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Patterns of Household Concentrations of Multiple Indoor Air Pollutants in China

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Most previous studies on indoor air pollution from household use of solid fuels have used either indirect proxies for human exposure or measurements of individual pollutants at a single point, as indicators of (exposure to) the mixture of pollutants in solid fuel smoke. A heterogeneous relationship among pollutant−location pairs should be expected because specific fuel-stove technology and combustion and dispersion conditions such as temperature, moisture, and air flow are likely to affect the emissions and dispersion of the various pollutants differently. We report on a study for monitoring multiple pollutants—including respirable particles (RPM), carbon monoxide, sulfur dioxide, fluoride, and arsenic—at four points inside homes that used coal and/or biomass fuels in Guizhou and Shaanxi provinces of China. All pollutants exhibited large variability in emissions and spatial dispersion within and between provinces and were generally poorly correlated. RPM, followed by SO_2, was generally higher than common health-based guidelines/standards and provided sufficient resolution for assessing variations within and between households in both provinces. Indoor heating played an important role in the level and spatial patterns of pollution inside homes, possibly to an extent more important than cooking. The findings indicate the need for monitoring of RPM and selected other pollutants in longer-term health studies, with focus on both cooking and living/sleeping areas.

Introduction

Almost three billion people worldwide rely on biomass (wood, charcoal, crop residues, and dung) and coal as their primary source of domestic energy (*1*, *2*). Hundreds of harmful chemical substances are emitted during the burning of biomass or coal in the form of gases, liquids (suspended droplets), or solids (suspended particulates), in particularly large quantities when burned in open or poorly ventilated stoves. These pollutants include carbon monoxide, nitrogen dioxide, particles in the inhalable range (below 10 μm in aerodynamic diameter), and other organic matter (predominantly composed of polycyclic aromatic hydrocarbons such as benzo[*a*]pyrene and other volatile organic compounds such as benzene and formaldehyde) (*3*−*5*). Combustion of coal in addition to the above pollutants may release oxides of sulfur and heavy metal contaminants including arsenic and fluorine (*6*).

Exposure to indoor air pollution (IAP) from the combustion of solid fuels has been implicated, with varying degrees of supporting evidence, as a causal agent of several diseases in developing countries including acute respiratory infections (ARI), chronic obstructive pulmonary disease (COPD), lung cancer (for coal smoke), asthma, nasopharyngeal and laryngeal cancers, tuberculosis, low birth weight, and diseases of the eye such as cataracts and blindness (*7*−*10*). Conservative estimates of global mortality due to indoor air pollution from solid fuels show that, in 2000, more than 1.6 million deaths and nearly 3% of the global burden of disease were attributed to this risk factor, making this risk the 11th leading cause of global mortality and eighth leading cause of global disease burden among selected major risk factors (*11*, *12*).

Most previous research, especially those projects that have simultaneously monitored exposure and health outcomes, have used either indirect proxies for human exposure (e.g., the use of solid fuels) or measurements of individual pollutants [e.g., particulate matter (PM) or carbon monoxide (CO)] as indicators of (exposure to) the mixture of pollutants in solid fuel smoke (*7*, *13*−*15*). Furthermore, the concentrations of these pollutants have been measured at a single point or small number of points in each household. The use of indirect or single-pollutant indicators will likely remain common in epidemiological studies and program evaluation, because of cost and difficulties associated with measurement of multiple pollutants at multiple points.

There are nevertheless a number of reasons for more in-depth measurement of pollution in selected studies (*16*). First, the relationship between different pollutants (e.g., CO and PM) has varied substantially across studies or even across various fuel−stove combinations within the same study (*17*−*19*). The heterogeneous relationship among pollutants is to be expected, because while these pollutants are mostly products of incomplete combustion, combustion conditions such as temperature, moisture, and air flow are likely to affect the emissions of the various pollutants differently (*17*). Second, even if the emissions of multiple pollutants are correlated on average, the combustion conditions may affect their relationships at specific moments during the combustion cycle (*17*, *19*, *20*). Third, the spatial dispersion of these pollutants inside the house may vary depending on whether they are particles or gases and on the physical characteristics of the house (e.g., whether the house consists of a single large room or multiple rooms separated by walls and doors). The heterogeneity of emissions and spatial dispersion means that multipollutant and multipoint monitoring can provide better quantification of exposure for the diverse health outcomes that may be associated with the different pollutants in solid fuel smoke, in turn resulting in designing more effective interventions. For example, while PM may be the appropriate indicator for acute respiratory infections (ARI)

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and chronic obstructive pulmonary disease (COPD), the effects of maternal exposure on birth weight may be affected by CO. Transition from wood to charcoal has been found to significantly reduce PM emissions with less benefits for CO concentrations (*19*). Measurements of both pollutants would allow evaluation of the complete benefits of interventions. An extreme case of the above issues arises in the case of pollutants that are specific to the type of fuel used (e.g., fluoride and arsenic from the combustion of some types of coal). Measurement of these pollutants, especially when coupled with data on the specific diseases that are affected by them (e.g., dental or skeletal fluorosis and arsenic poisoning) (*6*), would allow estimation of the risk associated with exposure and evaluation of the effectiveness of interventions in reducing them.

In addition to the above analytical benefits, multipollutant and multilocation measurements in a small number of in-depth studies can guide the choice of indicator pollutants in subsequent studies (*16*). Measuring some pollutants is more difficult or costly than others because of the equipment used or additional field and laboratory requirements. The operations of measurement instruments for various pollutants also interfere differently with household activities because of the size of equipment or their noise. Finally, field measurements of various pollutants entail different degrees of reliability based on the available technology and analytical techniques. These factors motivate selecting the subset of pollutants in a number of in-depth studies that can provide the most information on the concentrations of multiple pollutants at different points in the house, within the constraints such as cost and interference with household activities, for subsequent studies.

As a step toward better understanding the diversity of exposure determinants and patterns, this paper reports on one such in-depth study for monitoring multiple pollutants in coal-burning households in Guizhou and Shaanxi provinces in China. The above issues are particularly relevant in China, where nearly 80% of households rely on solid fuels (biomass and coal) for their domestic energy (*21*, *22*). The diversity of climate, housing, fuels and stoves, and sociocultural factors that influence food types and food preparation in China also motivate calibrating the indicator pollutants to local conditions. The study was the pilot phase of a larger ongoing study of household energy technology, IAP, and health in China. The study was conducted during winter heating season so that the role of both cooking and heating in patterns of indoor air pollution are examined.

Study Area

The study took place in Guiding County, Guizhou province, and Hanbin district of Ankang County, Shaanxi province (referred to as Guizhou province and Shaanxi province, respectively, hereafter) (Figure 1). Some important characteristics of the two provinces are summarized in Table 1, and they illustrate the relevance of IAP exposure from household energy use, including the importance of heating. Heating season in both provinces lasts approximately from November until March. Winter temperatures are, however, lower in Shaanxi, requiring longer daily heating hours and limiting air exchange (i.e., closing of windows and doors).

The most common house design in Guizhou consisted of 2–3 rooms, including cooking/living, sleeping, and entrance/storage, connected with doors. Although most houses had a separate cooking area, cooking was done almost entirely in one of the main rooms (cooking/living room), especially during the heating season (as was the case during the study period). This room, which also served for heating and living purposes, contained a stove (Figure 2a), with a chimney in most houses, used also for drying and smoking food (e.g., corn, chili, and pork) (Figure 2b). The separate cooking area was used primarily for large family events (e.g., Spring Festival) and for making animal feed. All houses had an attic above the cooking/living and sleeping rooms, used for food drying and storage, and at times containing an additional bed. The stove chimney ended in the attic in all houses. The lower rooms and the attic were separated by a porous ceiling (e.g., made of pieces of wood), which allows air flow between the two levels (Figure 2b). Most houses in Shaanxi province had a cooking area connected to the main house by a door, a living room with a ground stove (fire-pit) used for heating and boiling water (Figure 2c), and one bedroom which sometimes also has a ground stove. Most houses had a small attic area used for storage but not for sleeping.

FIGURE 1. Guizhou and Shaanxi provinces.

TABLE 1. Characteristics of the Two Study Regions

	Guizhou	Shaanxi
study county	Guiding	Ankang
altitude of study region, m	1100–1400	350–1000
avg summer temp, °C (daily min, max)	27 (19, 33)	28 (23, 41)
avg winter temp, °C (daily min, max)	8 (−1, 15)	5.5 (−8, 11)
avg rainfall, mm	1100–1400	1120
% of population rural	85	80
under-5 mortality (per 1000 live births)	32	39
main food staples	rice, corn	wheat, rice
other foods	chili	chili, corn
no. of sunny hours/year	~1100	~1500

Coal is a common fuel in both Guizhou and Shaanxi, used by nearly all households for heating and by many for cooking. The types of coal and combustion technologies are, however, different in the two provinces. Most of the coal used by the study households in Guizhou province is bituminous coal and/or anthracite, obtained from surface exposures (*6*). In parts of the province, including the study region, these coals have undergone mineralization, causing their enrichment in potentially toxic trace elements such as arsenic and/or fluorine (*6*). In the study households in Guizhou, coal is generally burned in metal stoves with limited insulation and ventilation (Figure 2a). In Shaanxi province study households, most of the coal used for household energy is stone-coal (also called bone-coal) (Figure 2c), with high concentrations of sulfur and in some locations fluoride and/or arsenic. (Note: The terms "bone-coal" and "stone-coal"

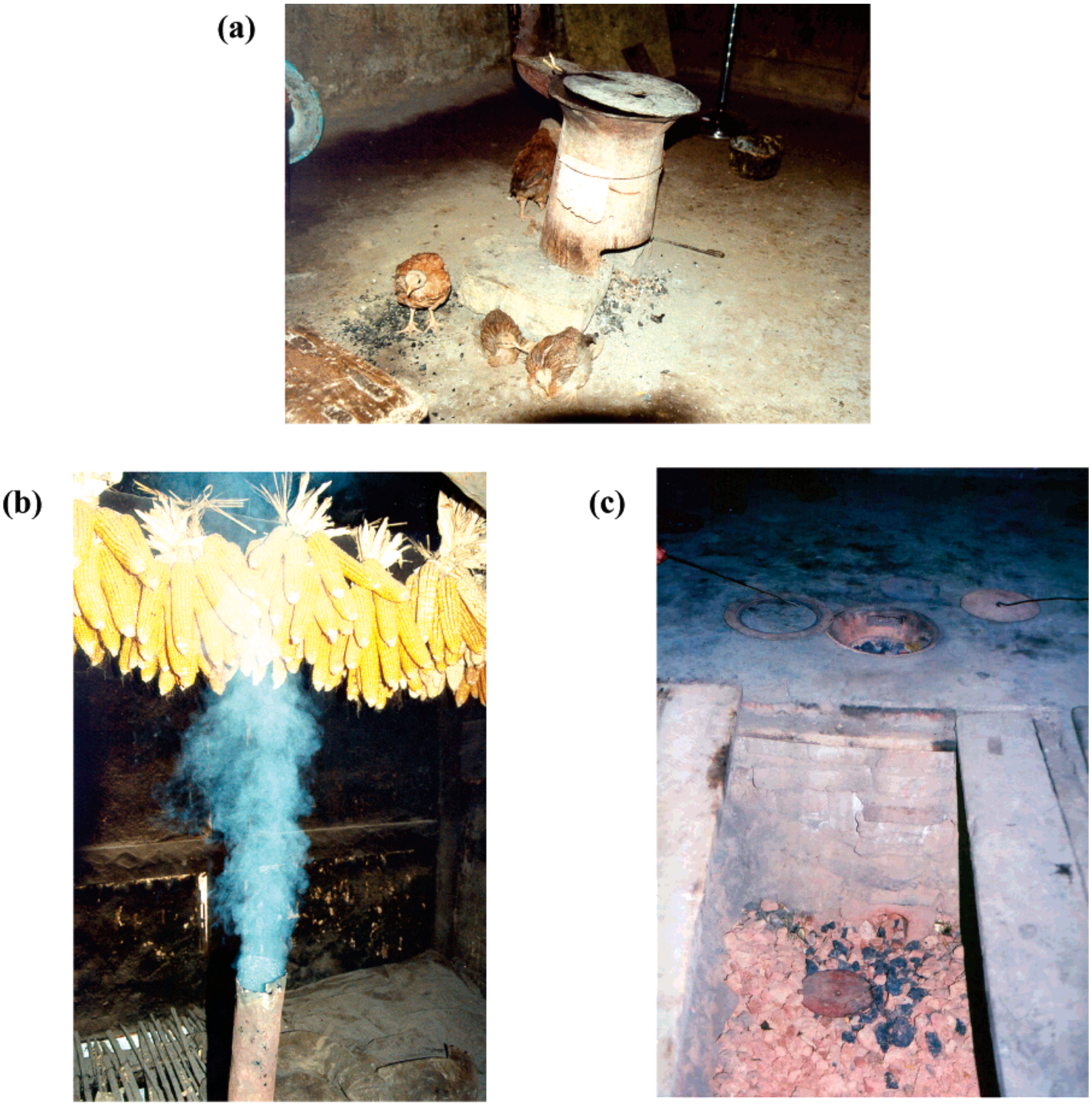

FIGURE 2. (a) Stove in cooking/living room in Guizhou province; (b) stove chimney used for drying food in the attic in Guizhou province; (c) ground stove in the heated living room in Shaanxi province.

are generally used for carbonaceous shales, organic-rich rocks with more than 50 wt % ash yield. Shaanxi Province is a major coal producer. The use of stone-coal, which is generally considered lower quality, nonetheless persists in the poorest households including those in the study area.) The coal stoves in Shaanxi province are simple combustion chambers dug in the floor (named ground stove or fire-pit) with a bottom outlet to remove the pieces of rock that remain after burning (Figure 2c).

Experimental Procedures

Selection of Households. Four households in each province were selected for monitoring. Measurements were taken at four different points in each house, over four consecutive days to also examine day-to-day variation. Because this was a detailed pilot study, there was an emphasis on selecting households whose housing and fuel-stove characteristics were representative of those in the larger study group. The selection of households was based on specific criteria that included the following:

(i) Houses were located both at the center and near the edges of the village.

(ii) Houses had structures (number and layout of rooms) that were typical of the village.

(iii) In three houses in Guizhou province, coal was the only fuel used on the days of measurement. This allowed monitoring the relationship among different pollutants in multiple houses that used the most common fuel in the province. The fourth household used both coal and biomass in the cooking/living room. This household was selected to monitor the indicator pollutants under another energy use pattern, likely to occur in some houses. In Shaanxi province, all households used a coal stove for heating in the living room and a combination of biomass and coal in the cooking room.

(iv) The stoves, especially the coal stoves, in all selected households were typical of those seen in other households in the village (Figure 2).

(v) The households joined the study voluntarily.

Measurements of Pollutants. *Measurement Locations and Times.* Measurements for multiple pollutants (Table 2) took place over four 24-h days, in late January 2003 in Guizhou province and early February 2003 in Shaanxi province, by teams consisting of investigators from the China National

TABLE 2. Number of Measurement Days and Households for Different Pollutants[a]

pollutant	no. of days	no. of households	total measurements (both provinces)	common standards/guidelines
respirable particles (RPM)[b]	4	4	128	50 μg/m^3 (24 h) U.S. EPA[c] 3000 μg/m^3 (8 h) ACGIH[c,d]
CO	4	4	128	10 ppm (8-hr) WHO 25 ppm (8 h) ACGIH
SO_2	4	4	128	0.04 ppm (24 h) WHO 5 ppm (15 min STEL) ACGIH
arsenic	1	2	16	10 μg/m^3 (8 h) ACGIH
fluoride	2	2	32	2500 μg/m^3 (8 h) ACGIH

[a] On each measurement day, measurements were taken at four points in each house as described in Experimental Procedures. The number of measurement days, households, and points was based on equipment availability. U.S. EPA, United States Environmental Protection Agency; WHO, World Health Organization; ACGIH: American Conference of Governmental Industrial Hygienists (all ACGIH limit values listed in the table are with reference to the occupational environment); STEL, short-term exposure limit. [b] Defined as PM_4, particulate matter with a median aerodynamic diameter of less than 4 μm. [c] For PM_{10} (particulate matter less than 10 μm in aerodynamic diameter) in ambient air. Current scientific literature based on studies in outdoor air indicates that there may be no threshold for health effects associated with particulate matter exposure (*23, 24*). [d] For PM_4 (particulate matter with a median aerodynamic diameter of less than 4 mm) not otherwise regulated in the occupational environment.

Center for Disease Control and Prevention (CDC) as well as provincial and county CDC staff. On each day, measurements for all pollutants (Table 2) began in mid or late morning and continued until the next day, as close to a full 24-h period as possible. Measurements were taken at four points in each house on each monitoring day. Measurements in Guizhou province were in the cooking/living room, bedroom, attic, and outside the main entrance of the house. Cooking/living room and outside measurements were taken at a height of approximately 1–1.5 m, corresponding to the sitting position of an adult or standing position of a child. Bedroom measurements were taken above the bed surface, and attic measurements as close to the chimney outlet as possible (within approximately 1 m). In Shaanxi province, measurements were in the cooking room, living room, bedroom, and outside the main entrance of the house, at a height of approximately 1–1.5 m.

Respirable Particles. Sampling for respirable particles was done according to The National Institute for Occupational Safety and Health, NIOSH, Protocol 0600, designed to capture particles with a median aerodynamic diameter of 4 μm (PM_4) (*25*). Samples were collected by use of a 10-mm nylon cyclone equipped with a 37-mm diameter poly(vinyl chloride) (PVC) filter (pore size 5 μm; supplied by SKC Inc.) at a flow rate of 2.5 L/min. Air was drawn through the cyclone preselectors by battery-operated constant-flow pumps (model PCXR8 supplied by SKC Inc.). All pumps were calibrated prior to and after each sampling day with a field minimeter, itself calibrated by a soap bubble meter in the laboratory. Pumps were also calibrated in the laboratory after each field exercise with the same minimeter. To maintain battery power throughout the sampling period, pumps were programmed to cover the 24-h interval through intermittent sampling (1 min out of every 4–6 min). One field blank was taken on each sampling day.

Gravimetric analyses were conducted at the laboratory of the National Institute for Environmental Health and Related Products Safety, China CDC, on an analytic microbalance (1/100 000, 2004 MP) calibrated against standards provided by the Bureau of National Technological Control. All filters (field blanks and samples) were conditioned for 24 h before weighing. Respirable dust concentrations were calculated by dividing the blank-corrected increase in filter mass by the total air volume sampled.

Carbon Monoxide and Sulfur Dioxide. Carbon monoxide (CO) and sulfur dioxide (SO_2) were measured with long-term diffusion tubes (manufactured by Gastec), with a detection range of 10–200 ppm over 24 h for CO and 2–100 ppm over 8 h for SO_2. For SO_2, multiple tubes were used during the 24-h sampling interval and replaced at the end of the recommended exposure time, with the 24-h average calculated as the time-weighted average of the short-term tubes.

Arsenic and Fluoride. Arsenic and fluoride were sampled on mixed cellulose acetate filters and treated pads (pore size 0.5 μm, supplied by SKC Inc.) by use of battery-operated pumps (PCXR8 supplied by SKC Inc.) according to NIOSH protocols 7300 and 7902, respectively (*25*). Analysis of the filters was carried out at the China CDC laboratory by GF-AAS (graphite furnace atomic absorption spectrometry) with auto-inject sample system (Unicam 989 QZ) for arsenic and ion-selective electrode for fluoride (Orion EA 940).

Results

Single Pollutant Concentrations. Figure 3 shows the distributions of RPM, CO, and SO_2 concentrations by measurement point in both provinces. RPM concentrations at all locations in both provinces were in excess of health-based standards for particulate matter in ambient (outdoor) environment (Table 2) and comparable to the 3000 μg/m^3 threshold limit value (TLV) for insoluble respirable particulates in occupational settings recommended by ACGIH (Table 2) in the cooking/living room, bedroom, and attic in Guizhou. Mean 24-h SO_2 concentrations were higher than the WHO guideline value of 0.04 ppm at all locations including outdoors (see below on spatial patterns). With the exception of a few measurements in the attic in Guizhou and in the coal-heated living room in Shaanxi, average concentrations of SO_2 were within the 15-min STEL ACGIH guideline value of 5 ppm at all locations. Since each measurement tube was exposed over a period of a few hours, peak concentrations over a shorter period may have exceeded this value, which would not be detectable in our data. The diversity of energy technology and housing in different regions of China, and the variations in measurement conditions (e.g., season, duration, and location) prevent direct comparisons with previous studies in China. Nonetheless, broad comparisons with previous studies, as summarized by Smith (*26*), illustrate that RPM concentrations in Guizhou were similar to the highest levels seen in previous work and those in Shaanxi were lower than those in previous research.

RPM concentrations were considerably higher in Guizhou than in Shaanxi, but SO_2 concentrations were higher in Shaanxi than the corresponding points in Guizhou (e.g., the area near the coal-burning ground stove in the heated living room in Shaanxi compared to the cooking/living room in Guizhou). Higher RPM concentrations in Guizhou may have been due to the fuel–stove combination or combustion conditions (amount/size of fuel added and air flow), and higher SO_2 concentrations in Shaanxi may have been due to

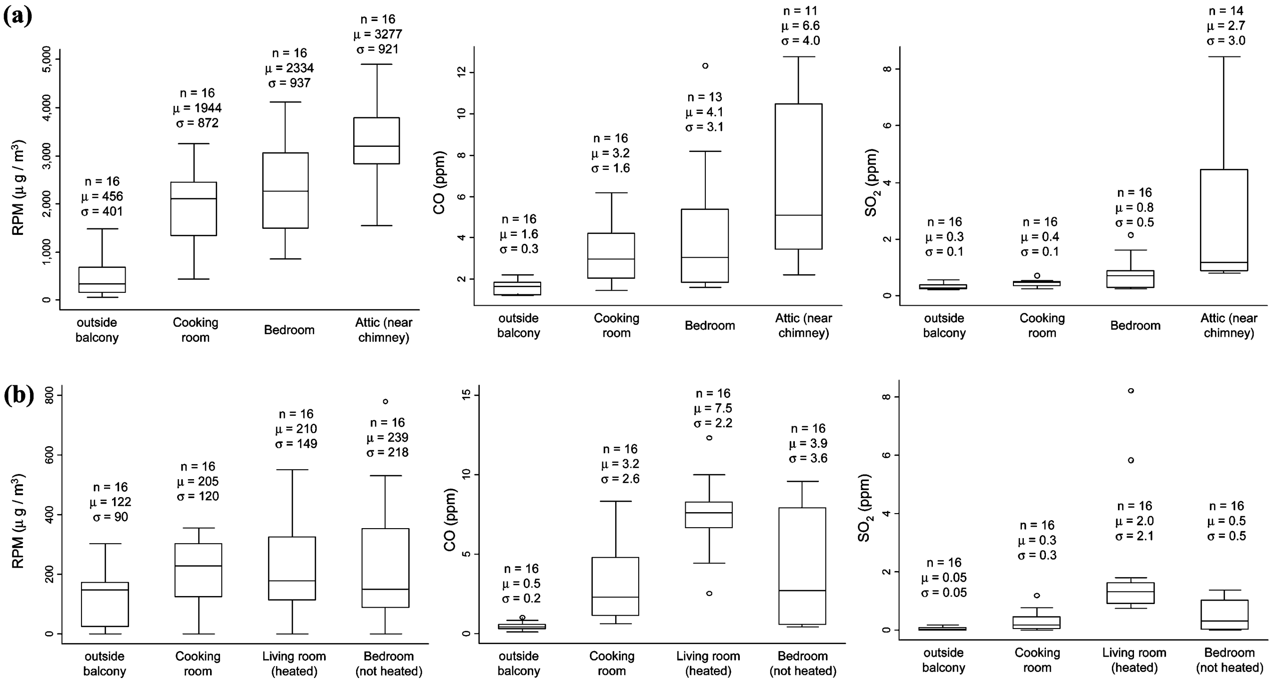

FIGURE 3. Average 24-h concentrations of RPM, CO, and SO_2 at different measurement points in (a) Guizhou province and (b) Shaanxi province. Note: If the concentrations of CO and SO_2 were higher than the detection range of tubes (200 ppm for CO and 100 ppm for SO_2), the following assumptions were made: (i) Those measurements that were only slightly higher than the measurement range were set to the maximum value. The number of measurements was (a) Guizhou, three measurements for CO (two in the bedroom and one in the attic) and two measurements for SO_2 (both in the attic); (b) Shaanxi, six measurements for CO (three in the heated living room and three in the bedroom) and one measurement for SO_2 (in the heated living room). (ii) Those measurements that were substantially higher than the measurement range were set to 150% of the maximum value. The number of measurements was (a) Guizhou, five measurements for CO (one in the bedroom and four in the attic) and zero measurements for SO_2; (b) Shaanxi, zero measurements. The overall results were not sensitive to this assumption because most of these measurements were at points where pollution was consistently highest (e.g., attic).

both type of coal and the use of chimney in Guizhou province (the SO_2 concentrations in the attic in Guizhou were higher than Shaanxi, possibly as a result of the venting chimney that ended in the attic; Figure 2b).

Average CO, arsenic, and fluoride concentrations were consistently below available health-based standards and guidelines and in some cases CO concentrations were close to the detection limits of the diffusion tubes. CO levels at all locations were consistently less than the WHO guideline values of 10 ppm and ACGIH guideline value of 25 ppm for 8-h exposures, respectively, and did not exceed the short-term exposure level (STEL) guideline values provided by the same agencies during cooking periods. The 24-h average concentration of arsenic in air ranged from 0.02 to 0.1 $\mu g/m^3$ in Guizhou province and from 0.10 to 0.92 $\mu g/m^3$ in Shaanxi province, all lower than the 8-h time-weighted average TLV values of 10$\mu g/m^3$ from ACGIH guidelines. The 24-h average concentration of fluoride in air ranged from 1.7 to 4.4 $\mu g/m^3$ in Guizhou province and from approximately 0 to 2.4 $\mu g/m^3$ in Shaanxi province, again substantially lower than the 8-h time-weighted average TLV values of 2500 $\mu g/m^3$ from ACGIH guidelines. Since these were 24-h concentrations, 8-h levels may have been higher in intervals around cooking, not observable in our data.

Spatial Patterns of Pollutant Concentrations. In Guizhou (Figure 3a), the upstairs attic, where the chimney ended, consistently had the highest concentrations of pollutants. A somewhat unexpected finding in Guizhou was that the concentrations in the bedroom—where there was no stove and which was connected by a door to the cooking/living room—were consistently similar to, or even slightly higher than, the cooking/living room where the stove was located. Although this phenomenon differed across pollutants, this implies that direct dispersion inside the house, and probably more importantly the transport of pollutants through the chimney and subsequent dispersion via the attic, make the bedroom an important microenvironment for exposure. Therefore, although the attic is not directly an important exposure microenvironment because of the time−location−activity patterns of household members (i.e., despite its high concentrations, little time is spent there), its porous floor creates an important role in dispersion of pollutants into the rooms on the main floor.

In Shaanxi (Figure 3b), the cooking room, the heated living room, and the bedroom all had relatively similar RPM concentrations. But the concentrations of CO and SO_2 were highest in the heated living room. The reason for this differential pattern is that biomass was the primary fuel in the cooking area and was used for a shorter time, resulting in high RPM concentration but limited contribution to CO and SO_2. The fuel used in the heated living room on the other hand was consistently coal, with high sulfur concentration and used for a longer time, therefore resulting in higher emissions of CO and SO_2. Pollutant concentrations in the bedroom had the largest variation across measurement days and households. Given that the households did not use a stove in the bedroom, pollution levels in this room are determined by dispersion from other locations and therefore highly dependent on the household-specific parameters that affect dispersion. The high concentrations in the heated living room and the nonnegligible levels in the bedroom illustrate the important role of heating as a source of exposure in winter.

Outside concentrations in both provinces were lower than all points inside the house, although nonnegligible for some households. In these rural settings, the outside concentrations are almost certainly a result of household energy use, and dispersion of indoor emissions to the outdoors through chimneys and other openings. It is not very likely that the indoor pollutant concentrations are greatly affected by the ambient levels, which are lower.

TABLE 3. Pairwise Correlation Coefficients for Long-Term 24 h RPM, CO, and SO_2 Concentrations across All Measurement Locations

	CO	SO_2
	(a) Guizhou Province	
RPM	0.48 ($p < 0.001$)	0.42 ($p < 0.001$)
CO		0.82 ($p < 0.001$)
	(b) Shaanxi Province	
RPM	0.29 ($p = 0.02$)	0.18 ($p = 0.15$)
CO		0.57 ($p < 0.001$)

IAP exposure depends on pollution concentrations as well as time−location−activity patterns of household members (i.e., where they are inside the house and at what times, in relation to pollution patterns). In both provinces, pollution from cooking affects primarily women who spend 2−4 h per day cooking human and animal food. In Shaanxi, where cooking takes place in a separate kitchen, exposure during cooking primarily affects women and children who may stay close to their mothers. In Guizhou, cooking food for household consumption (but not for animals) takes place in the living area, exposing all household members. In both provinces, heating is a source of exposure for all household members who spend time around the heating stove in the living area. In Guizhou, where the chimney in most houses extends only as high as the attic area, smoke disperses in the house through the porous separation of main floor and attic and hence affects household members during sleeping as well.

Correlations among Multiple Pollutants. Correlations between RPM, CO, and SO_2 were examined across households to evaluate whether RPM measurements, which involve equipment that is more costly, harder to operate in field conditions, and noisier, could be substituted by CO or SO_2 measurements. When data from all measurements (Table 2) were pooled, the pairwise correlations between RPM and each of these two pollutants were weak, although generally statistically significant (Table 3).

The comparison of the correlation coefficients between the two provinces shows that when all points are considered together, first, the concentrations of CO and SO_2 were better correlated than the concentrations of either pollutant with RPM, and second, the correlation between either CO or SO_2 and RPM was lower in Shaanxi than in Guizhou. There are two possible reasons for the findings on pollutant correlations. First, CO is a common product of coal combustion and SO_2 is a common outcome of combustion of coal with high concentrations of sulfur, which is the case in many parts of China. This is in contrast to RPM, which is more dependent on the fuel−stove combination and combustion conditions. Second, CO and SO_2, both of which are gaseous, have dispersion characteristics more similar to each other than to particles. As seen in Figure 3, on average, Shaanxi province homes had lower concentrations of RPM but generally higher concentrations of CO and SO_2, possibly due to the type of coal used and combustion conditions. Fuels and stoves used by the households in each province were relatively similar, to the extent observable under field conditions. Therefore, it is unlikely that stratification on stove and fuel type would improve the correlations considerably. Rather, it is likely that variations in combustion conditions, caused by stove handling behaviors and more subtle differences in fuels and stoves, are the cause of these findings, as also found in previous studies that have monitored PM and CO (biomass was the fuel in previous studies) (*14*, *17*, *19*).

Exposure to IAP depends on the location of household members with respect to the source and dispersion of pollutants. For example, cooking, heating, and sleeping activities would take place at different locations in the house. This motivates an assessment of the performance of indicator pollutants in each location. The relationship between daily average concentration of RPM and CO at the four measurement points is shown in the Supporting Information, as well as the pairwise correlation coefficients for RPM, CO, and SO_2. As seen in the Supporting Information, pollutant correlations become even weaker when compared at individual points. The correlations generally remain large between CO and SO_2 and for measurements near the chimney in Guizhou province. Beyond these special cases, the results for these three pollutants show that their average concentrations are not correlated in solid-fuel-burning households in these two Chinese provinces. Previous research has shown that instantaneous concentrations are even more poorly correlated than daily averages, possibly because the pollutants are affected by different combustion parameters, which are less likely to co-occur instantaneously than on average (*17*, *19*).

Discussion

Exposure to IAP is a complex function of energy (fuel type and stove characteristics), housing, and behavioral factors. Because unlimited measurements are not possible under field conditions, indicator pollutants must be selected in each field study that best characterize exposure to the important pollutants, given the purpose and context of each monitoring exercise (*16*). This study aimed to provide an example of systematic assessment and choice of indicator pollutants in China, where a diverse set of energy use, housing, and exposure patterns exists. The results of this study illustrate the following:

(i) RPM was the pollutant with the highest resolution for examining temporal and spatial variations in both provinces. Furthermore, RPM concentrations in most locations were above available health-based guidelines making it an important pollutant for assessing potential impacts of interventions.

(ii) For indoor measurements, analysis of variance (ANOVA) illustrated that the fraction of variance of RPM measurements that was explained by variability between different days in the same household was between 70% and 200% of the fraction explained by variability between different households in Guizhou province and between 30% and 400% in Shaanxi province. This indicates that, for some locations within the house, RPM concentrations had considerable temporal variations even between consecutive days, making a case for longitudinal monitoring of some households.

(iii) The lack of correlation between RPM concentration and CO and SO_2 concentrations implies that, in these households, the less expensive CO measurements cannot provide a robust indicator for the more costly, but more important pollutant from a respiratory health perspective, particulate matter. Since no or few health studies have used CO as a cause of respiratory health effects, it might seem reasonable not to measure this pollutant at all. Two other considerations, however, motivate measuring CO. First, CO poisoning is an important hazard, especially in heating season when doors and windows are closed. Second, because of the low cost of measuring CO, and because seasonal variations in CO concentrations may provide valuable information about total combustion, this pollutant should be measured at least in a limited subset of households. Since SO_2 levels were higher than health-based guidelines in approximately 40% of measurements in Shaanxi province, SO_2 measurement would likely be of value in Shaanxi province.

(iv) Although the attic in Guizhou province had very high pollution levels, it contributes little to people's exposures (residents reported spending the least amount of time in the attic). The cooking, living, and sleeping areas are likely to contribute most to people's exposures and are appropriate for 24-h monitoring of particulates in subsequent data collection.

(v) That the concentrations of arsenic and fluoride were below available guideline values at all locations across households was an unexpected finding. Coal here has been found to have high concentrations of fluorine (Guizhou) and arsenic (Guizhou and Shaanxi) (*6*). Previous studies and qualitative assessment by the authors also indicate relatively high prevalence of dental/skeletal fluorosis and arsenic toxicity in Guizhou. The low concentrations therefore mean that direct inhalation may not contribute significantly to exposures but may be mediated by bioaccumulation through secondary deposition on food (Figure 2). Another possible route of exposure is through water. This allows limiting the number of measurements of trace elements to a smaller subset of households to generate additional baseline data on environmental levels, while at the same time pointing out the need for biological monitoring and food assays to assess exposure and the potential benefits of interventions.

The challenge in IAP monitoring in developing countries is to optimize a set of measurements following validated protocols with sufficient resolution to provide pollution and exposure information in a manner that can be used to assess not only potential health impacts but also the efficacy or effectiveness of planned interventions (*16*). Detailed in-depth monitoring of multiple pollutants in multiple points inside households in two Chinese province has allowed the determination of pollutant−location combinations to be used in the households during the baseline and postintervention follow-ups. These studies will cover a larger number of households (approximately 75 in each province) on a cross-sectional basis, together with 5−6 households being monitored longitudinally to examine day-to-day and seasonal variations. The results also indicate that currently particulate measurements, despite being cumbersome and expensive, are likely to be required in most studies of indoor air pollution from solid fuels, as they may offer higher reliability for predicting respiratory health impacts. In addition, since few studies anywhere have monitored such a large number of pollutants simultaneously, the results have served to demonstrate correlations and choice of indicator pollutants for applications in other settings where solid fuel use is common. Beyond the specific implications for monitoring in these provinces, the results of the measurements provide strengthened evidence that IAP measurements—and subsequent interventions—in rural households require considerable regional customization.

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Supporting Information Available

Figure showing PM−CO relationships in different measurement points in Guizhou and Shaanxi provinces. This information is available free of charge via the Internet at http://pubs.acs.org.

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