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Household CO and PM measured as part of a review of China's National Improved Stove Program

Abstract In 2001–2003, a team of researchers from the United States and China performed an independent, multidisciplinary review of China's National Improved Stove Program carried out since the 1980s. As part of a 3500-household survey, a subsample of 396 rural households were monitored for particulate matter less than 4 μm (PM_4) in kitchens and living rooms over 24 h, of which 159 were measured in both summer and winter. Carbon monoxide was measured in a 40% subsample.The results of this indoor air quality (IAQ) component indicate that for nearly all household stove or fuel groupings, PM_4 levels were higher than – and sometimes more than twice as high as – the national PM_{10} standard for indoor air (150 μg PM_{10}/m^3). If these results are typical, then a large fraction of China's rural population is now chronically exposed to levels of pollution far higher than those determined by the Chinese government to harm human health. Further, we observed highly diverse fuel usage patterns in these regions in China, supporting the observations in the household survey of multiple stoves being present in many kitchens. Improved stoves resulted in reduced PM_4 from biomass fuel combinations, but still not at levels that meet standards, and little improvement was observed in indoor pollution levels when other unimproved stoves were present in the same kitchen. As many households change fuels according to daily and seasonal factors, resulting in different seasonal concentrations in living rooms and kitchens, assessing health implications from fuel use requires longitudinal evaluation of fuel use and IAQ levels, combined with accurate time-activity information.

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Key words: Indoor air pollution; Particulate matter; Carbon monoxide; Seasonal change in fuel use; mixed fuels.

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Practical Implications

Leaving aside the difficult issue of enforcement, it is uncertain whether Chinese household IAQ standards represent realistic objectives for current attainment given current patterns of energy consumption in rural China, which rely so heavily on unprocessed solid fuels. Even when used with chimneys, these fuels emit substantial pollution into the household environment. It is probable that low-emission technologies involving gaseous/liquid fuels or high combustion - efficiency biomass stoves need to be promoted in order to achieve these standards for the greater part of the population.

Introduction

China released a revision of it's health-based air quality standards in November 2002, to include the first set of standards governing indoor air quality (IAQ) in residences and offices (GB/T 1883–2002), which went into effect March 1, 2003. Among the pollutants covered, the standard for particulate matter (PM) less than 10 μm (PM_{10}) is 150 $\mu g/m^3$ for a 1-day average and the standard for carbon monoxide (CO) is 10 mg/m^3 for a 1-h average. These levels correspond to Class II (residential areas) ambient standards (GB 3905–1996). These standards represent a landmark in being among the first in the world to address IAQ inside residences[1].

These IAQ standards were promulgated just as the IAQ measurements reported here were being completed as part of an independent, multidisciplinary review[2] of China's improved rural household stove programs since the 1980s. This review follows from an initial review by Smith et al. (1993). Overall, nearly a billion rural Chinese citizens have benefited from improved fuel efficiency because of the improved stoves promoted from the 1980s through the 1990s by the China's National Improved Stove Program (NISP), the largest and most successful improved stove program ever implemented anywhere in the world, and similar successful programs initiated at the provincial and local levels (International Workshop, 2005). Although NISP was focused mostly on improving fuel efficiency, as only biomass stoves with chimneys were disseminated, it can be expected that IAQ was improved as well. Chinese studies in those periods showed that quite high indoor levels were common in homes using older technology (Sinton et al., 1995).

Thus, as part of the NISP review, we conducted a modest assessment of IAQ to evaluate the extent that improved biomass stoves lowered indoor air pollution, the effects other fuel and stove combinations in common usage had on IAQ, and whether improved stoves and similar technologies can be used as a practical measure in rural communities to conform to health-based standards.

The survey of indoor air pollution in association with the review of NISP also informs on the use of fuel use surveys and CO dosimeter tubes as indirect proxies of exposure in studies of health outcomes. Most epidemiologic investigations of health outcomes as a result of indoor solid fuel use have used indirect proxies for exposure (cooking fuel or fuel type). While there are model-based approaches that have been developed in India which allow better estimation based on incorporation of ventilation and other household-based characteristics (Mehta, 2002), these are not widespread and the use of indirect indicators will likely remain common in epidemiologic studies given current technologies. There are promising developments of cheap lightweight particle monitors; however, that are suited to this purpose that would allow longitudinal evaluation of the relationship with health effects (Edwards et al., 2006). Those studies that have measured indoor concentrations of PM have usually measured concentrations for 1 day during a single time period of the year. Studies that have used CO as a proxy for PM exposure have been successfully demonstrated in regions that use predominantly biomass fuels (Naeher et al., 2001), but have not been fully evaluated in situations with diverse fuel usage. Therefore, in the current article, we also examine the implications of the large number of stove fuel combinations that were encountered during the Indoor Air Pollution (IAP) assessment for health assessments.

Research methods

Sample selection

Three provinces, Zhejiang, Hubei, and Shaanxi, were selected for study to represent high, medium, and low adoption rates of improved stoves and improved fuels and a range of income levels. We undertook a clustered random household survey of about 3500 households in five counties of each province that included indirect measures of IAQ, health, stove performance, and questions on a range of socioeconomic factors (Sinton et al., 2004a). Because of resource and logistics limitations imposed by the amount of equipment carried by the monitoring team, IAQ monitoring was restricted to one or two counties per province.

Indoor air quality-monitoring data were collected in two phases. The first phase of fieldwork was in June to August 2002, and the second in December 2002 to January 2003. In the first phase, villages were chosen to ensure that the entire range within the household sample of stove, fuel, and ventilation conditions were represented. In Zhejiang, seven villages in two counties were selected, in Hubei eight villages in two counties were selected, and in Shaanxi eight villages in one county were selected. Within most villages, 12 households (with a low of 6 and a high of 18) were randomly selected among those who had been administered the household survey. As there is little winter heating in Zhejiang and therefore relatively small change in fuel use patterns, the second phase of monitoring was restricted to Hubei and Shaanxi, where stove and fuel use in winter is very different than in the summer. In all, 288 households were selected in the summer (98 in Zhejiang, 94 in Hubei, and 96 in Shaanxi), and 267 were selected in the winter (131 in Hubei and 136 in Shaanxi). One hundred and fifty-nine households were measured in both summer and winter seasons in Hubei and Shaanxi provinces. All households were monitored for PM less than 4 μm (PM_4) for a 24-h period, and household members answered a short questionnaire. In each village ambient PM_{10} was monitored for 24 h. Because of equipment constraints, only two of the households being monitored for PM_4 in each village (40% of homes) were selected for continuous CO monitoring for 24 h.

[1]Although not standards, Canada has had residential IAQ guidelines for many years - Canada Department of National Health and Welfare. Exposure Guidelines for Residential Indoor Air Quality. Ottawa. April 1987

[2]The objectives were to evaluate the methods used to promote improved stoves, to assess the development of commercial stove production and marketing organizations, and to measure the household impacts of the programs through surveys of health, stoves, and indoor air quality (Sinton et al., 2004). In this paper we discuss the indoor air quality portion of the study

Given the range of conditions found in China, this household survey does not provide a statistically representative picture of the national status of stoves or IAQ. Rather this study informs on typical situations in three important provinces at different levels of income and climate. Unexpected variability in numbers, types, and conditions of fuels and stoves, patterns of use, dwelling configurations, and ambient environments encountered in the survey prevent drawing strong conclusions about the IAQ implications of all fuel types encountered.

Sampling methods

Particulate matter. PM_4 was selected as the particulate measure as almost all the particle mass from residential combustion of solid fuels is contributed by particles less than 1 μm in diameter As the objectives were to assess indoor concentrations from residential fuel combustion, measurement of PM_4 would capture the majority of emissions from residential fuel combustion and allow comparison of indoor concentrations generated by residential fuel use to indoor air standards. PM_4 samples were collected on 5.0 μm pore size GLA 5000 PVC filters (SKC Inc., Eighty Four, PA, USA) with a GS-3 lightweight conductive plastic cyclone (SKC Inc.) that meet the ACGIH/ISO/CEN respirable curve with a 50% cut-point of 4.0 μm (bias within ISO/NIOSH requirements) at 2.75 l/min using standard calibrated PCXR8 air pumps (SKC Inc.). Flow rates were calibrated pre- and post-sampling using a 0.4–5.0 lpm rotameter (320 4A5, SKC Inc.), which was calibrated using a bubble flow meter primary standard [Chinese Air Pollution Methods (CAPM), China]. Filters were equilibrated for 72 h and weighed on a Mettler AE 260 delta range balance in an environmentally controlled weigh room. Temperatures during weighing were 20°C ± 2°C and relative humidities were 20% ± 3%. The number of field blanks was equivalent to 5% of the total number of filters collected. Uncertainties for PM_4 were ± 25 μg/m^3 based on blank measurements. Although measurements were nominally for 24 h, the logistics of shifting equipment between households dictated that most were somewhat less. In the summer, average sampling times were 22.1 ± 1.1 h. In winter, sampling times were 22.4 ± 1.2 h. In no case, however, was the total time less than 20 h.

Carbon monoxide. Carbon monoxide concentrations were monitored using HOBO® CO electrochemical sensors with logging ability (Onset computer corporation Pocasset, MA, USA). As individual monitors respond slightly differently to CO concentrations, CO monitors were compared with four concentrations (0.5, 10, 25 and 60 ppm) of calibration reference standards (Scott Specialty Gases, Philadelphia, PA, USA) prior

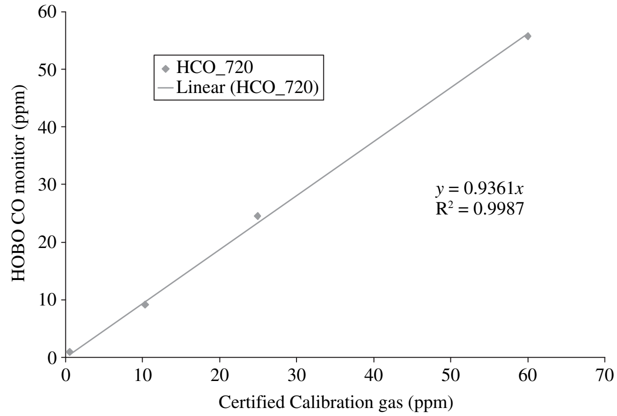

Fig. 1 Example of carbon monoxide electrochemical sensor reference comparison

to use and post-sampling to derive adjustment factors to correct to laboratory certified concentrations. Calibration gas was pumped in at 500 ml/min for 3 min through a rubber cup placed over one of the HOBO ports and immediately sealed with tape. The subsequent 2 min of 1-s CO concentrations were then averaged and compared with calibration gas concentrations. HOBO CO monitors employed in this study were well correlated to reference gas standards with average r^2 of 0.997 ± 0.002 with a slope of 0.987 ± 0.06 compared with calibration standards. Figure 1 shows an example comparison against the reference gases. A similar procedure was followed at the end of the sampling period to ensure linearity. HOBO CO statistics were then adjusted by the response factors derived from the gas standards to adjust to laboratory certified concentrations. Uncertainties were ± 0.44 ppm in mean CO based on side-by-side tests.

Carbon monoxide dosimeter tubes. Standard Gastec CO diffusion tubes, 1DL with a range of 0.4–400 ppm, were used for 24 h measurements for CO in living rooms and kitchens. Dosimeter tubes were capped and an initial reading of the stain taken on removal from the sampling site using a standard white background and ruler with millimeter scale. The length in millimeters was then used with a standard calibration curve to calculate the resultant concentration in ppm. Subsequently, the reading was independently confirmed in the field office by the field manager who read all tubes. Prior to sampling field teams were trained to standardize the reading of endpoints for the stain.

Breath carbon monoxide. Carbon monoxide breath analysis was performed with small (16.5 cm × 6.35 cm × 5.08 cm), lightweight (159 g) Micro CO breath analyzers (Micro Direct, Inc., Lewiston, ME, USA). The instruments feature fast response time, 1 ppm resolution and a range of 0–500 ppm within an operating range of 0–40°C. Reported hydrogen cross-sensitivity is <3% and sensor drift is <2% per month.

Questionnaire. A questionnaire was administered at the end of monitoring to ascertain pollution-related household activities during the monitoring period such as household fuels used, main fuel used for cooking, main fuel used for heating, smoking, garbage burning, socioeconomic variables, stove characteristics and performance, energy use and cost, and housing characteristics. A draft of the household survey instrument was administered during a pilot study in a rural area near Tianjin, and revised based on initial experience.[3]

Results and discussion

Household and province characteristics

The provinces typify the range of conditions found in China in a number of respects. The three provinces together have 11% of the nation's rural population. Average household size in Hubei (3.9 persons) and Shaanxi (4.0 persons) was slightly larger than the national average of 3.82 persons, while in Zhejiang it was significantly smaller (3.23 persons). Annual incomes diverge widely among the three provinces. In Zhejiang, average per capital annual income in 2001 was nearly twice the national average of 2366 yuan (US$286 in 2001) per person, while in Hubei it was almost exactly average, and in Shaanxi it was one-third below the average. In Zhejiang, average floor area per person was 47.8 m^2 per person, about twice the national average, and the amount constructed in concrete and steel was about three times the national average. In contrast, households in Hubei was somewhat above (31.19 m^2 per capita) and Shaanxi somewhat below (23.76 m^2 per capita) the national average for floor area (25.73 m^2 per capita). Although nearly 25% of rural households in Zhejiang owned gas or electric water heaters (for supplying hot water for washing), the proportion was only around 2% in Hubei and Shaanxi (Sinton et al., 2004).

In the subset of homes where indoor air pollution was monitored, almost all homes in Zhejiang had indoor kitchens, of which 70% had a full partition from the living area, and 30% were un-partitioned. In Hubei, 49% had an indoor kitchen including 30% with a full partition from the living area and 50% of homes had a separate indoor kitchen outside the home. Homes in Shaanxi were quite different and 17% had an outdoor kitchen and 62% had a separate indoor kitchen outside the home and only 20% had an indoor kitchen including 6% with a full partition from the living area. Median kitchen volume was 37 m^3 in Zhejiang, 44 m^3 in Hubei and 36 m^3 in Shaanxi.

Mean high and low July temperatures in the three regions as measured in three nearby cities to the monitoring areas (Xian, Wuhan and Ningbo) are somewhat similar (Shaanxi -mean high 33°C mean low 22°C; Hubei – mean high 33°C mean low 26°C; and Zhejiang – mean high 31°C mean low 24°C), but in winter Zhejiang tends to be warmer (mean high 12°C mean low 5°C) followed by Hubei (mean high 10°C mean low 2°C), with Shaanxi experiencing the coldest winters (mean high 6°C mean low −3°C).

Particulate matter and carbon monoxide comparisons between provinces

The PM_4 concentrations in 75% of kitchens and 73% of living rooms during the winter exceeded the PM_{10} standard of 150 $\mu g/m^3$ for a 24-h average. During the summer, 48% of kitchens and 46% of living rooms exceeded the standard. In both seasons, 62% of kitchens and 59% of living rooms exceeded the standard. If PM_{10} and not PM_4 had been measured, a greater fraction of homes would have exceeded the standard in both seasons.

Tables 1 and 2 show mean PM_4 and CO concentrations in kitchens and living rooms, respectively, stratified by aggregated fuel types used in the home (both heating and cooking). Fuels were aggregated based on

Table 1 Summary of 24-h PM_4 and CO concentrations in kitchens aggregating by fuel types

	Summer			Winter		
Kitchens	*n*	Mean	s.d.	*n*	Mean	s.d.
Wood logs and wood twigs[a]						
PM4 $\mu g/m^3$	39	164	110	NA	NA	NA
CO ppm	4	15	28	NA	NA	NA
Crop residues[a]						
PM4 $\mu g/m^3$	48	282.9	286	25	456.4	301.3
CO ppm	12	28.3	30.9	7	3.9	4.2
Coal[a]						
PM4 $\mu g/m^3$	33	141.9	83	56	289	325.5
CO ppm	6	13.1	26.4	6	16.3	26.5
Wood and crop residues[a]						
PM4 $\mu g/m^3$	33	192.5	107.1	9	198.4	57.1
CO ppm	3	2.9	2.4	4	0.6	0.3
Wood logs and coal[a]						
PM4 $\mu g/m^3$	10	118.7	51.2	16	290.3	215.2
CO ppm	3	2.8	2.2	4	2.1	3.2
Crop residues and coal[a]						
PM4 $\mu g/m^3$	48	111.6	80.6	60	346.3	431.6
CO ppm	3	3.1	2.1	7	15.4	24.1
Wood, crop residues and coal[a]						
PM4 $\mu g/m^3$	47	154.1	112.7	26	354.5	682.7
CO ppm	11	4.8	5.8	5	3.2	2.7

[a]Includes some houses using LPG or biogas in addition to fuels specified.

[3]A copy of the questionnaire and all the others used in the study may be found within the China Stoves portion of the website: http://ehs.sph.berkeley.edu/hem/page.asp?id=29

Table 2 Summary of PM_4 and CO concentrations in living rooms aggregating by fuel types

	Summer			Winter		
Living room	*n*	Mean	s.d.	*n*	Mean	s.d.
Wood logs and wood twigs[a]						
PM4 $\mu g/m^3$	40	150.4	92.5	NA	NA	NA
CO ppm	5	0.8	0.5	NA	NA	NA
Crop residues[a]						
PM4 $\mu g/m^3$	48	209.8	150.0	26	411.1	276.4
CO ppm	12	1.7	1.0	9	17.3	22.5
Coal[a]						
PM4 $\mu g/m^3$	35	157.0	86.3	54	371.5	670.1
CO ppm	6	2.3	1.6	7	17.2	24.4
Wood and crop residues[a]						
PM4 $\mu g/m^3$	34	162.0	97.9	8	176.3	74.9
CO ppm	3	1.8	0.8	4	0.4	0.3
Wood logs and coal[a]						
PM4 $\mu g/m^3$	10	104.6	61.1	16	211.2	117.9
CO ppm	3	2.6	2.1	4	0.4	0.3
Crop residues and coal[a]						
PM4 $\mu g/m^3$	44	110.3	66.1	60	588.4	871.4
CO ppm	2	1.4	0.6	10	21.3	22.5
Wood, crop residues and coal[a]						
PM4 $\mu g/m^3$	47	123.0	82.9	26	284.4	298.2
CO ppm	12	2.0	1.5	8	0.8	0.8

[a]Includes some houses using LPG or biogas in addition to fuels specified.

solid fuel use to form the following groups: (i) wood logs and wood twigs; (ii) crop residues; (iii) coal; (iv) wood and crop residues; (v) wood logs and coal; (vi) crop residues and coal; and (vii) wood, crop residues and coal. Houses that also had some liquefied petroleum gas (LPG) and biogas use in addition to the specified solid fuels were aggregated into the groups as these fuel types were estimated to have relatively little impact on indoor concentrations relative to the solid fuels in these homes. Although use of gaseous fuels in these homes might be expected to reduce PM_4 concentrations from solid fuels (because of reduced energy requirements from solid fuels as a result of some tasks being performed with gaseous fuels), we were not able to discern any significant differences in PM_4 concentrations for solid fuel combinations where gaseous fuels were and were not used. Thus, we estimate that potential misclassification of fuel types leading to modification of the relative differences in levels of PM_4 and CO between fuel types, was minimal in these cases. Although these different fuels represent different stages along the energy ladder (Smith et al., 1994) and transitional stages of technology adoption have been recognized where gaseous and other improved technologies co-exist with the traditional technology (Masera et al., 2000), we did not observe transitional stages from an indoor air pollution perspective in these provinces in China. We were not able to identify clear trends of reduced particle pollution when gaseous fuels were also used in addition to solid fuels for coal, biomass, and wood fuel combinations. High variability between households, however, may have obscured these trends within these sample sizes and more accurate accounting of emissions and energy use would be required. We speculate, however, that this may also be the result of two factors: (i) emissions of PM_4 from solid fuel use are high during the initial phases of the fire and may outweigh reductions in emissions from reduced cooking tasks once the fire is established. Thus, as overall efficiencies of even improved stoves are low (~20%), the energy that escapes and does not go into the pot may exceed the energy saved using the gaseous fuels; (ii) often both stoves would be lit simultaneously, and the length of time the solid fuel stove is lit is not necessarily related directly to cooking tasks (i.e. after a task is completed, a solid fuel stove frequently continues to smolder until the remaining fuel is consumed).

Because of large inter-household variability within provinces, we did not observe large differences in indoor air pollution levels between provinces for specific fuel combinations. Rather we did observe significant differences in fuel use patterns between provinces, which resulted in overall differences in mean particulate and CO concentrations between provinces. Figure 2 shows the average PM_4 concentrations kitchens and in living rooms in each province stratified by season. Significant differences in non-parametric Kruskal–Wallis tests (asymptotic significance <0.000) were observed between houses in Zhejiang and Hubei and between Zhejiang and Shaanxi during the summer for both living rooms and kitchens. Homes using biomass fuels typically had more elevated concentrations. Quite interestingly, although Zhejiang was selected for its high rates of adoption of improved stoves relative to Hubei (medium) and Shaanxi (low), PM_4 concentrations were significantly elevated in Zhejiang and exceeded PM_{10} standards in both living rooms and kitchens. Although improved stoves had flues that vent pollution outdoors, the elevated indoor concentrations were likely the result of additional open fires being lit in the kitchens for other cooking or water-heating tasks and increased use of crop residues. The signs of these fires were frequently observed during home visits and Figure 3 shows two examples from Hubei. Predominantly biomass using households in 53 homes in Inner Mongolia and 78 homes in Gansu were also reported to have higher PM_4 concentrations compared with 83 predominantly coal using houses in Guizhou and 81 in Shaanxi (Jin et al., 2005). The majority of homes with PM_4 concentrations below the standard in the summer when no heating was present were homes in Hubei and Shaanxi that used coal or a mixture of coal and biomass fuels. In the winter, Hubei and Shaanxi had approximately equal numbers of homes that were below the standard. The characteristics of these homes, however, were quite different. In Shaanxi, these homes were characterized as those using coal fuels for cooking and heating. In Hubei, the homes were characterized as

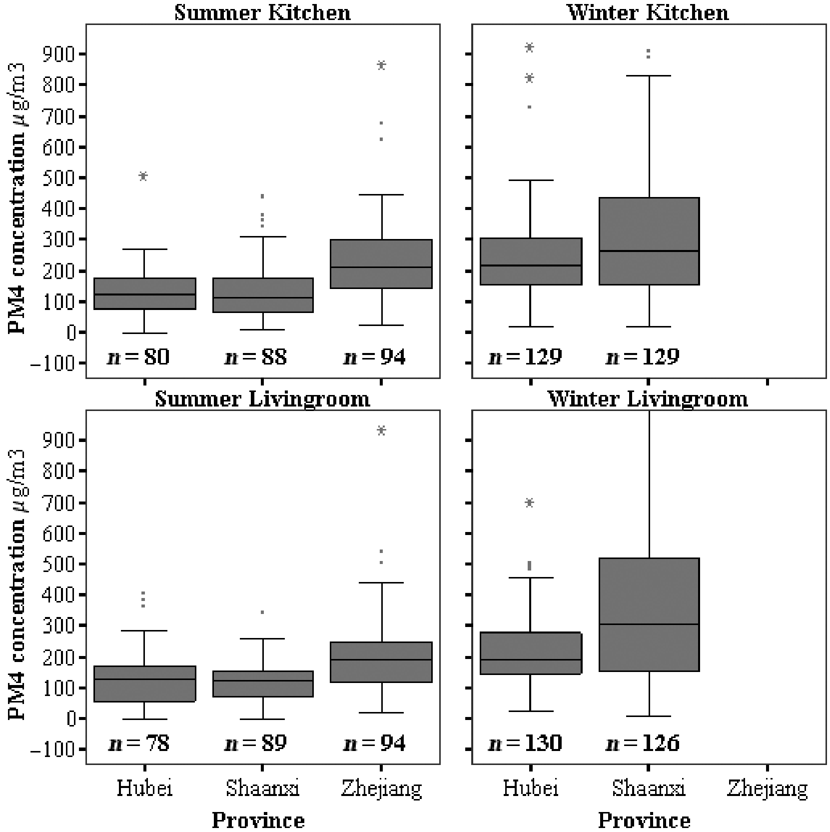

Fig. 2 Twenty-four-hour mean indoor air concentrations of PM_4 in kitchens and living rooms by province and season

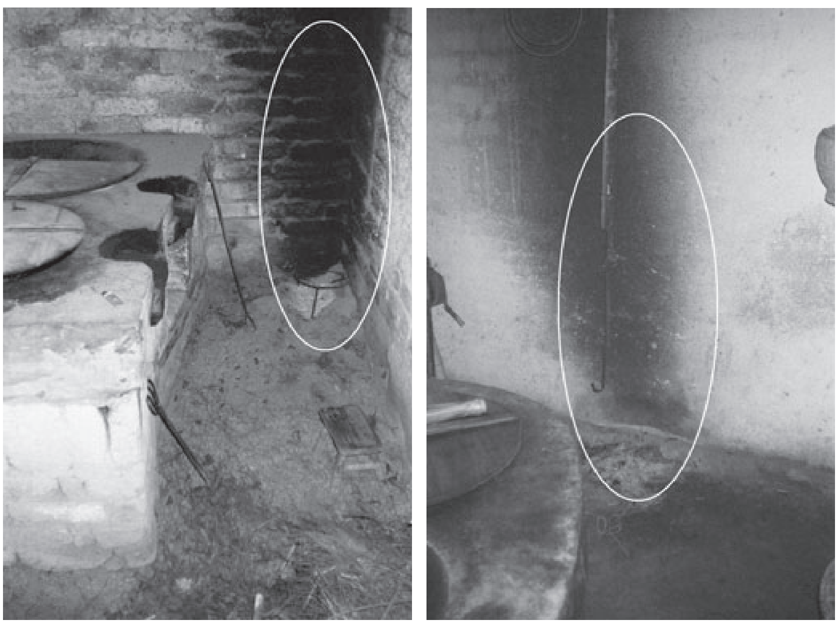

Fig. 3 Open fires for boiling water, used in addition to other stoves, Hubei

those using no heating fuel where two-thirds used coal and biomass for cooking and one-third used biomass fuels.

Significant differences in PM_4 concentrations in both kitchens and living rooms were also observed between Shaanxi and Hubei during the winter (Figure 2). These

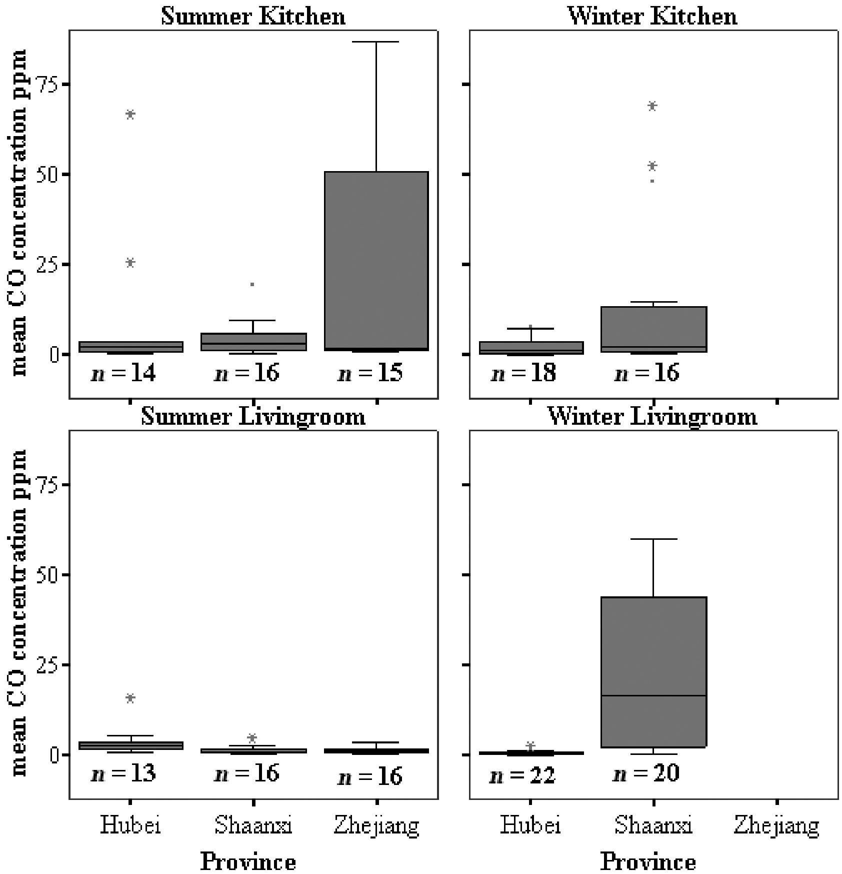

Fig. 4 Twenty-four-hour mean indoor air concentrations of CO in kitchens and living rooms by province and season

are the result of unvented coal stoves being used for space and bed heating in Shaanxi, and are discussed below. In addition to differences between provinces, significant differences were also observed between winter and summer concentrations for Shaanxi and Hubei (Zhejiang had no winter measurements), with significantly higher PM_4 concentrations during the winter. The range of concentrations measured in 2002 in Shaanxi during the NISP IAP review were of a similar magnitude to those measured in 2003 in the same province by Jin et al. (2005), who reported mean concentrations of 187–306 $\mu g/m^3$ for different rooms and months. In the current study, the mean concentration measured in the kitchens in Shaanxi during the winter was 359 $\pm$ 386 $\mu g/m^3$ ($n = 129$) and during the summer was 134 $\pm$ 102 $\mu g/m^3$ ($n = 88$). Similarly, for living rooms PM_4 concentrations were 494 $\pm$ 748 $\mu g/m^3$ ($n = 126$) during the winter and 118 $\pm$ 69 $\mu g/m^3$ ($n = 89$) during the summer.

A somewhat different picture emerges for mean 24-h CO concentrations by province and season although sample numbers are much lower in these comparisons (Figure 4). While kitchens in Zhejiang that are characterized by use of biomass fuels show large variability during the summer mean concentrations are lower than Hubei and Shaanxi. In the winter, however, similar patterns are observed and concentrations in Shaanxi are much higher than those for Hubei because of the coal use during winter seasons. CO concentrations in the living rooms during the winter were generally higher than levels in the kitchens in Shaanxi, although the converse was true during the summer. Interestingly, the opposite was the case in Hubei, and summer concentrations were higher than winter concentrations presumably because of decreased use of crop residues. Although during the NISP IAP review, we did not observe higher CO concentrations in the predominantly biomass using households in contrast to Jin et al. (2005), the range of CO concentrations measured in Shaanxi were of similar magnitude as those measured by Jin et al. (2005) who reported 2.0–13 ppm in different rooms and months in Shaanxi. Mean CO concentrations in kitchens, during the NISP IAP review, were 4.5 $\pm$ 4.9 ppm ($n = 16$) during the summer and 13.5 $\pm$ 22.3 ppm ($n = 16$) during the winter, respectively. For living rooms, CO concentrations were 1.5 $\pm$ 1.1 ppm ($n = 16$) during the summer and 24.2 $\pm$ 22.7 ppm ($n = 20$) during the winter, respectively. Even in summer, however, in all provinces many households measured in the NISP IAP review experienced CO levels several times the national IAQ standard of 10 mg/m^3 for a 1-h average (equivalent

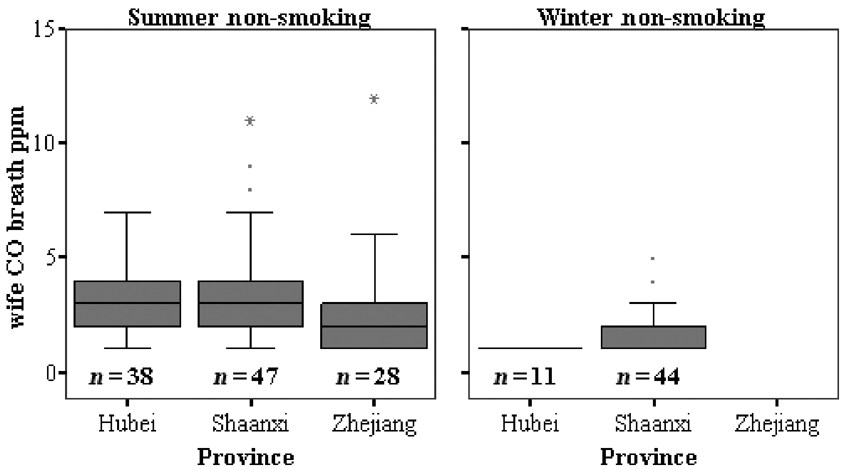

Fig. 5 CO breath concentrations for wife of household head after indoor air sampling

to 9 ppm), and in the winter the situation was much worse, particularly for households using agricultural residues and unvented coal stoves for heating.

Breath carbon monoxide

The level of CO in exhaled breath is an intermediate between an indicator of exposure and an indicator of health. It reflects exposure to CO over the previous 5–15 h, but also is a measure of the degree to which the haemoglobin in the blood has taken up CO, thereby depriving the body of oxygen carrying capacity. In addition to acute effects associated with short-term exposure to high levels of CO, long-term exposures to elevated but subacute levels of CO are associated with chronic health impacts.

Figure 5 shows CO breath concentrations for non-smoking wives of the head of household after the

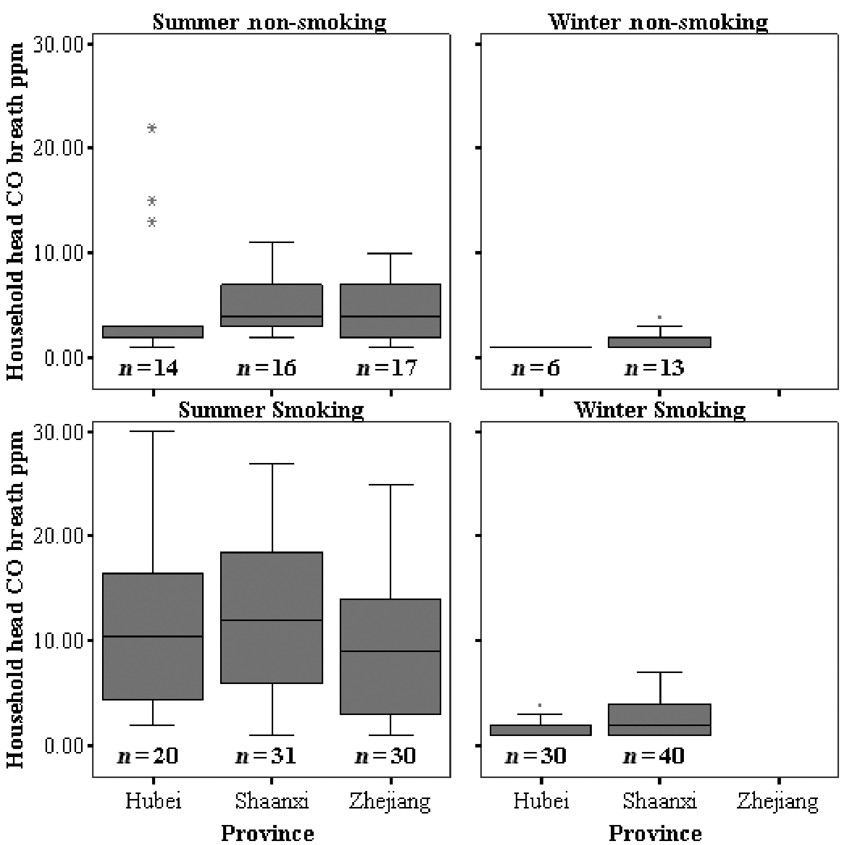

Fig. 6 CO breath measurements for household head before indoor air sampling (Similar results were obtained after indoor air sampling but n is larger here)

24-h indoor air sampling. Interestingly, in contrast to PM_4 indoor air concentrations, CO breath concentrations were much lower in Zhejiang compared with Hubei and Shaanxi. Because of the large variation in indoor CO concentrations in biomass burning households in Zhejiang and the low sample numbers, trend analysis with CO breath concentrations is not possible although the mean concentrations show a similar pattern to CO breath levels. Perhaps more surprisingly, however, the CO breath concentrations in the winter are significantly lower than in the summer in both Hubei and Shaanxi, in spite of significantly higher PM_4 concentrations in kitchens during this period. It is likely that these differences are the result of an additional source of exposure during the summer periods or a change in behavior during these seasons, which results in different relationships between indoor concentrations and personal exposures, and highlights a potential source of exposure error in using indoor air concentrations as proxies of exposure in health-based epidemiological investigations, and the need for personal exposure measures.

Figure 6 shows CO breath concentrations for both smoking and non-smoking heads of household. For non-smoking heads of household CO breath concentrations followed a similar pattern to the wives, and were considerably lower in the winter in spite of higher indoor air concentrations of PM_4 in both kitchens and living rooms during this season. Again it is likely that a significant source of exposure is not accounted for in indoor air concentrations during the summer. For smokers, there is also a similar trend between seasons and CO breath concentrations for smokers also tend to be much higher during the summer, although self-reported cigarette consumption was not significantly different between seasons.

Figure 7 shows breath CO concentrations of the children of the head of household. Similar to the husbands and wives the children also show lower breath CO concentrations in Zhejiang and higher breath CO during the summer compared with the winter in Shaanxi, although sample numbers in the winter were low.

Table 3 Correlations between PM_4 and CO dosimeter tubes in Shaanxi

Location	Number of samples	Pearson correlation coefficient (r)
Kitchen	34	0.49 ($P < 0.003$)
Living room	35	0.52 ($P < 0.002$)
Combined	69	0.49 ($P< 0.000$)

Table 4 Correlations between PM_4 and CO dosimeter tubes for Hubei, Shaanxi and Zejiang

Location	Fuel type	Number of samples	Pearson correlation coefficient (r)
Kitchen	Crop residues	21	0.50 ($P < 0.02$)
	Coal	12	0.58 ($P < 0.05$)
	Crop residues + coal	13	0.68 ($P < 0.01$)
	Wood + crop + coal	20	0.78 ($P < 0.00$)
	Combined	85	0.27 ($P < 0.01$)
Living room	Crop residues	20	0.83 ($P < 0.00$)
	Coal	13	0.66 ($P < 0.02$)
	Crop residues + coal	12	0.54 ($P < 0.07$)
	Wood + crop + coal	20	0.84 ($P < 0.00$)
	Combined	86	0.38 ($P < 0.00$)
Combined	Combined	171	0.29 ($P < 0.00$)

Biomass only fuel types lacked sufficient samples for comparison.

Correlations between measures

In general, correlations between PM_4 and CO tubes for aggregate kitchen and living room measurement locations in Shaanxi (35 homes, 69 pairs) were considerably higher than those reported by He et al. (2005) in a study of four homes in Shaanxi across four different measurement locations. Further, we found similar correlations when performed separately for kitchen

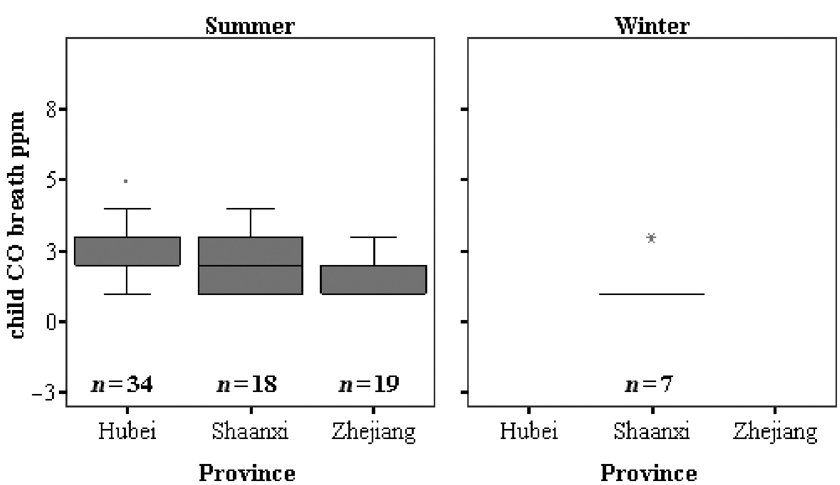

Fig. 7 Child CO breath before indoor air sampling (non-smoking)

and living room locations (Table 3). Although correlations improved with stratification by fuel type or by main cooking fuel, the reduction in sample numbers makes these estimates not robust.

Table 4 shows correlations between PM_4 and CO dosimeter tubes for all provinces Zhejian, Hubei and Shaanxi for all fuels and stratified by aggregate fuel types (again gaseous fuels were included in aggregate fuel type measures). Similar correlations were observed between 24-h mean electrochemical CO concentrations and PM_4. In general, correlations for all fuel types and locations combined were weak, and were similar when all fuel types were stratified by room location. When stratified by fuel type but not by room location correlations were often slightly improved, except for crop residues. This was the result of homes in Zhejiang where CO and PM_4 levels correlated well in living rooms ($r = 0.76$ $P < 0.01$ $n = 10$) but not in kitchens. When restricted to Hubei and Shaanxi and stratified by fuel type but not room location correlations were again slightly improved. While He et al. (2005) have suggested that poor correlations between PM and CO dosimeter tubes across measurement locations could be the result of different dispersion characteristics of particles and gaseous pollutants, this was not observed in the current study between kitchens and living rooms. This was possibly as a result of multiple stoves in different locations, but more probably the result of much greater sample sizes in the current study. Further stratification on aggregate fuel type resulted in significantly improved correlations again highlighting the importance of accurate estimates of fuel use in homes. This has important implications if CO tubes are to be used as indirect proxies of particulate exposure in health-based investigations in these three provinces in China. Clearly, interpretation of dosimeter tubes would have to be in conjunction with detailed surveys of fuel use in households.

Carbon monoxide breath concentrations for the wife of the head of household were not correlated to indoor measures when smokers were excluded, even when stratified by province. Associations with CO breath concentrations and indoor air measures tended to be negative in Hubei and Shaanxi, although not significant. The lack of correlation is not surprising given that the indoor air concentrations and breath concentrations in the summer suggest that there is an additional source of exposure that is not accounted for in indoor concentrations.

Smoking

Two-thirds (66%) of houses monitored for PM_4 reported smoking indoors. Many analyses were not possible when only homes with no smoking were selected because of the large reduction in sample numbers. Table 5 demonstrates the impact of smoking on IAP levels for fuel combinations where there were sufficient samples for comparison. Because of the magnitude of PM_4 concentrations as a result of the solid fuel sources, and resultant variability, we were not able to discern contributions of smoking to indoor air PM_4 concentrations. Clearly, however, as cleaner fuels become more predominant the smoking contribution may play a much more significant role and potentially confound measurement of indoor air levels. Future studies should incorporate nicotine dosimeter badges to control for smoking contributions as part of measurement scenarios.

Table 5 PM_4 concentrations ($\mu g/m^3$) stratified by smoking status limited to fuel combinations where n was sufficient for comparison

		Kitchen			Living room		
Fueltype	Smoking	*n*	Mean	s.d.	*n*	Mean	s.d.
Summer							
Wood logs + wood twigs + crop residues + coal + biogas	Yes	12	236	146	10	151	109
	No	3	139	92	2	32	35
Wood twigs + crop residues + Coal	Yes	6	91	51	7	85	102
	No	3	78	91	3	203	81
Wood twigs + crop residues + LPG	Yes	21	226	98	21	188	96
	No	2	219	169	2	178	111
Wood twigs + LPG	Yes	9	170	125	9	132	73
	No	5	116	57	5	98	74
Crop residues	Yes	11	378	513	10	191	129
	No	5	216	141	5	138	126
Crop residues + coal	Yes	29	115	93	28	112	69
	No	14	95	56	11	125	65
Crop residues + LPG	Yes	19	232	135	20	240	189
	No	11	321	243	11	205	104
Coal	Yes	10	116	89	12	134	100
	No	11	153	55	11	152	83
Winter							
Wood logs + crop residues + coal	Yes	14	242	158	14	247	290
	No	5	273	106	5	236	77
Wood logs + coal	Yes	4	401	476	4	193	72
	No	4	389	119	4	306	159
Wood twigs + crop residues	Yes	4	200	72	4	179	56
	No	3	197	56	3	206	91
Wood twigs + crop residues + coal	Yes	15	478	888	15	316	367
	No	8	191	105	8	235	202
Wood twigs + coal	Yes	6	404	286	6	254	151
	No	6	196	121	6	176	66
Crop residues	Yes	17	478	335	18	428	281
	No	3	228	68	3	232	211
Crop residues + coal	Yes	37	418	532	36	622	869
	No	16	188	93	17	420	479
Coal	Yes	29	263	205	28	477	906
	No	15	451	532	14	313	242
Coal + LPG	Yes	6	126	56	6	118	94
	No	4	176	51	4	253	104

Differences between main cooking and heating fuels

Figure 8 shows average PM_4 concentrations for main cooking fuel in households who reported no heating fuel use. Although there is considerable variability, in most cases the indoor air concentrations in the winter are considerably higher than those in the summer in both kitchens and living rooms. This is probably the

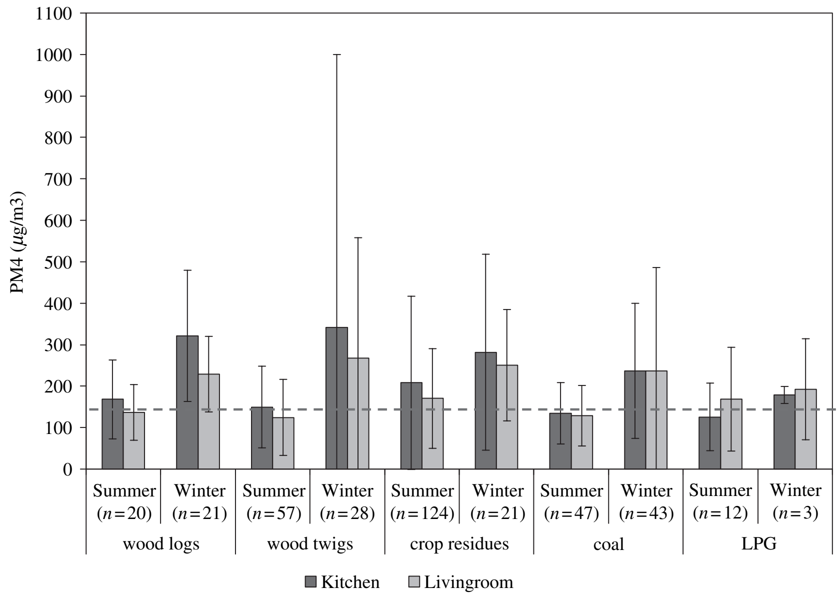

Fig. 8 Average PM_4 concentrations for main cooking fuel in households who reported no heating fuel use

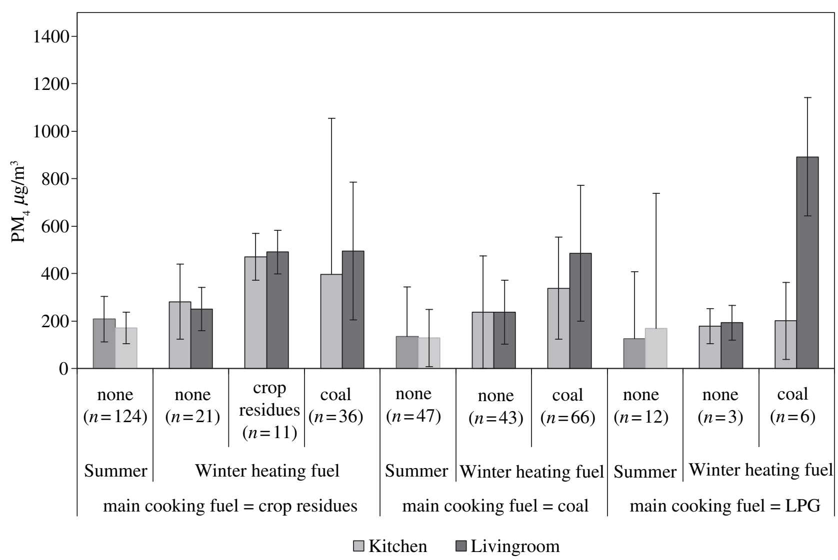

Fig. 9 Average PM_4 concentrations for main cooking fuel in households with heating fuel use (Hubei and Shaanxi)

result of decreased ventilation in the homes combined with increased fuel use during the winter. During the winter indoor PM_4 concentrations are considerably above the Chinese indoor standard for PM_{10}. (Of course, if we had been measuring PM_{10} concentrations and not PM_4, indoor concentrations would have exceeded the standard by an even greater margin.) During the summer average indoor PM_4 concentrations in both kitchens and living rooms are approximately equivalent to the indoor standard for PM_{10}, although there is considerable variability and many homes clearly exceed these levels. When heating fuels are used in the homes (Figure 9), the differences are more pronounced. While it is not possible to draw any conclusions about differences in PM_4 concentrations between seasons in houses that used LPG as the main cooking fuel, it is immediately apparent that PM_4 concentrations are much higher than would be expected with just LPG use during both seasons. In these homes, although the main cooking fuel is reported to be LPG, the contribution of other solid fuels used during other cooking tasks are significant and in many cases lead to concentrations that exceed the indoor standard. It probably also indicates the impact of pollution penetrating from outside, because of 'neighborhood pollution' in the village (Smith et al., 1994). This highlights the potential misclassification and error for gaseous fuels that may occur in health studies if

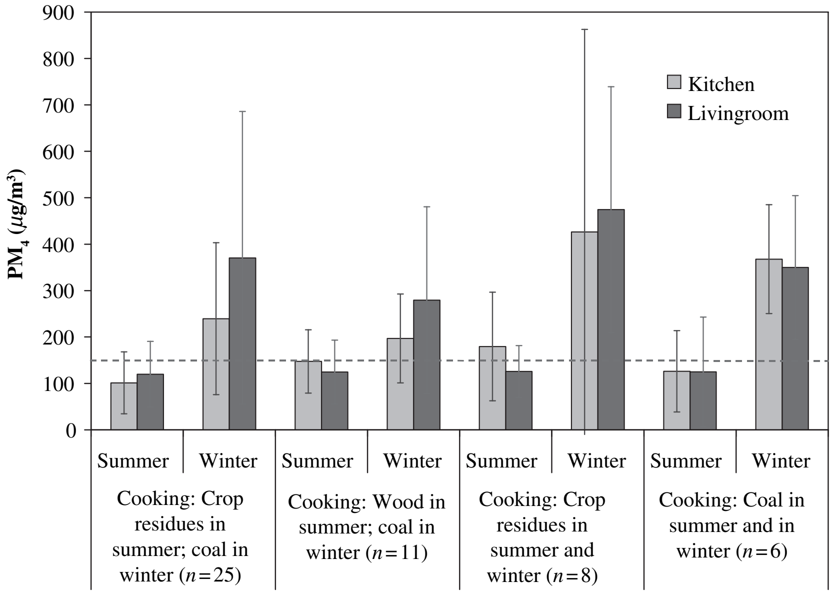

Fig. 10 Average PM_4 for households who used unvented coal stoves in the living room for heating (Hubei and Shaanxi)

only information on main cooking fuel is collected in questionnaires.

Although these analyses were performed based on main cooking fuel, and some misclassification may result because of use of additional fuels in the households, when the analysis is restricted to matching all of the specified fuels used in each home similar trends are observed, although the number of houses and statistical power are reduced.

Unvented coal-heating stoves

Figure 10 shows average PM_4 concentrations in living rooms and kitchens for houses that were measured during both seasons and used unvented coal stoves for heating in the living room during the winter. For all combinations of fuel types shown here, average PM_4 concentrations were higher during the winter than in the summer. Although average summer levels for these fuel combinations were close to the new national standard for IAQ for PM_{10} (150 μg PM_{10}/m^3), average winter levels in living rooms and many kitchens were often double the levels of the national standard. This is of particular concern given the increased amounts of time families spend indoors in winter. This was true for participants who used the same fuels (coal) for cooking during the summer and the winter and for heating the household, and for participants who changed cooking fuels during different seasons. Thus, the differences in levels likely result from tighter closing of houses and increased fuel use during the winter.

For those households who changed cooking fuels from biomass to coal in the winter, the kitchen PM_4 levels were somewhat lower than the living room levels in the winter. Although this perhaps indicates the performance of some cooking tasks in the living room, as kitchen levels are lower than when the cooking fuel remains the same between seasons, this demonstrates the significant impact of unvented coal stoves on IAQ in these homes.

Further assessment of the relative contributions of these sources to the exposure of the individuals is important given that health effects vary depending on the nature of the particles and the composition of organic and inorganic pollutants. Thus, as the fuels burnt for cooking and heating may not be the same in the kitchen and the living room, and ventilations rates, the particle distribution and the composition of organic and inorganic pollutants may vary between these two environments, estimation and modeling of health impacts requires source apportionment of personal exposures, possibly based on measurement of the time spent in each location by different family members.

Seasonal changes in fuel use

The IAQ results accord with the highly diverse fuel usage patterns in these provinces in China, and are

Table 6 Seasonal change in cooking fuel for houses with heating in the winter (Hubei and Shaanxi)

	Cooking fuel in winter				
Houses with heating in winter	Wood	Crop residue	Coal	LPG	Total
Cooking fuel in summer					
Wood	1	3	5	0	9
Crop residue	0	11	25	0	36
Coal	0	6	6	1	13
Total	1	20	36	1	58

Table 7 Seasonal change in cooking fuel for houses with no heating in the winter (Hubei and Shaanxi)

Houses with no heating in winter	Cooking fuel in winter				
	Wood	Crop residue	Coal	LPG	Total
Cooking fuel in summer					
Wood	7	1	1	0	9
Crop residue	1	2	2	0	5
Coal	3	2	11	2	18
LPG	0	1	2	0	3
Total	11	6	16	2	35

supported by the observations in the larger household survey (3500 homes) of multiple stoves being present in many kitchens. In the summer monitoring, an average of 2.6 types of fuels were used per household, which decreased to 1.9 in the winter as the number of household burning biomass and LPG fell, and the number using coal rose. So pronounced was the variety of fuel combinations used that in houses where IAQ was measured, 28 different fuel combinations were used in kitchens in the winter and 34 were used in the summer. Not only were multiple fuels used, but there was also a shift in the fuel and stove usage patterns in many individual houses measured in both seasons (Shaanxi and Hubei). Table 6 shows changes in cooking fuels in homes measured in both seasons that used space heating in the winter where ~70% of the homes reported changing their main cooking fuel type between seasons. Table 7 shows changes in cooking fuels for homes that did not use space heating in winter where ~43% of homes reported changing their main cooking fuel type between seasons. Although creating considerable complications for analysis, this documents a little-explored component of fuel use in these rural settings, namely the seasonal changes in fuel use patterns and the use of multiple fuels within the same home. This has important implications for epidemiological investigations into health impact, as fuel use changes could lead to considerable misclassification. In addition, if particulate measurements are made in association with health assessments, significant errors may result if seasonal variability and differences in particle types and concentrations as a result of different fuel use between kitchens and living areas are not taken into account. This reinforces the need for longitudinal surveys of fuel use combined with measurements of indoor air pollution levels. The extent of fuel and stove variations, even with measurements of 396 homes, resulted in sample sizes that were often not sufficient to make definitive statistical statements about the resulting IAQ differences between many fuel combinations.

Differences between improved and traditional biomass stoves

Given the extent and duration of the massive development and promotion of stove improvements conducted by NISP and subnational programs starting in the early 1980s, as well as the rise in commercial stove activity starting in the 1990s, partly because of government incentives, the distinction between 'improved' and 'traditional' stoves is not a sharp one today, but related to the historical process and local expectations in any one place. In many locations, there have also been several generations of 'improved' stoves during this period. This led us to apply our own structural definition of 'improved' and 'traditional' based on observed stove characteristics in each house, which allowed us to be consistent and seemed to accord to general perceptions and historical technological development. An improved stove is defined here as one with a chimney and combustion-chamber grate, no matter when built.

In houses using combinations of biomass fuels including wood logs, wood twigs and agricultural residues, improved stoves resulted in significantly lower average PM_4 and median CO kitchen concentrations than traditional stoves in the summer in both parametric (t-test) and non-parametric tests (Wilcoxon rank sum and Kruskal–Wallis). (Table 8). For houses that used coal or LPG and biomass in the same house no significant differences were observed. During the winter, when space heating was present, there were no significant differences in PM_4 or CO kitchen concentrations between houses using improved and traditional stoves with wood log, wood twig, and agricultural residues, or for houses that used coal or LPG and biomass in the same house. This was not surprising because of the variance introduced by heating of interior spaces and bed platforms (Kang) and variance introduced by house conditions and types, and multiple stoves and fuels used during these cold periods. Results were not affected by exclusion of homes where smoking was present.

Table 8 Improved biomass stoves compared with traditional biomass stoves using wood logs, wood twigs and agricultural residues in the summer. See definition of: 'improved' in text

Measurement	Stove type	Number stoves	Kitchen concentration	Average difference	Kruskal–Wallis sig.	t-test sig
PM_4 24 h concentration ($\mu g/m^3$)	Traditional	88	268 (95% CI 208–328)	116	<0.0001	0.0005
	Improved	63	152 (95%CI 133–172)			
HOBO CO 24 h median (ppm)	Traditional	7	1.85 (95%CI 1.08–2.62)	1.15	0.0064	0.0042
	Improved	6	0.70 (95%CI 0.32–1.07)			

Conclusions and implications

Although it is not possible now to identify sharp distinctions among stove generations, it seems clear that the types of non-chimney stoves once common in rural China were much more polluting of living spaces than those today. This can be seen in the many dozens of IAQ studies carried out by Chinese investigators in the 1980s, which found quite high levels. Unfortunately, however, these studies generally did not collect sufficient household information or use standard monitoring methods that would allow general conclusions about population exposures at that time (Saksena et al., 2003; Sinton et al., 1995)

Although pollution levels have decreased from the high levels because of traditional stoves, there is still need to reduce them further to meet Chinese health-based standards including the new national indoor air pollution standard. Although mean indoor concentrations of PM_4 in Hubei and Shaanxi were close to the national standard for PM_{10} in summer, hourly concentrations of CO were often several times the national air quality standard in a large fraction of homes. Mean concentrations in Zhejiang were considerably above the standard in the summer in both kitchens and living rooms. In the winter, PM_4 levels were often double the levels of the national standard and hourly CO concentrations were much worse, particularly for households using biomass. If these results are typical, then a large fraction of China's rural population is now chronically exposed to levels of pollution far higher than those set by the Chinese government IAQ standards to protect human health. Although improved biomass stoves resulted in reduced PM_4 from biomass fuel combinations, little improvement was observed in indoor pollution levels when other unimproved stoves are present in the same kitchen, and an improved biomass stove for cooking may not be a sufficient strategy for reducing indoor air levels if more polluting space-heating stoves using coal are also being used.

These findings have implications for the assessment of exposures in epidemiological investigations. As many households change fuels according to daily and seasonal factors, resulting in different seasonal concentrations in living rooms and kitchens, health implications from fuel use require longitudinal evaluation changing household fuels and corresponding IAQ levels, combined with accurate time-activity information to determine the duration that family members spend in each environment. Given that CO breath concentrations suggest that there is an additional source of exposure in the summer not captured by indoor air concentrations of CO or PM_4, personal exposure samples would be recommended.

There are also implications for ambient air pollution impacts. Plumes of pollution downwind of these rural areas may vary in both concentration and composition during different seasons. To ascertain the overall impact for regional pollution or global warming scenarios, therefore the seasonal variation as a result of changing fuel use would have to be incorporated into models of atmospheric impacts.

Leaving aside the difficult issue of enforcing household IAQ standards, there is clearly a need for more systematic monitoring of existing IAQ in rural as well as urban areas to plan and target remedial measures. It is uncertain, however, whether the current standards represent realistic objectives for current attainment given the extensive use of solid fuels in Chinese rural populations in these provinces. It is well established that in all but the cleanest of communities where when combustion sources have been removed from the residential environment (Koistinen et al., 2004), indoor concentrations are often significantly higher than outdoor concentrations, and particularly in solid fuel using households. Our work and that of others raise the practical question whether these standards are achievable given current patterns of energy consumption in rural China, which rely so heavily on solid fuels. Even when used with chimneys, these fuels emit substantial pollution into the household environment. The question is what cleaner-burning solid-fuel technologies need to be implemented or whether fossil or biomass-based liquid and gaseous fuels could be promoted that would achieve these standards for the greater part of the population.

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