22.8
Coal Radiator	25	230	656	Heating	373.4
Coal Furnace	4	25	62	Heating	68.7
Coal Honeycomb Briquette Stove	14	79	248	Heating[a]	187.4

[a] Mainly used for space heating, occasionally for cooking.

Table 4
Emission factors of $PM_{2.5}$ (g/kg) from coal and biomass chosen in this study.

	Honeycomb Briquette[b]	Loose or chunk coal[b]	Biomass
Shen et al., EST, 2013	0.59	8.9	8.3
Zhang et al., AE, 2000[a]	0.21	0.17	1.54
Li et al., Ecol. Model., 2015	NA	NA	3.6
Zhi et al., EST, 2008	7.33	13.2	NA
Chen et al., AE, 2015	3.58	4.93	NA
Average	2.93	6.80	4.48
Chosen in this study	6.33		4.48

NA: Not available.
[a] Measured in TSP.
[b] Using EFs of bituminous coal.

3.2. Ambient air $PM_{2.5}$ concentration and meteorological covariates

Table 1S shows the results of gravimetric calibration of the DustTrak monitors conducted in January 2017. The calibration factors of the two DustTrak were 4.89 and 5.60, respectively; an average calibration factor for the winter season was 5.18. Fig. 3b illustrates the hourly ambient air $PM_{2.5}$ concentrations from January 9th to March 10th after gravimetric correction. The hourly averaged $PM_{2.5}$ concentration during the sampling period was 139 ± 107 μg/m³ (mean ± standard deviation), and hourly $PM_{2.5}$ concentrations also displayed a diurnal periodic pattern, strong autocorrelations, and higher values at night than during the day.

Fig. 4 illustrates the time series of hourly mean meteorological conditions, including temperature (T), pressure (P), relative humidity (RH), northwest wind speed (NWWS) and southwest wind (SWWS). Fig. 3S illustrates the scatter plots of total $PM_{2.5}$ EPH (X_t), log-transformed ambient air $PM_{2.5}$ concentration ($\ln(Y_t)$) and meteorological variables at each hourly point. The scatter plots in Fig. 3S show that RH and EPH seem to be positively correlated with $\ln(Y_t)$, while temperature appeared to be negatively correlated. Moreover, the absolute value of wind speed was negatively associated with $\ln(Y_t)$. Wind direction manifested interesting patterns; the southwest wind reduced $\ln(Y_t)$ more slowly than the northwest wind did, while the southeast wind reduced $\ln(Y_t)$ more slowly than the northeast wind (Fig. 3S, first row fourth and fifth panel). This implies that the north wind had a stronger effect on improving air quality in EHZ village.

3.3. Multivariate regression between household emissions and ambient air pollution

Equation (3–1) shows the results of 1-h lagged multivariate regression of $PM_{2.5}$ EPH and other meteorological variables with log-transformed hourly ambient air $PM_{2.5}$ concentration; the estimates of coefficients and standard errors are listed in Table 5.

$$\ln(Y_t) = 0.4461x_{t-1} - 0.0251P_t + 0.0293T_t + 0.0343RH_t + 0.1380NWWS_t + 0.3145\,SWWS_t + \varepsilon_t$$

(Equation 3 −1)

The adjusted R-square of this lagged linear regression is 0.59 and p < 0.05 for all the variables in the model, indicating that 59% of the total variance of log-transformed hourly ambient $PM_{2.5}$ concentrations can be explained by T, P, RH, NWWS, SWWS and household $PM_{2.5}$ EPH. Additionally, this lagged linear regression analysis implies that during the heating season we monitored, on average, an increase of 50 g of primary $PM_{2.5}$ EPH from household solid fuel combustion in EHZ village at hour t-1 was associated with a 2% (95% confidence interval: 1.5%, 2.9%) increase of hourly ambient $PM_{2.5}$ level at hour t, adjusting for meteorological effects. On average, during the heating season, 39% (95% CI: 26%, 54%) of the ambient $PM_{2.5}$ concentration was associated with household emissions over the previous hour, after adjusting for meteorological effects.

3.4. Time series analysis and regression

To test the relationship between $PM_{2.5}$ emissions from household fuel use and local ambient pollution with even more conservative assumptions, we applied an ARIMA model that essentially eliminates the ambient $PM_{2.5}$ concentration spikes that occur nightly due to stove use as being due to autocorrelation, which of course we do not believe, and tests whether stove use at other times is related to ambient pollution. This highly conservative assumption still shows an effect, albeit less strong as could be expected, which adds to the confidence that what we show with the multivariate regression is real. See SI material part 3 for more in-detailed for ARIMA analysis results, model selection and model diagnosis.

Time series of PM2.5 EPH from Biomass and Coal

a)

Time series of ambient PM Concentration

b)
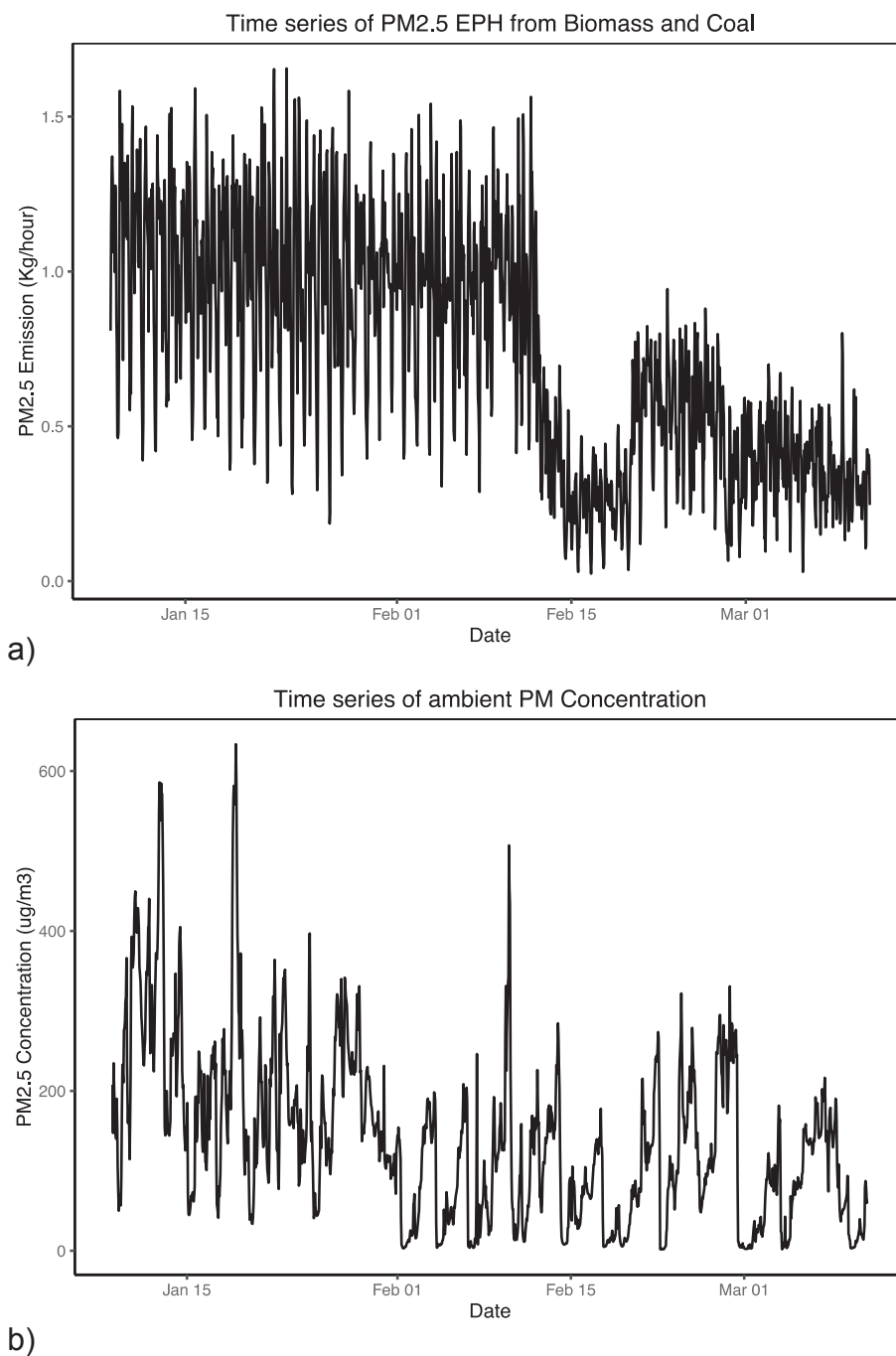
Fig. 3. aTime Series of PM$_{2.5}$ Emission per Hour (EPH) from Biomass and Coal Use by EHZ Households from January 9th to March 10th, 2013. b. Hourly Ambient Air PM$_{2.5}$ Mass Concentration from January 9th to March 10th, 2013.

4. Discussion

4.1. Coal and biomass use for household heating

By using SUMs to measure household cookstove and heating device use over time during the heating season, we characterized household fuel use patterns in peri-urban Beijing in detail. In EHZ village, we observed a high prevalence of coal and biomass use for space heating, which is consistent with other studies in northern China (Li et al., 2015; Shan et al., 2015; Zhang and Koji, 2012; Zhi et al., 2015). Similar to findings from studies in Yunnan, Hebei,

and Hubei provinces in China, EHZ households used a combination of improved and solid fuels for cooking; EHZ households, however, used electricity and LPG for a greater proportion of cooking tasks, with biomass mainly used as a supplemental fuel for specific tasks such as water boiling and cooking animal food (Baumgartner et al., 2011; Peng et al., 2010; Zhi et al., 2015). During the heating season from January to March, over 90% of primary PM$_{2.5}$ emissions from EHZ households were due to space heating, most of which resulted from coal combustion in coal furnaces or in fixed-location honeycomb coal briquette stoves with chimneys.

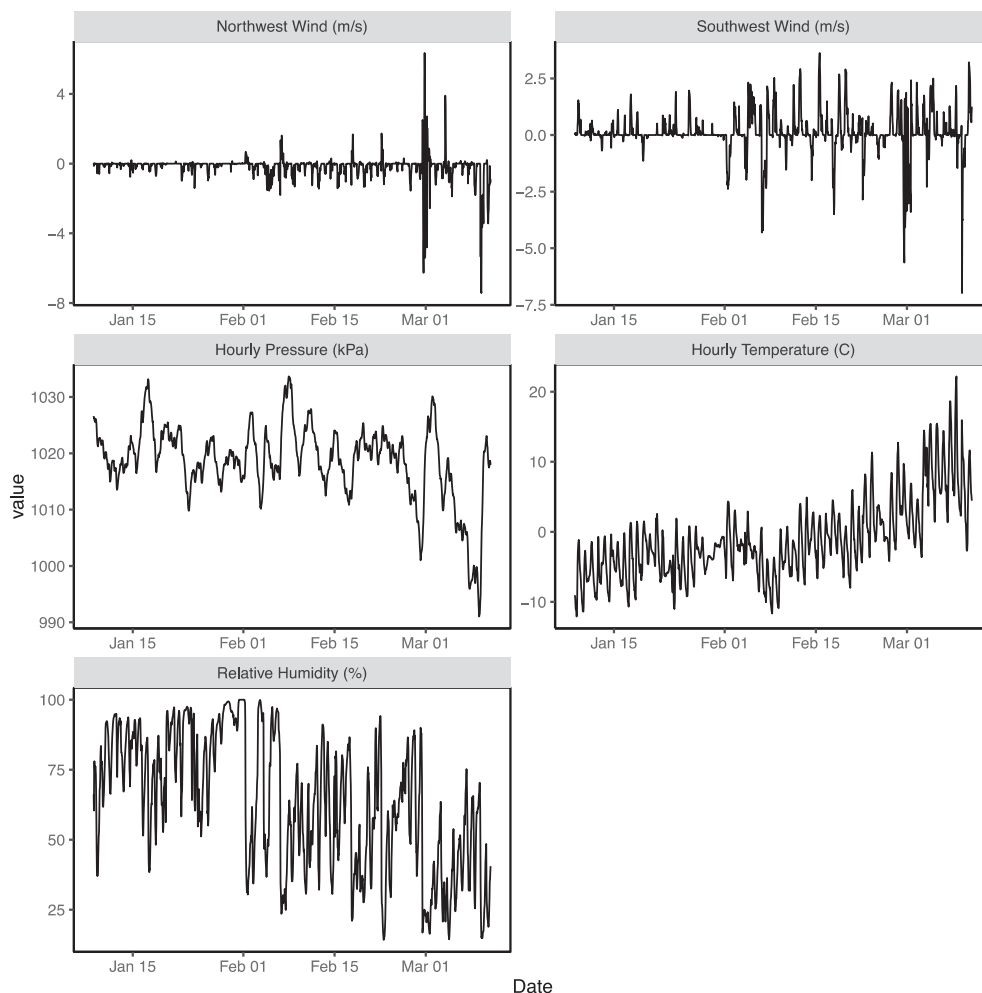This finding suggests that household coal burning for space

Fig. 4. Time series plot of meteorological variables.

Table 5
Coefficient of parameters in multivariate regression models.

Parameters	Estimate of coefficient	Standard error	P value
Lagged regression model			
Temperature	0.0293	0.0070	<0.001*
Pressure	−0.0251	0.0045	<0.001*
Relative Humidity	0.0343	0.0014	<0.001*
Northwest Wind	0.1380	0.0296	<0.001*
Southwest Wind	0.3145	0.0238	<0.001*
1 h lagged PM$_{2.5}$ Emission	0.4461	0.0707	<0.001*

*$p < 0.05$.

heating remains an important and dominant source of PM$_{2.5}$ emissions in peri-urban and rural areas in the North China Plain, including EHZ village. Compared to other sources of primary PM$_{2.5}$ emissions, household emissions from solid fuel use are poorly controlled, and PM$_{2.5}$ end-of-pipe EFs for household solid-fuel devices are generally orders of magnitude higher than those for industrial sources such as coal-fired power plants, where various controlling technologies are often installed (Lei et al., 2011). Most of the observed coal furnaces and fixed stoves, as well as the biomass *kang* and floor heating systems, were equipped with chimneys. While chimneys can reduce indoor air pollution levels, they usually direct household combustion emissions into ambient air without emission controls (Carvalho et al., 2016; Smith et al., 2011). This

movement of emissions — from the indoor environment to the near home, ambient environment — poses concerns for ambient air quality and population health, especially in areas with a high density of coal and biomass combustion (Carvalho et al., 2016).

Previous studies related to household fuel use in China relied mainly on household surveys or broad census questions about cooking and space-heating fuel use. Surveys, interviews and time-activity journals are all subject to recall bias and generally cannot provide detailed longitudinal information on fuel use frequency or durations. Recently, Carter et al. used continuous indoor PM$_{2.5}$ monitoring to estimate daily hours of biomass combustion for cooking and heating in households in the Tibetan Plateau in China with greater objectivity and temporal resolution (Carter et al., 2016). In this study, we complemented household surveys with cookstove and heating device SUMs monitoring to establish hourly solid fuel use patterns. Compared to other published studies in northern China, our study offered a finer time scale and a more objective quantification of fuel use in households, enabling us to estimate primary PM$_{2.5}$ emissions per hour (EPH) from household solid fuel use and to assess the associated impact on local ambient air quality. Our EPH estimates are dependent on the accuracy of SUMs peak selection algorithms, as well as the assumption that each type of cookstove or heating device used a similar amount of fuel within a given time period. SUMs peak selection methodologies were originally used to analyze data collected in Guatemalan and Indian households (Mukhopadhyay et al., 2012; Pillarisetti

et al., 2014; Ruiz-Mercado et al., 2012). We adapted elements of these studies, but developed new algorithms to accommodate a broader range of cookstove and heating device types, and performed visual checks to ensure the accuracy of the algorithms for all devices. While stoves and heating devices consume fuels differently, the majority of the solid fuel used by EHZ households during the heating season study period was coal for space heating in furnaces, so fuel and device type were comparable among households. Thus, this assumption will have a negligible impact on the accuracy of the emissions estimates, and bias the results towards the null hypothesis making it harder to find the relationship between emission and ambient air pollution.

4.2. PM$_{2.5}$ concentration in peri-urban Beijing

During our sampling period, we observed high concentrations of PM$_{2.5}$ in EHZ village. During 85% of the monitored heating season days, daily PM$_{2.5}$ concentrations exceeded the World Health Organization's (WHO) 24-h standard of 25 μg/m^3; on 74% of days, measured concentrations exceeded the PM$_{2.5}$ daily average interim air quality guideline of 75 μg/m^3, which is also the new Chinese national 24-h ambient air quality standard (Ministry of Environmental Protection of P.R. China and General Administration of Quality Supervision, Inspection and Quarantine of P.R.China, 2012; World Health Organization, 2005). Recognizing that these levels occur commonly during the year, a more appropriate comparison is probably with the annual Chinese standard of 35 μg/m^3. This was exceeded on 90% of days. between January 9th to March 10th, 2013.

Despite the popular belief that air quality in rural areas and villages outside large Chinese cities is better than that in city centers and the focused research effort on the air pollution in mega cities (Chan and Yao, 2008), our study found that, during the heating season, average ambient PM$_{2.5}$ concentrations measured in EHZ village in peri-urban Beijing were in the range or even higher than the reported average PM$_{2.5}$ concentrations for urban Beijing. Though the Beijing Municipal Environmental Protection Bureau (BJEPB) started monitoring PM$_{2.5}$ data in 2012 at < 35 stations and publishing an air quality index online, historic PM$_{2.5}$ concentrations collected during the heating season we monitored were not available (BJEPB, 2016). Thus, we used hourly PM$_{2.5}$ data monitored at the US Embassy in northeast urban Beijing for comparison (U.S. Department of State, 2016). The average ambient PM$_{2.5}$ concentration at the US Embassy site between January 9th and March 10th, 2013 was 99 ± 75 μg/m^3, lower than the measured concentration in EHZ village over the same period (139 ± 107 μg/m^3). Other studies conducted in northern China found that the average annual ambient PM concentration was comparable between rural villages and urban centers, with significantly lower PM concentrations found only in remote rural field sites away from residential areas (Li et al., 2014). Even though, depending on wind direction, local PM$_{2.5}$ may have been occasionally influenced by transport from the city, our work indicates that locally-generated ambient PM$_{2.5}$, is an important concern for China's growing peri-urban areas, as well as its urban centers.

4.3. Impact of household fuel combustion on ambient air pollution

Through multivariate regression evaluating the relationship between log-transformed hourly PM$_{2.5}$ concentration and 1-h lagged EPH and meteorological variables, we found a statistically significant association between PM$_{2.5}$ EPH from households and the log-transformed local ambient PM$_{2.5}$ concentration, after adjusting for meteorological conditions. Our study suggests that 39% (95% CI: 26%, 54%) of hourly averaged ambient PM$_{2.5}$ concentration was associated with total PM emissions from household space heating during the most recent hour at a local site. Our findings are consistent with other recent studies on the contribution of residential fuel use to ambient air pollution in China (Lelieveld et al., 2015; Liu et al., 2016; Xiao et al., 2015a). In contrast to our study's findings, Carter et al. reported that household heating deteriorated indoor air quality but not ambient air quality in China's Tibetan Plateau (Carter et al., 2016). This is likely due to lower population density in Beichuan County on the Eastern Tibetan Plateau than that in the North China Plain where our study was conducted. Our finding that household space heating negatively affects ambient air quality complements other studies on the impact of household cooking with solid fuel on ambient air quality (Chafe et al., 2014) and central heating systems on ambient air quality (Chen et al., 2013). We highlight that uncontrolled household emissions from space heating contribute significantly to ambient air pollution in China even if this sector accounts for a relatively small proportion of coal consumption in China (China Energy Statistical Yearbook, 2012, 2013).

Our study additionally demonstrates autocorrelation of local ambient PM$_{2.5}$ concentrations and the strong influence of meteorological conditions. All the meteorological variables − including T, P, RH, SWWS and NWWS − monitored at EHZ showed a statistically significant relationship with log-transformed ambient air PM$_{2.5}$ concentration in multivariate regression model. This is consistent with other studies on daily variations of air pollution with meteorological conditions (Liang et al., 2015; Liu, 2009; Zhou et al., 2015).

Because this study did not include measurement of background ambient PM$_{2.5}$ levels or source apportionment, we cannot quantify the relative contributions of other pollution sources to ambient PM$_{2.5}$ concentrations in EHZ, such as traffic, industrial sources, transportation from other sources in city and secondary particle formation. EHZ village is located 3 km outside of Beijing's 6th ring road and 1 km from the adjacent G5 national expressway. Though there is a brick factory just north of EHZ, which was not in operation during the winter study period, it is not located near any major industrial pollution sources. Motor vehicle emissions are a relatively large contributor to secondary aerosol formation, but much less to ambient primary PM$_{2.5}$ concentrations during winters in Beijing (Zhang et al., 2013). Another limitation of this study is that we could not conduct sensitivity analysis, due to the difficulties of consistently estimating variability of each parameter in the model.

Because of the high PM$_{2.5}$ EFs (Chen et al., 2015; Shen et al., 2010; Zhang et al., 2000; Zhi et al., 2008) and relatively large fuel consumption (Zhang and Koji, 2012), emissions from household solid fuel combustion were an important contributor to the severe ambient air pollution we observed in EHZ village. During the winter heating period, PM air pollution levels in the North China Plain often reach high levels, especially in unfavorable meteorological conditions (Rohde and Muller, 2015; Xiao et al., 2015a). For example, over a 12-month period, Li et al. found a mean PM$_{10}$ concentration of 182 ± 152 μg/m^3 in rural villages in northern China (Li et al., 2014). This suggests that pollution from outside EHZ village could be transported or dispersed to EHZ and pollution emitted within EHZ village could also be dispersed to other areas. Given the small size of EHZ village and the fine temporal scale of our air pollution data, it is reasonable to believe these effects will be minimal. However, transport of pollution in and out of the village made it harder for us to characterize the relationship between household emissions and ambient air pollution. Despite these limitations, our study showed a significant correlation between household space heating emissions and log-transformed ambient air PM$_{2.5}$ concentrations in a multivariate linear regression model. Overall, our results indicate that comprehensive pollution control strategies for Beijing, the surrounding North China Plain, and other

similar areas should address emissions from household solid fuel combustion, particularly during the winter heating season. Our findings also emphasize the need to monitor particle air pollution in peri-urban and rural areas in order to understand ambient pollution in China.

5. Conclusion

By conducting household surveys and continuous monitoring of cookstoves, heating devices, air quality, and meteorological conditions in EHZ village in peri-urban Beijing, we found empirical evidence suggesting that emissions from household solid fuel combustion negatively impact local ambient air quality during the winter heating season. During the heating season in EHZ, we observed high concentrations of ambient $PM_{2.5}$ and high emissions of $PM_{2.5}$ resulting from household solid fuel use, mostly from coal combustion for space heating. Using a multivariate linear regression model, we found a statistically significant relationship between 1-h lagged local $PM_{2.5}$ EPH from household sources, and ambient air $PM_{2.5}$ concentrations, after adjusting for meteorological effects.

The methods we developed and applied to estimate $PM_{2.5}$ emissions from solid fuel combustion we employed can be utilized in other household air pollution as well as ambient air pollution studies. Our findings suggest that uncontrolled household emissions, particularly from space heating with coal, remain a large and significant source of ambient air pollution in rural and peri-urban China during winter months. Any comprehensive pollution control strategy for northern China should address the full range of pollution sources, including uncontrolled emissions from household solid fuel combustion, particularly during the winter heating season. To reduce household air pollution in peri-urban and rural areas of northern China, technological and policy resources should be leveraged to ensure use of cleaner energy sources for space heating and/or installation of solid fuel pollution controls at the household level.

Acknowledgements

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2017.05.053.

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