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Geographical, Spatial, and Temporal Distributions of Multiple Indoor Air Pollutants in Four Chinese Provinces

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Exposure to indoor air pollution from household energy use depends on fuel, stove, housing characteristics, and stove use behavior. We monitored three important indoor air pollutants—respirable particles (RPM), carbon monoxide (CO), and sulfur dioxide (SO_2)—for a total of 457 household-days in four poor provinces in China (Gansu, 129 household-days; Guizhou, 127 household-days; Inner Mongolia, 65 household-days; and Shaanxi, 136 household-days), in two time intervals during the heating season to investigate spatial and temporal patterns of pollution. The two provinces where biomass is the primary fuel (Inner Mongolia and Gansu) had the highest RPM concentrations (719 μg/m^3 in the single cooking/living/bedroom in Inner Mongolia in December and 351−661 μg/m^3 in different rooms and months in Gansu); lower RPM concentration were observed in the primarily coal-burning provinces of Guizhou and Shaanxi (202−352 μg/m^3 and 187−361 μg/m^3 in different rooms and months in Guizhou and Shaanxi, respectively). Inner Mongolia and Gansu also had higher CO concentrations (7.4 ppm in the single cooking/living/bedroom in Inner Mongolia in December and 4.8−11.3 ppm in different rooms and months in Gansu). Among the two primarily coal-burning provinces, Guizhou had lower concentrations of CO than Shaanxi (1.2−1.8 ppm in Guizhou vs 2.0−13.3 ppm in different rooms and months in Shaanxi). In the two coal-burning provinces, SO_2 concentrations were substantially higher in Shaanxi than in Guizhou. Relative concentrations in different rooms and provinces indicate that in the northern provinces heating is an important source of exposure to indoor pollutants from energy use. Day-to-day variability of concentrations within individual households, although substantial, was smaller than variation across households. The implications of the findings for designing environmental health interventions in each province are discussed.

Introduction

Biomass fuels (wood, charcoal, crop residues, and dung) and coal are the primary source of domestic energy for one-half of the world's population (*1*). Combustion of biomass and coal, especially in open or poorly ventilated stoves, emits hundreds of health damaging pollutants (*2*) that cause a number of diseases (*1*, *3*, *4*). Indoor air pollution (IAP) from household use of solid fuels resulted in more than 1.6 million deaths and nearly 3% of the global burden of disease in 2000 (*1*, *5*).

IAP exposure has multiple technological (e.g., fuel and stove), environmental (e.g., housing characteristics), and behavioral (e.g., energy use behavior and time−location−activity budgets) determinants. Although most indoor air pollutants are products of incomplete combustion, combustion conditions such as temperature, moisture, and air flow are likely to affect the emissions of the various pollutants differently. The spatial dispersion of pollutants inside the house may also vary, depending on whether they are particles or gases and 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). Finally, there may be day-to-day or longer term (e.g., seasonal) changes in emissions of any pollutant or pollutant groups because of changes in energy use patterns or housing conditions.

A number of studies have examined temporal and spatial patterns of IAP, or the relationship between multiple pollutants (*6*−*13*), but none has considered the combinations of spatial, geographical, day-to-day, and seasonal variability for multiple pollutants using comparable measurement methods. An understanding of the patterns of exposure to different mixtures of pollutants is required to design and evaluate interventions. In this paper, we use data collected in mid- and late-heating season (December and March) in multiple households in four poor provinces in China to assess the spatial and temporal distributions of three important indoor air pollutants (respirable particles, carbon monoxide, and sulfur dioxide). The results are being used to generate a baseline profile for the patterns and determinants of exposure to indoor air pollution from household energy use and to design more effective intervention technologies in the world's most populous nation. The implications of findings for interventions are discussed.

Study Location

More than 70% of China's households rely on solid fuels (biomass and coal) for their domestic energy (*1*, *14*). Indoor air pollution caused an estimated 500 000 annual deaths in the developing countries of the Western Pacific region (approximately 85% of the region's population lives in China) in the year 2000, making IAP the fourth leading cause of regional mortality and fifth leading cause of regional disease burden (*1*, *5*). The diversity of factors that affect multiple indoor air pollutants is particularly relevant in China. These factors include climate and geography, housing, locally available fuels and stoves, and socio−cultural factors such

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TABLE 1. Housing Characteristics of Study Populations[a]

	Gansu ($n = 463$)	Guizhou ($n = 476$)	Inner Mongolia ($n = 323$)	Shaanxi ($n = 479$)
Construction Material				
mud, wood, and tile	70.2	41.8	37.5	82.5
brick, wood, and tile	26.8	41.6	37.5	5.8
other	3.0	16.6	25.1	11.7
Location Where Cooking Usually Takes Place				
specialized kitchen[b]	88.8	24.3	54.7[c]	94.6
in bedroom	11.2[d]	4.0	45.3[d]	0.0
in living room	NA[d]	71.7[e]	NA[d]	5.4
Other Characteristics				
houses with gaps between wall and roof	69.5	18.2	11.1	1.7
kitchens with window	90.3	73.8	72.8	60.3
kitchens with ventilation fans	1.3	0.6	14.9	4.8

[a] Numbers show % households. [b] Specialized kitchen is separated by a wall or glass from the sleeping/living room. The two areas have separate entrances in Gansu and are connected with doors in Guizhou and Shaanxi. In Inner Mongolia a specialized kitchen may either have a separate entrance or be connected to the sleeping/living room with a door. [c] Given the changing house construction in Inner Mongolia, a specialized kitchen may be fully or partially separated from the living/sleeping area. In 4.8% of the study households, it was connected to the living/sleeping room with a partial separation wall; in 49.5% of the study households, it was connected to the living/sleeping room with a full separation wall (with a door or glass windows); in 0.4% of the study households, it was outside the house. [d] In Gansu and Inner Mongolia, living and sleeping areas are usually the same, in one room. [e] Most households in Guizhou also have a specialized kitchen with a biomass stove which is generally used for cooking animal feed or for large events (e.g., Spring Festival) (see also Table 2).

as income, food types, and food preparation. Although until the 1980s and 1990s biomass was the dominant source of household energy in China, economic development and deforestation—and policies to reduce and reverse it—have compelled many rural residents to switch to coal (*14*, *15*). China has also implemented an ambitious program to disseminate improved (high-efficiency and low-emissions) stoves (*15*, *16*). A recent evaluation of the program has illustrated that the design and performance of the new stoves was highly variable, with many of the stoves labeled as "improved" lacking flues or other characteristics necessary for reducing exposure (*15*). The study took place in Gansu, Guizhou, Shaanxi, and Inner Mongolia provinces of China. Heating season in all provinces lasts approximately from early November until late March. Winter temperatures are, however, lower in northern, colder provinces and the "intensity" of home heating is higher, particularly in Inner Mongolia; or windows may be open in Guizhou during the heating season but they are closed and sealed in Inner Mongolia. Detailed information on the study provinces, and on selection of study households, is provided in Supporting Information.

Housing. Table 1 provides basic information on housing in the study households. Most houses in Gansu province have a kitchen separated by a wall from the sleeping/living room, with separate entrances. In Guizhou most houses have 2−3 rooms—cooking/living, sleeping, and entrance/storage—connected with doors. Most houses have a separate cooking area, but cooking is done almost entirely in one of the main rooms (cooking/living room), especially during the heating season; the separate cooking area is used primarily for large family events (e.g., Spring Festival) and for making animal feed. Houses in Guizhou have an attic above the cooking/living and sleeping rooms, used for food drying and storage, and at times containing an additional bed. The lower rooms and the attic are separated by a porous ceiling (e.g., made of pieces of wood) which allows air flow between the two levels. Most houses in Shaanxi province have a cooking area connected to the main house by a door, a living room with a ground stove (fire-pit), and one bedroom which sometimes also has a ground stove. Older homes in Inner Mongolia are constructed inside a cavelike structure with a single room used for cooking, living, and sleeping. This room contains a "bed-stove" configuration (a cooking stove connected to a bed for heating). Some homes have an additional room used for sleeping or storage. Newer homes are made with modern construction material with similar structure. The newest homes in the study area have a wall with windows and door between the cooking and sleeping/living areas.

Energy Technology. Table 2 shows the summary of fuel and stove use in the study households. Multifuel use and multistove use are common features of household energy in China because of fuel availability and multiple uses of energy (e.g., cooking and heating) (see also ref *15*). In particular, although coal is the nearly universal fuel for heating in Guizhou and Shaanxi, 18% and 52% of study households in the two provinces, respectively, used biomass as their main cooking fuel.

Methods

Measurement Locations and Times. For each province and for each pollutant, Table 3 shows the number of households and measurement days, measurement points, and time periods. All measurements took place by teams consisting of investigators from the Chinese National Center for Disease Control and Prevention (CDC) assisted by provincial and county CDC staff and health workers. On each day, measurements for all pollutants began in mid- or late-morning and continued until the next day, as close to a full 24-h period as possible.

Measurements took place in two different time periods: March through early April 2003 (late part of the heating season) and December 2003 through January 2004 (peak of the heating season); we refer to these two measurement periods as March and December throughout the paper. Therefore, the results of this study illustrate exposure during the heating season; exposure during summer months is expected to be substantially lower because less energy is used and because windows may be open more frequently. The households monitored in December were a subset of those from March. Repeated measurements were conducted in a small number of households to examine day-to-day variability of pollution.

Due to the cost of measurements, an optimal combination of pollutants and measurement locations was selected to best characterize the exposure conditions of each province, based on a pilot study in January 2003 which examined the relationship between different pollutants and different measurement points (*17*) and a survey on household energy

TABLE 2. Energy Technology in the Study Households (Numbers Reported as % Households unless Otherwise Stated)[a]

	Gansu ($n = 463$)	Guizhou ($n = 476$)	Inner Mongolia ($n = 323$)	Shaanxi ($n = 479$)
Main Cooking Fuel				
coal	1.6	81.4	8.0	48.2
biomass (wood and crop residue)	98.4	17.9	91.7	51.8
liquefied petroleum gas (LPG)	0.0	0.1	0.3	0.0
biogas	0.0	0.6	0.0	0.0
Main Heating Fuel				
coal[b]	21.7	97.3	90.8[c]	97.4
biomass (wood and crop residue)[b]	78.3	2.7	98.7[c]	2.5
LPG and biogas	0.0	0.0	0.2	0.1
Commonly Used Cooking Stove(s)[d]				
biomass stove[e]	98.8	40.8	95.2	66.1
coal stove[f]	2.7	75.5	44.0	57.1
fire pan[g]	15.4	0.2	7.9	0.1
open fire	8.0	0.3	0.6	0.1
other	0.1	0.6	1.1	0.1
Commonly Used Heating Stove(s)[d]				
biomass stove[h]	42.5	7.2	91.8	4.3
coal stove[f]	32.6	94.4	8.2	96.7
fire pan[g]	35.8	0.7	0.4	0.1
open fire	6.3	0.7	0.4	0.3
other	3.0	0.9	1.1	0.3
Ventilation Characteristics of Wood Stoves[i]				
with flue	96.7	30.3	97.1	11.6
flue going out of the house	94.4	18.4	94.4	6.7
flue higher than eave	22.8	8.6	82.6	6.1
Ventilation Characteristics of Coal Stoves[i]				
with chimney	48.4	91.6	97.2	2.7
chimneys going out of the house	46.0	14.5	93.9	2.3
chimney higher than eaves	5.6	6.7	63.5	1.4

[a] See Figure S-2 in the Supporting Information for stove images. [b] The types of coal and combustion technologies are different in Guizhou and Shaanxi. Most of the coal used by the study households in Guizhou is bituminous coal and/or anthracite, obtained from surface exposures (*21*). In parts of the province, which include the study region, these coals have undergone mineralization, and contain potentially toxic trace elements such as arsenic and/or fluorine (*21*). In Shaanxi, most of the coal used for household energy is stone-coal (also called bone-coal), with high concentrations of sulfur, and in some locations fluoride and/or arsenic. Crop residues in each province were from the local crops (see Table S-1 in the Supporting Information for the main food staples) or from smaller tree branches and leaves. [c] Biomass is the primary fuel in Inner Mongolia and is used during the day for cooking. The bed-stove configuration is the most prevalent method of combustion for heating, in which biomass fuel is used. At the same time, many households add coal to their biomass stove during the night, because coal burns more slowly, and hence continues heating for a longer time. [d] Some households use more than one stove for heating or cooking, hence the numbers add to more than 100%. [e] Biomass cooking stoves in Guizhou and Shaanxi are brick or clay stoves. Most biomass stoves are "unimproved stoves" (i.e., without a chimney or with a chimney that does not go outside the house or beyond the eave) (see also ref *15*) for a discussion of "improved" and "unimproved" stoves in China). Biomass cooking stoves in Gansu are similar but larger. Most biomass stoves are without a chimney or with a chimney that does not go outside the house or beyond the eave. In Inner Mongolia, a cooking biomass stove is made from brick or clay and also connected to the bed acting as a bed-heating stove. [f] In Guizhou more than 60% of the coal stoves are made from a simple, enclosed metal container with limited insulation and no door, and the remaining less than 40% are iron-made "air circular" stoves. Most coal stoves in Guizhou have a chimney, but few have a chimney that goes outside the house and beyond the eave. In Shaanxi, coal stoves for heating are built underground. An unimproved coal stove is an underground stove without a chimney, and an improved coal stove is the underground stove with a chimney that goes out of the house and above the eave. [g] In Gansu a fire pan is used for either heating or cooking. [h] Biomass heating stove is a bed stove (burning biomass under the bed) with an opening inside or outside the room. [i] All numbers given as a proportion (percent) of all stoves of the specified type.

use behaviors and time−activity budgets (*18*). On the basis of the pilot study, pollutants were measured at 2−3 points in the cooking, living, and sleeping areas which are the main exposure microenvironments. In a small number of homes and for selected pollutants, measurements were made at additional points for comparison.

Respirable Particles (RPM). Respirable particles were measured 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) (*19*). Samples were collected using a 10-mm nylon cyclone equipped with a 37-mm diameter poly(vinyl chloride) (PVC) filter (pore size 5μm supplied by SKC Inc., U.S.A.) at a flow rate of 2.5 l/min. Air was drawn through the cyclone preselectors using battery-operated constant flow pumps (model PCXR8 supplied by SKC Inc., U.S.A.). All pumps were calibrated prior to and after each sampling day using a field minimeter, itself calibrated by a soap bubble meter in laboratory. Pumps were also calibrated in the laboratory after each field exercise using 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 using an analytic microbalance (1/100,000, Sartorius 2004 MP, Germany) 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 using long-term diffusion tubes (manufactured by GASTEC, U.S.A.), with detection ranges of 10−200 or 50−1000 ppm for CO and 2−100 ppm for SO_2.

TABLE 3. Number of Households and Days for Measurement of Different Pollutants

pollutant	March 2003: households with 1 measurement day: no. of households	no. of points[a]	March 2003: households with multiple measurement days: no. of households	no. of days	no. of points[a]	Dec 2003: households with 1 measurement day: no. of households	no. of points[a]	Dec 2003: households with multiple measurement days: no. of households	no. of days	no. of points[a]
Gansu										
RPM[b]	72	2	6	4	2	17	2	6	2–3	2
CO	72	2	6	4	2	17	2	6	2–3	2
SO_2	1	1	2	4	1–2					
Guizhou										
RPM[b]	76	2	7	2–4	2	16	2	6	2–3	2
CO	1	2	7	2–6	1–2	16	2	6	2–3	2
SO_2			7	3–6	2	16	2	6	2–3	2
Inner Mongola										
RPM[b]						49	2	4	3	2
CO						49	2	4	4	2
SO_2						7	2			
Shaanxi										
RPM[b]	75	2	6	4	3	18	2	6	3	3
CO			6	4	3	18	2	6	3	3
SO_2	75	2	6	4	3	18	2	6	3	3

[a] Measurement points were the cooking and living/sleeping rooms in Gansu, living/cooking and sleeping rooms in Guizhou, living and sleeping rooms in Shaanxi, and living/sleeping/cooking room in Inner Mongolia (two points in the same room). There were a small number of additional measurements in the attic, where the chimney ends, in Guizhou (23 for RPM, 80 for CO, and 88 for SO_2). Cooking and living room 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 each province and room with stove, distances to stoves were standardized. In bedrooms without stoves, the distances from beds were standardized. Measurements in households with multiple measurements were taken at the same point in all measurement days. Despite these efforts for standardization, the fact that home arrangements vary, may be the source some of variation in observed pollutant concentrations. Because the aim of this study is to characterize human exposure (vs stove properties), this variability of concentrations is itself a determinant of human exposure because the measurement points were selected based on their potential as indicators of human exposure. [b] Defined as PM_4, particles with a median aerodynamic diameter of 4 μm (see Methods).

Results and Discussion

Figures 1–3 show the distributions and descriptive statistics for the measured pollutants by measurement point, measurement period, and province. [For Figures 1–3, note that if the concentrations of CO and SO_2 were outside the detection range of the tubes (10–200 or 50–1000 ppm for CO and 2–100 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) Gansu (March), 20 measurements for CO (10 in the cooking room and 10 in the living/bedroom); Gansu (December), 6 measurements for CO (3 in the cooking room and 3 in the living/bedroom); (b) Guizhou (March), 0 measurements for CO and 0 measurements for SO_2; Guizhou (December), 0 measurements for SO_2; (c) Shaanxi (March), 5 measurements for CO (0 in the cooking room, 3 in the living room and 2 in the bedroom); Shaanxi (December), 30 measurements for CO (7 in the cooking room, 14 in the living room and 9 in the bedroom); (d) Inner Mongolia (December), 27 measurements for CO (13 in the cooking/living/bedroom point 1 and 14 in the cooking/living/bedroom point 2). (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) Gansu (March), 16 measurements for CO (7 in the cooking room and 9 in the living/bedroom); Gansu (December), 4 measurements for CO (1 in the cooking room and 3 in the living/bedroom); (b) Guizhou (March), 0 measurements for CO and 0 measurements for SO_2; (c) Shaanxi (December), 7 measurements for CO (1 in the cooking room, 2 in the living room and 4 in the bedroom); (d) Inner Mongolia (December), 19 measurements for CO (11 in the cooking/living/bedroom point 1 and 8 in the cooking/living/bedroom point 2). (iii) Those measurements that were lower than the measurement range were set to the minimum value. The number of measurements was (a) Gansu (March), 13 measurements for SO_2 (9 in the cooking room and 4 in the living/bedroom); (b) Shaanxi (March), 3 measurements for CO (1 in the cooking room, 1 in the living room and 1 in the bedroom) and 86 measurements for SO_2 (34 in the cooking room, 1 in the living room and 51 in the bedroom); Shaanxi (December), 21 measurements for SO_2 (11 in the cooking room, 4 in the living room and 6 in the bedroom). The overall results and conclusions of the analysis were not sensitive to this assumption.]

Pollutant Concentrations in Different Provinces. RPM concentrations in all provinces were in excess of health-based standards and guidelines for particulate matter in ambient (outdoor) environment (e.g., the U.S. EPA requires the 24-h mean concentration of $PM_{2.5}$ to be below 65 μg/m^3 and annual mean concentration below 15 μg/m^3). The two provinces where biomass is the primary fuel (Inner Mongolia and Gansu) had the highest RPM concentrations (>700 μg/m^3 at both points in the cooking/living/bedroom in Inner Mongolia; 351–661 μg/m^3 in different rooms and months in Gansu); the higher concentration in Inner Mongolia reflect its colder temperature and longer heating hours, with a stove and housing arrangement that combines heating and cooking. Lower concentrations were observed in the primarily coal-burning provinces of Guizhou and Shaanxi (202–352 μg/m^3 in different rooms and months in Guizhou; 187–361 μg/m^3 in different rooms and months in Shaanxi). RPM concentrations in Guizhou were significantly lower than those measured in the pilot study (*17*) in four homes in a single village. This difference is partly because the pilot measurements were intended for selecting appropriate sampling points and pollutants for the larger study, rather than being from a representative sample households.

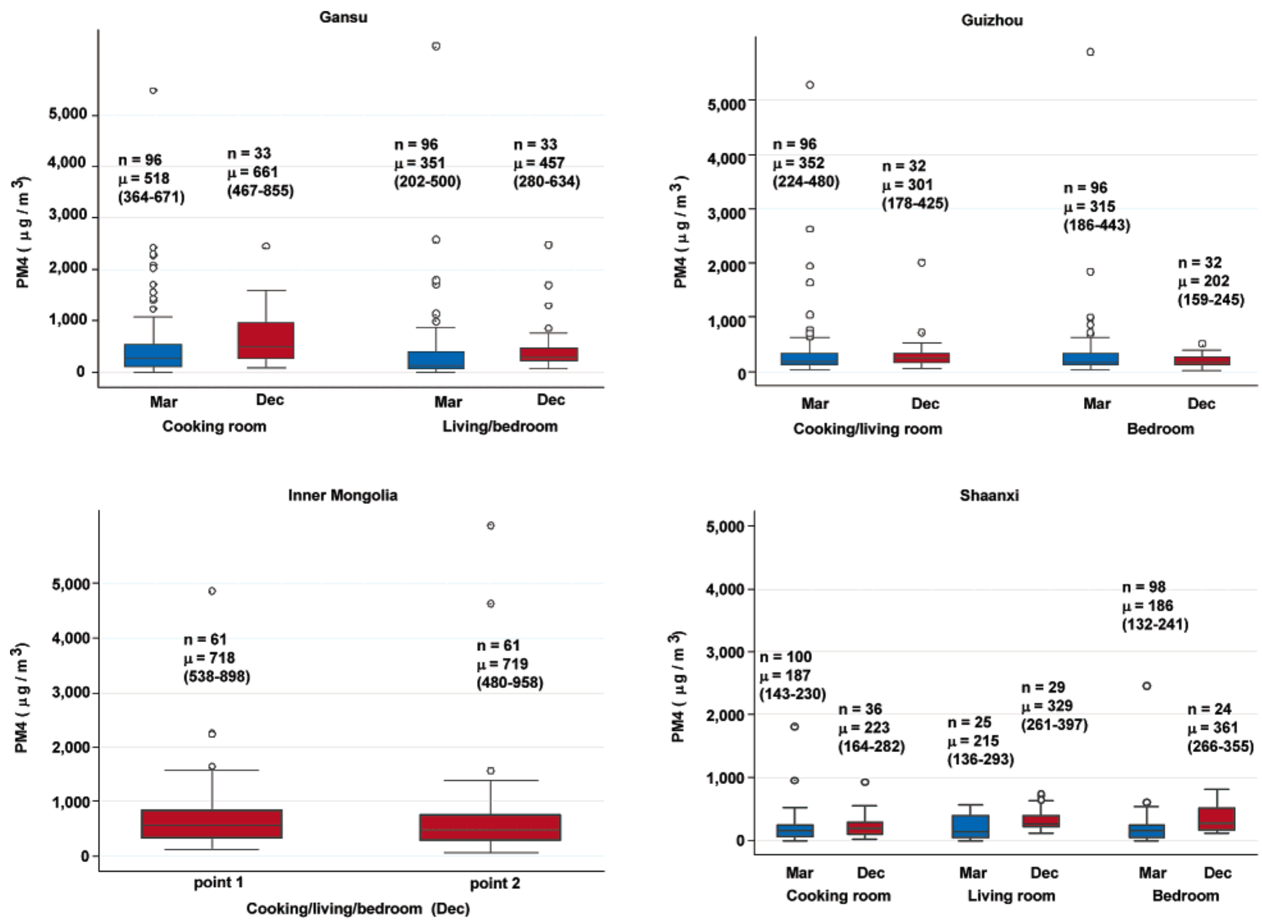

FIGURE 1. Average 24-h concentration of RPM at different measurement points and measurement periods (March and December) in Gansu, Guizhou, Shaanxi, and Inner Mongolia provinces. *n*: number of observations. *μ*: mean. Numbers in brackets give the 95% CI for the mean.

Except for a small number of observations in Gansu, Inner Mongolia, and Shaanxi, 24-h mean CO concentrations were consistently below available health-based standards and guidelines (e.g., WHO guideline values of 10 ppm and American Conference of Governmental Industrial Hygienists, ACGIH, guideline value of 25 ppm for 8-h exposures), and in some cases CO concentrations were close to the detection limits of the diffusion tubes. Since these were 24-h concentrations, concentrations may have been higher during cooking or when doors and windows were closed at night, not observable in our data. Guizhou had the lowest CO concentrations (<2 ppm), partly due to the specific type of coal and stove, and partly because the attic, where the chimney ends, provides ventilation for removing CO from the house (the chimney and the porous attic provide better ventilation for CO which is gaseous compared to particles). Mean 24-h concentrations were 3.7−13.8 ppm at difference points and months in the attic.

Due to cost, we measured SO_2 only in the two coal-burning provinces of Guizhou and Shaanxi, where detectable concentrations were expected based on the result of the pilot study (*17*) (a small number of measurements in Gansu showed significantly lower concentrations of SO_2; in Inner Mongolia SO_2 concentrations in a small number of measurements were comparable to those in Shaanxi and Guizhou because coal was used for heating at night). SO_2 concentrations were higher than the WHO guideline value of 0.04 ppm at all locations in both provinces. SO_2 concentrations were substantially 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 had a mean 24-h concentration of 0.97−1.44 ppm compared with 0.16−0.20 ppm in the cooking/living room in Guizhou). Higher SO_2 concentrations in Shaanxi are likely due to both the type of coal and the use of a chimney in Guizhou (mean 24-h SO_2 concentrations in the attic in Guizhou were higher than those in Shaanxi, 1.08−3.40 ppm at difference points and months).

Pollutant Concentrations at Different Measurement Points. The concentrations of pollutants in different rooms were generally determined by whether a stove was used in that room for cooking versus heating and by housing characteristics that affect dispersion. The role of these factors also varied across pollutants. In Gansu, the cooking room had a higher RPM concentration than the bedroom (518 vs 351 $\mu g/m^3$ in March and 661 vs 457 $\mu g/m^3$ in December; the differences between the two points were not significant at $p = 0.05$). CO concentrations, however, did not show the same consistent ordering, and the relative levels in the two rooms changed in March and December. If outlier measurements (>99th percentile) are dropped, the mean CO concentrations in March were 5.5 and 4.8 ppm in the cooking room and bedroom, respectively; in December they were 8.4 and 9.3 ppm. The reason for this differential pattern of the two pollutants may be that shorter periods of more intense combustion, intermittent with longer periods of no combustion, in the cooking room result in similarly high emissions of RPM and CO. In the bedroom, the heated bed contains hot ashes for many hours with closed windows, and RPM emissions are lowered more than CO. As a result RPM concentrations are consistently higher in the cooking room than the bedroom, but those of CO are comparable between the two rooms.

In Guizhou, the concentrations of all pollutants 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 only slightly lower than, the cooking/

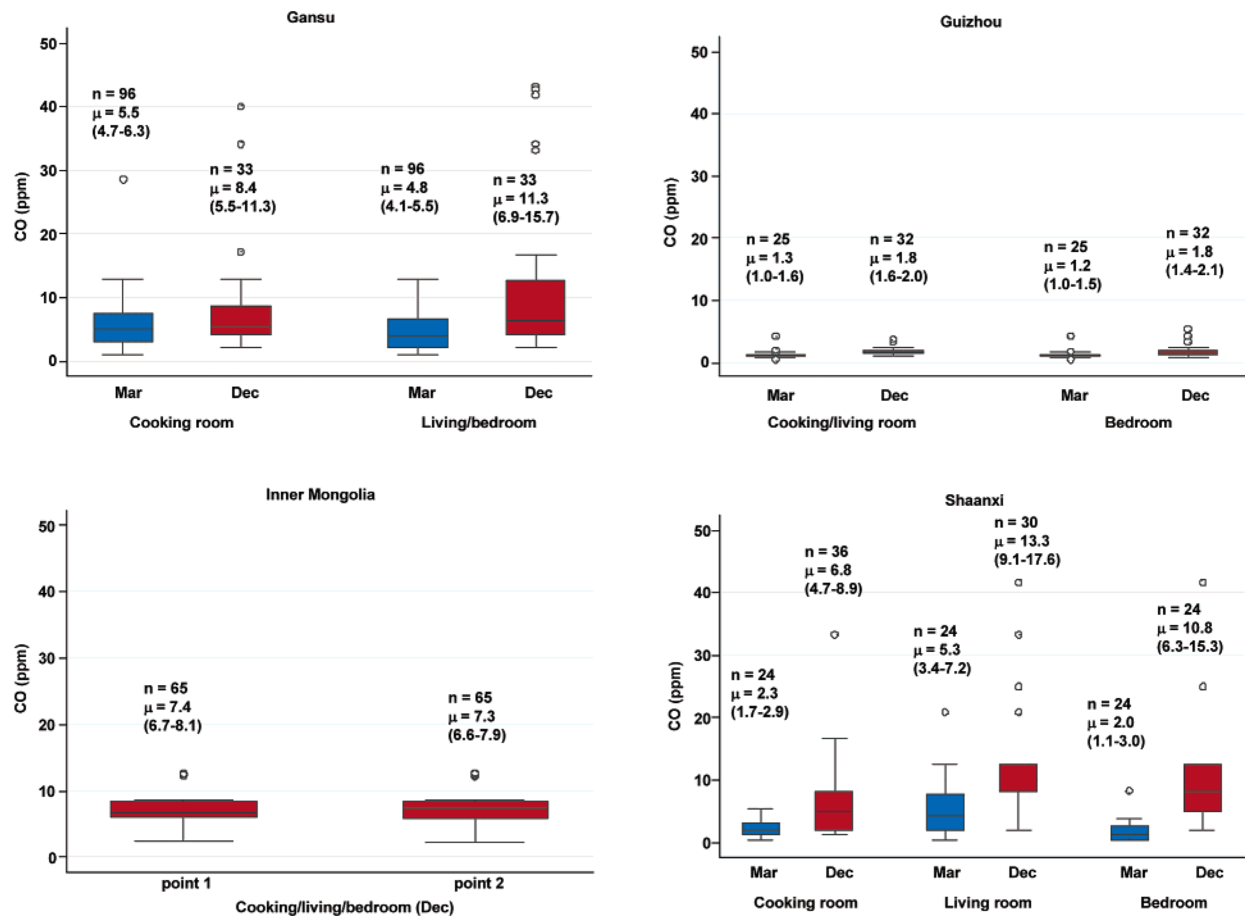

FIGURE 2. Average 24-h concentration of CO at different measurement points and measurement periods (March and December) in Gansu, Guizhou, Shaanxi, and Inner Mongolia provinces. *n*: number of observations. *μ*: mean. Numbers in brackets give the 95% CI for the mean.

living room where the stove was located. This result illustrates 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 exposure microenvironment. The small number of measurements in the attic shows that it consistently had the highest concentrations of all three pollutants (RPM, 453−649 $\mu g/m^3$; CO, 3.7−13.8 ppm; SO_2, 1.08−3.40 ppm in different months). Therefore, although the attic, where the chimneys ended, 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 Inner Mongolia, cooking and heating take place in the same room, making it the main exposure microenvironment. In Shaanxi, 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 the differential patterns is that biomass was the primary cooking fuel, and was used for a shorter time, resulting in high RPM concentration but limited contribution to CO and SO_2. In the heated living room on the other hand, coal with a high sulfur concentration was used for longer durations, therefore resulting in higher emissions of CO and SO_2. Pollution in the bedroom was determined by a combination of direct emission and dispersion from other locations (a stove was used in 9 out of 24 household-days of measurement in the bedroom in December; 89 out of 98 household-days of measurement in the bedroom in March) and was therefore highly dependent on household-specific characteristics. 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.

Pollutant Concentrations in Mid- and Late-Heating Season. The data in this study did not provide evidence on differences between pollution levels in mid- and late-heating seasons (December vs March) that are generalizable across provinces. In Gansu and Shaanxi, the concentrations of all measured pollutants were higher in December than in March, at all measurement points (Figures 1−3) (many of the differences were not statistically significant at $p = 0.05$). In Guizhou, RPM concentrations were nearly equal in December and March (352 vs 301 $\mu g/m^3$) in the cooking/living room; bedroom concentrations in December were substantially, and significantly, lower than March (202 vs 315 $\mu g/m^3$) (without outliers (>99th percentile) the concentrations in the two seasons would be 202 vs 256 $\mu g/m^3$). Concentrations of CO and SO_2 were however higher in December than in March in both the cooking/living and bedroom. The more consistent pattern of mid- and late-heating season in Gansu and Shaanxi may be because in both provinces energy is used for home heating in winter. Although heating is also important in Guizhou, winter in Guizhou is characterized with high humidity, creating a need for keeping stored food dry (*17*, *18*). Therefore, warmer temperatures in March may have systematically resulted in lower energy use (and pollution) in the living rooms in Shaanxi and Gansu but not those in Guizhou (Table 4; the differences are not statistically significant). Because of the logistics of the project initiation, data collection in Inner Mongolia began only in December 2003, and hence no similar measurements from March were available for comparison.

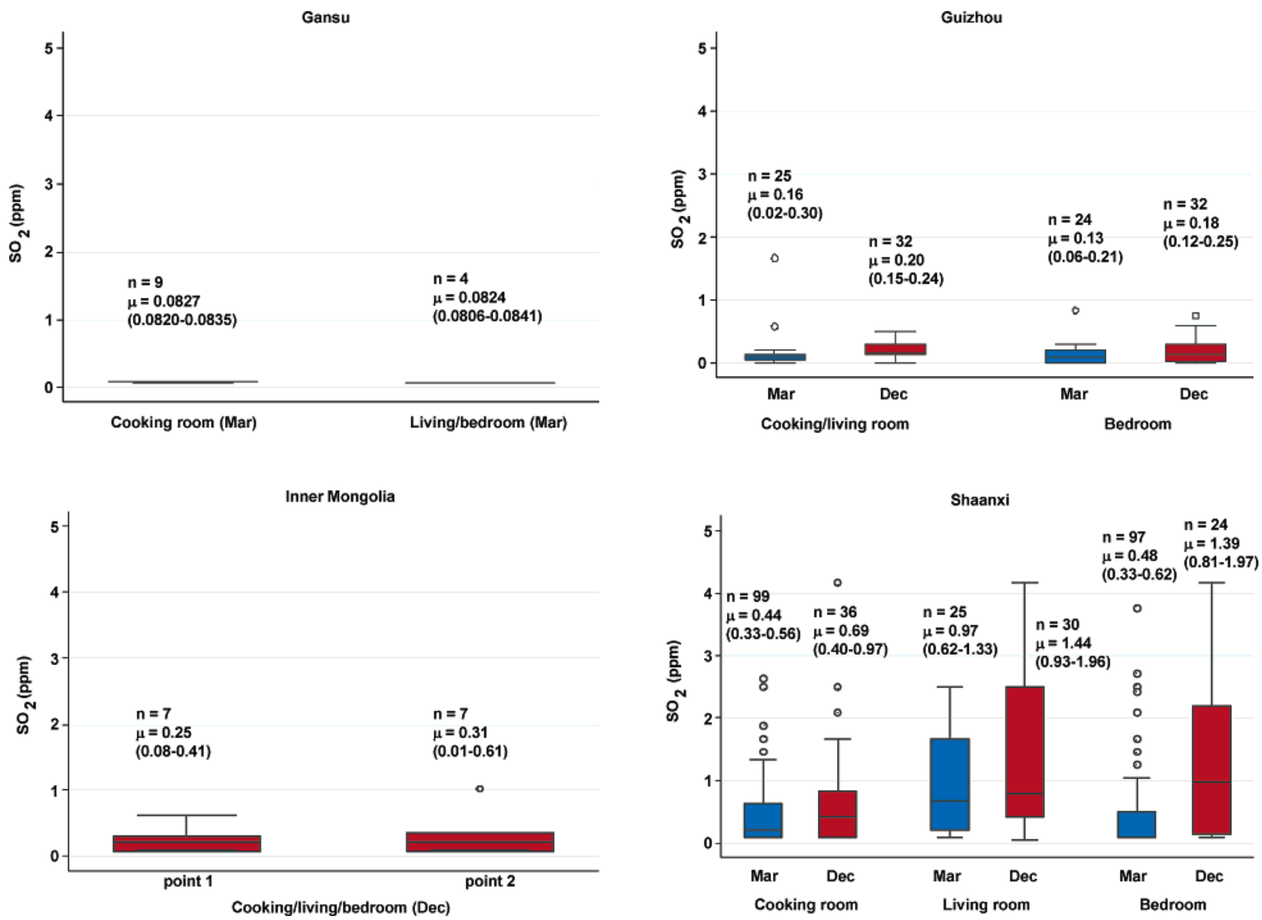

FIGURE 3. Average 24-h concentration of SO_2 at different measurement points and measurement periods (March and December) in Gansu, Guizhou, Shaanxi, and Inner Mongolia provinces. *n*: number of observations. *μ*: mean. Numbers in brackets give the 95% CI for the mean.

TABLE 4. Number of Hours of Stove Use in Mid- and Late-Heating Season (December and March)[a]

	mean hours stove used (95% CI)	
	March	Dec
Gansu		
cooking room	3.0 (2.7−3.3) ($n = 96$)	2.7 (2.1−3.4) ($n = 33$)
living/bedroom	2.3 (1.2−3.5) ($n = 96$)	4.1 (1.5−6.8) ($n = 33$)
Guizhou		
cooking/living room	16.5 (15.8−17.2) ($n = 96$)	15.3 (14.0−16.5) ($n = 32$)
Inner Mongolia		
cooking/living/bedroom		7.3 (6.1−8.4) (n = 65)
Shaanxi		
cooking room	8.4 (6.6−10.3) ($n = 100$)	9.6 (6.5−12.6) ($n = 36$)
living room	16.8 (13.7−19.9) ($n = 25$)	18.2 (16.1−20.3) ($n = 30$)
bedroom	8.7 (6.7−10.7) ($n = 98$)	6.8 (2.8−10.8) ($n = 24$)

[a] *n* shows the number of household-days of observation.

Pollutant Concentrations across Multiple Measurement Days. We examined day-to-day variation in pollution using data from those households with multiple measurements in each province and in each monitoring period (Table 3). For each province and each measurement point, Table 5 shows minimum and maximum concentrations in the multiple measurement days, calculated separately for each household with multiple measurements and then averaged over all such households. Table 5 also shows the coefficient of variation (defined as standard deviation divided by mean) for multiple measurements in each household, calculated separately for each household and then averaged over all such households. Table 5 shows that, on average, pollutant concentrations varied by a factor of 2−10 across measurement days, for different pollutants, measurement points, and provinces. Standard deviations of multiple measurements in the same household varied between 10% and 100% of their mean. With the exception of SO_2 in Shaanxi, the variation was consistently smaller in December than in March, possibly because longer hours of stove use in December reduced heterogeneity across different measurement days.

Because of measurement difficulty and cost, most epidemiological studies rely on indirect indicators of exposure (e.g., fuel, stove, energy use behaviors, and time−activity budgets) or a small number of actual measurements of pollution and/or exposure. It is therefore important to compare the variability of pollution across households (i.e., interhousehold variation) with variability across different

TABLE 5. Minimum and Maximum Concentrations and Coefficient of Variation in Households for the Multiple Measurement Days, Averaged over All the Households[a]

RPM	min−max (*n*) March	min−max (*n*) Dec	coefficient of variation (*n*) March	coefficient of variation (*n*) Dec
Gansu				
cooking room	127.4−984.9 (6)	340.8−831.5 (6)	0.8 (6)	0.4 (6)
living/bedroom	76.4−585.6 (6)	180.0−382.9 (6)	0.8 (6)	0.4 (6)
Guizhou				
cooking/living room	79.2−303.2 (6)	191.8−551.6 (6)	0.5 (6)	0.4 (6)
Inner Mongolia				
cooking/living/bedroom		600−1867.8 (4)		0.4 (4)
Shaanxi				
cooking room	45.8−525.1 (6)	102.8−220.8 (6)	0.8 (6)	0.4 (6)
living room	87.5−363.9 (6)	204.2−501.4 (6)	0.9 (6)	0.4 (6)
bedroom	83.3−287.5 (6)	247.2−448.6 (6)	0.7 (6)	0.3 (6)

CO	min−max (*n*) March	min−max (*n*) Dec	coefficient of variation (*n*) March	coefficient of variation (*n*) Dec
Gansu				
cooking room	3.9−10.2 (6)	5.5−8.0 (6)	0.4 (6)	0.2 (6)
living/bedroom	2.2−5.5 (6)	10.1−17.7 (6)	0.4 (6)	0.4 (6)
Guizhou				
cooking/living room	0.8−2.1 (6)	1.5−2.2 (6)	0.4 (6)	0.2 (6)
Inner Mongolia				
cooking/living/bedroom		5.8−11.4 (4)		0.3 (4)
Shaanxi				
cooking room	1.3−3.8 (6)	5.1−10.8 (6)	0.6 (6)	0.3 (6)
living room	2.5−9.0 (6)	14.2−17.4 (6)	0.5 (6)	0.1 (6)
bedroom	1.1−3.8 (6)	5.3−15.1 (6)	0.4 (6)	0.4 (6)

SO_2	min−max (*n*) March	min−max (*n*) Dec	coefficient of variation (*n*) March	coefficient of variation (*n*) Dec
Guizhou				
cooking/living room	0.0−0.4 (6)	0.1−0.2 (6)	1.0 (6)	0.5 (6)
Shaanxi				
cooking room	0.3−0.7 (6)	0.4−0.8 (6)	0.3 (6)	0.4 (6)
living room	0.6−1.4 (6)	0.9−1.5 (6)	0.6 (6)	0.3 (6)
bedroom	0.1−0.4 (6)	0.7−1.5 (6)	0.3 (6)	0.4 (6)

[a] *n* is the number of households with multiple measurements (see also Table 3).

measurement days (interday variation) in the same household. For households with multiple measurements, Table 6 shows the ratio of proportion of variance explained by interhousehold variation to interday variability. In all cases, except CO in the cooking/living room in Guizhou in March, there was more variation in pollution across households than within households. This result indicates that the duration of stove use, quantity of fuel, ventilation, and stove use behavior that determine pollutant concentrations are likely to vary more across households than from day to day in individual households.

Implications for Interventions. In China, where rapid economic growth and infrastructure expansion have contributed to near-universal access to electricity for lighting (*20*), more than 70% of households continue to use coal or biomass as their primarily fuel for cooking and heating (*1*). Because rapid transition to clean fuels is not a feasible short-term option, there is a need for interventions that lower emissions by modifying current fuel−stove−housing−behavior combinations. Understanding exposure routes will inform the design of more effective interventions. Some of the main exposure paths in these four Chinese provinces are summarized below:

• In households with separate cooking and living/sleeping areas, and distinct cooking and heating stoves (most households in Gansu and Shaanxi), the cooking stove is a source of exposure throughout the year for women, and their young children, who may spend time near their mothers. The duration of exposure from cooking is a few hours per day, and stove improvement alone can reduce exposure. Even in these households, smaller cooking tasks (e.g., making tea) take place in the living area, for example on a fire pan or ground stove, which results in IAP exposure for all household members. In these households heating is a source of exposure, possibly to a larger extent than cooking, for all household members in winter. Therefore, reducing exposure requires improvements in the heating stove above and beyond that used for cooking.

• In households where cooking and living areas are the same, and there is no physical distinction between cooking and heating stoves (most households in Inner Mongolia), cooking is a source of exposure throughout the year for all household members. In winter, heating requires stove use, and hence causes exposure, for a period as long as or longer than cooking. In principle, stove improvement can reduce indoor concentrations and exposure. Stove improvements may also have to be accompanied with housing changes that separate the main stove body from the living area (i.e., create a specialized kitchen). With this alternative housing design,

TABLE 6. Proportion of Variance in Pollutant Concentration Explained by Interhousehold Variability, Relative to the Proportion Explained by Interday Variability in the Same Households[a]

	RPM		CO		SO_2	
	March	Dec	March	Dec	March	Dec
Gansu						
cooking room	5.0	10.0	3.8	36.9	0.1	
living/bedroom	2.8	10.3	11.8	8.9	0.0	
Guizhou						
cooking/living room	1.6	2.5	0.5	3.8	2.6	9.3
Inner Mongolia						
cooking/living/bedroom		1.9		4.0		
Shaanxi						
cooking room	1.2	19.2	10.9	12.3	28.3	8.3
living room	20.1	3.8	2.1	113.9	19.7	344.9
bedroom	2.7	22.4	8.0	9.2	6.2	35.0

[a] The ratio is for the fraction of variances explained by each variable *alone*, obtained using sequential analysis-of-variance, ANOVA.

the stove's heating function is performed only by heat transfer to the bed.

• In most households in Guizhou, a separate cooking area and stove exist, but it is used only occasionally. Most cooking takes place in the living area, combined with heating in winter. These households in practice have the same exposure patterns as those with combined cooking/living areas and cooking/heating stoves. The stove in Guizhou can be improved without housing changes. Because the porous ceiling between the ground floor and the attic allows pollutant dispersion back into the main floor, stove improvements must be accompanied with increased chimney length to limit the dispersion of pollutants inside the house and reduce exposure.

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Supporting Information Available

Study provinces, characteristics of study provinces, demographic and economic characteristics of the study households and respondents, and photographs of stoves from the four study provinces. This material is available free of charge via the Internet at http://pubs.acs.org.

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