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## Combustion-derived nanoparticle exposure and household solid fuel use in Xuanwei and Fuyuan, China

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### Abstract

Combustion-derived nanoparticles (CDNPs) have not been readably measurable until recently. We conducted a pilot study to determine CDPN levels during solid fuel burning. The aggregate surface area of CDPN ( $\mu\text{m}^2/\text{cm}^3$ ) was monitored continuously in 15 Chinese homes using varying fuel types (i.e. bituminous coal, anthracite coal, wood) and stove types (i.e. portable stoves, stoves with chimneys, firepits). Information on fuel burning activities was collected and  $\text{PM}_{2.5}$  levels were measured. Substantial exposure differences were observed during solid fuel burning (mean:  $228.1 \mu\text{m}^2/\text{cm}^3$ ) compared to times without combustion (mean:  $14.0 \mu\text{m}^2/\text{cm}^3$ ). The observed levels during burning were reduced by about four-fold in homes with a chimney (mean:  $92.1 \mu\text{m}^2/\text{cm}^3$ ;  $n = 9$ ), and effects were present for all fuel types. Each home's CDPN measurement was only moderately correlated with the respective  $\text{PM}_{2.5}$  measurements ( $r^2 = 0.43$ ;  $p = 0.11$ ). Our results indicate that household coal and wood burning contributes to indoor nanoparticle levels, which are not fully reflected in  $\text{PM}_{2.5}$  measurements.

### Keywords

coal; biomass; wood; stove; nanoparticle; respiratory

### 1. Introduction

Household solid fuel, including coal and wood, is used by approximately three billion people worldwide for cooking and heating (World Resources Institute 1996). Daily exposure to high levels of domestic fuel combustion products has led indoor air pollution (IAP) attributed to solid fuel use to be estimated as the eighth largest risk factor for global disease and deemed a major public health problem in developing countries (Ezzati 2004).

The widespread use of coal and wood for heating and cooking in Xuanwei, China, has provided an opportunity to study household solid fuel exposures. Xuanwei is a coal-rich

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semi-mountainous county on a high plateau in north-eastern Yunnan Province. Traditionally, 90% of its residents are farmers with minimal industrial and automotive air pollution exposure (Mumford et al. 1987; Lan et al. 2002). There are three different types of major fuels for domestic cooking and heating in Xuanwei: “smoky coal” (bituminous coal), “smokeless coal” (anthracite coal), and wood. Xuanwei smoky coal has relatively high aromatic and low sulfur content, and, as its name implies, smokes heavily on combustion. Smokeless coal, on the other hand, has low aromatic but high sulfur and ash content and does not produce significant smoke levels. Prior to the 1990s, fuel burning for cooking and heating typically took place in a simple, unvented firepit or stove in the family room. Currently, many homes use a chimney for ventilation. Through these burning scenarios, Xuanwei residents are exposed to high levels of IAP (Mumford et al. 1987; Lan et al. 2002). This exposure, coupled with low automotive and industrial air pollution exposures, provides a unique opportunity to study exposures and risks of diseases associated with IAP from household coal burning.

Epidemiological studies of using solid fuel often use questionnaire-based exposure assessments. For example, a recent systematic review of studies exploring the association between lung cancer and household coal use found that studies used either qualitative (i.e. questions with yes/no responses, such as do you burn coal at home?) or quantitative (i.e. number of years of exposure or amount of coal use) questions to measure subjects’ coal exposures (Hosgood et al. 2011). Only a few studies have incorporated quantitative exposure assessment methods of IAP attributable to solid fuel use, such as PM<sub>2.5</sub> (Park and Lee 2003; Siddiqui et al. 2009). To date, no study has reported on combustion-derived nanoparticles (CDNPs) associated with household solid fuel burning in China.

Combustion-derived nanoparticles are particles that range from 1 to 1000 nm in size, and have been ubiquitous as ambient exposures since the advent of fire (Avakian et al. 2002); however, they have not been readily detectable up till about 10 years ago (Sgro et al. 2003). Due to their small size and high surface to volume ratio, CDNP are more highly reactive than their larger counterparts. Therefore, for smaller particles, dose metrics other than mass, such as particle counts or surface area, are likely to be more relevant to disease (Oberdorster et al. 2001; Valavanidis et al. 2008).

Recently, technology has been developed that is able to estimate, in real-time, the surface area of nanoparticle aerosols in the ambient air. We employed a new nanoparticle detection methodology, able to quantify the surface area equivalent dose of nanoparticles, in 15 homes participating in our ongoing exposure survey in Xuanwei and Fuyuan, China, to determine the range in nanoparticle exposure levels present in homes during solid fuel burning. Further, we collected parallel PM<sub>2.5</sub> samples to explore the correlation between nanoparticle exposure and the traditional gravimetric exposure metric.

## 2. Methods

Homes in this pilot study were from our ongoing exposure assessment study that was designed to quantify IAP exposure levels associated with household solid fuel use. Briefly, homes were eligible to participate if they burned solid fuel and used the same cooking and

heating equipment for the past five years. Of the homes that participated in the comprehensive exposure assessment study in Xuanwei and Fuyuan, Yunnan Province, China, a subset of 15 homes received the additional CDNP exposure assessment, which is reported here. This subset of 15 homes represents a variety of fuel types and stove types used in Xuanwei and Fuyuan homes. For all homes, informed consent was obtained from a female study participant residing in the home.

The AEROTRAK™ 9000 nanoparticle aerosol monitor, a portable diffusion charger +electrometer, was used to monitor real-time the ambient air around the stoves burning solid fuels to assess exposure to combustion-based nanoparticles. The AEROTRAK™ 9000 nanoparticle aerosol monitor estimates the surface area equivalent dose of particles in the size range of 10–1000 nm. The AEROTRAK™ 9000 monitor was calibrated prior to use in this study by the equipment manufacturer. The calibration procedure includes an internal electronics integrity verification and challenge to a known concentration to sodium chloride that was verified using a reference particle counter. Zero calibrations were performed weekly. The average surface area of nanoparticles theoretically deposited in the tracheo-bronchial region of the respiratory tract per milliliter of air ( $\mu\text{m}^2/\text{cm}^3$ ) was monitored real-time in 10 s intervals throughout two calendar days in 15 homes using different fuel types (i.e. smoky coal, smokeless coal, wood) and a variety of stove types (i.e. portable stove, stove with chimney, firepit/brick stove). Some homes in this region utilize multiple stoves, for example, home 1 of this report used primarily a high stove as well as a portable stove on occasions.

Stationary particulate measurements ( $\text{PM}_{2.5}$ ) were collected parallel to the AEROTRAK™ 9000 on a 37 mm Teflon filter using a cyclone with an aerodynamic cut-off of  $2.5 \mu\text{m}$  (model BGI, GK 2.05SH) at a flow rate of 3.5 L/min ( $\pm 20\%$ ). The AEROTRAK™ 9000 and  $\text{PM}_{2.5}$  measurement setup were placed between 1 m and 2 m from the main combustion source, although placement varied somewhat because of limited allowable space.

During the sampling time, the homes' solid fuel burning activities were recorded. The activity questionnaire assessed what stove type was used (low stove, high stove, brick stove, firepit, portable stove), what fuel type was used (bituminous coal, anthracite coal, beehive (honeycomb-shaped briquette usually composed by mixing coal with sand or clay), wood), and what activities were performed (i.e. cooking human food, cooking animal food, heating) for all times during the sampling period. This activity log was then linked to the nanoparticle data by time, allowing the average nanoparticle levels to be determined for not only the entire sampling period, but also for periods of solid fuel burning and periods of no fuel burning and kindling.

To assess the similarities between the standard method of assessing particulate measurements and the nanoparticle measurements, the correlation between the  $\mu\text{m}^2/\text{cm}^3$  and  $\mu\text{g}/\text{m}^3$  results was evaluated by the Spearman correlation coefficient. Correlations were evaluated by comparing the nanoparticle average for the home's total sampling period, which was on average 1044 min per home, with the average of the respective daily  $\text{PM}_{2.5}$  measurements, with each sampled for an average of 1385 min.

### 3. Results

A majority of the homes sampled in this pilot study burned bituminous coal (66.7%), with a few burning anthracite coal (20%), and one burning wood for heating and cooking (Table 1). Most homes used either a low (33.3%) or high stove (26.7%) with a chimney, while the rest of the stove types were without a chimney. All homes with a chimney burned bituminous coal, except for one that burned anthracite coal (Table 1).

Combustion-derived nanoparticles were detected in all 15 homes. When linking the burning activities to the nanoparticle measurements, we observed a clear association between cooking and heating activities and elevated nanoparticle levels (examples: Figures 1–4). These patterns were observed in all homes, regardless of type of fuel burned, type of stove used, or presence of chimney. Interestingly, the surface area fluctuated within similar activities, which may be the result of the fire's ignition, stoking, or other characteristics.

Average nanoparticle levels were  $228.1 \mu\text{m}^2/\text{cm}^3$  while burning solid fuel and  $14.0 \mu\text{m}^2/\text{cm}^3$  while no fuel was burned. Again, these observations were seen for all types of fuel burned and all types of stoves used (Table 2). The presence of a chimney in the home reduced nanoparticle levels by a factor of  $\sim 4$  (Table 2). The reduction of nanoparticles during solid fuel burning by the presence of a chimney was seen when burning bituminous coal [with chimney (mean:  $103.6 \mu\text{m}^2/\text{cm}^3$ ); without chimney (mean:  $471.0 \mu\text{m}^2/\text{cm}^3$ )] and anthracite coal [with chimney (mean:  $46.0 \mu\text{m}^2/\text{cm}^3$ ); without chimney (mean:  $230.6 \mu\text{m}^2/\text{cm}^3$ )]. Differences were less dramatic when comparing different stove types with chimneys burning bituminous coal [high stove (homes 1–3; Table 1); low stove (homes 4–8; Table 1)].

The parallel sampling data indicated that the average CDNP measurement ( $\mu\text{m}^2/\text{cm}^3$ ) was moderately correlated with the  $\text{PM}_{2.5}$  average ( $r^2 = 0.43$ ;  $p = 0.11$ ).

### 4. Discussion

A critical obstacle to studying the health effects of CDNP in humans is that exposure assessment methods have not been readily available until recently for use in epidemiological studies. We had the opportunity to overcome this hurdle and incorporate a nanoparticle aerosol monitor into an ongoing exposure assessment study in Xuanwei and Fuyuan. We measured the average surface area of CDNP continuously in coal and wood burning homes, along with the homes' fuel burning activities and  $\text{PM}_{2.5}$  levels. Our data suggest that solid fuel burning for heating and cooking elevates household nanoparticle exposures, regardless of fuel or stove type used, and that these levels are reduced dramatically in the presence of a chimney. We observed that having a chimney in the home reduced nanoparticle levels by a factor of  $\sim 4$ . This is the first attempt, to our knowledge, to measure CDNP in homes burning solid fuels in China.

Combustion-derived nanoparticles have been measured in other scenarios using the same methods as this study. For example, investigators observed that the surface area of nanoparticles increased inside an automobile while driving and in a home during incense burning (Qi et al. 2008; Ji et al. 2010). Additional studies have also continuously measured IAP attributable to solid fuel use, focusing on particulate matter (Park and Lee 2003;

Siddiqui et al. 2009). These studies found PM<sub>2.5</sub> levels to be increased in Pakistani and Costa Rican homes when wood was being burned for the residents' cooking needs, supporting our preliminary observations (Park and Lee 2003; Siddiqui et al. 2009). Further, the observed reduction in nanoparticle exposure in the presence of a chimney is consistent with previous reports showing a reduction in solid fuel combustion by-products with the introduction of ventilation (Albalak et al. 2001; Naeher et al. 2001; Zuk et al. 2007).

Currently, monitoring for airborne particulates uses primarily mass per unit volume (i.e. micrograms per cubic meter). Since the incorporation of these gravimetric methods, airborne exposures have been typically measured by PM<sub>2.5</sub> and PM<sub>10</sub>. For smaller particles, dose metrics other than mass may be more relevant. Due to their small size, gravimetric measures may not be appropriate for particles in the nano- and ultrafine size fraction since they contribute little to the overall mass but contribute significantly to the overall number of the airborne particles present (Andersen et al. 2007). Therefore, particle counts or surface area may be a better measure of exposure for CDNP. The question remains, however, as to whether measurements from methods for particle counting CDNP are independently informative, and not strongly correlated, with those of more conventional gravimetric measures like PM<sub>2.5</sub>. Investigators have explored these issues in studies of urban air pollution. One study conducted in Denmark found that nanoparticles correlated moderately with PM<sub>2.5</sub> ( $r=0.40$ ) and PM<sub>10</sub> ( $r=0.39$ ), allowing for the evaluation of the health effects associated with each size fraction (Andersen et al. 2008). Similarly, a study conducted in three European cities found that PM<sub>2.5</sub> poorly correlated with the number concentrations of ultrafine particles in the aggregate, but highly correlated in one city (de Hartog et al. 2005; Boogaard et al. 2010). Our observed moderate correlation ( $r^2=0.43$ ;  $p=0.11$ ) between CDNP and PM<sub>2.5</sub> was similarly observed by a study measuring these exposures in homes burning biomass in India (Sahu et al. 2011).

While our results provide the suggestion that environmental nanoparticle aerosols should be monitored using nanoparticle-specific metrics, in addition to PM<sub>2.5</sub>, further research is needed. First, the full extent that nanoparticle exposure measurements (e.g. particle counts, surface area) are correlated with traditional, gravimetric exposure methodologies must be determined. This will involve measuring the diameter, density, surface area, and count of the airborne nanoparticles within various size fractions, along with particle mass measurements. Such a comprehensive analysis would expand on our observed moderate correlation between the nanoparticle and PM<sub>2.5</sub> measures. Further research is also needed to determine the components that are contributing to our measured nanoparticle exposures, such as polycyclic aromatic hydrocarbons (PAHs) and black carbon, which have been found to be elevated during coal burning in Xuanwei (Mumford et al. 1987, 1995).

Research into the development of methods to provide reliable assessments of exposure to nanoparticles is critical to an understanding of the potential health risk. The proper measurement of nanoparticles in epidemiological studies is important since exposure to fine particles is known to cause respiratory disease, and there is evidence that toxicity increases with decreasing particle size (Valavanidis et al. 2008). For example, urban air pollution in the ultrafine size fraction has been found to induce the greater levels of inflammation and oxidative damage than its larger counterparts (Li et al. 2003; Vinzents et al. 2005). It has

also been suggested that due to their small diameter, nanoparticles are capable of penetrating epithelial cells, entering the bloodstream from the lungs (BeruBe et al. 2007). Research to date on CDNP has focused on the toxicity associated with nanoparticles in diesel soot (Hirano et al. 2003), welding fume (McNeilly et al. 2004), carbon black (Renwick et al. 2004), and coal fly-ash (Gilmour et al. 2004a). Animal studies have found links with CDNP and adverse health effects in the lung, kidney, liver, and spleen. Specifically, pro-inflammatory effects have been seen in rats after inhalation exposure (Gilmour et al. 2004b). Similarly, when evaluating cell lines, pro-inflammatory gene transcription has been observed as a result of oxidative stress associated with CDNP (Shukla et al. 2000). In mice, carbon nanoparticles have been found to exacerbate airway inflammation and alter cytokine expression (Inoue et al. 2006). Combustion-derived nanoparticles have also been found to be capable of inducing cellular reactive oxygen species (ROS) production and toxic oxidative stress (Xia et al. 2006). Researchers have begun to explore the human health effects associated with CDNP (Mills et al. 2011; Sgro et al. 2011); however, much research is still needed.

We have taken initial steps to be able to integrate nanoparticle exposure assessments into epidemiological studies on IAP and malignant and non-malignant respiratory disease outcomes. We found that solid fuel burning elevates nanoparticle exposures, regardless of fuel or stove type used, and that these levels are reduced dramatically in the presence of a chimney. Given the moderate correlation between the nanoparticle and PM<sub>2.5</sub> measures, however, the extent that nanoparticle metrics yield information beyond those of traditional gravimetric metrics remains to be seen until further explored in a more comprehensive evaluation of exposure assessment metrics for fine particulates. Should nanoparticle metrics ultimately yield unique information about IAP exposure, the associations of these measures with adverse health outcomes may be explored in epidemiological studies.

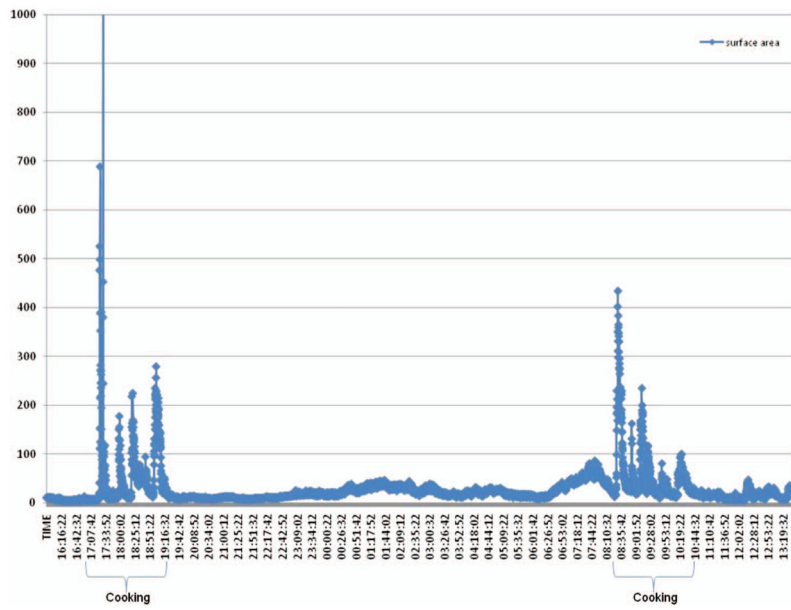
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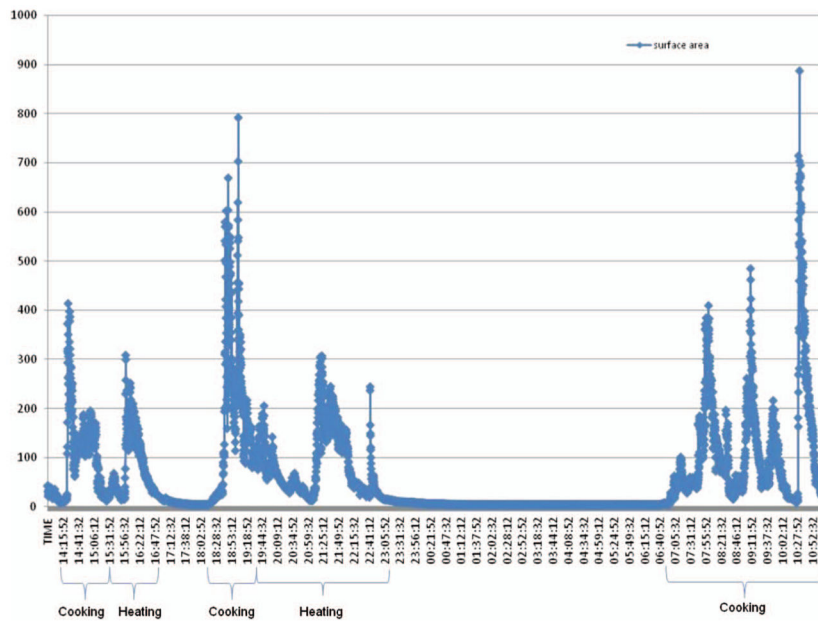
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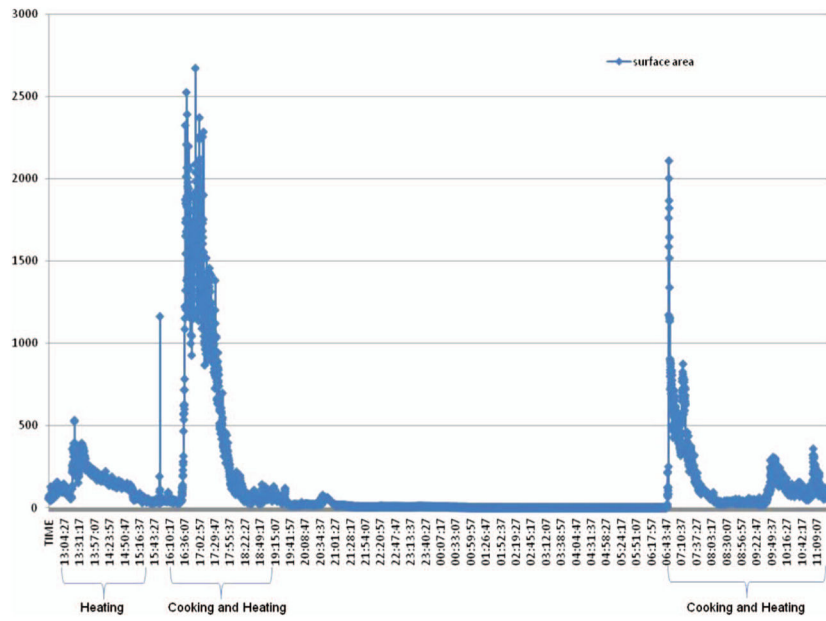




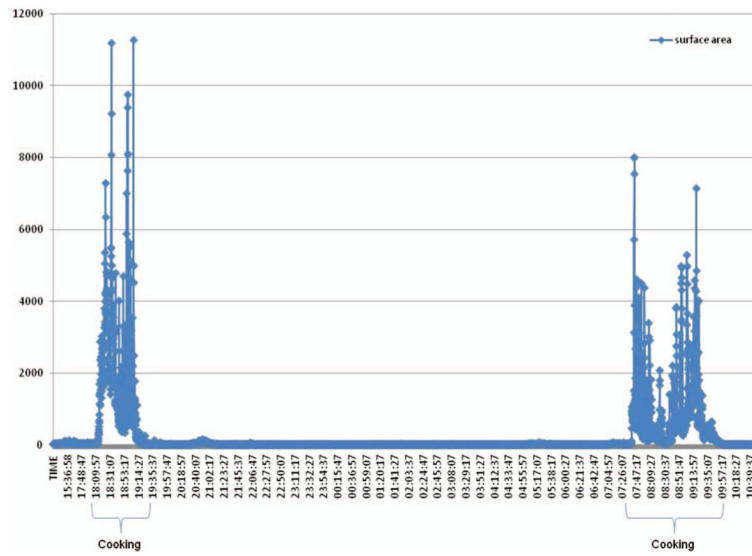
**Figure 1.** The surface area of combustion-derived nanoparticles (CDNP) ( $\mu\text{m}^2/\text{cm}^3$ ) in a home burning bituminous coal in a high stove with a chimney\*. Notes: \*y-axis represents surface area of CDNP ( $\mu\text{m}^2/\text{cm}^3$ ), with a range from 0 to 1000; x-axis represents time (h:min:s).



**Figure 2.** The surface area of combustion-derived nanoparticles (CDNP) ( $\mu\text{m}^2/\text{cm}^3$ ) in a home burning bituminous coal in a low stove with a chimney\*. Notes: \*y-axis represents surface area of CDNP ( $\mu\text{m}^2/\text{cm}^3$ ), with a range from 0 to 1000; x-axis represents time (h:min:s).



**Figure 3.** The surface area of combustion-derived nanoparticles (CDNP) ( $\mu\text{m}^2/\text{cm}^3$ ) in a home burning bituminous coal in a firepit without a chimney\*. Notes: \*y-axis represents surface area of CDNP ( $\mu\text{m}^2/\text{cm}^3$ ), with a range from 0 to 3000; x-axis represents time (h:min:s).



**Figure 4.** The surface area of combustion-derived nanoparticles (CDNP) ( $\mu\text{m}^2/\text{cm}^3$ ) in a home burning wood in a portable stove without a chimney\*. Notes: \*y-axis represents surface area of CDNP ( $\mu\text{m}^2/\text{cm}^3$ ), with a range from 0 to 12,000; x-axis represents time (h:min:s).

Table 1

Description of each home sampled for combustion-derived nanoparticles in Xuanwei and Fuyuan, China.

House	Stove type	Chimney present?	Fuel type	Duration of nanoparticle measurement (min)	Average nanoparticle measurement ( $\mu\text{m}^2/\text{cm}^3$ )	Average $\text{PM}_{2.5}$ ( $\mu\text{g}/\text{m}^3$ )	Figure
1	High stove	Yes	Bituminous coal	1320	24.2	108.7	-
2	High stove	Yes	Bituminous coal	600	13.8	45.6	-
3	High stove	Yes	Bituminous coal	1230	27.2	142.3	1
4	Low stove	Yes	Bituminous coal	1020	144.3	254.0	-
5	Low stove	Yes	Bituminous coal	1050	21.7	131.0	-
6	Low stove	Yes	Bituminous coal	1290	34.0	97.4	-
7	Low stove	Yes	Bituminous coal	1260	17.7	66.1	-
8	Low stove	Yes	Bituminous coal	1200	61.6	108.2	2
9	Portable stove	No	Bituminous coal	1215	490.4	483.7	-
10	Portable stove	No	Anthracite coal	1215	216.9	95.2	-
11	Firepit	No	Bituminous coal	1290	131.7	588.7	3
12	High stove	Yes	Anthracite coal	780	46.0	33.1	-
13*	Brick stove	No	Anthracite coal	435	269.1	72.6	-
14	Firepit and portable stove	No	Beehive**	717	104.9	122.3	-
15	Portable stove	No	Wood	1052	246.2	220.9	4

Notes:

\* sampling occurred during only one day;

\*\* a beehive is a honeycomb-shaped briquette.

**Table 2**

The average surface area of combustion-derived nanoparticles ( $\mu\text{m}^2/\text{cm}^3$ ) by stove and fuel characteristics, and solid fuel burning.

Characteristic	<u>Not during solid fuel burning</u>		<u>During solid fuel burning</u>	
	Mean	10th, 90th percentile	Mean	10th, 90th percentile
Stove type				
Low stove	11.6	2.2, 25.0	110.1	10.0, 263.6
High stove	11.6	1.8, 26.4	58.6	8.8, 96.8
Portable stove	21.9	10.6, 37.8	507.2	59.5, 1112.4
Firepit and portable stove	25.9	5.6, 23.6	168.9	37.1, 340.6
Brick stove	n/a*	n/a*	269.1	139.9, 467.8
Firepit	2.6	1.6, 5.3	184.9	9.5, 435.7
Chimney Present?				
Yes	11.6	2.0, 25.6	92.1	9.6, 221.9
No	19.9	2.1, 31.2	351.9	34.7, 796.6
Fuel type				
Bituminous coal	12.2	1.8, 25.9	227.7	9.5, 634.8
Anthracite coal	n/a*	n/a*	171.4	34.3, 392.1
Beehive	25.9	5.6, 23.6	168.9	37.1, 340.6
Wood	19.4	10.2, 34.2	969.1	32.1, 2679.9
All homes	14.0	2.0, 26.2	228.1	13.8, 540.8

Notes:

\* Not applicable. Three homes burned anthracite coal for heating and cooking during entire sampling period. One home used a brick stove, one a portable stove, and one a high stove with a chimney.