Fine Particle ($PM_{2.5}$) Composition of Indoor and Outdoor Air Samples Collected in Xuanwei County, China

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Abstract

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Indoor air pollution is a cause of many adverse health outcomes especially in developing countries where high levels of pollutants are generated by burning solid fuels for cooking and heating purposes. In China, the lung cancer rates are highest in Xuanwei County and the burning of coal in the home is thought to be a primary cause -- especially among women who are generally responsible for domestic cooking activities and have low smoking rates. Fine particulate matter ($PM_{2.5}$) samples were collected in Xuanwei County during the launch of an epidemiological study investigating the relationship between birth outcomes and $PM_{2.5}$ exposure in collaboration with Kunming Medical University. PM$_{2.5}$ was measured in nine households across three villages for three consecutive days. At each home, two 12-hr samples (approximating daytime and nighttime periods) were collected indoor concurrently with a 24-hr outdoor measurement. Continuous fine particle sampling was also conducted indoors using a low-cost particle sensor. Samples were analyzed for total PM$_{2.5}$ mass concentration and selected components (levoglucosan, 1-nitropyrene, and benzo[a] pyrene) indicative of source and potential carcinogenicity. Concentration of PM$_{2.5}$ mass and selected components varied considerably between time periods sampled indoors (daytime range 25.2 – 4371 μg/m^3; nighttime range 21.7 – 156 μg/m^3) and between indoor and outdoor compartments (indoor range 23.4 – 2264 μg/m^3; outdoor range 16.4 – 41.1 μg/m^3). Continuous PM$_{2.5}$ particle monitoring confirmed the episodic nature of the fine particle generation with peaks observed mainly during typical hours of cooking activities. None of the outdoor samples were above Chinese regulatory standards for ambient air quality. Several indoor concentrations of PM$_{2.5}$ were well above recommended WHO guidelines and benzo[a]pyrene (BaP) concentrations were above Chinese National Criteria standards. Future studies may benefit from longitudinal sampling to assess temporal variations that may exist within households, particularly those that have choices in cooking and heating fuels.

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Background and Significance

Air Pollution Exposure

Air pollution is a complex mixture of many gaseous chemicals and solid components, known as particulate matter (PM), that vary in size and composition. Major components of PM include metals, ions (sulfate, nitrate), organic compounds, minerals, reactive gases, and biologic materials (Valavanidis et al., 2008; Kampa et al., 2008). PM is classified as coarse, fine, or ultrafine particles depending on their mean aerodynamic diameter. Coarse particles (PM_{10}) have a diameter larger than 2.5 micrometers (µm), fine particles ($PM_{2.5}$) have a diameter less than 2.5 µm, and ultrafine particles are a subset of fine particles that have a diameter less than 0.1 µm. Air pollutants can be naturally or anthropogenically derived. Major sources of natural air pollutants include forest fires, dust storms and volcanic eruptions, but these occurrences are sporadic and transient and are not associated with long-term public health outcomes (Godish, 2004). Anthropogenic air pollution comes from the combustion of biomass and fossil fuels, primarily associated with transportation, industrial processes, waste disposal, and domestic burning. Air pollutants that are generated at the time of combustion are termed primary pollutants. Primary pollutants can react with each other or atmospheric chemicals to produce secondary air pollutants, including ozone and particulate matter (Godish, 2004).

Humans are predominantly exposed to air pollutants via inhalation. The respiratory system is composed of the upper respiratory tract (oral and nasal cavities, pharynx, larynx) and the lower respiratory tract (trachea, primary bronchus and lungs). The average adult respiration rate (at low activity) is 1.5 m^3/hour resulting in over 600,000 m^3 of inhaled air over a 70 year lifetime (USEPA, 2011). When air containing PM is inhaled, the larger particles impact (are trapped) in the upper airways where they are usually swallowed or expelled. In contrast, fine particles are capable of penetrating deeply into lower airways and interacting with the alveolar sacs (the site of gas exchange) and potentially pass directly into the bloodstream (Mader, 2008). For this reason, fine particles are termed 'inhalable' or 'respirable' and are of primary concern for human health.

The term "air pollution" generally describes the ambient (outdoor) air pollution measured at a fixed location and averaged over a period of time. Estimates of regional air quality are made by interpolating between the network of fixed air pollution monitoring stations for that time period. Ambient air pollution is measured and regulated as though it is evenly distributed across the region it is intended to represent. Many studies have found, however, that air pollution level can vary considerably in space, even over a very small area (Jerrett et al., 2005; Beeleen et al., 2009). Similarly, indoor air pollution (IAP) levels can vary dramatically within a building or between buildings, even in the same community. This variation results from differences in building characteristics and from the activities of the building's occupants. Combustion of fossil and biomass fuels inside the home (including smoking) are the primary sources of IAP and can substantially raise the concentration of IAP inside. For this and other reasons related to particle deposition and indoor air circulation, PM concentrations indoors are generally higher than outdoors (USEPA, 2004).

Combustion indoors can influence the ambient air pollution levels in the surrounding area, especially in stagnant air conditions as can be seen with woodsmoke accumulation in areas during an inversion. Chafe et al. (2014) quantified the contribution of household biomass combustion to outdoor $PM_{2.5}$ levels. They estimated that household biomass combustion attributed to cooking with solid fuels contributed to 10% of ambient $PM_{2.5}$ in East Asia (including China) and 12% of the global ambient $PM_{2.5}$. However, this study was examining the biomass combustion produced from cooking only, suggesting that estimates would be higher had indoor biomass combustion for heating also been considered.

Combustion indoors will cause elevated levels of $PM_{2.5}$ during and for some time after the end of burning. Depending on the frequency and intensity of domestic burning activities, indoor air pollution--also called household air pollution (HAP)--levels can be alarmingly high, exposing residents to unhealthful levels of PM. It is estimated that 3 billion people worldwide use some form of solid, carbon-based fuel (wood, dung, crop residues, charcoal, coal) as their primary source of energy for cooking and heating (Balakrishnan et al., 2011). Indoor burning is often observed as a burden borne by poor populations who rely on solid fuels and traditional stoves for cooking and heating. A survey performed by Ellegard (1996) suggests lifetime exposure to indoor air pollutants generated by cooking fuels is the product of the fuel type, duration of daily cooking, intensity of burning, and number of years spent cooking indoors. A HAP exposure model used in the World Health Organization's Global Burden of Disease (GBD) project in 2010 estimated daily average $PM_{2.5}$ exposures of 285 $\mu g/m^3$, 337 $\mu g/m^3$, and 204 $\mu g/m^3$ for children, women, and men, respectively, in India (Balakrishnan et al., 2014). These results suggests that gender and age may be additional determinants of exposure to indoor air pollution in certain households and communities.

For many people, the majority of their time is spent indoors at the home. Unfortunately, not much is known about the air pollution levels in the home as sampling is not easily or consistently conducted, nor is it regulated. In 2001 Koistinen et al. compared $PM_{2.5}$ concentrations indoors and outdoors to the personal air pollution exposure of 201 people sampled in a Finnish study and found that indoor concentrations were the best predictor of personal $PM_{2.5}$ exposure.

This suggests that understanding more about the air pollution sources and concentrations inside the home is important to understanding personal exposure to $PM_{2.5}$ air pollution.

Several studies have shown that inhalation exposure to $PM_{2.5}$ is associated with increased morbidity and mortality from respiratory, cardiovascular, and autoimmune diseases, such as lung cancer (Pope et al., 1995; Pope et al., 2002; Brook et al, 2010; IARC, 2010; Koike et al., 2014; WHO, 2014). The World Health Organization (WHO) estimated that 4.3 million people died prematurely in 2012 as the result of illnesses related to exposure to indoor air pollution and that 50% of all premature deaths among children under 5-years-old were specifically caused by the inhalation of household products of incomplete combustion (WHO 2012; WHO, 2014). More recently, it was estimated that 17% of the world's annual premature lung cancer deaths in adults are attributable to household burning of solid fuels (IARC, 2010).

Health standards and governmental regulations for air pollution typically use an absolute measurement of $PM_{2.5}$ concentration with consideration for only 1 or 2 components such as lead or benzo[a]pyrene (BaP), yet this measurement does not capture how the composition of $PM_{2.5}$ is affecting health outcomes. The toxicity of inhalable PM depends on its size, surface area, and chemical composition (Nel, 2005; Cassee et al., 2013). Distinct adverse health outcomes have been associated with particulate matter constituents. Several *in vitro* and *in vivo* studies have concluded that toxicity induced from exposure to PM is related to its chemical composition, suggesting that cytotoxic and proinflammatory effects are associated with chemical and physical properties of its constituents (Zelikoff et al., 1993; Erdal et al., 1999; Osornio-Vargas et al., 2003; Steenhof et al., 2011). A review by Valavanidis et al. (2008) further distinguishes that organic compounds contribute to mechanisms of cytotoxicity whereas transition metals contribute to the initiation of oxidative DNA damage. In animal studies, polycyclic aromatic hydrocarbons (PAHs), such as BaP and 1-Nitropyrene, are known to produce carcinogenic, mutagenic, and genotoxic effects (Deutsch-Wenzel et al., 1983; Kim et al., 2005). The International Agency for Research on Cancer (IARC) confirms that emissions from coal combustion are known to be carcinogenic, emitting several toxic air pollutants including fluorine, arsenic, lead, selenium, and mercury, but while the toxicities of many individual PM constituents are well studied, the health effects associated with exposure to characterized mixtures of air pollutants is still unclear (Naeher et al., 2007).

An analogy can be made to the health effects of smoking cigarettes or inhaling traffic fumes. Each scenario represents exposure to a complex mixture of components with potential health consequences. In the case of cigarette and diesel exhaust exposure, it was the combination of the released chemicals that were eventually regulated and classified by IARC as human carcinogens. In additional, animal studies have demonstrated that inhalation of metals oxides (arsenic, cadmium, lead, and selenium) can cause acute toxicity and have the potential to accumulate in the liver, kidney, and pulmonary tissue (Rhoads & Sanders, 1985; Hughes, 2002). The presented toxicology data along with emerging epidemiological findings continue to suggest that PM composition is an important measurement for discerning health implications of air pollution exposure.

Air Pollution & Lung Cancer in Xuanwei, China

In China, nearly twice as many premature deaths are caused by exposure to indoor sources of fuel combustion compared to outdoor air pollution sources (Zhang and Smith, 2007). In most rural communities, traditional indoor burning practices are still employed and it remains customary for women to be responsible for the majority of cooking and housekeeping activities. As a result, it is the women in rural China who tend to be exposed to high concentrations of HAPs. Xuanwei

County, located in Yunnan province, has the nation's highest incidence of lung cancer, especially for women, regardless of smoking status, and the domestic burning of coal has been long implicated as the cause (Barone-Adesi et al., 2012; Chapman et al., 1988; He et al., 1991; Kim et al., 2014; Lan et al., 2002; Mumford et al., 1987; Pope et al., 2002).

In Xuanwei County, coal is the most common solid fuel used for heating and cooking, although alternative fuel sources--wood, corn husks, and tobacco plant stems--do exist. Multiple studies conducted in the 1990's found that coal smoke exposure was associated with lung cancer (Wu-Williams et al., 1990; Xu et al., 1991; Tao et al., 1991; Smith et al., 1994). However, not all households in Xuanwei County use coal as a fuel source. Many use biomass sources such as wood, agricultural waste, and dried dung. In contrast to coal smoke, exposure to biomass smoke is associated with nonmalignant disorders, including acute lower respiratory illness in children, chronic respiratory illness, pulmonary hypertension, heart disease, lung function changes and possibly eye disorders and adverse pregnancy outcomes (Sandoval et al., 1993; Ellegard et al., 1996; Smith et al., 2000).

In 2009, Lv et al. conducted air sampling in Xuanwei and Fuyuan County homes burning coal and found that indoor concentrations of BaP--a known carcinogenic particle-bound PAH--were, on average, 95-times higher than the indoor maximum permissible risk level of 1.0 ng/m^3 as regulated by Chinese standard GB/T18883-2002. The investigators also noted that bituminous coal emitted more PAHs than anthracite coal or wood and so the specific fuel used may explain some of the variation in concentrations. Around the same time, Lan et al. (2008) and Lin et al. (2012) were observing that lung cancer rates among bituminous coal users varied (up to 20-fold) by the patient's address in Xuanwei County. Since then, researchers have observed that residents of Xuanwei County tend to purchase coal from the coal mine nearest to them, which can be either anthracite (“smokeless”) or bituminous (“smoky”) coal (Downward et al., 2014). Measurements of particle-bound PAHs emitted from bituminous coal burning collected by Downward et al. (2014) indicated that mean personal exposure to BaP was nearly two magnitudes higher than the Chinese national criteria for BaP exposure in indoor environments (measured: 74.4 ng/m^3; standard: 1 ng/m^3). Differences in the coal composition--either due to geological formation or at-home coal preparation (such as briquetting)--were suggested to be causal factor in the unusual lung cancer rates observed in this region of China (Downward et al., 2014).

Opportunity for Research

Research studies on fine PM exposure are increasingly using continuous monitoring sensors that innumerate individual particles in hopes of understanding how PM levels vary throughout the exposure period. While these studies will help elucidate gaps in understanding temporal aspects of exposure, these studies do not take into consideration the source--by determining the composition--of the PM. Measuring the frequency and amplitude of fine PM exposure could potentially miss crucial information regarding composition and toxicity of the particles and may skew the relationship between exposure to fine PM and adverse health outcomes.

Based on what we know about the components of incomplete combustion, it is reasonable to assume that the presence and abundance of carcinogenic compounds found in indoor air would influence the toxic response of inhalable PM. It is important to consider that composition of PM may explain the elevated lung cancer rate observed in Xuanwei County. Ongoing interventions addressing solid fuel use (including coal) in China are trying to reduce exposure to indoor air pollution by changing the type of fuel being used and updating stove designs and technologies to include more complete combustion and improved ventilation (Zhang and Smith, 2007). The extent of interventions and adoption rates are not known in Xuanwei, but based on the published literature included herein solid fuels and are known to still be used in this region for cooking and heating. Theoretically, complete combustion of carbon-based fuels should yield only water and carbon dioxide, but coal or biomass burned in simple household stoves will convert up to 10 - 38% of the carbon fuel into potentially harmful products of incomplete combustion (PICs) (Zhang and Smith, 2007). To truly understand personal exposure to air pollution, further characterization of PM composition in Xuanwei, China is needed.

A commencing epidemiological birth cohort study is using fine particle count information derived from instruments called PUWPs (Personal University of Washington Particle monitors) developed by Dr. Edmund Seto of the University of Washington's Department of Environmental and Occupational Health Sciences. In June 2014, Dr. Seto was invited to meet with collaborators from Kunming Medical University in Xuanwei, China and accompany them on site visits to several of the villages where recruiting for the study would occur. This trip afforded the opportunistic collection of indoor and outdoor air samples from rural villages in Xuanwei County from which $PM_{2.5}$ mass and composition could be analyzed. Due to the opportunistic nature of this sampling scheme, several resource and time constraints were imposed upon the study design. The original sampling was planned as paired 24-hr samples inside and outside of approximately 12-15 homes in 3-4 villages in rural Xuanwei County. The intent was to sample a cross section of homes representing the spectrum of socioeconomic status and cooking/heating fuel use.

Specific Aims

This study aims to analyze samples of indoor and outdoor $PM_{2.5}$ for total mass and component concentrations indicative of source and potential toxicity. It is hypothesized that $PM_{2.5}$

composition--in addition to concentration--is highly variable among households in rural villages in Xuanwei County relating to occupant habits and fuel used for cooking and heating; information not gathered by studies of PM counts and mass alone. Specifically, this opportunistic study attempts to:

1. Measure $PM_{2.5}$ and select components indicative of source or carcinogenicity – levoglucosan (Lg), 1-nitropyrene (1-Np), BaP -- inside and outside of households in villages in Xuanwei County, China;
2. Report the enrichment of $PM_{2.5}$ and components by time period sampled indoors or by compartment sampled using ratios of mass concentration and mass fraction comparing day versus night indoors and indoor versus outdoors at each household; and
3. Compare the two time periods sampled indoors (daytime vs nighttime) and the compartments sampled (indoor vs outdoor) for each component across all households using non-parametric statistical tests.

Materials and Methods

Study Area

This study collected data from rural Xuanwei County, Quijing Prefecture, Yunnan Province, China. Samples were collected over a 3-day period commencing on the 25th of July, 2014 and ending on the 27th of July, 2014 (Figure 1). Each village was sampled for 24 hours starting with Jiu Bao, followed by Qi Long, and ending with Tang Tang. The three sampling locations in each village were selected to represent the spectrum of living conditions, represented in each village and generally included a household relying on solid fuel combustion for cooking, a household that primarily used electricity, but had the option to use solid fuel, and a household that only used electricity.

The three villages varied in their setting and proximity to major roads and other PM sources. Jiu Bao is located on one side of a valley on the main road into and through the prefecture. This road experiences periodic heavy diesel traffic and the village residences are generally clustered around this road. Qi Long village, in contrast, is off of the main transit route and sits in a quiet valley with only local traffic. Tang Tang village is the commercial center for the area, appears to have the most infrastructure, the largest population and the most traffic of the three villages. It is located in a river valley at the confluence of two tributaries.

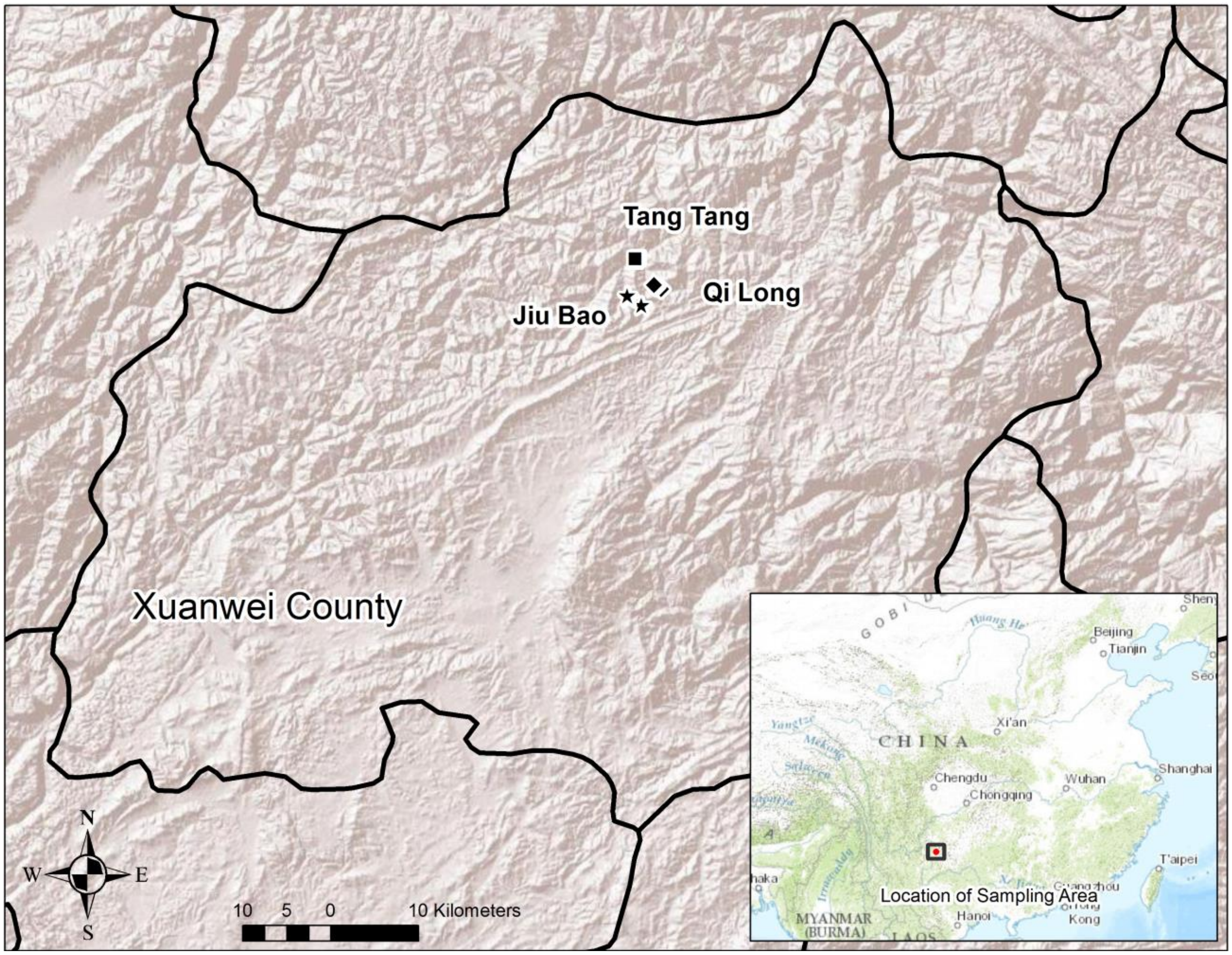


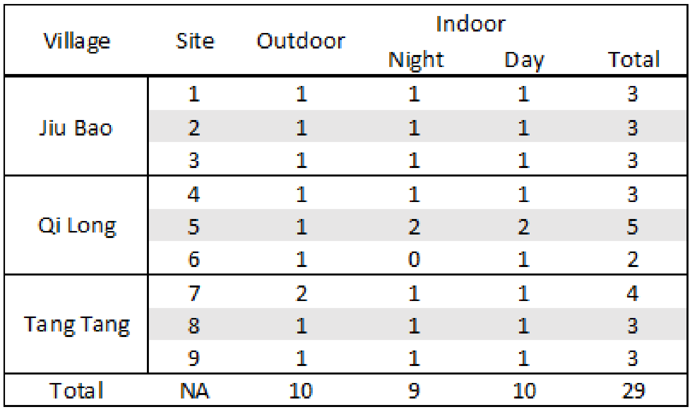
Figure 1. $PM_{2.5}$ sampling villages in northern Xuanwei County, Yunnan Province, China

Sampling and Analysis

Teflon filters (Pall Scientific, 37mm, 1µm pore size) were allowed to off-gas and conditioned to constant temperature and humidity in an environmental chamber, located at the University of Washington, before being weighed by a microbalance (Metler-Toledo Inc., Columbus, OH) such that two successive weighings were within 5 µg. The filters were then assembled into Harvard Personal Exposure Monitors (HPEMs) with an aerodynamic cut point of 2.5 µm at a flow rate of 1.8 L/min. The assembled monitors were wrapped in aluminum foil, sealed in plastic bags and transported to the three rural Chinese villages.

These samples were set up in pairs of indoor and outdoor samples at three residences per village, where one village was sampled on each of three successive days. The samples were run at flow rates of 1.8 L/min (±5%) using battery powered personal sampling pumps (TSI SidePak). Flow rates were verified by a DryCal (Defender 530) primary gas flow standard. The airflow was measured before and after sampling and averaged to determine the air volume sampled. Samples were collected in two 12-hour periods for indoor samples (Day: ~7:00 am to 7:00 pm; Night: ~7:00 pm to 7:00 am) and one 24-hour period for outdoor samples at each site. The number of samples collected are presented in Table 1.

Table 1. Number of samples collected at each sampling location.



Village	Site	Outdoor	Indoor		
			Night	Day	Total
Jiu Bao	1	1	1	1	3
	2	1	1	1	3
	3	1	1	1	3
Qi Long	4	1	1	1	3
	5	1	2	2	5
	6	1	0	1	2
Tang Tang	7	2	1	1	4
	8	1	1	1	3
	9	1	1	1	3
Total	NA	10	9	10	29

To evaluate the consistency of results, duplicate samples were collected at one indoor location (site 5) and one outdoor location (site 7). Blank samples were designated in one outdoor and one indoor environment to ensure filters were not being contaminated during assembly or transport to the field. Blank samples were exposed to the ambient environment for 5 seconds before being resealed.

Continuous $PM_{2.5}$ particle monitoring was also conducted in each household of this study using a Portable University of Washington Particle (PUWP) sampler developed by the Seto lab and employing a Shinyei fine particle sensor mounted to a microprocessor and written to microSD storage media. The design of the PUWP is essentially the same as that used in a recent study of ambient air quality in Xi'an, China, and technical details on the PUWP can be found in that study (Gao et al., 2015). These sensors were deployed in approximately the same location as the PTFE filter cassettes and pumps and for the same duration. These units collect data in units of sensor response which can be converted to $PM_{2.5}$ particle counts and then to mass, but for the purposes of this study only serve to indicate time periods of $PM_{2.5}$ generating activity and relative magnitude of fine particle generation and so are left in the native sensor response units.

Upon completion of sampling, the filters were wrapped in aluminum foil, placed in sealed plastic bags, and returned to the environmental chamber located at the University of Washington. Filters were conditioned back to constant temperature and humidity before being weighed in the same manner as previously mentioned. The filters were then shipped to an outside lab, Cooper Environmental Services LLC, (Beaverton, OR) for XRF analysis of metals (results not presented) according to EPA compendium IO 3.3 (USEPA, 1999). After this non-destructive analysis, the filters were returned to the UW where they were analyzed for high molecular weight (particle bound) PAHs. Filters were extracted with dichloromethane by closed-vessel microwave-accelerated liquid extraction (USEPA, 2007). The microwave (CEM MARS 5) method was provided by CEM (CEM Corp.). The analysis was done by stable-isotope dilution GC-MS analysis (USEPA, 1989). Levoglucosan was analyzed using the method as described by Simpson *et al.* (2004). Specific nitro-PAHs (1-nitropyrene, 2-nitropyrene, and 2-nitrofluoranthene) were analyzed according to the method described by Miller-Schultz *et al.* (2007).

The XRF results provided mass/cm^2 of filter for 48 metals. The nitro group PAH analysis provided mass/filter for 1-Nitropyrene, 2-Nitropyrene, and 2-Nitrofluoranthene. The additional high molecular weight PAH analysis provided mass/sample for 10 PAHs; fluoranthene, pyrene, benz[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, dibenz[a,h]anthracene, and benzo[g,h,i]perylene.

Statistical Analysis

Compilation and data analysis were performed in R (version 3.2.2). Results from each analysis were combined into a single data set by the filter ID, the unique identifier for each sample. A time-weighted average PM$_{2.5}$ concentration was calculated by dividing the change in filter weight in micrograms by the volume of air sampled through the filter. Time-weighted average concentrations were also calculated for the results of each subsequent analysis after converting the reported mass per filter to micrograms. Data distributions were evaluated with descriptive statistics. Comparisons of concentrations between groups--indoor time period (day vs night) and location (indoor vs outdoor)--were evaluated using the paired nonparametric Wilcoxon Signed-Rank test. We compared outdoor concentrations between villages using the nonparametric Kruskal-Wallis test. For statistical tests an alpha of 0.05 was used, and p-values less than 0.05 were deemed significant.

Results

Samples Collected and Quality Control

The concentration and composition of indoor and outdoor $PM_{2.5}$ from the nine air sampling locations are reported here. In order to ensure that the filters did not become overloaded by high indoor $PM_{2.5}$ concentrations, sampling in the indoor environment were split into two 12-hr sampling periods; daytime starting from ~ 07:00 and ending ~ 19:00 and nighttime starting from ~ 19:00 and ending at ~ 07:00 the next morning. Site 6 experienced a pump failure during the indoor nighttime sample and so no values are available for this sample. Accounting for the loss of one sampling pump, a total of 29 samples were collected: 19 indoor (9 nighttime, 10 daytime) and 10 outdoor. All samples (n = 29) collected sufficient mass to calculate $PM_{2.5}$ concentration with > 5µg difference between post and pre-weights.

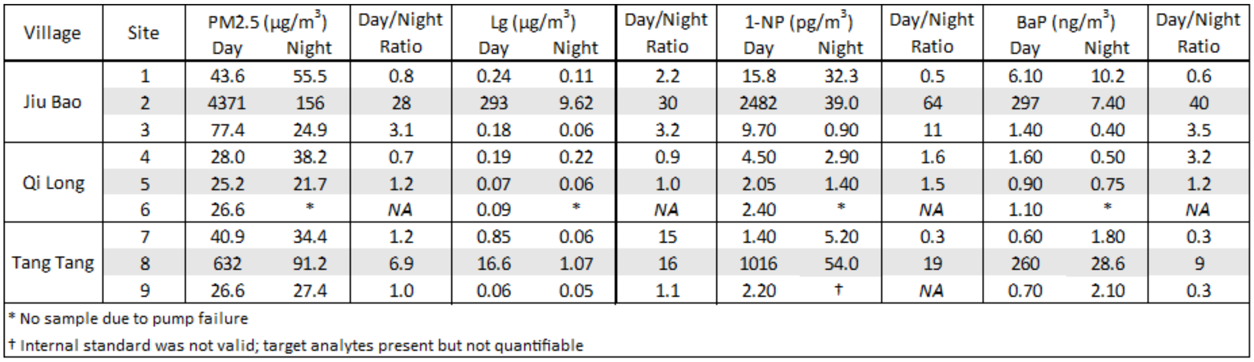
For the composition analysis, all outdoor samples were above the limit of quantitation (LOQ) for each analyte except for the sample at Site 5 for dibenz(a,h)anthracene. Indoor daytime and nighttime samples were all above the LOQ with the exception of: BaP at Site 3; benzo[k]fluoranthene at Sites 3, 4, 7, 9; Lg and 1-Np at Sites 3, 5, 7, 9; fluoranthene at Sites 3, 4, 5, 7, 9; and pyrene, benz[a]anthracene, chrysene, and dibenz[a,h]anthracene at Sites 3, 4, 5, 6, 7, and 9.

Precision was assessed by collecting duplicate outdoor samples at Site 7 and duplicate indoor samples at Site 5. The two samples collected from Site 7 agreed well with concentrations for $PM_{2.5}$, Lg, and other high molecular weight (HMW) PAHs with a 5, 9, and 6% difference, respectively. The two Site 7 samples did not agree as well for nitro-PAHs with 45% relative difference between these samples. Duplicate indoor samples collected at Site 5 showed similar precision, but with greater variation between results for these samples. Duplicate samples from Site 5 collected at night had good agreement for Lg and other HMW PAHs, with observed relative differences of 11 and 10%, respectively. Other components showed poorer agreement with $PM_{2.5}$ and nitro-PAHs observed relative differences between the two nighttime samples of 25 and 125% respectively. Duplicate indoor samples collected at Site 5 during the day had good agreement for $PM_{2.5}$ and Lg with relative differences of 3 and 11% difference, respectively. Less agreement was observed in daytime samples at Site 5 for nitro-PAHs and other HMW PAHs with relative differences of 46 and 24% between samples, respectively.

Indoor PM2.5 Mass Concentration

Here, results from total $PM_{2.5}$ and three selected components are reported; results from component analysis not listed here are provided in Appendix 1. Lg is presented to represent cellulose combustion, 1-Np is presented to represent diesel fuel combustion, and BaP is presented due to its known toxicity as a human carcinogen. Table 2 provides total $PM_{2.5}$ mass concentration as well as concentrations of Lg, 1-Np, and BaP from indoor samples. In general, daytime concentrations were more variable than nighttime concentrations. Daytime concentrations were typically greater than nighttime concentrations for all components.

Table 2. Total $PM_{2.5}$, Lg, 1-Np and BaP mass concentrations indoors for day and night sampling periods including day to night concentration ratios by village and site number.



Village	Site	PM2.5 ($\mu g/m^3$) Day	PM2.5 ($\mu g/m^3$) Night	Day/Night Ratio	Lg ($\mu g/m^3$) Day	Lg ($\mu g/m^3$) Night	Day/Night Ratio	1-NP (pg/m^3) Day	1-NP (pg/m^3) Night	Day/Night Ratio	BaP (ng/m^3) Day	BaP (ng/m^3) Night	Day/Night Ratio
Jiu Bao	1	43.6	55.5	0.8	0.24	0.11	2.2	15.8	32.3	0.5	6.10	10.2	0.6
	2	4371	156	28	293	9.62	30	2482	39.0	64	297	7.40	40
	3	77.4	24.9	3.1	0.18	0.06	3.2	9.70	0.90	11	1.40	0.40	3.5
Qi Long	4	28.0	38.2	0.7	0.19	0.22	0.9	4.50	2.90	1.6	1.60	0.50	3.2
	5	25.2	21.7	1.2	0.07	0.06	1.0	2.05	1.40	1.5	0.90	0.75	1.2
	6	26.6	*	*NA*	0.09	*	*NA*	2.40	*	*NA*	1.10	*	*NA*
Tang Tang	7	40.9	34.4	1.2	0.85	0.06	15	1.40	5.20	0.3	0.60	1.80	0.3
	8	632	91.2	6.9	16.6	1.07	16	1016	54.0	19	260	28.6	9
	9	26.6	27.4	1.0	0.06	0.05	1.1	2.20	†	*NA*	0.70	2.10	0.3

* No sample due to pump failure

† Internal standard was not valid; target analytes present but not quantifiable

Total $PM_{2.5}$

For total $PM_{2.5}$, daytime concentrations ranged from 25 to 4371 $\mu g/m^3$ at Sites 5 and 2, respectively. Nighttime concentrations ranged from 22 to 156 $\mu g/m^3$ at Sites 5 and 2, respectively. Daytime concentrations were higher than nighttime concentrations at the majority (62.5%) of the sites: higher for Sites 2, 3, 5, 7, and 8, but equal at Site 9. Daytime concentrations were lower at Sites 1 and 4. Overall, daytime concentrations were not significantly different than nighttime concentrations ($p = 0.3$). Day to night concentration ratios are all calculated as being close to 1, except for Sites 2, 3 and 8, which have ratios of 28, 3.1 and 6.9, respectively. Ratios greater than 1 indicate the magnitude by which daytime concentrations exceed nighttime concentrations at these sites.

Levoglucosan

Measurements of indoor Lg concentrations followed a similar pattern to total $PM_{2.5}$ (Table 2). Daytime concentrations of Lg ranged from 0.06 µg/m^3 at Site 9 to 293 µg/m^3 at Site 2. Nighttime concentrations of Lg ranged from 0.05 µg/m^3 at Site 9 to 9.62 µg/m^3 at Site 2. In general, daytime concentrations were higher at all sites except 4 and 5 where the concentrations were approximately equal. Lg measurements collected during the day were significantly higher than samples collected at night ($p = 0.039$). Overall, the day to night concentration ratios were higher than the day to night ratios for total $PM_{2.5}$. It is noteworthy that Sites 1, 7, and 8 had substantially high indoor levels of Lg, with Lg day to night ratios greater than their total $PM_{2.5}$ ratios.

1-Nitropyrene

Concentrations of 1-Np were much lower than Lg (on the order of picograms), but followed a similar relative concentration pattern, with Sites 2 and 8 having the highest concentrations during both day and night (Table 2). Daytime concentrations ranged from 2 to 2482 pg/m^3 at Sites 5 and 2, respectively. Nighttime concentrations ranged from 0.9 to 54 pg/m^3 at Sites 3 and 8, respectively. However, nitro-PAH results from Site 9's nighttime were inconclusive due to a laboratory error during analysis. Daytime concentrations of 1-Np were higher than nighttime concentrations at Sites 2, 3, 4, 5 and 8, but nighttime concentrations were higher at Sites 1 and 7. Compared to total $PM_{2.5}$, day to night concentration ratios for 1-Np were generally higher, with the highest at Sites 2 and 8. For instance, the daytime concentration of 1-Np at Site 3 was 11-fold higher than the nighttime concentration, differing greatly from its ratio for total $PM_{2.5}$ or Lg. On the other hand, the day to night 1-Np concentration ratio at Site 1 was similar to total $PM_{2.5}$ (0.5 compared to 0.8, respectively), but lower than the ratio observed for Lg (0.5 compared to 2.2, respectively). At Site 7, the day to night 1-Np concentration ratio was also lower than the day to night Lg concentration ratio (0.3 compared to 15, respectively). Across the entire dataset, daytime 1-Np concentrations were higher than nighttime concentrations at 71.4% of the sites, but not significantly so ($p = 0.37$).

Benzo[a]pyrene

Daytime concentrations for BaP were lowest at Site 7 and highest at Site 2, ranging from 0.6 and 297 ng/m^3, respectively. Nighttime concentrations were lowest at Site 3 and highest at Site 8, ranging from 0.4 and 28.6 ng/m^3, respectively. Across the entire dataset, the day to night BaP concentration ratio was greater than 1 at most sites with the exception of 1, 7, 9, which had ratios of 0.6, 0.3, and 0.3, respectively. The day to night BaP concentration ratio was highest at Sites 2 and 8, with ratios of 40 and 9, respectively. Overall, the daytime indoor concentrations of BaP were not significantly different than the nighttime indoor concentrations ($p = 0.74$).

Indoor vs Outdoor PM2.5 Concentration

To facilitate comparison between the indoor and outdoor environments, the two 12-hour indoor samples at each site were averaged to create a 24-hour mean indoor concentration at each site (with the exception of Site 6 as previously noted). Indoor and outdoor data are presented for total PM$_{2.5}$, Lg, 1-Np, and BaP in Table 3.

Table 3. Total $PM_{2.5}$, Lg, 1-Np and BaP mass concentrations for indoor and outdoor compartments including indoor to outdoor concentration ratios by village and site number.

Village	Site	Total $PM_{2.5}$ ($\mu g/m^3$)		In/Out	Lg ($\mu g/m^3$)		In/Out	1-NP (pg/m^3)		In/Out	BaP (ng/m^3)		In/Out
		Indoor	Outdoor	Ratio	Indoor	Outdoor	Ratio	Indoor	Outdoor	Ratio	Indoor	Outdoor	Ratio
Jiu Bao	1	49.5	25.3	2.0	0.17	0.10	1.7	24.1	9.50	2.5	8.15	2.70	3.0
	2	2264	24.8	91	151	0.18	823	1260	3.80	332	152	1.30	117
	3	51.1	23.0	2.2	0.12	0.10	1.2	5.30	3.10	1.7	0.90	1.90	0.5
Qi Long	4	33.1	19.9	1.7	0.20	0.12	1.7	3.70	5.00	0.7	1.05	1.40	0.8
	5	23.4	16.4	1.4	0.07	0.06	1.2	1.73	3.50	0.5	0.83	1.00	0.8
	6	26.6	17.2	1.6	0.09	0.05	1.7	2.40	3.30	0.7	1.10	1.00	1.1
Tang Tang	7	37.6	30.3	1.2	0.46	0.08	6.0	3.30	3.10	1.1	1.20	1.75	0.7
	8	362	41.1	8.8	8.82	0.20	43	535	11.9	45.0	144	6.30	23
	9	27.0	29.4	0.9	0.05	0.05	1.1	2.20	7.30	0.3	1.40	1.80	0.8

Total $PM_{2.5}$

Indoor total $PM_{2.5}$ concentrations were highly variable compared to outdoor concentrations (Table 3). Indoor concentrations ranged from 23 to 2264 $\mu g/m^3$, whereas outdoor concentrations ranged from 16 to 41 $\mu g/m^3$. The highest concentrations were observed at Sites 2 and 8--as seen previously in the comparison of indoor samples collected during the day and night. The outdoor samples did not follow this same trend. Sites 7 and 8 were observed to have the highest outdoor concentration. The indoor to outdoor $PM_{2.5}$ concentration ratios were all greater than 1 with the exception of Site 9, where the outdoor was slightly higher than the indoor concentration. The highest indoor to outdoor ratio was observed at Sites 2 and 8, where the indoor samples were 91- and 8.8-fold higher than their corresponding outdoor samples. Overall, total $PM_{2.5}$ concentration was significantly higher indoors than outdoors ($p = 0.008$). Outdoor concentrations varied significantly between villages ($p = 0.03$) and were highest in Tang Tang Village and lowest in Qi Long village.

Levoglucosan

Indoor and outdoor Lg concentrations were highest at Site 2 (151 and 0.18 $\mu g/m^3$, respectively) and lowest at Site 9 (0.05 and 0.05 $\mu g/m^3$, respectively). Indoor concentrations of Lg followed a similar pattern to that of total $PM_{2.5}$, with Sites 2 and 8 having the highest Lg concentrations both indoors and outdoors (Table 3). The indoor to outdoor Lg concentration ratios ranged between 1 and 2, except for Sites 2, 7, and 8. At these sites, the indoor Lg concentration was appreciably higher than outdoor Lg concentrations, with ratios of 823, 6, and 43, respectively. Indoor to outdoor ratios of Lg concentration followed a similar pattern to total $PM_{2.5}$ but on a different order of magnitude, where enrichment of Lg indoors was many-fold higher at Sites 2 and 8 than was observed for total $PM_{2.5}$. Overall, the Lg concentration indoors was significantly higher than outdoors ($p = 0.004$) as was observed for total $PM_{2.5}$. In contrast to $PM_{2.5}$, outdoor Lg concentrations were not significantly different among villages ($p = 0.53$).

1-Nitropyrene

Concentrations of 1-Np were measured on the order of picograms (pg/m^3). Indoor 1-Np concentrations range from 1.7 to 1260 pg/m^3 at Sites 2 and 5, respectively, and outdoor 1-Np concentrations range from 3 to 12 pg/m^3 at Sites 7 and 8, respectively. Similar to Lg and total $PM_{2.5}$, indoor concentrations of 1-Np are much higher at Sites 2 and 8, resulting in indoor to outdoor ratios that are magnitudes higher than at other sites. At 4 of the 9 sites, outdoor 1-Np concentrations are higher outdoors than indoors resulting in an indoor to outdoor ratio less than 1. These ratios are expected as 1-Np is a marker of diesel combustion and no diesel combustion was expected indoors. Surprisingly, the other five sites have indoor to outdoor ratios greater than 1, with ratios ranging from 2 to 332. Overall, indoor concentrations of 1-Np are not significantly higher than outdoor concentrations ($p = 0.43$) and outdoor 1-Np concentrations are not significantly different among the three villages ($p = 0.79$).

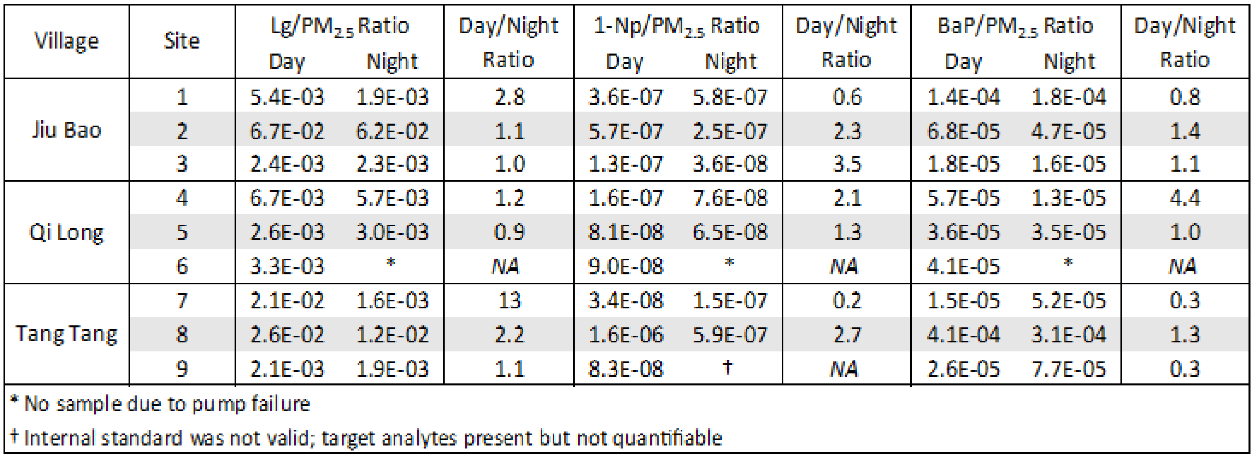
Benzo[a]pyrene

Concentrations of BaP were measured on the order of nanograms (ng/m^3). Indoor BaP concentrations range from 0.8 to 150 ng/m^3 at Sites 5 and 2, respectively, and outdoor BaP concentrations range from 1 to 6 ng/m^3 at Sites 5 and 8, respectively. The highest concentrations of BaP are seen indoors at Sites 2 and 8, with indoor to outdoor ratios of 117 and 23, respectively. Only Site 1 is also observed as having a higher indoor concentration BaP, with an indoor to outdoor ratio of 3. At all other sites, indoor BaP concentration is either similar to outdoor concentrations or slightly enriched indoors (indoor to outdoor ratios ranging from 0.3 to 1.1). Overall, the indoor BaP concentrations do not differ significantly from outdoor concentrations ($p = 0.82$), nor do the outdoor BaP concentrations differ significantly among villages ($p = 0.11$).

Indoor Component Mass Fraction

Mass fraction results for the three selected components (Lg, 1-Np, and BaP) are reported below. The mass fraction was calculated by dividing the total $PM_{2.5}$ mass (µg) by the mass of the component (µg). Table 4 provides the ratio of indoor Lg, 1-Np, and BaP concentration to total indoor $PM_{2.5}$ concentration, also referred to as the mass fraction. In general, the data show that daytime ratios were higher than the nighttime ratios, but the daytime and nighttime mass fractions were similar to each other and had less variability than the mass concentrations results presented above.

Table 4. Ratio of components of interest-- Lg, 1-Np and BaP-- mass concentrations indoors for day and night sampling periods including day to night concentration ratios by village and site number.



Village	Site	$Lg/PM_{2.5}$ Ratio		Day/Night Ratio	$1\text{-}Np/PM_{2.5}$ Ratio		Day/Night Ratio	$BaP/PM_{2.5}$ Ratio		Day/Night Ratio
		Day	Night		Day	Night		Day	Night	
Jiu Bao	1	5.4E-03	1.9E-03	2.8	3.6E-07	5.8E-07	0.6	1.4E-04	1.8E-04	0.8
	2	6.7E-02	6.2E-02	1.1	5.7E-07	2.5E-07	2.3	6.8E-05	4.7E-05	1.4
	3	2.4E-03	2.3E-03	1.0	1.3E-07	3.6E-08	3.5	1.8E-05	1.6E-05	1.1
Qi Long	4	6.7E-03	5.7E-03	1.2	1.6E-07	7.6E-08	2.1	5.7E-05	1.3E-05	4.4
	5	2.6E-03	3.0E-03	0.9	8.1E-08	6.5E-08	1.3	3.6E-05	3.5E-05	1.0
	6	3.3E-03	*	*NA*	9.0E-08	*	*NA*	4.1E-05	*	*NA*
Tang Tang	7	2.1E-02	1.6E-03	13	3.4E-08	1.5E-07	0.2	1.5E-05	5.2E-05	0.3
	8	2.6E-02	1.2E-02	2.2	1.6E-06	5.9E-07	2.7	4.1E-04	3.1E-04	1.3
	9	2.1E-03	1.9E-03	1.1	8.3E-08	†	*NA*	2.6E-05	7.7E-05	0.3

* No sample due to pump failure

† Internal standard was not valid; target analytes present but not quantifiable

Levoglucosan

The mass fraction of indoor Lg measured during daytime sampling ranged from 2.1x10^{-3} at Site 9 to 6.7x10^{-2} at Site 2. The mass fraction of indoor Lg measured during nighttime sampling ranged from 1.6x10^{-3} at Site 7 to 6.2x10^{-2} at Site 2. In general, the mass fraction of Lg found in daytime samples were higher than for nighttime samples, although 5 of the 8 samples had approximately equivalent mass fractions day and night. The Lg mass fraction collected during the day was observed to be significantly different than samples collected at night ($p = 0.039$). Across the nine sites, the day to night ratio of mass fractions ranged from 0.9 at Site 5 to 13 at Site 7. These day/night ratios were tempered in comparison to the ratios for mass concentration, where day to night ratios were as high as 823 for Lg at Site 2.

1-Nitropyrene

The mass fraction of indoor 1-Np measured during daytime sampling ranged from 3.4 x10^{-8} at Site 7 to 1.6x10^{-6} at Site 8. The mass fraction of indoor 1-Np measured during nighttime sampling ranged from 3.6x10^{-8} at Site 3 to 5.9x10^{-7} at Site 8. In general, the mass fraction of 1-Np found in daytime samples were higher than nighttime samples with the exception of Sites 1 and 7. The mass fraction of 1-Np collected during the day did not differ significantly than samples collected at night ($p = 0.46$). Across the nine sites, the day to night ratio of mass fraction ranged from 0.2 at Site 7 to 3.5 at Site 3. Similar to Lg mass fraction, the day/night ratios of 1-Np mass fraction were subdued compared to day/night ratios for 1-Np concentration.

Benzo*[a]pyrene*

The mass fraction of indoor BaP measured during daytime sampling ranged from 1.8×10^{-5} at Site 3 to 4.1×10^{-4} at Site 8. The mass fraction of indoor BaP measured during nighttime sampling ranged from 1.3×10^{-5} at Site 4 to 3.1×10^{-4} at Site 8. The mass fraction of BaP found in daytime samples were higher at sites 2, 3, and 6 and lower at sites 1, 7, and 9. Overall, the BaP mass fraction of daytime samples was not significantly different than samples collected at night ($p = 0.84$). Across the nine sites, the day/night ratio of mass fractions ranged from 0.3 at sites 7 and 9 to 4.4 at site 4. Similarly the lowest day/night ratio of BaP concentration was observed at 0.3 at sites 7 and 9, although was as high as 40 at Site 2.

Indoor vs Outdoor Component Mass Fraction

Table 5 provides the mass fraction of Lg, 1-Np, and BaP to total $PM_{2.5}$ concentration for both indoor (day and night combined) and outdoor samples.

Table 5. Comparison of indoor and outdoor ratios of components of interest--Lg, 1-Np and BaP--by village and site number.

Village	Site	$Lg/PM_{2.5}$ Ratio		In/Out Ratio	$1\text{-}Np/PM_{2.5}$ Ratio		In/Out Ratio	$BaP/PM_{2.5}$ Ratio		In/Out Ratio
		Indoor	Outdoor		Indoor	Outdoor		Indoor	Outdoor	
Jiu Bao	1	3.5E-03	4.1E-03	0.9	4.9E-07	3.8E-07	1.3	1.6E-04	1.1E-04	1.5
	2	6.7E-02	7.4E-03	9.0	5.6E-07	1.5E-07	3.6	6.7E-05	5.2E-05	1.3
	3	2.4E-03	4.3E-03	0.5	1.0E-07	1.3E-07	0.8	1.8E-05	8.2E-05	0.2
Qi Long	4	6.1E-03	6.1E-03	1.0	1.1E-07	2.5E-07	0.4	3.2E-05	7.0E-05	0.5
	5	2.8E-03	3.4E-03	0.8	7.4E-08	2.1E-07	0.3	3.5E-05	6.1E-05	0.6
	6	3.3E-03	2.9E-03	1.1	9.0E-08	1.9E-07	0.5	4.1E-05	5.8E-05	0.7
Tang Tang	7	1.2E-02	2.5E-03	4.9	8.8E-08	1.0E-07	0.9	3.2E-05	5.8E-05	0.6
	8	2.4E-02	4.9E-03	4.9	1.5E-06	2.9E-07	5.1	4.0E-04	1.5E-04	2.6
	9	2.0E-03	1.7E-03	1.2	8.1E-08	2.5E-07	0.3	5.2E-05	6.1E-05	0.8

Levoglucosan

The mass fraction of Lg ranged from 2.0×10^{-3} at Site 9 to 6.7×10^{-2} at Site 2 for indoor samples, and 1.7×10^{-3} at Site 9 to 7.4×10^{-3} at Site 9 for outdoor samples. The indoor to outdoor Lg mass fraction ratios showed an enrichment of the Lg fraction indoors at sites 2, 7, and 8 and enrichment outdoors at sites 3 and 5, with the rest of the sites having approximately equivalent Lg fractions indoors compared to outdoors with a range of indoor to outdoor ratios of 0.9 to 1.2. Across the nine sites sampled, the mass fraction of Lg measured indoors was not significantly different than outdoors ($p = 0.43$).

1-Nitropyrene

The mass fraction of 1-Np ranged from 7.4×10^{-8} at Site 5 to 1.5×10^{-6} at Site 8 for indoor samples, and 1.0×10^{-7} at Site 7 to 3.8×10^{-7} at Site 1 for outdoor samples. The indoor to outdoor 1-Np mass fraction ratios showed enrichment of the 1-Np fraction outdoors at sites 3, 4, 5, 6 and 9 with indoor to outdoor ratios of 0.8, 0.4, 0.3, 0.5, and 0.3, respectively. Enrichment indoors was observed at sites 1, 2 and 8 with indoor to outdoor ratios of 1.3, 3.6, and 5.1, respectively. Site 7 had approximately equivalent 1-Np fractions indoors and outdoors with an indoor to outdoor ratio of 0.9. Across the nine sites sampled, the mass fraction of 1-Np measured indoors was not significantly different than outdoors ($p = 0.91$).

Benzo[a]pyrene

The mass fraction of BaP ranged from $1.8\text{x}10^{-5}$ at Site 3 to $4.0\text{x}10^{-4}$ at Site 8 for indoor samples, and $5.2\text{x}10^{-5}$ at Site 2 to $1.5\text{x}10^{-4}$ at Site 8 for outdoor samples. The indoor to outdoor BaP mass fraction ratios ranged from 0.2 to 2.6, but the general trend was enrichment of the BaP fraction outdoors at most sites with only moderate enrichment indoors at sites 1, 2, and 8. Across the nine sites sampled, the mass fraction of BaP measured indoors was not significantly different than outdoors ($p = 0.65$).

Continuous PM2.5 Sampling Indoors

Continuous indoor fine particle sampling by the PUWP samplers deployed at each site are presented in Figures 1 through 9 below. These results are reported as plots of sensor response over the time period sampled. These plots show fine particle generation in each home is episodic and occurs at distinct times of the day. The peaks of particle levels are exclusively during the daytime sample period except at Site 4 where periodic particle levels occurred overnight between 02:00 and 06:00.

The number and magnitude of peaks was greatest at sites 1, 2, 3, 4, 7, and 8 while the rest of the sites had fewer and smaller peaks of particle levels as measured by these sensors. These sites correspond to the same sites that have higher mass concentrations as measured by filter samples. Plot peaks, representing periods of $PM_{2.5}$ concentration increase were generally observed in the evening, morning and around midday. The sensor at site 2 stopped recorded data around

12:00 during a period of fine particle formation in this home. This could have been due to an overwhelming of the sensor or a power outage at this site.

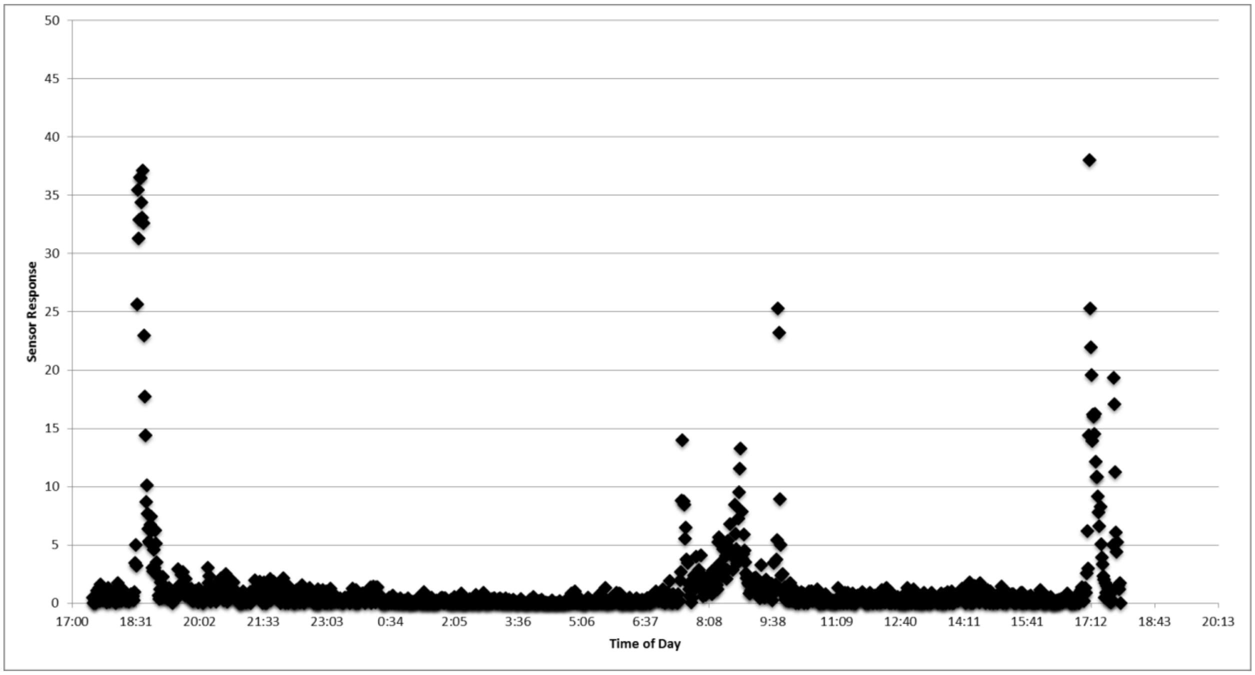


Figure 2. Site 1 PUWP sensor response during the period sampled.

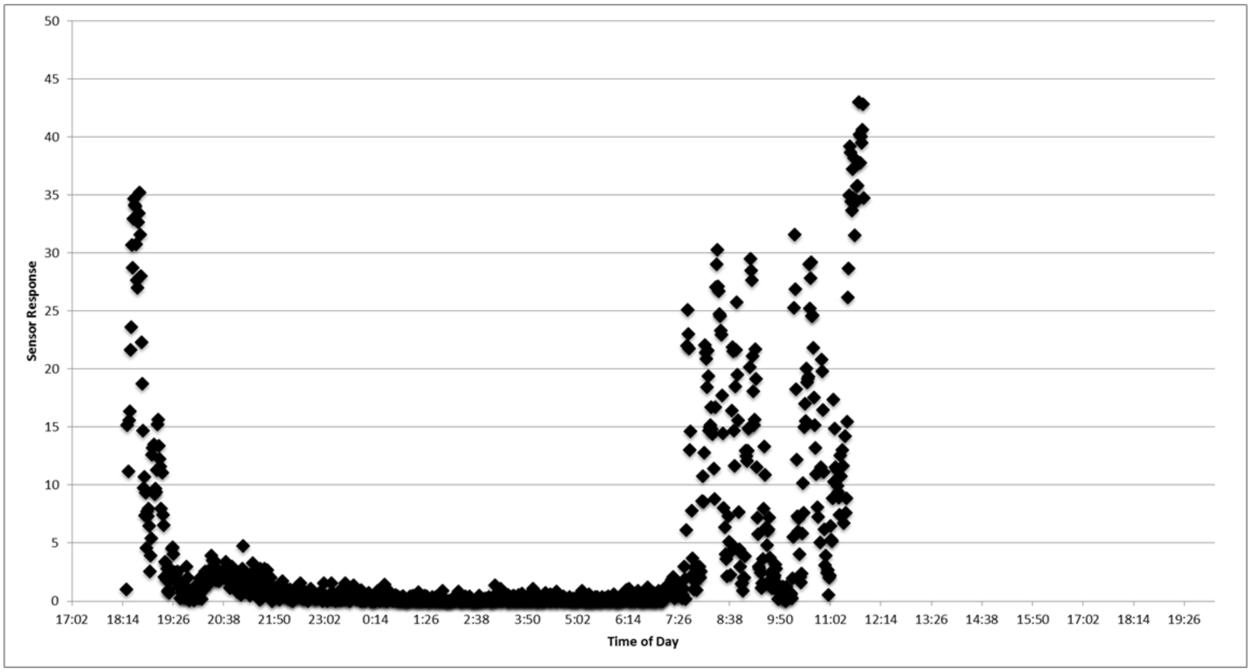


Figure 3. Site 2 PUWP sensor response during the period sampled.

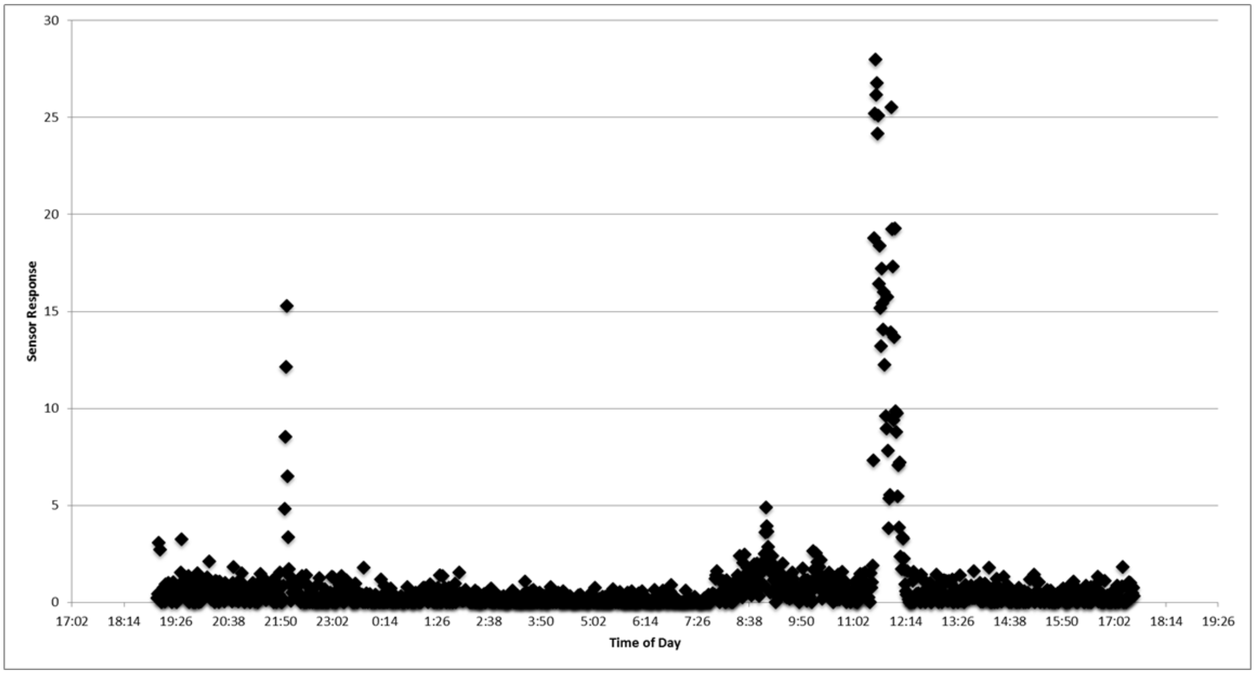


Figure 4. Site 3 PUWP sensor response during the period sampled.

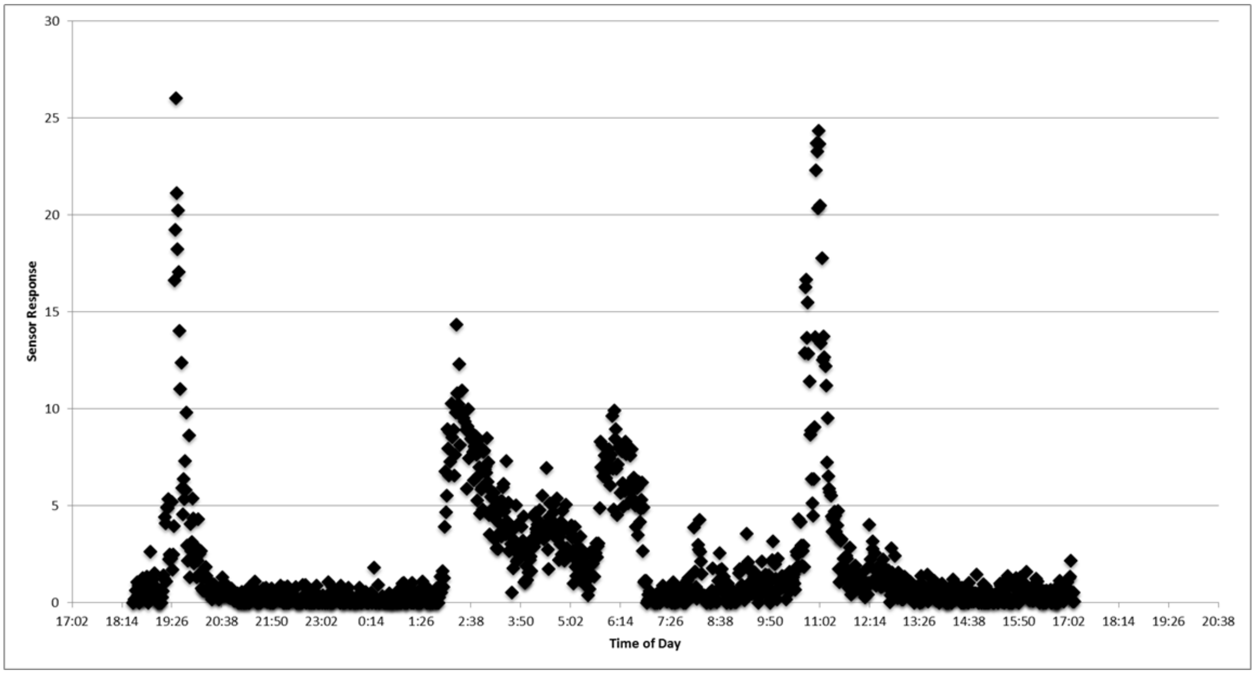


Figure 5. Site 4 PUWP sensor response during the period sampled.

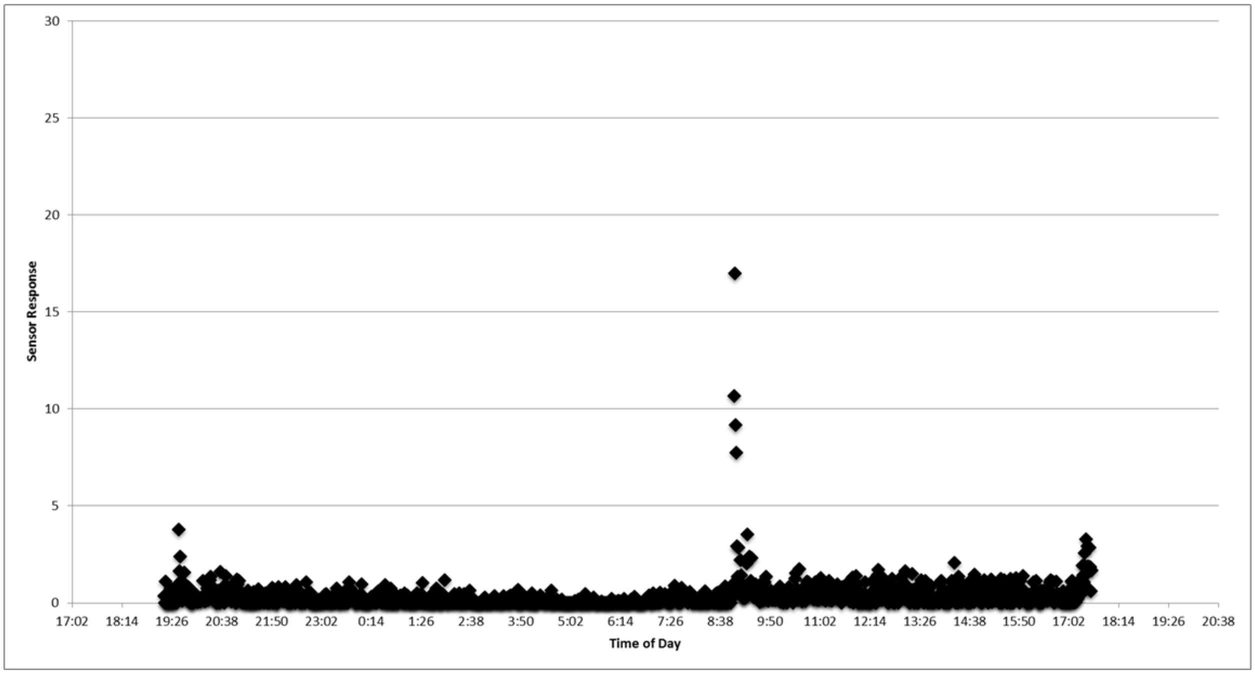


Figure 6. Site 5 PUWP sensor response during the period sampled.

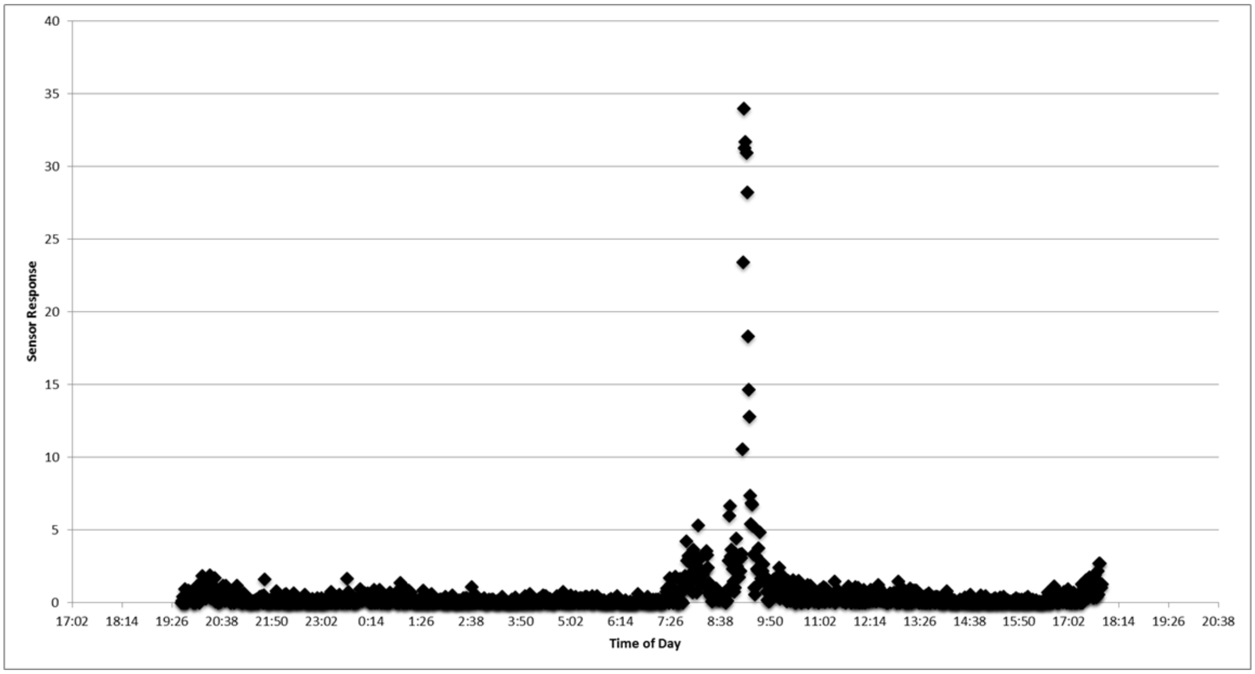


Figure 7. Site 6 PUWP sensor response during the period sampled.

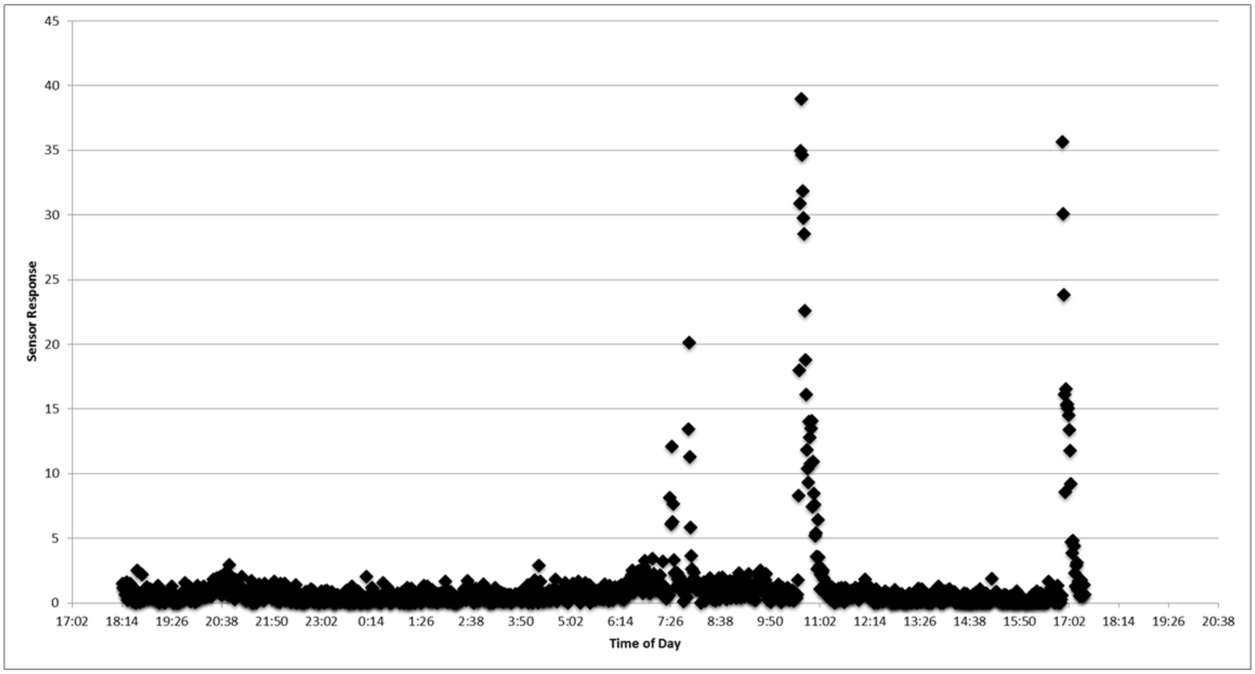


Figure 8. Site 7 PUWP sensor response during the period sampled.

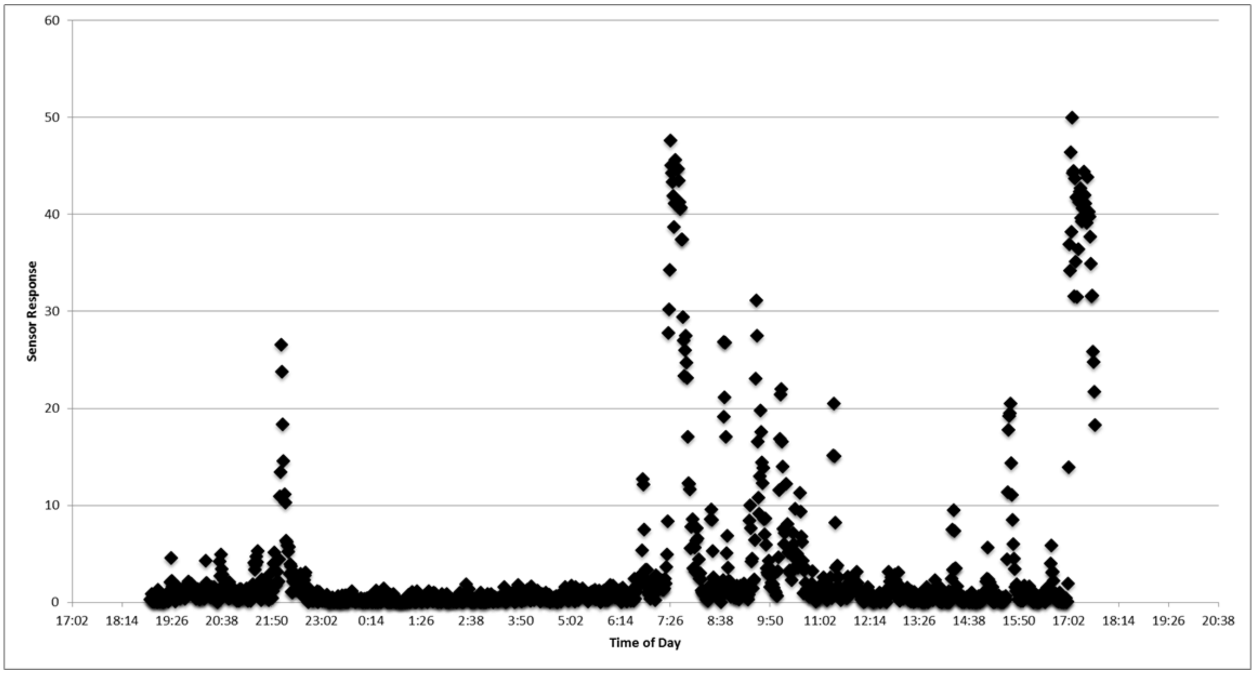


Figure 9. Site 8 PUWP sensor response during the period sampled.

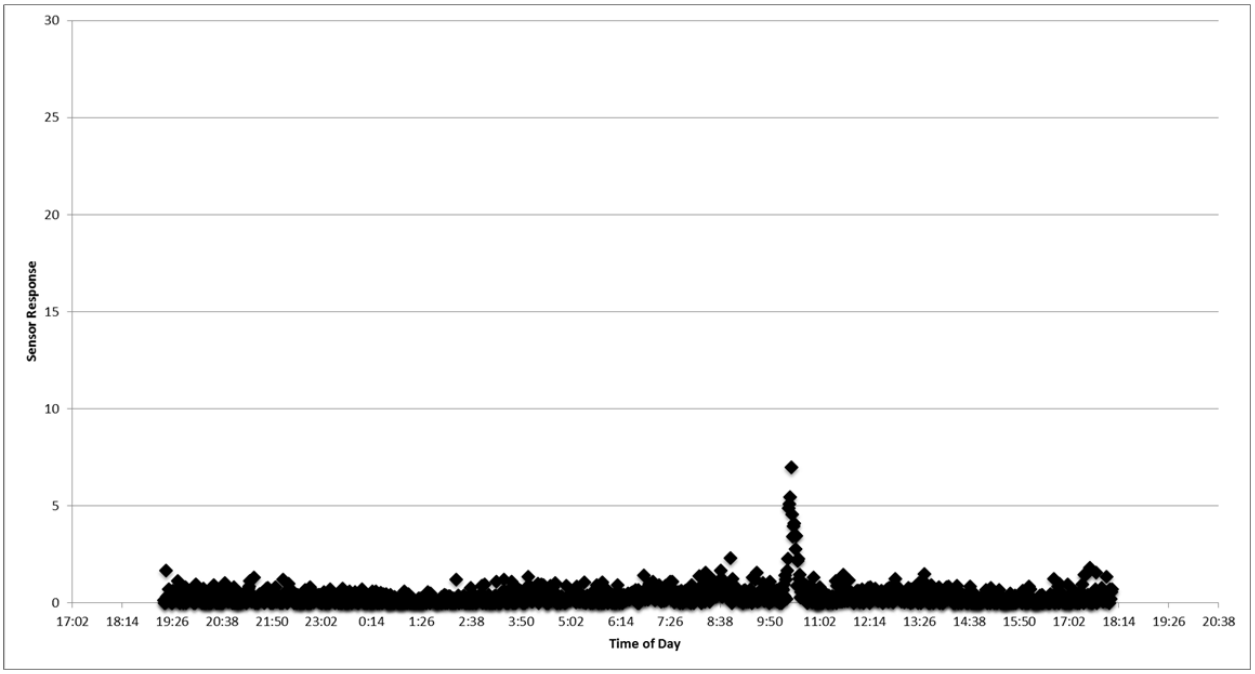


Figure 10. Site 9 PUWP sensor response during the period sampled.

Discussion of Results

Revision of Sampling Strategy

The initial sampling plan was to sample 12-15 households spread across 4-5 villages. We traveled to these villages as guests of our collaborators on the particle count and birth outcome study previously mentioned and so could not dictate the travel schedule or time spent in the villages. Time limitations ultimately caused us to amend the sampling to 9 households spread across the three villages mentioned herein; leaving us with extra samples to use at these sites. We were limited by the number of sampling pumps, but we decided to sample for 2 12-hr periods indoors to limit the potential for filter overloading and pump failure. This improvised sampling scheme allowed us to sample during two distinct time periods indoors, roughly representing a period of high occupant activity (7am-7pm) and a period of low activity (7pm-7am). The actual activities of the residences was not assessed in this study as it was an opportunistic environmental sampling pilot project and no questionnaires were administered or interviews conducted. These samples were collected in the summer, and no indication of fuel use for heating was observed in any household. The disparity in $PM_{2.5}$ and other component concentrations between these sites is assumed to be due to differences in indoor behavior of the occupants related to personal habits (smoking, incense burning, etc.), cooking activities, or household factors (e.g., ventilation).

Discussion of PM2.5 Mass Concentration Results

Differences in $PM_{2.5}$ concentrations were observed between daytime and nighttime sampling periods, and between indoor and outdoor sampling compartments at most sites. Sites 2 and 8 had alarmingly high daytime concentrations of $PM_{2.5}$ at 4371 and 632 µg/m^3, respectively. These two sites along with Site 3 were between 3 and 28-fold higher than nighttime concentrations at the same site. The increased concentration of fine particles during indoor daytime sampling indicate a human activity derived source of $PM_{2.5}$. Results of the PUWP samplers confirm that fine particle generation indoors is very episodic and peak particle formation times roughly align with times associated with cooking (morning, midday and evening), with time of low particle formation in between. During deployment of equipment at sites 2 and 8 wood burning was observed in these homes and is the likely culprit for the elevated daytime levels. Site 3 was not observed to be burning any fuel, but did have indoor levels high enough to suspect that some fuel burning or smoking did occur in the area of the indoor sample during the daytime period. The PUWP monitors captured a strong particle formation event around midday at this site and could have been the single event that lead to elevated daytime concentrations. In contrast, sites 1, 4, and 9 had higher nighttime indoor $PM_{2.5}$ concentrations. The PUWP monitoring results show several periods of increased particle formation overnight at Site 4 between 02:00 and 06:00, explaining the increased nighttime concentration at this site. As this sampling was conducted in July, and nighttime temperatures were very mild, no indication of burning fuel for heat was observed. Sampling during the winter may yield very different results with overnight fuel use for heat creating the potential for high indoor $PM_{2.5}$ concentrations overnight as well.

Comparing indoor to outdoor $PM_{2.5}$ concentration over the 24-hr sampling period required averaging the two 12-hr indoor samples. Indoor to outdoor ratios (I/O) are a useful metric to assess if indoor and outdoor air pollution is approximately equal; representing an I/O ratio of 1. Our results show I/O ratios were greater than 1 at all sites except Site 7, indicating higher indoor than outdoor $PM_{2.5}$ concentrations. The highest I/O ratios were at sites 2 and 8 where indoor $PM_{2.5}$ concentrations were very high and dominated by daytime $PM_{2.5}$ particle formation, as previously discussed. All other sites had average indoor $PM_{2.5}$ concentrations slightly higher or equivalent to outdoor concentrations with I/O ratios between 0.9 and 2.2, suggesting no major indoor sources of fine particulate matter at these sites. For almost all of our sites, nighttime indoor concentrations were generally still higher than outdoor 24-hr concentrations, which may be important as all members of the household are likely to be exposed to evening levels as they sleep, whereas women and children may be more likely to be exposed to daytime levels as has been found in studies in India (Balakrishnan et al., 2014).

Comparison to Other PM2.5 Studies

Hu et al. (2014) reported results on the indoor sampling of $PM_{2.5}$ concentrations in Xuanwei households. The geometric mean concentration of $PM_{2.5}$ was reported to be 144 ug/m^3 in homes burning smoky coal, 96 µg/m^3 in homes burning smokeless coal, and 327 µg/m^3 in homes burning wood. The $PM_{2.5}$ concentrations from the coal-burning homes reported by Hu et al. (2014) are greater than the $PM_{2.5}$ concentrations measured at the household in our study observed to be burning coal (49.5 µg/m^3). This could be due to difference in ventilation, amount of fuel burned, or the behavior of lighting the coal briquette outside before transferring it inside into the stove (an action not observed in this study, but known to be used in the Xuanwei area). Our measurements of $PM_{2.5}$ concentrations from wood-burning sites were similar to those observed by Hu et al. (2014) at one site (362 µg/m^3), but much higher at another (2264 µg/m^3). The wood-burning site in our study with extremely high $PM_{2.5}$ concentration did not have stove indoors and was using an open fire pit to burn wood. This fire pit had several logs actively smouldering during our sampling period. In addition to fuel and stove type, concentration of $PM_{2.5}$ indoors is greatly influenced by the characteristics of the home and the behavior of the residents.

Discussion of PM Component Results

The mass fraction of each component was calculated to assess the relative enrichment of each component between the two indoor time periods and the indoor and outdoor compartments sampled.

Levoglucosan

In Xuanwei County, most of the indoor air pollution exposure research focuses on sources of and exposure to coal combustions, but results presented here indicate that some homes in this region are still using wood or other cellulose-based fuel for cooking and possibly heating. Levoglucosan (Lg) is often used as a marker of wood combustion, however, it is also emitted by the combustion of other cellulose containing materials such as incense, cigarettes, and agricultural byproducts (Locker, 1998). An elevated mass fraction of Lg to $PM_{2.5}$ indicates that the combustion of wood or other cellulose containing material may be a dominating source of indoor air pollution. In China, incense burning is a common yet inconsistent practice. The frequency and intensity of indoor incense burning could affect the level of Lg detected indoors and may explain some of the variation observed between daytime and nighttime samples.

The results showed indoor Lg enrichment to be significantly higher in the daytime compared to nighttime, suggesting that wood was being burned inside homes during the daytime sampling period. Sites 2, 7 and 8 were found to have the greatest daytime Lg enrichment indoors, which, based on the observation that sites 2 and 8 were burning wood as fuel, is an expected result. Site 7 was observed to be burning incense during the deployment of the sampling equipment, but only low levels of daytime indoor $PM_{2.5}$ were measured. Comparison of indoor to outdoor samples yields similar conclusions and provides further evidence that indoor $PM_{2.5}$ concentrations in homes with a high fraction of Lg are more likely to be forming combustion from cellulose-containing fuels.

1-Nitropyrene

1-Nitropyrene (1-Np) can be used as a marker of diesel fuel combustion from vehicles, but can also indicate the burning of heavy solid fuels for cooking or indoor heating (Kim et al., 2005). In this sample population, the enrichment of 1-Np reveals insightful, yet puzzling, patterns among the sites. Most sites had greater 1-Np enrichment during daytime samples compared to night, which is expected considering this component is thought to be generated outdoors by diesel traffic passing through the villages during the daytime. However, two of the sites sampled (sites 1 and 7) measured higher enrichments of 1-Np in the nighttime samples compared to the daytime samples. These sites are in the two villages that have a major thoroughfare running through the village, but the sampled houses are not situated any closer to the road than the other sampled houses in these villages. These outlying results could be a consequence of unusual residential activity--such as a diesel vehicle being started or left idling near the sampling equipment late at night.

This study found the enrichment of 1-Np to be greater in most of the outdoor samples compared to indoor samples and higher in the villages with main roads running through them. These results are expected given that 1-Np tends to be derived from the exhaust of diesel vehicles. However, the 1-Np fraction was found to be enriched indoors at the three sites (sites 1, 2, and 8) observed to be burning solid fuel inside the home during the deployment of the sampling equipment. Differences among sites 1, 2, and 8 are likely explained by differences in ventilation and fuel source. For instance, site 1 was likely burning a coal briquette, but only had slight indoor enrichment of 1-Np. An explanation of this could be attributed to the ventilation of their stove. Sites 2 and 8, on the other hand, had 3- to 5-folds the enrichment of 1-Np indoors, but both appeared to be burning wood. Although there was no indication of kerosene oil use in these home, kerosene is known to emit 1-Np and could be a potential source of indoor 1-Np (Ohnishi et al 1985). Wood combustion is not known to emit 1-Np, so it is possible that nitrogenation is occurring after sample collection on the filters with very high soot loads. This was the case for the two most concentrated samples collected during the daytime at sites 2 and 8. The aforementioned observations from this study raise some concerns regarding the use of 1-Np as a marker of diesel combustion in settings with high $PM_{2.5}$ concentrations.

Benzo[a]pyrene

Benzo[a]pyrene concentrations were measured in all air samples because of its known carcinogenicity (IARC, 2010). Indoor concentration of BaP was higher during the day at all sites except 1, 7 and 9; following the trend observed for other components and indicating that increased BaP-creating activities are occurring during daytime hours.

The nighttime samples were nearly all lower in BaP concentration except at site 1, 7, and 9. We expect the activities that generate BaP to be primarily associated with indoor fuel combustion and so lower nighttime levels were expected. These sites with higher nighttime concentrations indicate activities that either happen outside of the daytime time period for this study 7am -7pm or are enriching indoors overnight from penetration by outdoor air.

Average 24-hr indoor concentrations of BaP were observed to be higher at sites 1, 2, and 8 with between 3 and 117 fold higher indoor compared to outdoor BaP concentrations. All other sites had either equivalent or slightly higher outdoor BaP concentrations. The three sites with exceptionally high BaP concentrations were again the sites with active burning observed indoors during sample collection. Outdoor BaP concentrations were less variable and indicate a regional or ubiquitous source of this component. This could be formed from regional power plants or from the many households using fuel for cooking in these villages.

Indoor samples were enriched in BaP (as a fraction of total $PM_{2.5}$ mass) at sites 1, 2, and 8, but nowhere near in magnitude to the difference in concentration between these sites. Indoor daytime BaP mass fraction is slightly higher than the nighttime mass fraction at sites 2 and 8, but is 4.4 fold higher at site 4. Site 4 was not observed to be burning any fuel while sampling occurred, but this sample was collected in kitchen set apart from the main living quarters and the cooking in this room, even with an electric stove, could be the source of the different level of BaP enrichment between the daytime and nighttime samples. Sites 7 and 9 are enriched in BaP in the nighttime samples compared to daytime samples and may indicate a penetration of outdoor air during the nighttime sampling period. These two sites are in the center of the main village commerce center at the bottom of the river valley as opposed to site 8 in the same village, which on the opposite side of the river and not on the main thoroughfare.

Comparison to Other PM Component Studies

Review of the online literature did not yield results that would suggest indoor or outdoor air concentrations of Lg or 1-Np have been reported for Xuanwei County. On the other hand, multiple reportings of BaP concentrations measured in the Xuanwei area were discovered. This is likely due to its suspected involvement in the high levels of lung cancer seen in this area (Lan et al., 2002; Lv et al., 2009; Downward et al., 2014).

Our study measured BaP concentrations to be 152 and 144 ng/m^3 in wood-burning homes and 8.15 ng/m^3 in the home observed to be burning coal. In 2014, Downward et al. collected 266 personal PAH exposure samples of residents in Xuanwei and Fuyuan counties that used solid fuel, and found geometric mean concentrations of BaP of 58.2 ng/m^3 for wood burning, 44.7 ng/m^3 for smoky coal, and 10.6 ng/m^3 for smokeless coal. Results presented here are similar to that of Downward et al. (2014) for the coal burning site--if the house in our study was using the smokeless (anthracite) coal--but are 2- to 3- folds higher than those reported for wood burning. This difference could be due to the different types of sampling used (personal versus area), however results suggest that BaP exposure could be likely from wood burning in addition to burning smoky coal.

Lv et al. (2009) reported BaP concentrations ranging from 7.7 to 380.3 ng/m^3 (indoor) and 0.9 to 271.5 ng/m^3 (outdoor). Results from Lv et al. (2009) are similar to, but generally higher than, BaP concentrations reported here. Inconsistencies in results are likely due to the differing site selection processes. Our study sampled the indoor and outdoor environment of a diverse cross section of homes using diverse fuel types, whereas Lv et al. (2009) deliberately sampled indoor and outdoor locations burning smoky coal. Comparing our results to those of Lv et al. (2009) provides further evidence that wood burning has the potential to emit BaP within the same concentration range as smoky coal, and air pollution studies should be sampling combustion formed by all solid fuel types in order to adequately evaluate exposure to BaP.

BaP was also measured indoors in Xuanwei County in 1995 and reported by Lan et al (2002). Lan et al. (2002) reported the findings of PM_{10} samples collected from households burning coal. Mean indoor BaP concentration was 1660 ng/m^3 for unvented stoves compared to 250 ng/m^3 for vented stoves. These results are higher than BaP concentrations measured in this study, but could be due to Lan et al (2002) only reporting data from households actively burning coal. Our results from household burning wood are similar in concentration to those reported for vented coal burning indoors with sites 2 and 8 having average BaP concentrations of 144 and 152 ng/m^3.

Influential Factors of Household Characteristics

During deployment of sampling equipment, burning of solid fuel was observed at sites 1, 2, and 8 where some form of solid fuel was being burned to heat the contents of a kettle (Image 1). The indoor sampling area of Site 1 appeared to be using a coal stove ventilated to the outside while, sites 2 and 8 appeared to be burning wood in unventilated stoves. Sites 2 and 8 differed in that the wood in site 2 appeared to be gathered logs and twigs and the fire was smoldering rather than burning, whereas site 8 appeared to have wood from larger trees stacked and chopped to appropriate size and was being burned at a temperature above smoldering. Site 5 also had evidence of a recent open fire in the room sampled, but no fire was burning at the time of equipment deployment and the low measured concentration in this residence indicate that no burning occurred during sampling.

Sites 3, 4, and 6 all had a solid fuel stove designed to burn processed and formed coal briquettes, but only the stove at Site 4 had a hood with ventilation to the outdoors and no recent evidence of burning indoors (Image 2). Sites 3 and 6, in contrast, had dark markings on the wall indicative of stove use, although how recently the stoves were used could not be assessed. No fuel combustion was observed at these homes, but the relatively high indoor concentration at Site 3 indicate that either the stove was used during the sampling period or there was another source of $PM_{2.5}$ inside the home such as smoking or incense use. Sites 7 and 9 did not appear to ever use solid fuel having all electric appliances available. Site 7 did have a coal stove similar to Site 4, although the area was being used for storage and did not appear to be recently used for cooking. There was incense burning at Site 7 and this may be the cause for higher indoor concentration at this site compared to Site 9. Site 9 did not appear to have any capability to burn solid fuel in the home.

Images 1 and 2 illustrate the large varying levels of social economic status as reflected by the housing and conditions and cooking situations in homes in this study. One of the homes (Site 2) did not have electricity, and had the highest indoor $PM_{2.5}$ concentrations. The homes in Tang Tang village were generally of higher quality visually than those in other villages, which perhaps reflects their proximity to the main township and commerce center. Yet, there was not clear indication from the measured indoor air quality that the Tang Tang households had lower concentrations than homes in other villages. For example, Site 5 corresponded to a rather poor home with a dirt floor and rather primitive cooking on this floor, yet had the lowest concentrations of indoor $PM_{2.5}$.

Figure 11. Indoor sampling area at sites 1, 2, 5, and 8 (clockwise from top left image).

The solid fuel burning at site 1 did not appreciably raise the indoor $PM_{2.5}$ concentration as was the case for sites 2 and 8. This likely has to do with the ventilation, the fuel being used and the temperature/completeness of combustion. Other sources of $PM_{2.5}$ at these locations, besides those observed for cooking, could also have contributed to the higher concentration observed at these sites. Cigarette smoking was observed outside of many of the homes during the deployment of the equipment, but only one residence (Site 8) had an adult smoking a pipe inside the home during deployment of the equipment.

Figure 12. Indoor sampling area at sites 3, 4, and 6 (clockwise from top left image).

Comparison to Air Quality Standards

Air sampling is critical for monitoring, evaluating, and enforcing regulated air quality standards. As in the United States, regulatory standards in China exist only for outdoor (ambient) air quality. Currently, China is phasing-in new standards by 2016, which will include a $PM_{2.5}$

annual average limit of 35 μg/m^3 and a 24-hr average of 75 μg/m^3 (Transport Policy, 2014). None of the outdoor samples collected during this study exceeded the 24-hr regulatory limit. Although, if outdoor air concentrations remained consistent over the year, or if additional emissions occur during the wintertime (heating) season, then ambient $PM_{2.5}$ in Tang Tang village may exceed maximum air pollution limits.

The World Health Organization (WHO) has developed indoor air quality guidelines to direct efforts in reducing the burden of disease caused by exposure to high levels of indoor air pollution. These guidelines recommend indoor $PM_{2.5}$ concentrations below 25 μg/m^3 measured as a 24-hr sample (WHO, 2014). To compare indoor air samples to the WHO's 24-hr mean concentrations guidelines, the $PM_{2.5}$ concentrations measured over the two 12-hr indoor samples at each residence were averaged. This resulted in general lowering of overall concentrations because the inclusion of the nighttime monitoring tempered the extreme indoor concentrations (most evident in results from sites 2 and 8).

Regardless, all sites (except Site 5) were observed to have a mean 24-hr indoor $PM_{2.5}$ concentrations above the WHO guideline of 25 μg/m^3. Three of the nine sites sampled were observed to be actively burning solid fuel during this study. These results indicate that even in homes not known to be burning solid fuel for cooking, the levels of $PM_{2.5}$ pose a risk to the health of the human occupants. This risk of adverse health effects is especially high for residents of sites 2 and 8 where active, unvented wood burning inside the home was observed. Site 2 had only two very old occupants, whereas Site 8 had a plethora of occupants, some as young as toddlers. The nearly ubiquitous cigarette smoking by adult males in this region as well as the use of incense may also be contributing to the levels of $PM_{2.5}$ in these homes. During daytime sampling, all sites were observed to be above the World Health Organization (WHO) air quality guidelines 24-hr mean concentration of 25 µg/m^3. During nighttime sampling, all sites--except sites 3 and 5--were above the WHO guideline level. No information exists for site 6 during this period due to a pump failure.

China's new ambient air quality guidelines include limits for BaP of 1 ng/m^3 as an annual average and 2.5 ng/m^3 for a 24-hr average. All outdoor samples were above the 1 ng/m^3 regulatory standard and sites 1 and 8 had concentrations above the 2.5 ng/m^3 standard. China also has national criteria standards for indoor BaP concentrations of 1 ng/m^3 (Chinese National Criteria, 2002). All sites except 5, 7, and 9 were observed to have daytime levels above the Chinese maximum allowable indoor concentration standard. While these levels are only 12-hr samples the concentrations observed at these three sites are alarmingly high and indicate a strong potential carcinogenicity of indoor air in these homes. BaP concentrations were notably high indoors at sites 2 and 8 where wood was observed being burned. The enrichment of BaP in $PM_{2.5}$ samples was also highest at sites 1, 2, and 8, all sites where either coal or wood was known to be burning during sampling. The greatest enrichment was observed indoor at site 1 and outdoor at site 8, suggesting that BaP is created by the activities in and around these homes. These results indicate that indoor and outdoor BaP concentrations in these villages are above levels where a risk of adverse health effects exist. The WHO Indoor Air Quality guidelines do not provide recommendations for Lg or 1-Np, although it is stated that no safe level of BaP exists (WHO, 2014).

Study Limitations

As guests of our collaborators and traveling for the launch of an epidemiological cohort study we collected opportunistic indoor and outdoor samples in as many homes as we could. As such, only a small cross-sectional dataset was produced. Sample size was limited by time to recruit and access households willing to participate in this small scale data collection endeavor. The small number of homes sampled in this study reduced our ability to make meaningful comparison between and among the results. Furthermore, the small sample size limited our ability to gather enough samples above the limit of quantitation (LOQ) and run statistical analyses on the XRF analysis results or conduct a principal component analysis. Without IRB approval, we were unable to interview or administer questionnaires to the occupants; therefore, we could not ascertain the characteristics of the home, its occupants, or exactly what $PM_{2.5}$-generating activities were occurring during the sampling period. The nature of this sampling opportunity also prevented multiple or longer term sampling. We were limited by only sampling each home once. Further, we were unable to sample all homes on the same day in order to control for day-to-day variability in ambient (outdoor) samples. Our limited time and resources also precluded us from measuring other, more volatile, components of fine PM or elemental and organic carbon, which may have helped differentiate fine PM sources and potential carcinogenicity.

Conclusion

Overall, this study found higher concentrations of $PM_{2.5}$ and greater variation in both concentration and composition of indoor samples compared to outdoor samples. Indoor air concentrations sampled during the day were generally higher and more variable than concentrations measured at night. These observations were confirmed by use of a continuous $PM_{2.5}$ sensor in each home and showed most $PM_{2.5}$ generating activity occurred during the day and were highly variable in nature. We noted visual evidence of wood and coal combustion in the homes with the highest levels of fine particles and observed enrichment of 1-nitropyrene, levoglucosan, and benzo[a]pyrene in the sampling results from these homes as well. At several sites, indoor concentrations of $PM_{2.5}$ and benzo[a]pyrene were above levels recommended by the World Health

Organization, suggesting that long term exposure at these levels could lead to adverse health outcomes for the residents.

Our observations confirm that interventions targeting indoor air quality have not reached every household in this region. Many homes would benefit from interventions to provide improved stoves, increased ventilation, or cleaner fuel by reducing overall concentrations of $PM_{2.5}$. Households in Xuanwei would continue to benefit from smoking cessation programs, although this would not address exposure to byproducts of combustion of fuel used for cooking and heating.

Future studies attempting to associate fine PM exposure with health outcomes should consider collecting samples for analysis of composition. This study chose to investigate PM composition, but the small sample size hindered potential analyses. The three components of fine PM discussed here (1-Np, Lg, and BaP) are helpful in identifying sources of air pollution and targeting interventions that reduce or remove the source of these components. Despite the fact that this study was limited by only analyzing three components, results support that there is high variability in PM composition among households in Xuanwei--even ones located in the same village. Results also support that PM constituents may be produced by multiple sources (originating indoor and outdoor). From review of the literature, we know that the toxicity of these components varies greatly as well. Therefore, having data that reveal not only PM particle counts or mass concentrations, but fractional analysis of PM components may help elucidate the link between air pollution exposure and health outcomes of interest. It will be important for future air sampling studies, focused in Xuanwei and beyond, to include robust composition analysis in their results.

Also, the continuous particle measurements illustrate the episodic nature of PM formation within households. This suggests that future studies may benefit from longitudinal sampling to assess temporal variations that may exist within households, particularly those that have choices in cooking and heating fuels. Note that the samples in the current study were only collected during the summertime, and further work would benefit from comparing these summertime (primarily from cooking) data with the PM levels and composition during wintertime where both cooking and heating activities are likely to be conducted. Future longitudinal studies would benefit from detailed monitoring of time activity patterns of residents because it affects exposure, as well as tracking of cooking behaviors, as the data clearly show that concentrations of PM increase dramatically in response to combustion events within the home.

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Appendix A

A1. Comprehensive results of PM2.5, levoglucosan and nitro PAHs.

Village	Site Number	Compartment	Sample Period	Sampled Air Volume (m^3)	PM$_{2.5}$ Concentration (µg/m^3)	Levoglucosan (µg/m^3)	2-nitropyrene (pg/m^3)	2-nitropfluorene (pg/m^3)	1-nitropyrene (pg/m^3)
Jiu Bao	1	Indoor	12-hr Night	1.61	55.5	0.11	35.4	37.1	32.3
		Indoor	12-hr Day	1.06	43.6	0.24	36.7	41.0	15.8
		Outdoor	24-hr	2.62	25.3	0.10	15.8	22.1	9.50
	2	Indoor	12-hr Night	1.51	156	9.62	53.2	37.2	39.0
		Indoor	12-hr Day	1.04	4371	293	838	1349	2482
		Outdoor	24-hr	2.28	24.8	0.18	7.20	11.6	3.80
	3	Indoor	12-hr Night	1.40	24.9	0.06	9.20	7.50	0.90
		Indoor	12-hr Day	1.09	77.4	0.18	15.4	14.2	9.70
		Outdoor	24-hr	2.43	23.0	0.10	7.10	13.5	3.10
Qi Long	4	Indoor	12-hr Night	1.33	38.2	0.22	5.30	4.30	2.90
		Indoor	12-hr Day	1.12	28.0	0.19	14.6	20.0	4.50
		Outdoor	24-hr	2.48	19.9	0.12	7.30	11.6	5.00
	5	Indoor	12-hr Night	1.31	18.9	0.06	5.40	5.10	2.30
		Indoor	12-hr Night	1.32	24.5	0.07	1.40	1.10	0.50
		Indoor	12-hr Day	1.14	24.9	0.07	12.1	19.5	3.00
		Indoor	12-hr Day	1.11	25.5	0.06	9.90	15.1	1.10
		Outdoor	24-hr	2.35	16.4	0.06	6.50	9.90	3.50
	6	Indoor	12-hr Day	1.14	26.6	0.10	13.3	16.5	2.40
		Indoor	12-hr Night	*	*	*	*	*	*
		Outdoor	24-hr	2.39	17.2	0.05	7.10	9.90	3.30
Tang Tang	7	Indoor	12-hr Night	1.44	34.4	0.06	10.6	12.4	5.20
		Indoor	12-hr Day	1.09	40.9	0.85	7.10	8.50	1.40
		Outdoor	24-hr	2.49	29.6	0.07	5.40	5.10	2.10
		Outdoor	24-hr	2.42	31.1	0.08	6.80	8.40	4.10
	8	Indoor	12-hr Night	1.39	91.2	1.07	59.6	35.3	54.0
		Indoor	12-hr Day	1.10	632	16.6	350	313	1016
		Outdoor	24-hr	2.46	41.1	0.20	16.9	17.2	11.9
	9	Indoor	12-hr Night	1.36	27.4	0.05	†	†	†
		Indoor	12-hr Day	1.10	26.6	0.06	4.90	7.50	2.20
		Outdoor	24-hr	2.42	29.4	0.05	9.10	11.1	7.30

* No sample due to pump failure † Internal standard was not valid; target analytes present but not quantifiable

A2. Comprehensive results of high molecular weight particle bound PAH analysis.

Village	Site Number	Compartment	Sample Period	Sampled Air Volume (m^3)	Fluoranthene ($\mu g/m^3$)	Pyrene ($\mu g/m^3$)	Benz (a) anthracene	Chrysene ($\mu g/m^3$)	Benzo (b) fluoranthene	Benzo (k) fluoranthene	Benzo (a) pyrene	Indeno (1,2,3-cd) pyrene (µg/m3	Dibenz (a,h) anthracene	Benzo (g,h,i) perylene ($\mu g/m^3$)
Jiu Bao	1	Indoor	12-hr Night	1.61	0.70	0.70	4.00	3.50	13.50	6.30	10.20	10.90	2.90	16.00
		Indoor	12-hr Day	1.06	1.20	1.20	2.60	2.50	8.30	3.80	6.10	6.70	1.50	9.30
		Outdoor	24-hr	2.62	0.80	0.80	1.30	1.30	4.10	1.80	2.70	3.10	0.60	4.40
	2	Indoor	12-hr Night	1.51	1.50	2.10	3.80	3.00	7.40	4.00	7.40	8.20	0.80	8.70
		Indoor	12-hr Day	1.04	19.7	31.8	63.5	55.5	261	169	297	175	16.0	176
		Outdoor	24-hr	2.28	0.70	0.60	0.70	0.80	2.10	1.00	1.30	1.50	0.30	2.00
	3	Indoor	12-hr Night	1.40	0.40	0.40	0.40	0.40	0.50	0.40	0.40	0.60	0.40	0.60
		Indoor	12-hr Day	1.09	0.50	0.50	0.80	0.70	1.70	1.00	1.40	3.20	0.60	3.60
		Outdoor	24-hr	2.43	0.70	0.70	0.80	0.90	2.60	1.20	1.90	2.10	0.30	2.50
Qi Long	4	Indoor	12-hr Night	1.33	0.40	0.40	0.40	0.40	0.70	0.40	0.50	0.80	0.40	1.20
		Indoor	12-hr Day	1.12	0.80	0.70	0.50	0.80	2.30	1.10	1.60	2.20	0.40	2.70
		Outdoor	24-hr	2.48	0.70	0.60	0.70	0.80	2.20	0.90	1.40	1.60	0.30	2.40
	5	Indoor	12-hr Night	1.31	0.40	0.40	0.40	0.40	0.90	0.50	0.60	0.80	0.40	1.40
		Indoor	12-hr Night	1.32	0.40	0.40	0.40	0.40	1.10	0.50	0.90	1.00	0.40	1.60
		Indoor	12-hr Day	1.14	1.10	0.80	0.60	0.90	1.80	0.70	1.10	1.50	0.40	1.60
		Indoor	12-hr Day	1.11	1.00	0.70	0.50	0.60	1.50	0.50	0.70	1.10	0.50	1.40
		Outdoor	24-hr	2.35	0.80	0.70	0.60	0.70	1.70	0.60	1.00	1.10	0.20	1.50
	6	Indoor	12-hr Day	1.14	0.50	0.40	0.40	0.40	1.70	0.70	1.10	1.80	0.40	3.10
		Indoor	12-hr Night	*	*	*	*	*	*	*	*	*	*	*
		Outdoor	24-hr	2.39	0.80	0.70	0.60	0.70	1.70	0.80	1.00	1.30	0.30	1.80
Tang Tang	7	Indoor	12-hr Night	1.44	0.60	0.60	0.80	0.70	2.50	1.30	1.80	2.40	0.50	2.80
		Indoor	12-hr Day	1.09	0.50	0.50	0.50	0.50	0.80	0.50	0.60	1.00	0.50	1.70
		Outdoor	24-hr	2.49	0.60	0.60	0.80	0.70	2.50	1.10	1.70	2.00	0.40	2.60
		Outdoor	24-hr	2.42	0.60	0.60	0.90	0.80	2.60	1.20	1.80	2.10	0.40	2.80
	8	Indoor	12-hr Night	1.39	4.20	5.30	13.1	11.2	27.8	15.0	28.6	24.6	3.80	28.1
		Indoor	12-hr Day	1.10	86.4	125	89.8	79.7	246	136	260	202	25.5	224
		Outdoor	24-hr	2.46	0.90	1.10	2.40	2.10	7.70	3.70	6.30	6.70	1.20	9.60
	9	Indoor	12-hr Night	1.36	0.70	0.70	0.90	0.80	2.70	1.20	2.10	2.40	0.40	3.00
		Indoor	12-hr Day	1.10	0.50	0.50	0.50	0.50	1.00	0.50	0.70	1.10	0.50	1.90
		Outdoor	24-hr	2.42	0.70	0.70	0.90	0.80	2.60	1.10	1.80	2.10	0.50	2.90

* No sample due to pump failure