

**Exposure and Health Assessments of the Effects of Agricultural Field  
Burning in Young Adults with Asthma Living in Pullman Washington**

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A final report to Grant Pfeifer of the Air Quality Program  
Washington State Department of Ecology

Final Report  
April 6, 2005

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## Executive Summary

Agricultural (Ag) burning is a cost-effective method of cleaning and preparing the field for the succeeding growth season. However, smoke from Ag burning may contain various air pollutants, which may cause or exacerbate respiratory disease. However, the short-duration excursions of Ag burning smoke often do not violate the National Ambient Air Quality Standards at locations where air quality monitors are situated. Although a limited number of studies documented potential health effects from Ag burning smoke, there is a paucity of literature characterizing community residents' exposure to Ag burning smoke and the associated health effects.

In the past several years, Ag burning has been subject to intense public health debate and in several cases law suits were filed in eastern Washington and Idaho. This study, funded by the EPA's Northwest Center for Particulate Matter and Health, the Washington State Department of Ecology and the U.S. EPA Region 10, aimed to assess the short-term exposure and health effects of Ag burning.

This study was conducted in Pullman, WA during the fall 2002 prescribed agricultural field burning season. This study consisted of 32 young adults with asthma (aged 18-52, median 24) and 2 randomly assigned monitoring sessions for each subject, including an active session and an on-call session. The active session required 16 participants to perform in-lab measures of on-line eNO (Sievers, Boulder CO), coached spirometry (microDL) and complete symptom questionnaires at the same time of day every Monday, Wednesday and Friday during a 30-day period. The on-call session occurred during the remaining 30-days of the 60-day monitoring session for these 16 individuals. During a declared episode, the on-call subjects would be paged in to have all health measures performed with the active subjects over the 3 successive days from the initial called episode. During our study, there were one sham episode and 4 real episodes, defined as more than three consecutive 30-min  $PM_{2.5}$  averages exceeding  $40 \mu g/m^3$ .

Air quality measurements at the central site included continuous PM<sub>2.5</sub>, PM<sub>10</sub>, carbon dioxide, nitrogen oxides, and meteorological conditions and 12-hour integrated PM<sub>2.5</sub>, elemental and organic carbon (EC/OC), and levoglucosan (LG)-a marker for biomass combustion. The personal exposure measurements were collected from 16 subjects, inside of all but four residences, and outside of 6 residences. Exposure estimates included personal exposure measurements of PM<sub>2.5</sub>, EC/OC, LG, calculated exposure to PM<sub>2.5</sub> of outdoor origin, and calculated exposure to PM<sub>2.5</sub> from Ag burning.

This report consists of three chapters of manuscripts. The first chapter characterizes the air quality during Ag burning episode and non-episode periods. Two source apportionment methods, including the Chemical Mass Balance model (CMB-8) and the Positive Matrix Factorization (PMF), were utilized to apportion the measured PM<sub>2.5</sub> mass concentrations to major sources found in Eastern Washington. The second chapter investigates personal exposure to PM and utilizes the CMB results in Chapter 1 to estimate personal exposure to PM originating from ambient sources and Ag burning activities. The third chapter utilizes the air quality measurements and exposure estimates to assess acute health effects from exposure to PM<sub>2.5</sub> originated from Ag burning smoke.

During the study period, the observed 1-h average PM<sub>2.5</sub> concentrations ranged between 0.3 and 59.6 µg/m<sup>3</sup>, averaging 13.0±9.2 µg/m<sup>3</sup>. Major contributions of PM<sub>2.5</sub> included soil (38%), vegetative burning (35%), and sulfate aerosol (20%) based on the CMB analysis. The PMF generated profiles were consistent with those selected for CMB modeling. In addition, the PM<sub>2.5</sub> mass concentration estimates from the two models were significantly correlated for individual sources. LG, PM<sub>2.5</sub> from biomass combustion and soil PM<sub>2.5</sub> mass concentrations (both derived from receptor modeling) were all significantly higher during the episodes than during non-episode days, while other measurements including NO<sub>x</sub>, CO<sub>2</sub>, OC, and EC were relatively similar on episode and non-episode days. Although we successfully identified Ag burning episodes with a higher contribution of PM<sub>2.5</sub> from vegetative burning, an equal or higher contribution from airborne soil dust to the real-time PM<sub>2.5</sub> measurements could not be ignored.

The observed mean personal exposure to  $PM_{2.5}$  was  $13.8 \pm 11.1 \mu\text{g}/\text{m}^3$ , which was on average  $8.0 \mu\text{g}/\text{m}^3$  higher during the Ag burning episodes ( $19.0 \pm 11.8 \mu\text{g}/\text{m}^3$ ) than non-episodes ( $11.0 \pm 9.7 \mu\text{g}/\text{m}^3$ ). The personal LG exposure also was higher during the episode than non-episode periods. The ambient contribution fraction, which propagates central site measurements to personal exposure, ranged between 0.28 and 2.21. The correlation between the central-site and personal LG was 0.75. We combined the CMB and total exposure modeling results in a model to predict  $PM_{2.5}$  exposure originated from Ag burning for individual subjects ( $E_{ab}$ ). The estimated  $E_{ab}$  ranged from 2.0 to  $7.1 \mu\text{g}/\text{m}^3$  (mean= $3.5 \pm 1.3 \mu\text{g}/\text{m}^3$ ) and correlated with personal LG measurements ( $r=0.53$ ). Uncertainties in the  $E_{ab}$  estimates were due in part to the dependence on the ambient contribution fraction for total  $PM_{2.5}$  as a surrogate for biomass burning related PM mass. We found significant between-subject variation between episodes and non-episodes in both the  $E_{ab}$  estimates and subjects' activity patterns. This suggests that the LG measurements at the central sites may not be representative of individual exposure to Ag burning smoke.

We hypothesized that adults with mild-moderate asthma who are not using anti-inflammatory medication would show a positive association of eNO and negative association of  $FEV_1$  and maximal mid-expiratory flow (MMEF) with the peak 1-hour average of  $PM_{2.5}$  during the previous 24 hours. We further refined our health assessment by using individual specific exposure estimates originated from Ag burning. Health measures included 594 on-line exhaled nitric oxide (eNO) and 591 coached spirometry tests. These health effects were assessed with a GEE model that included fixed covariates for gender, age, BMI, exposure estimates, an interaction term between medication use and exposure, and adjusted for temperature and relative humidity. There was no significant effect of peak 1-hour  $PM_{2.5}$  on measures of eNO among those not prescribed anti-inflammatory medications:  $-0.35$  ppb (95% CI:  $-1.70, 1.01$ ) per  $10 \mu\text{g}/\text{m}^3$  increase in  $PM_{2.5}$  or those prescribed controller medications:  $1.68$  ppb (95% CI:  $-1.51, 4.87$ ) per  $10 \mu\text{g}/\text{m}^3$  increase of  $PM_{2.5}$ . Similar null effects of peak  $PM_{2.5}$  exposure were noted for spirometric measures of MMEF and  $FEV_1$ . Sensitivity analyses that assessed Ag burning

related exposure using LG, indoor concentration of Ag burn related PM, or estimated exposure to ambient or Ag burning originated PM did not change our null results.

Our study had several strengths that added to the validity of the results. These included repeated in-lab measures of sub-clinical effects (eNO and spirometry), inclusion of a sham Ag burn episode to control for non-agricultural PM related changes in pulmonary measures and symptoms measures, detailed exposure measures that included residential indoor and personal measures in a community where agricultural burning represented a relatively high fraction of total PM<sub>2.5</sub>. Although the frequency and peak levels of Ag burn related PM<sub>2.5</sub> were low, they were representative of the recent Ag burning related PM. Since the 1999 Memorandum of Understanding between the wheat growers and the WA Department of Ecology, and the subsequent implementation of Ag burn control strategies by Ecology, acreage burned and Ag related PM emission have been decreasing over the years. The low and infrequent exposures were observed in the present 2002 Ag burn study as well as in the previous 2 years (2000 and 2001).

Although the null results may be true, the following factors could have contributed to the absence of effect in our study: the selection of a relatively non-susceptible study population, non-linear effects of agricultural field burn PM on eNO and spirometric measures, timing of our health assessments, inability to accurately capture the spatial and temporal variation of PM, inability to accurately measure agricultural combustion contributions to the PM mass, and an equal or greater contribution to peak PM<sub>2.5</sub> from airborne dust during the Ag burning episodes.

In conclusion, the observed PM<sub>2.5</sub> levels and excursions were typical of those of previous years. Although we did not find an association between peak PM<sub>2.5</sub> from field burning and decrements in pulmonary function or increases in eNO in young adults with asthma, we cannot rule-out health effects from field burning in more susceptible populations or at higher PM concentrations. We recommend future studies that measure sub-clinical effects on children with asthma, older individuals with cardiac disease, or farm workers exposed to potentially greater agricultural PM concentrations.

## **Acknowledgements**

We thank the subjects and Collen Marquist, Ranil Dhammapala, Mark Hoffman, Dennis Finn, Lee Bamesberger, Kristie Schumaker, and Sara Jarvis who devoted much of their time to our agricultural burning air and health monitoring work. We thank Mr. Grant Pfeifer and Ms. Karen Wood of the Washington Department of Ecology (Ecology) for their support and assistance in study design. We appreciate comments provided by the Ecology's Expert Panel. This study was funded by the Ecology (with support from US EPA Region 10) Interagency Agreement C03000099 and the EPA Northwest Center for Particulate Air Pollution and Health grant R827355.

## Chapter 1. Atmospheric Characterization

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## **1.1 Abstract**

Agricultural burning has been subject to intense debate in Eastern Washington. Rural communities are concerned about health impacts related to smoke exposure from field burning. However, the short-duration excursions of smoke often do not violate air quality standards at locations where air quality monitors are situated. The purpose of this study was to characterize the air quality in Pullman, WA during the fall 2002 prescribed field burning season, as part of a larger study conducted to examine community exposure to agricultural burning smoke and the related short-term health effects. Data collected included continuous PM<sub>2.5</sub>, PM<sub>10</sub>, CO<sub>2</sub>, nitrogen oxides, and 12-hour integrated PM<sub>2.5</sub>, OC, EC, and levoglucosan. Four episodes were defined when three consecutive 30-min PM<sub>2.5</sub> averages exceeded 40 µg/m<sup>3</sup>. Two source-receptor models; the Chemical Mass Balance model (CMB) and Positive Matrix Factorization (PMF) were used to estimate smoke intrusion from regional agricultural burning. During this study, the average PM<sub>2.5</sub>, OC, and EC were similar during the daytime and nighttime, while LG was twice as high during the night. The CMB results showed major contributions of PM<sub>2.5</sub> from soil (38%), vegetative burning (35%), and sulfate aerosol (20%), and much less from vehicles (2%) and cooking (1%). The 3-source profiles generated by PMF were consistent with those selected for CMB modeling. The PM<sub>2.5</sub> estimates from these two models were highly correlated for individual sources. In addition, the LG, NO<sub>x</sub>, CO<sub>2</sub>, OC, and apportioned PM<sub>2.5</sub> from vegetative burning and soil were all significantly higher during the episodes than during non-episode days, while EC and PM<sub>2.5</sub> from secondary sulfate, vehicles, and cooking sources were similar throughout the study. We characterized the episodes of agricultural field burning with elevated LG, OC, and biomass burning contribution.

**KEYWORDS:** biomass burning, smoke impact in rural communities, source apportionment, exposure assessment.

## **1.2 Introduction**

In the past decades, agricultural burning has been subject to intense analysis, discussion and public debate in Eastern Washington (Jimenez, 2002). Neighboring

communities from rural areas have noticed an impact on air quality from farmers' burning practices, and citizens have been concerning about possible health problems related to smoke exposure, as well as potentially negative impacts on tourism and economic activities (Roberts and Corkill, 1998). In addition to smoke from field burning, Eastern Washington, a semi-arid region, can have significant levels of particles in the air from a variety of sources. These sources mainly include fugitive dust from roads and fields (Claiborn et al., 2000; Kim et al., 2003), agricultural field burning and forest fires (Jimenez, 2002).

Smoke from biomass burning contains several chemical compounds including carbon monoxide (CO), hydrocarbons (HC), volatile organic compounds (VOC's), benzene, polycyclic aromatic hydrocarbons (PAH's) and PM<sub>2.5</sub> (Jenkins et al., 1996). Some of these compounds are known carcinogens, such as benzene and certain polycyclic aromatic hydrocarbons (Roberts and Corkill, 1998). Others, such as PM<sub>2.5</sub>, can have a potentially detrimental impact on human health from acute exposure. Ambient PM concentrations have been linked to elevated morbidity and mortality in several epidemiological studies (US EPA, 2003). Jacobs et al. (1997) studied rice straw burning and asthma hospitalizations in Butte County, CA and found a relationship between acreage burned and the risk of hospitalization. However, the authors noted, the sparsely located ambient monitors might not capture the short-term smoke episodes from rice straw burning, and hence no correlations were found between acreage burned and measurements of O<sub>3</sub>, CO and PM<sub>10</sub>. Long and coworker (1998) reported from their questionnaire survey in Winnipeg, Canada that individuals with asthma or chronic bronchitis were more likely to be affected by straw burning. More recently, Tirigoe et al. (2000) found a relationship between PM<sub>10</sub> and children with asthma attacks, with the rise of PM<sub>10</sub> most likely from rice straw burning in Niigata, Japan.

Due to the scarcity of monitors in rural Eastern Washington and the relatively short-term durations of high air pollutant concentrations, (usually less than 6 hours), smoke from agricultural field burning does not often lead to violations of the NAAQS in eastern Washington (Jimenez, 2002). Moreover, although there is evidence that short-

term excursions in  $PM_{2.5}$  result in acute health effects (Romieu et al., 1996; Pekkanen et al., 1997; Peters et al., 1997; Vedal et al., 1998; Roemer et al., 2000; Yu et al., 2000; Ostro et al., 2001; Delfino et al., 2002; Delfino et al., 2003), short-term  $PM_{2.5}$  excursions are not currently regulated. The purpose of this study was to characterize air quality in Pullman, an eastern Washington town, during a period of prescribed agricultural field burning, as part of a larger study examining community exposure to agricultural burning smoke and the related health effects. Two source-receptor models were used to estimate smoke intrusion from regional agricultural field burning in the observed  $PM_{2.5}$  mass concentrations in Pullman.

## **1.3 Methods**

### **1.3.1 Monitoring period and site selection**

This study was conducted between September and November 2002 based on the historical evidence of greater amounts of acres burned per day and more smoke episodes reported by citizens in the fall burning season in Eastern Washington (Jimenez, 2002). The air quality measurements analyzed in this paper were collected primarily at one central monitoring site located at the rooftop of the Washington State University (WSU) Engineering Building (elevation= 770 m) to represent the ambient air quality at the WSU Campus (average elevation= 768 m) and other general Pullman area (average elevation= 774 m).

### **1.3.2 Air quality sampling**

Air quality data collected included continuous  $PM_{10}$  and  $PM_{2.5}$  from Tapered Element Oscillating Microbalance (TEOM) monitors (30-min averages, Series 1400a, Rupprecht & Patashnick Co., Inc), a light scattering nephelometer (10-min averages, M903, Radiance Research, Seattle, WA), a DataRAM with a  $PM_{2.5}$  size-selective inlet (10-min averages), and 12-h integrated  $PM_{2.5}$  samples (8:00 to 20:00 and 20:00 to 8:00) from collocated and triplicate single-stage 10-LPM Harvard Impactors ( $HI_{2.5}$ ) (Air Diagnostics INC., Naples, ME). Two  $HI_{2.5}$  sampled  $PM_{2.5}$  onto 37-mm Teflon filters, and

the third sampled onto a quartz filter. The nephelometer was calibrated against the  $HI_{2.5}$  measurements. Other air constituents included continuous carbon dioxide (Telaire 1050 Engelhard, Goleta, CA), nitrogen oxides (model 42 Thermal Environmental Instruments, Inc), and carbon monoxide (Model 9830 Monitor Labs, Inc). Meteorological parameters including temperature, humidity, wind speed and wind direction were also recorded using a small weather station (WeatherLink, Davis Instruments Corp. Hayward, CA 94545).

The PM collected on the Teflon media underwent gravimetric analysis using a Mettler-Toledo UMT2 at the University of Washington laboratory at constant temperature ( $22.2 \pm 1.8^\circ\text{C}$ ) and relative humidity ( $34.8 \pm 2.5\%$ ) for at least 24 hours prior to weighing (Allen et al., 2001). Filters were then analyzed for 55 inorganic elements using X-ray Fluorescence (XRF) at the Chester Lab (Chester LabNet, Tigard, Oregon). One set of the duplicate Teflon filters were extracted by ultrasonication in ethylacetate/triethylamine for LG analysis using Gas Chromatography – Mass Spectrometry (GC-MS) (Simpson et al., 2004). Sections of the quartz filters ( $1 \text{ cm}^2$ ) were analyzed for OC and EC via Thermal Optical Transmittance (Sunset Laboratory, Inc. Tigard, OR) using a modified version of the NIOSH 5040 method (Pang et al., 2002).

### **1.3.3 Episode definition**

An episode was declared when three or more 30-minute average  $PM_{2.5}$  concentrations exceeded  $40 \mu\text{g}/\text{m}^3$  during any 24-hour period according to the central site  $PM_{2.5}$  TEOM, DataRAM, and/or nephelometer measurements. This threshold value was selected based on the frequency of historical (2000 and 2001) hourly  $PM_{2.5}$  observations exceeding this magnitude at the downtown Pullman Nephelometer monitoring site operated by the Washington State Department of Ecology (DOE) (see Table 1). Our previous study (Jimenez, 2002) found a link between these exceedances and vegetative burning smokes episode in Pullman.

Episode determination was also aided by visual observations of agricultural burning smoke that might not be registered at the central site monitors due to wind directions; current and predicted meteorological conditions that may favor the occurrence

of an episode; and the WA DOE's daily morning burn calls for neighboring regions. This study also included a single-sided blind sham episode during a period of relatively low PM<sub>2.5</sub> levels to compare the subject health effect responses with any true episode periods. A declared episode triggered three consecutive days of intensive health effect monitoring (for more details on the exposure and health assessments, see Wu et al., 2005; Sullivan et al., 2005).

#### **1.3.4 Quality Control**

Field blanks and duplicates were deployed so that they comprised at least 10% of the total HI<sub>2.5</sub> sample size. The precision (1.2 µg/m<sup>3</sup>) and accuracy (3%) of the HI<sub>2.5</sub> have been reported in a previous paper (Liu et al. 2003). Filters were analyzed for LG by batch of approximately 20 filters. In each batch, two laboratory blanks and 4 spiked samples (with d<sup>7</sup>-levoglucosan in the extracts) were analyzed. The overall recovery based on the spiked samples was 75 ± 11%. The analytical precision based on 10-15% of samples within each batch that were analyzed in duplicate was 24%. Among the 104 samples analyzed for LG, 9 samples were below the limit of detection (LOD ~ 0.02 µg/mL) and two samples had fatal analytical errors (unacceptably low recoveries, failure to derivatize, or chromatographic interferences).

#### **1.3.5 Data analysis**

The concentration data for PM, gaseous pollutants, and particulate carbonaceous species at the central site were tested for differences between the episode and non-episode periods with a two-tailed two-sample t-test. The carbon fractions (OC and EC) in PM<sub>2.5</sub> were intended to be used to identify smoke impact and secondary organic aerosols. Particulate organic carbonaceous species may be emitted directly as primary particles or formed in the atmosphere from chemical reactions of semi-volatile organic compounds, while EC aerosol or soot is only emitted as primary PM (Cao et al., 2003). Source apportioned PM<sub>2.5</sub> mass concentrations (described below) were also compared between the episode and non-episode periods.

Data used for source apportionment analysis were subjected to several constraints and consistency checks.  $HI_{2.5}$  measurements were compared to those from the  $TEOM_{2.5}$  and to the reconstructed fine mass (RCFM), which is defined as the sum of the individual components of fine PM fractions from each chemical analysis; i.e. OC, EC and inorganic elements (Malm et al., 1994)

$$RCFM = 1.6[OC] + [EC] + [sulfate] + [nitrate] + [soil] + [others] \quad \dots\dots(1)$$

where the brackets denote mass concentrations ( $\mu\text{g}/\text{m}^3$ ) of each component. “Soil” corresponds to the sum of elements predominately associated with soil (Fe, Al, Si, Ca, and Ti), plus oxygen for the oxidized state of these elements ( $Al_2O_3$ , SiO, CaO, FeO,  $Fe_2O_3$ ,  $TiO_2$ ):

$$Soil = 2.20[Al] + 2.49[Si] + 1.63[Ca] + 2.42[Fe] + 1.94[Ti] \quad \dots\dots(2)$$

“Others” represents all other elements that were analyzed. RCFM was calculated for all 12-hour samples based on the XRF and EC/OC results and compared to the observed  $HI_{2.5}$  mass concentration.

For source apportionment, we used the US EPA’s Chemical Mass Balance receptor model Version 8 (CMB-8), which consists of a solution to linear equations that expresses each receptor chemical species concentration as a linear sum of products of the mass fraction of a chemical or a tracer in the emissions from each source type, and source contributions (Watson et al., 2001). The main inputs to the CMB-8 model were the  $PM_{2.5}$  chemical composition data, the mass fraction of the chemical species in the source profiles, and the uncertainties of individual species. Chemical species with large uncertainties have less influence in the solution because in the fitting procedure they are not weighed as much as those more precisely measured species or unique tracers. CMB-8 also provides internal performance measures to evaluate the results through the use of several fit indices.

The sources of PM<sub>2.5</sub> selected in the CMB-8 model included windblown dust (soil), vegetative burning smoke, secondary sulfate aerosol, vehicular traffic, and cooking fumes. The soil and sulfate aerosol profiles were derived from the Spokane dust profile (Core et al., 1982) and a previous source apportionment study in Spokane (Kim et al., 2003). The vegetative burning smoke profile was derived from a previous source apportionment study in Spokane (Hoffman, 2002) and other documented studies of emission factors for wood smoke reporting LG, which have a 5-fold variation (422 – 23,299,000 µg LG/ kg of wood) depending on the plant species (Oros and Simoneit, 2001; Oros and Simoneit, 2001b). We also included profiles documented in the receptor model source composition library (U.S. EPA-450/4-85-002) for vehicular traffic (Cass and McRae, 1981) and cooking fumes (Hildemann et al., 1991). The chemical tracers considered for modeling were Al, Br, Ca, Cl, Cu, OC, EC, Fe, K, Mn, S, Si, SO<sub>4</sub>, Ti, Zn and LG. We excluded elements with more than 70% samples below detection limit (i.e. Cr, Na, Ni, Pb, V).

In addition to the CMB model, we also applied the positive matrix factorization model (PMF) for source apportionment. The PMF model is a statistical model that adopts the least-squares approach to solve the factor analysis problem without requiring source profiles as the input variables (Paatero, 1997). We followed the procedure of Polissar et al. (2001) and Maykut et al. (2003) to generate the PMF model inputs and assign uncertainties to each measurement and developed three-source (3S) and four-source (4S) PMF models. We tested various FPEAK parameters, ranging from -0.8 to 0.8 with an increment of 0.1 to minimize the rotational ambiguity (Paatero, 1997; Maykut et al., 2003). The ‘Q values’ indicated that the FPEAK values between -0.4 and 0.1 provided the optimum solutions for the 3S and 4S models. After the factors were retrieved from the PMF models, they were regressed against the total PM<sub>2.5</sub> mass concentrations to obtain the source-specific PM<sub>2.5</sub> mass concentrations. These PMF generated source profiles were then compared with those used for the CMB-8.

## 1.4 Results

### 1.4.1 Air quality measurements

The TEOM<sub>2.5</sub>, TEOM<sub>10</sub>, and nephelometer PM<sub>2.5</sub> data (September - October 2002) are shown in Figure 1. Several TEOM<sub>2.5</sub> measurements spiked above 40 µg/m<sup>3</sup> and triggered two episode calls, including the periods of Sept 11-15 during which two consecutive episode calls were made, and Oct 17-19. Two potential episodes were not declared during Sept 25-26 and Oct 24-26. A sham episode was declared for Oct 9-11. The TEOM<sub>2.5</sub> was not functioning from 9/28 to 10/17, during which period the nephelometer data (calibrated against the HI<sub>2.5</sub> measurements) were used instead. The nephelometer was calibrated against the collocated HI<sub>2.5</sub> measurements (intercept=  $0.134 \times 10^{-5} \text{ m}^{-1}$ , slope=  $0.243 \times 10^{-5} \text{ m}^2/\mu\text{g}$ ,  $R^2 = 0.83$ , N= 99) and was lower than the heated TEOM<sub>2.5</sub> measurements. The discrepancy in peak values between TEOM<sub>2.5</sub> and neph is discussed later. Table 2 summarizes the measurements of PM and gaseous pollutants (CO<sub>2</sub>, CO and NO<sub>x</sub>). The mean PM<sub>10</sub> and PM<sub>2.5</sub> levels were 44.6, 15.1 (TEOM), and 11.3 µg/m<sup>3</sup> (nephelometer), respectively, with TEOM<sub>2.5</sub> exceeding 40 µg/m<sup>3</sup> for 61 30-min periods, while the nephelometer only recorded 9 30-min periods exceeding 40 µg/m<sup>3</sup>. Note that due to spatial variation in PM<sub>2.5</sub> (Wu et al. 2005), there were only 4 exceedances recorded by the nephelometer located in downtown Pullman (Table 1). The number of exceedances in 2002 as recorded by the nephelometer in 2002 was typical of the previous two years.

Table 3 summarizes the 12-h integrated PM<sub>2.5</sub>, OC, EC, LG, and the trace elements used in the source apportionment analyses. The means of the nighttime measurements for PM<sub>2.5</sub>, OC, EC, LG and some of the trace elements were higher than those of the daytime measurements, with a significant difference observed for PM<sub>2.5</sub> (p<0.01), LG (p<0.01) and the trace elements Si, Al, S, Ca, K and Mg (p<0.01). The day- and nighttime differences could be due to the effect of nighttime inversion with limited atmospheric mixing and/or nighttime residential wood burning.

The average ratio of RCFM to HI<sub>2.5</sub> was  $0.98 \pm 0.22$  (N= 123), showing good consistency between the reconstructed PM<sub>2.5</sub> and the actual gravimetric PM<sub>2.5</sub> mass

concentrations. Our LG measurements (mean= 74 ng/m<sup>3</sup>, range 2-327 ng/m<sup>3</sup>) were comparable to measurements obtained in Israel, mean ~ 73 ng/m<sup>3</sup> (Graham et al., 2004) and higher than those observed in Brazil, 1.65-7.45 ng/m<sup>3</sup> and 0.15-28.42 ng/m<sup>3</sup> (Santos et al., 2002; Santos et al., 2004), and Nigeria, 0.04- 3.3 ng/m<sup>3</sup> (Stanley and Simoneit, 1990; Simoneit et al., 1988). However, our observations were lower than those detected in other U.S. urban areas, 280-4860 ng/m<sup>3</sup> and 200-1200 ng/m<sup>3</sup> (Simoneit et al., 1993, 1999; Nolte et al., 2002; Fraser and Lakshmanan, 2000), and far below those observed during severe episodes of biomass smoke pollution in Southeast Asia, 1400-40240 ng/m<sup>3</sup> (Radzi Bin Bas et al., 2004). The differences were probably due to the proximity to sources as well as the extent of biomass burning that took place on a given day.

The correlations between air pollutants are summarized in Table 4. The Pearson's correlation coefficients were high for the TEOM<sub>10</sub> and TEOM<sub>2.5</sub> (r= 0.84), TEOM<sub>2.5</sub> and HI<sub>2.5</sub> (r= 0.80), and Neph and HI<sub>2.5</sub> (r=0.81). The lower correlation between the Neph and TEOM<sub>2.5</sub> (r= 0.68) was most likely due to the higher variation in the 30-min PM<sub>2.5</sub> concentrations, especially during spikes. The nephelometer may respond differently based on the nature of the episodes and thus aerosol characteristics (Liu et al., 2002). NO<sub>x</sub>, OC, and LG were also correlated with HI<sub>2.5</sub> (r>0.44, p< 0.01), suggesting that some of the PM<sub>2.5</sub> were related to vegetative combustion sources upwind. Concentrations of EC and CO<sub>2</sub> during episodes were not different from the non-episode levels and showed the least correlations with other parameters.

#### **1.4.2 Source apportionment**

Figure 2 shows the chemical profiles as dark textured bars of the five sources included in the CMB model. LG was used as a unique tracer for vegetative burning smoke to allow for a better separation from other combustion sources. The average PM<sub>2.5</sub> contribution from airborne soil was 4.55 ± 0.03 µg/m<sup>3</sup> (38%); from vegetative burning 3.96 ± 0.13 µg/m<sup>3</sup> (35%); from sulfate aerosol 2.22 ± 0.06 µg/m<sup>3</sup> (20%); from vehicular traffic 0.19 ± 0.01 µg/m<sup>3</sup> (2%); from cooking 0.12 ± 0.04 µg/m<sup>3</sup> (1%); and 0.42 ± 0.24 µg/m<sup>3</sup> (4%) from unexplained sources (Figure 3). Table 5 shows comparisons in the average source contributions to fine aerosol mass concentrations between Spokane (Kim et al., 2003) and Pullman. The Spokane study was conducted from 1995 through 1997,

and PMF was used for source apportionment. The average vegetative burning PM<sub>2.5</sub> was slightly higher in Spokane than Pullman. However, the average airborne soil PM<sub>2.5</sub> observed in Pullman was four times larger than that in Spokane. Sources of airborne soil could be a direct effect from the agricultural fields as well as unpaved roads surrounding Pullman. In addition, the contribution of PM<sub>2.5</sub> from vehicles was greater in Spokane than in Pullman, which is consistent with the greater amount of vehicular traffic in Spokane. The average contribution of sulfate aerosol to PM<sub>2.5</sub> was similar in both cities, suggesting a regional source of sulfate aerosol.

The observed relatively low PM<sub>2.5</sub> (HI<sub>2.5</sub> mean 11.5 µg/m<sup>3</sup>) and the uncertainties in the LG emission factors for wheat straw burning were initially considered as limitations for CMB to separate vegetative burning smoke from other combustion sources. During the study we found a significant inverse correlation ( $r = -0.6$ ,  $p < 0.01$ ) between LG and ambient temperature, which could be due to limited atmospheric mixing or more frequent residential wood burning at lower temperatures. Note that we could not distinguish LG emitted from residential wood burning from agricultural burning.

The 3-source PMF model identified vegetative burning, windblown dust, and secondary sulfate sources, with similar source profiles (shown as grey bars in Figure 2) to those of CMB. This consistency suggests that the CMB source profiles, which were compiled from studies in nearby cities, were suitable for the Pullman airshed. Note that LG was a significant component in only one profile (Veg burning) in the PMF modeling results, supporting the use of LG as a unique tracer for vegetative burning. The source-specific PM<sub>2.5</sub> mass concentrations estimated from the two models were highly correlated ( $p < 0.05$ ), with a Pearson's correlation coefficient of 0.70, 1.00, and 0.78 for vegetative burning, windblown dust, and secondary sulfate, respectively (Figure 4).

The 4-source PMF model identified one more source, "others," in addition to the three identified by the 3-source PMF model. However, the use of PMF in this study is limited by the small sample size ( $N = 123$ ). When we forced the PMF to produce 4 sources, we artificially introduced errors. The 3-source PMF model results provided results that were more consistent with the CMB results than the 4-source PMF model, so the 4-source model was not analyzed further. We also determined that the CMB estimates

were more reliable than the 3S PMF results based on the consistent findings between our CMB results and those in the Spokane study (Kim et al. 2003), no sample size constraints for CMB analysis, and the ability to include uncertainties in the CMB source estimates. Ultimately, the PMF results served as an independent method to further confirm the CMB results through the similar source profiles, identification of LG as a unique tracer for biomass burning, and high correlations between source estimates between the two methods.

### 1.4.3 Episode calls

Table 6 compares pollutant concentrations during episodes vs. non-episode days, including the sham episode. By definition, the continuous PM observations from the nephelometer were significantly higher ( $p < 0.01$ ) during all episodes than those during non-episode days. LG,  $\text{NO}_x$ ,  $\text{CO}_2$ , and OC were significantly higher during episode than non-episode days, with or without controlling for temperature using ANOVA. EC, on the other hand, was not elevated during episode days.

The CMB-estimated source contributions for episode vs. non-episode days are also shown in Table 6. PM from vegetative burning was significantly higher during episode days (average  $5.2 \mu\text{g}/\text{m}^3$ ) than non-episode days (average  $3.0 \mu\text{g}/\text{m}^3$ ). However, we also found a larger contribution of  $\text{PM}_{2.5}$  from soil during episode ( $6.9 \mu\text{g}/\text{m}^3$ ) than non-episode days ( $2.8 \mu\text{g}/\text{m}^3$ ). As expected, PM from other sources (secondary sulfate, vehicle, and cooking) did not differ between episodes and non-episode days. This study was conducted during the fall dry season, which also experienced enhanced dust intrusion from nearby roads and adjacent fields upwind of Pullman. Thus, it was likely that the real-time  $\text{PM}_{2.5}$  measurements were enhanced by both the presence of airborne soil dust and vegetative burning smoke. Although an episode declaration based solely on the criterion of exceeding a threshold  $\text{PM}_{2.5}$  value of  $40 \mu\text{g}/\text{m}^3$ , the mass measurements alone could not distinguish the soil from the biomass burning contribution. We also analyzed the effects of burn calls and total acreage burned during episode vs. non-episode days. These attempts failed because of the difficulties in collecting burn calls in the region across two states and inaccurate records of acreages burned. We also conducted back

trajectory analyses to track movement of the air mass during episodes but encountered similar difficulties in locating the exact field burn sites on any specific day.

## **1.5 Conclusion**

This study characterized the air quality in Pullman during the 2002 fall burning season. We found that the average  $PM_{2.5}$ , OC and EC concentration in Pullman were very similar during the daytime and nighttime, while LG was significantly higher during the nighttime due to trapping inversions and/or possibly residential wood burning. Good correlations between  $PM_{2.5}$  and  $NO_x$  as well as with OC, LG and  $HI_{2.5}$  suggested that the some of the observed  $PM_{2.5}$  originated from combustion sources upwind of the monitoring site.

Vegetative burning was found to be the second largest source of  $PM_{2.5}$  (35%), after airborne soil (38%). Our CMB results were consistent with those found previously in Spokane. In addition, the a priori CMB source profiles were similar to those identified by the PMF algorithm. Furthermore, the source-specific  $PM_{2.5}$  mass concentrations estimated by CMB were highly correlated with the contributions from the subset of sources identified by PMF. The PMF results confirmed that the CMB source profiles compiled from studies in other nearby cities were applicable to the Pullman airshed.

In this study, four smoke episodes were identified. LG,  $NO_x$ ,  $CO_2$ , OC, and apportioned biomass burning  $PM_{2.5}$  were all significantly higher during episode compared to non-episode days. EC was not significantly elevated during the episode periods. On the other hand, airborne dust also showed elevated levels during the defined field burning episodes.  $PM_{2.5}$  measurements alone could not be used to distinguish biomass combustion contribution from soil contribution during these episodes because the study was conducted in the dry season with soil enhancement.

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## **1.7 Figure Captions**

Figure 1. Continuous 30-min average PM<sub>2.5</sub> and PM<sub>10</sub> concentrations measured by TEOM and nephelometer and episode calls during the two-month study.

Figure 2. Source profile selected for CMB and predicted by 3s PMF analysis for the samples collected at the central site in Pullman during the two months study.

Figure 3. CMB source apportionment for five sources of PM selected for Pullman during months of September and October 2002.

Figure 4. Correlation between CMB and PMF 3S apportionment for the major sources of fine PM in Pullman.

## **1.8 Table Headings**

Table 1. Historical PM<sub>2.5</sub> hourly nephelometer measurements at the downtown Pullman monitoring site operated by the Washington State Department of Ecology.

Table 2. Summary of statistics for PM and gaseous pollutant observations at the central site during the two-month study period.

Table 3. Summary statistics for the 12-hour integrated PM samples at the central site during the two-month study period.

Table 4. Summary of 12-hour average correlation (number of samples) between PM, NO<sub>x</sub>, CO<sub>2</sub>, EC, OC and levoglucosan (LG) observed during the study.

Table 5. Summary of average source contributions to fine particle mass concentration found in Spokane and Pullman.

Table 6. Summary of results from t-test comparing episodes vs. the non-episode for the continuous samplers.

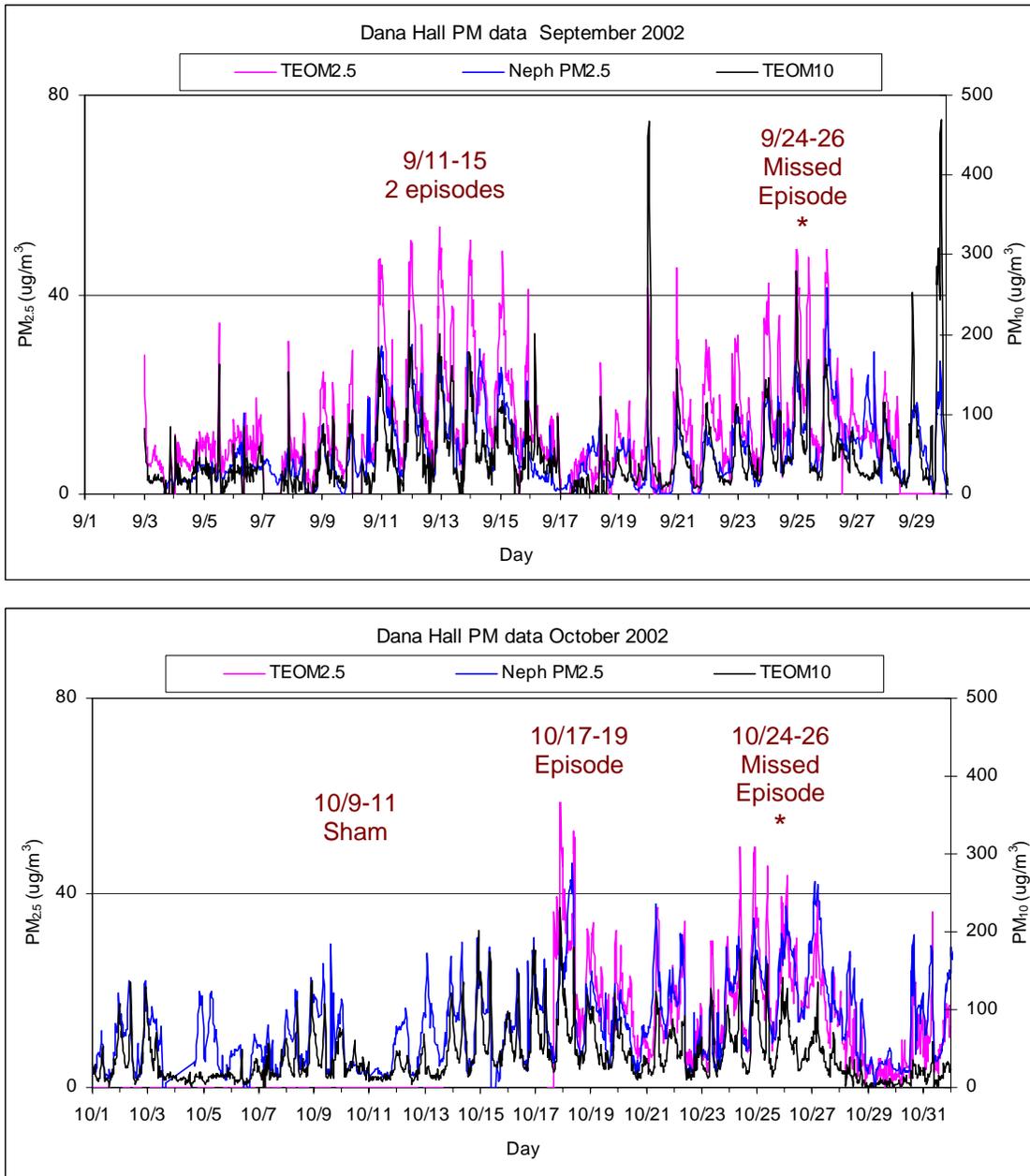


Figure 1.

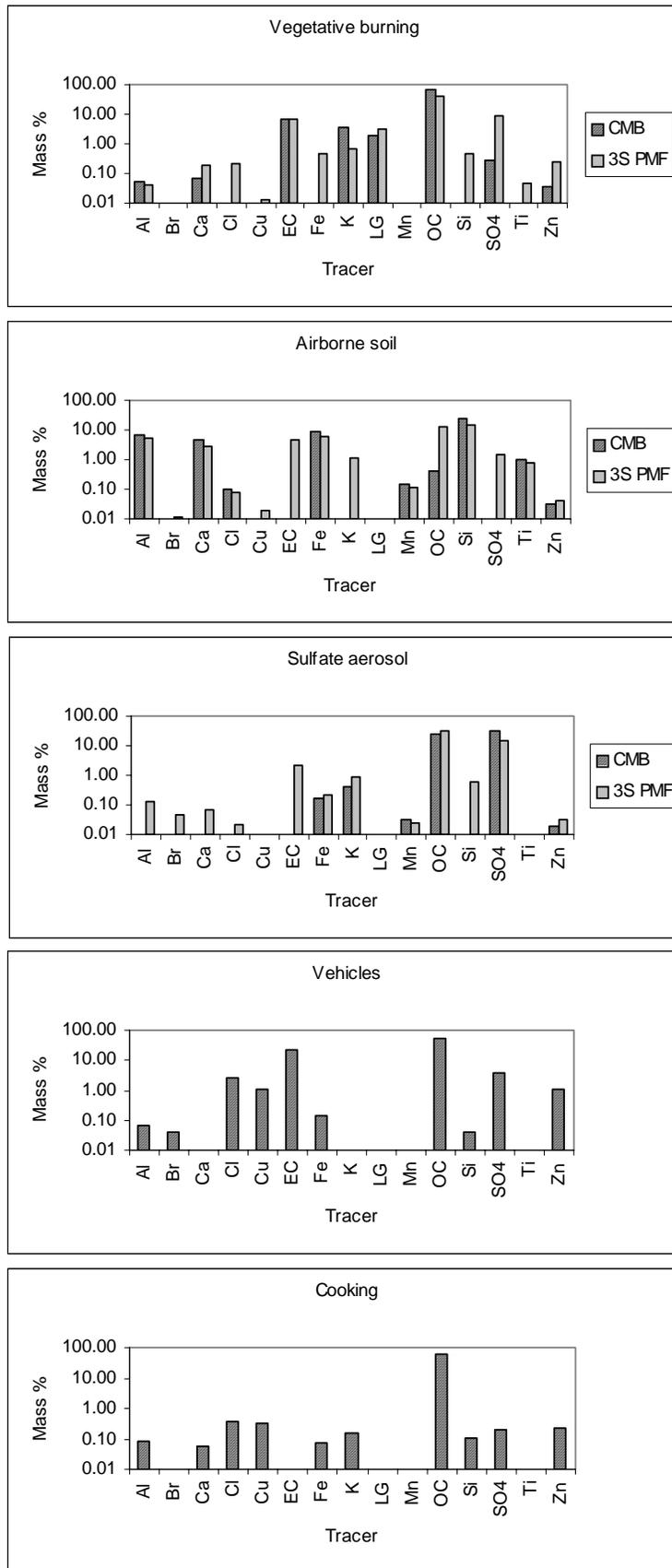


Figure 2.

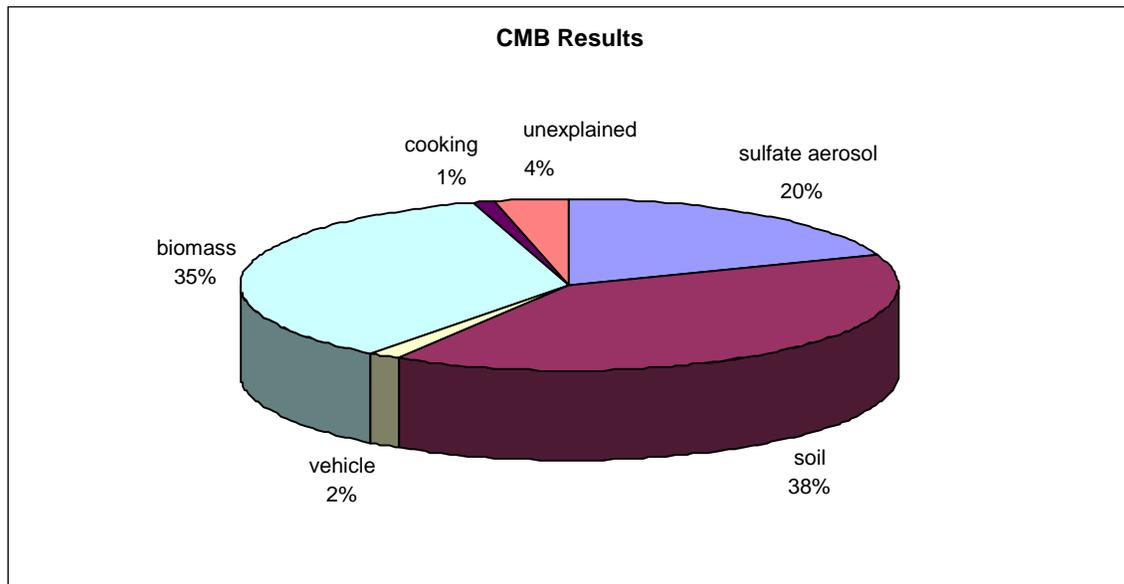


Figure 3.

Figure 4.

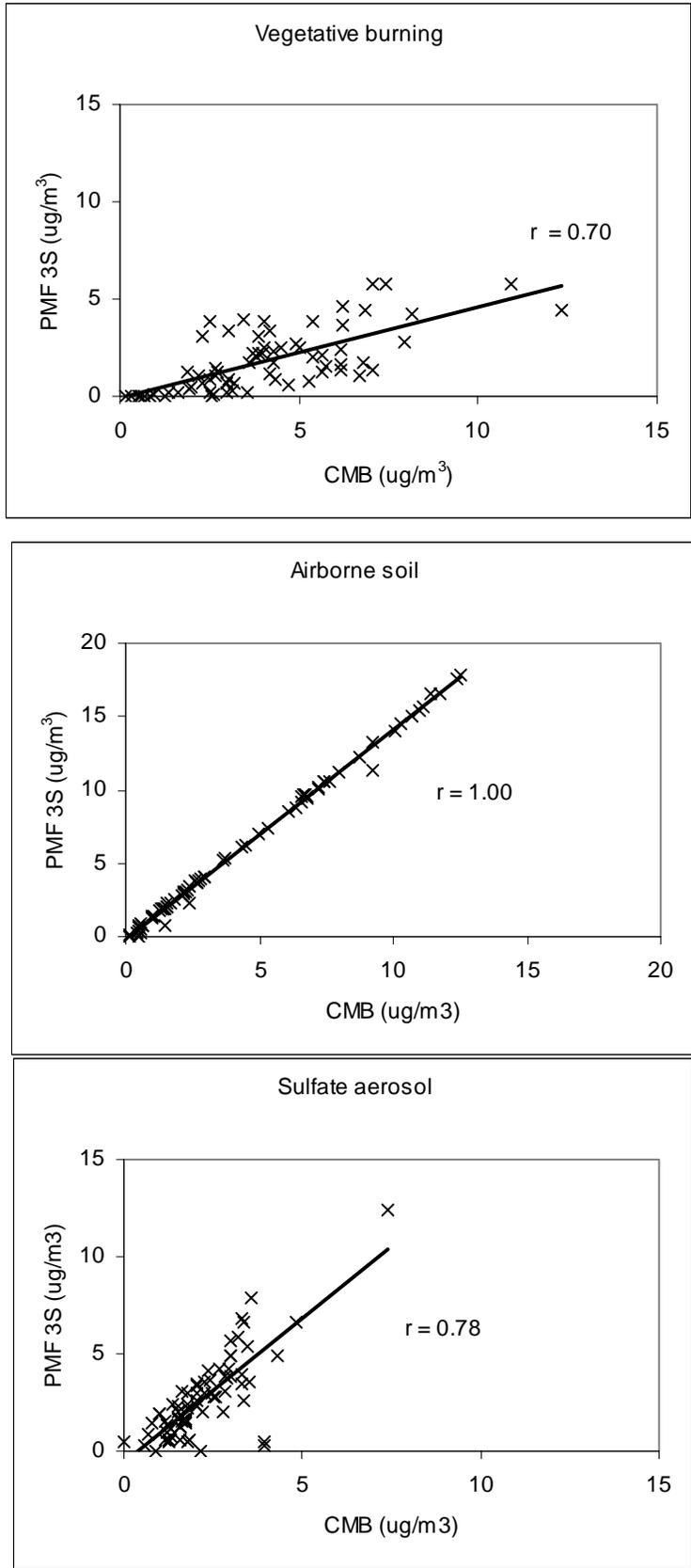


Table 1. Historical PM<sub>2.5</sub> hourly nephelometer measurements at the downtown Pullman monitoring site operated by the Washington State Department of Ecology.

Range of PM <sub>2.5</sub>	September - October		
	Year 2000 Frequency	Year 2001 Frequency	Year 2002 Frequency
0 < PM <sub>2.5</sub> < 10 µg/m <sup>3</sup>	1113	992	753
10 < PM <sub>2.5</sub> < 40 µg/m <sup>3</sup>	332	117	635
PM <sub>2.5</sub> > 40 µg/m <sup>3</sup>	13	4	4

Table 2. Summary of statistics for PM and gaseous pollutant observations at the central site during the two-month study period.

Parameter	TEOM <sub>10</sub> (µg/m <sup>3</sup> )	TEOM <sub>2.5</sub> (µg/m <sup>3</sup> )	Neph PM <sub>2.5</sub> (µg/m <sup>3</sup> )	CO (ppm)	NOx (ppb)	CO <sub>2</sub> (ppm)
Mean	44.6	15.1	11.3	0.51	34.9	465
Standard Deviation	42.2	10.1	8.0	0.56	38.1	29
Minimum	5.1	1.2	0.1	0.04	0.3	405
Maximum	469.2	67.4	48.5	3.21	294.6	568
25 percentile	17.8	7.8	4.9	0.09	9.8	438
Median	31.5	12.5	9.3	0.32	20.1	464
75 percentile	56.6	19.5	15.9	0.71	45.5	483
Observations (N)	2665	1852	2788	784	2857	2841
Frequency PM <sub>2.5</sub> > 40 µg/m <sup>3</sup>	-	61	9	-	-	-

30-minutes average integration time

Table 3. Summary statistics for the 12-hour integrated PM samples at the central site during the two-month study period.

Species	Day					Night				
	Mean	Median	Min	Max	N	Mean	Median	Min	Max	N
<sup>1</sup> HI <sub>2.5</sub> <sup>++</sup>	10.0	9.6	2.9	21.9	58	13.0	11.3	2.1	31.0	59
<sup>1</sup> OC	3.8	3.6	1.1	7.1	56	3.9	3.77	1.0	7.8	57
<sup>1</sup> EC	0.4	0.2	0.0	3.3	56	0.5	0.4	0.0	1.9	57
<sup>2</sup> LG <sup>++</sup>	50	31	2	327	45	96	76	3	318	50
<sup>2</sup> Si <sup>++</sup>	413	361	20	2088	60	996	967	13	2721	63
<sup>2</sup> Al <sup>++</sup>	137	121	0	692	60	363	313	0	958	63
<sup>2</sup> S <sup>++</sup>	252	252	68	675	60	206	177	62	416	63
<sup>2</sup> Ca <sup>++</sup>	77	71	4	333	60	210	148	0	533	63
<sup>2</sup> K <sup>++</sup>	83	71	19	254	60	121	118	13	287	63
<sup>2</sup> Na	46	30	0	315	60	52	20	0	272	63
<sup>2</sup> Mg <sup>++</sup>	12	3	0	87	60	38	19	0	186	63
<sup>2</sup> Ba	6	0	0	44	60	8	0	0	64	63
<sup>2</sup> Zn <sup>+</sup>	6	4	0	28	60	8	6	0	31	63
<sup>2</sup> Cl	6	2	0	72	60	7	4	0	45	63

<sup>1</sup>Units of mass concentration in  $\mu\text{g}/\text{m}^3$  <sup>2</sup>Units of mass concentration in  $\text{ng}/\text{m}^3$

<sup>++</sup>p< 0.01; <sup>+</sup>p< 0.05

Table 4. Summary of 12-hour average correlation (number of samples) between PM, NO<sub>x</sub>, CO<sub>2</sub>, EC, OC and levoglucosan (LG) observed during the study.

	TEOM <sub>10</sub>	TEOM <sub>2.5</sub>	Neph PM <sub>2.5</sub>	HI <sub>2.5</sub>	CO <sub>2</sub>	NO <sub>x</sub>	OC	EC	LG
TEOM <sub>10</sub>	1.00								
TEOM <sub>2.5</sub>	0.84 <sup>++</sup> (1699)	1.00							
Neph PM <sub>2.5</sub>	0.50 <sup>++</sup> (2607)	0.68 <sup>++</sup> (1792)	1.00						
HI <sub>2.5</sub>	0.76 <sup>++</sup> (104)	0.80 <sup>++</sup> (70)	0.81 <sup>++</sup> (102)	1.00					
CO <sub>2</sub>	0.19 <sup>++</sup> (2593)	0.23 <sup>++</sup> (1775)	0.16 <sup>++</sup> (2840)	-0.02 (104)	1.00				
NO <sub>x</sub>	0.31 <sup>++</sup> (2585)	0.48 <sup>++</sup> (1769)	0.40 <sup>++</sup> (2704)	0.51 <sup>++</sup> (104)	0.30 <sup>++</sup> (2765)	1.00			
OC	0.29 <sup>++</sup> (118)	0.45 <sup>++</sup> (81)	0.61 <sup>++</sup> (116)	0.64 <sup>++</sup> (104)	-0.07 (118)	0.24 <sup>++</sup> (118)	1.00		
EC	0.31 <sup>++</sup> (118)	0.36 <sup>++</sup> (81)	0.40 <sup>++</sup> (116)	0.42 <sup>++</sup> (104)	-0.05 (118)	0.47 <sup>++</sup> (118)	0.41 <sup>++</sup> (118)	1.00	
LG	0.16 (95)	0.30 <sup>+</sup> (62)	0.61 <sup>++</sup> (93)	0.44 <sup>++</sup> (95)	-0.03 (95)	0.25 <sup>+</sup> (95)	0.47 <sup>++</sup> (95)	0.18 (95)	1.00

<sup>++</sup>p < 0.01; <sup>+</sup>p < 0.05

Table 5. Summary of average source contributions to fine particle mass concentration found in Spokane and Pullman.

Average source contribution in Spokane *Kim et al. (2003)			Average source contribution in Pullman		
	Mass contribution (µg/m <sup>3</sup> )	%		Mass contribution (µg/m <sup>3</sup> )	%
Vegetative burning	5.28 ± 0.14	44	Vegetative burning	3.96 ± 0.13	35
Airborne soil	1.01 ± 0.04	8	Airborne soil	4.55 ± 0.03	38
Sulfate aerosol	2.30 ± 0.04	19	Sulfate aerosol	2.22 ± 0.06	20
Motor vehicle	1.29 ± 0.04	11	Motor vehicle	0.19 ± 0.01	2
Nitrate aerosol	1.04 ± 0.05	9	Cooking	0.12 ± 0.04	1
Chlorine-rich	0.68 ± 0.03	6	Unexplained	0.42 ± 0.24	4
Metal processing	0.29 ± 0.01	3			

The Spokane study was conducted from 1995 through 1997, and PMF was used for source apportionment

Table 6. Summary of t-test results comparing various air pollution measurements and estimates between real episode (regardless of declaration) and non-episode (including sham) days.

Variable	All episodes Mean $\pm$ SD (N)	Non episodes Mean $\pm$ SD (N)
Neph PM <sub>2.5</sub> ( $\mu\text{g}/\text{m}^3$ )	16.76 $\pm$ 8.63 <sup>++</sup> (1442)	9.86 $\pm$ 8.51 (6832)
LG (ng/m <sup>3</sup> )	106 $\pm$ 115 <sup>+</sup> (19)	59 $\pm$ 70 (86)
NO <sub>x</sub> (ppb)	42.33 $\pm$ 43.31 <sup>++</sup> (461)	33.94 $\pm$ 37.01 (2370)
CO <sub>2</sub> (ppm)	470 $\pm$ 36 <sup>++</sup> (483)	463 $\pm$ 30 (2362)
OC ( $\mu\text{g}/\text{m}^3$ )	4.75 $\pm$ 1.94 <sup>++</sup> (24)	3.14 $\pm$ 1.48 (94)
EC ( $\mu\text{g}/\text{m}^3$ )	0.59 $\pm$ 0.53 (24)	0.41 $\pm$ 0.53 (94)
Vegetative burning ( $\mu\text{g}/\text{m}^3$ )	5.22 $\pm$ 2.96 <sup>++</sup> (19)	2.97 $\pm$ 1.80 (78)
Airborne soil ( $\mu\text{g}/\text{m}^3$ )	6.92 $\pm$ 3.86 <sup>++</sup> (19)	2.83 $\pm$ 2.73 (78)
Sulfate ( $\mu\text{g}/\text{m}^3$ )	2.41 $\pm$ 0.75 (19)	2.21 $\pm$ 1.13 (78)
Vehicles ( $\mu\text{g}/\text{m}^3$ )	0.22 $\pm$ 0.19 (19)	0.15 $\pm$ 0.17 (78)
Cooking ( $\mu\text{g}/\text{m}^3$ )	0.09 $\pm$ 0.19 (19)	0.11 $\pm$ 0.29 (78)

<sup>++</sup>p < 0.01; <sup>+</sup>p < 0.05

## Chapter 2. Exposure Assessment

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## 2.1 Abstract

Several studies have documented potential health effects due to agriculture (Ag) burning smoke. However, there is a paucity of literature characterizing community residents' exposure to Ag burning smoke. This study assesses personal exposures to PM<sub>2.5</sub> (particulate matter with aerodynamic diameters < 2.5 μm) from agriculture burning smoke (E<sub>ab</sub>) for 33 asthmatic adults in Pullman, WA. The study took place in September-October of 2002, which corresponds to the primary field-burning season. PM<sub>2.5</sub> concentrations were measured on 16 subjects, inside of all but four residences, outside of 6 residences, and at a central site. The mean personal exposure to PM<sub>2.5</sub> was 13.8±11.1 μg/m<sup>3</sup>, which was on average 7.9 μg/m<sup>3</sup> higher during the Ag burning episodes (19.1±12.1 μg/m<sup>3</sup>) than non-episodes (11.1±9.7 μg/m<sup>3</sup>). The levoglucosan (a unique marker for biomass burning PM) on personal filter samples also was higher during the episodes than non-episodes. We applied the random component superposition model on central and home indoor PM measurements, and estimated an ambient contribution fraction that propagates central site measurements to personal exposure, which includes both spatial variation and infiltration efficiencies, ranging from 0.28 to 2.21. The correlation between the central-site LG and personal LG was r=0.75. We constructed a model for predicting E<sub>ab</sub> for individual subjects by combining the chemical mass balance and total exposure modeling results with the subjects' time-place-activity information. The estimated E<sub>ab</sub> ranged from 2.0 to 7.1 μg/m<sup>3</sup> (mean=3.5±1.3 μg/m<sup>3</sup>) and correlated with personal levoglucosan measurements with an r of 0.53. Uncertainties in the E<sub>ab</sub> estimates were due in part to the reliance on the ambient contribution fraction for total PM<sub>2.5</sub> as a surrogate for biomass burning related PM mass. We also found significant between-subject variation between episodes and non-episodes in both the E<sub>ab</sub> estimates and subjects' activity patterns. This suggests that the LG measurements at the central sites may not be representative of individual exposure to Ag burning smoke. We recommend collecting either personal exposure or microenvironmental samples in future studies for better characterization of E<sub>ab</sub>.

**KEYWORDS:** biomass burning, smoke impact, personal exposure, random component superposition model, recursive model, spatial variation.

## **2.2 Introduction**

Agriculture (Ag) burning (i.e. burning field stubble or waste rice straw) is a cost-effective way of cleaning and preparing the field for the succeeding growth season (Mazzola et al., 1997). However, smoke from Ag burning may contain various air pollutants, including particulate matter (PM), nitrogen dioxide, carbon monoxide, and a series of semi-volatile and volatile organic compounds (Bouble, 1969; Jenkins et al., 1996; Jacobs et al., 1997). Exposure to these pollutants may cause or exacerbate respiratory disease (Brunekreef and Holgate, 2002; Delfino, 2002). Since agriculture burning typically occurs in rural locations such as Eastern Washington, there is often a limited number of ambient air quality monitors located nearby, so that air pollution excursions may not be detected. Moreover, because the smoke episodes usually last only a few hours, the average 24-h NAAQS for PM<sub>2.5</sub> and PM<sub>10</sub> may not be violated. The combination of these factors results in many limitations in the few studies that have examined the health effects from exposure to agriculture burning smoke (Jacobs et al., 1997; Roberts and Corkill, 1998; Torigoe et al., 2000). For example, the air quality data in these studies were collected at centrally located monitoring sites, which may not represent the community residents' actual exposures (Ebelt et al., 2000; Liu et al., 2003). Previous studies also did not link the health effects specifically to smoke from Ag burning. Furthermore, we know of no existing publications that characterize community residents' exposure to Ag burning smoke.

This paper assesses personal exposures to PM<sub>2.5</sub> from outdoor sources ( $E_{ag}$ ) and from agriculture burning smoke ( $E_{ab}$ ) during the fall Ag burning season in Pullman, WA. In the first paper of this series (Jimenez et al., 2005), we characterized the air quality and Ag burning smoke, and estimated that 35% of the PM<sub>2.5</sub> during the study period was from biomass burning. In this paper, we apply a random component superposition model (Ott et al., 2000), a recursive mass balance model (Allen et al., 2003; Allen et al., 2004), and a total exposure model (Wilson et al., 2000; Williams et al., 2003; Allen et al., 2004; Wu et al., 2004) to estimate the  $E_{ab}$  using the PM<sub>2.5</sub> measurements collected at the central site and inside the study subjects' residences, as well as their time-place-activity information. In the final paper of this series (Sullivan et al., 2005), we examine the associations between health effects and exposure to smoke for 33 asthmatic adults.

## **2.3. Methods**

### **2.3.1 Study design**

This study was conducted in Pullman, WA, from September 3, 2002 to November 1, 2002. The fall burning season was selected because historically it involves more acreage burned and higher short-term PM<sub>2.5</sub> concentrations than those observed in the spring (Jimenez, 2002). Pullman is located in eastern WA (population ~ 25,000), approximately 80 miles south from Spokane, WA and 10 miles west from Moscow, ID. It was selected due to its relatively large potentially exposed population and its proximity to many regional Ag burning activities in Eastern Washington and Northern Idaho. Thirty-three adult subjects with asthma (mean age=27, min=18, max=52) were recruited to participate in this study. These subjects were either students or staff at Washington State University (WSU), and were typically on the WSU campus during the day. A central site was set up on top of the Dana Hall on WSU. Various PM<sub>2.5</sub> measurements were collected at the central site, including continuous PM<sub>2.5</sub> mass concentration using a Tapered Element Oscillating Microbalance (TEOM, Series 1400a, Rupprecht & Patashnick Co., Inc), light scattering as an indirect measure of particulate matter using a nephelometer (Radiance Research, Seattle, WA), and integrated 12-hr (starting at 8 AM and 8 PM) PM<sub>2.5</sub> samples from two collocated Harvard Impactors (HI<sub>2.5</sub>) (Air Diagnostics and Engineering, Inc., Naples, ME). One of the HI<sub>2.5</sub> was equipped with a Teflon filter for gravimetric and XRF analysis, and the other with a pre-fired quartz filter for elemental and organic carbon (EC/OC) analysis (Pang et. al. 2002; NIOSH, 2003). The PM collected on Teflon filters were also extracted and analyzed with GC/MS for levoglucosan (LG), a tracer for biomass burning smoke (Simpson et al., 2004). The detection limit of the LG analysis method is 0.02 µg/ml and the precision is 24%.

Home indoor PM was monitored at 13 subjects' residences during the first 4 weeks and the rest of the subjects during the second 4 weeks, using either the Radiance Research nephelometer or the personal DataRAM (pDR, Thermo-Andersen, Smyrna, GA). Four subjects did not have indoor PM measurements due to the lack of available instruments. The precision of the nephelometer is 3~8%, and for the pDR it is 12% under 10 µg/m<sup>3</sup> and approximately 5% above 10 µg/m<sup>3</sup> (Liu et al., 2002). Each week, two subjects volunteered for personal exposure monitoring for 5 consecutive days (starting

from late afternoon on Monday). Each subject kept a time-activity diary (TAD) with a 10-min resolution. Nominal 24-hr personal PM<sub>2.5</sub> samples were collected using two collocated Harvard Personal Environmental Monitors (HPEM<sub>2.5</sub>, Harvard School of Public Health, Boston, MA), each connected to its own personal pump (BGI AFC 400S, Waltham, MA) with a mass flow controller operated at 4 LPM. The HPEM<sub>2.5</sub> is a single stage inertial impactor with a 50% cut point of  $2.4 \pm 0.1 \mu\text{m}$  (Sioutas et al., 1999). The precision ( $2.2 \mu\text{g}/\text{m}^3$ ) and accuracy ( $0.4 \mu\text{g}/\text{m}^3$ ) of the HPEM<sub>2.5</sub> have been determined in our previous study (Liu et al., 2003). One of the HPEMs contained a Teflon filter for gravimetric analysis and the other contained a quartz filter for EC/OC analysis. The Teflon filters were further analyzed for XRF (N=30) and/or LG (N=48).

A spatial pattern was observed (between the central and B16 sites) during the first half of the study period. Thus additional outdoor HPEM<sub>2.5</sub> measurements were taken at 5 home sites during the second half of the study for examination of the spatial variation of PM<sub>2.5</sub>. These sites were selected to represent typical topographic characteristics in Pullman, which consists of four major hills separated by two major cross roads in the valleys (Figure 1). Each home outdoor sample was collected over approximately 25-hr intervals during two Ag burning “episodes” (Oct 9-11 and 17-19) and over 1 week (144-170 hrs) otherwise. In addition to the filter samples, continuous PM<sub>2.5</sub> was also measured by a Radiance Research nephelometer at one of the subject’s residences in northwest Pullman (Figure 1, B16) from Oct 12 to Nov 2 and at the Washington State Department of Ecology (WDOE) site (Figure 1) over the entire study. Episodes of elevated smoke from Ag burning were declared based on the combination of TEOM measurements at the central site, visual observations, and burn calls from the WDOE and Idaho Department of Environmental Quality (Jimenez et al., 2005). There were four real episodes, among which two were declared during the field campaign and the other two were identified later during the data analysis phase. One “sham” episode was declared during the field campaign (Jimenez et al., 2005) and was used as a control for the health study.

### **2.3.2 Data Analysis**

Data analyses were conducted to characterize spatial variation of PM<sub>2.5</sub> and personal PM exposure. The general linear model (GLM) was used to test the between-subject effects on various personal PM<sub>2.5</sub> measures, including mass concentration, LG,

and EC/OC. When there was no significant between-subject effect, subjects were pooled and the differences in PM exposures during Ag burn episodes and non-episodes were tested using GLM. In addition, individual personal exposures to PM of ambient or Ag burning origins were estimated for each subject.

A spatial regression model was constructed to examine factors affecting the PM<sub>2.5</sub> concentrations at the home sites. The dependent variable was the home outdoor HPEM measurements (5 sites) and the nephelometer measurements at the B16 and the DOE sites with averaging time matching those of the HPEM measurements. The predictors examined included the matched central-site nephelometer measurements ( $\mu\text{g}/\text{m}^3$ ), distance from outdoor sites to the central site (km), elevation (binary variable: below or above the central site), and the temperature at the central site ( $^{\circ}\text{F}$ ).

Personal exposures to ambient and Ag burning PM were estimated via three steps: (1) estimating particle infiltration efficiency ( $F_{\text{inf}}$ ) for each home using the Random Component Superposition (RCS) model (Ott et al., 2000; Williams et al., 2003); (2) using the total exposure model to separate ambient ( $E_{\text{ag}}$ ) and non-ambient exposures (Allen et al., 2004; Wu et al., 2004); and (3) estimating  $E_{\text{ab}}$  by proportioning  $E_{\text{ag}}$  to Ag burning related PM based on the Chemical Mass Balance (CMB) modeling results for each 12-h monitoring period (Jimenez et al., 2005).

*Estimate the particle infiltration efficiency.* The RCS model estimates  $F_{\text{inf}}$  by regressing outdoor PM<sub>2.5</sub> ( $C_o$ ) against indoor PM<sub>2.5</sub> ( $C_i$ ):

$$C_i = b_0 + b_1 * C_o \quad (1)$$

where  $b_1$  represents  $F_{\text{inf}}$  from the immediate home outdoor to indoor environments and  $b_0$  represents indoor sources and the measurement errors. Since home outdoor measurements were not available for most homes in this study, we used the 12-hr HI<sub>2.5</sub> measurements at the central site ( $C_c$ ) as  $C_o$  in Eq (1). The  $C_i$  is the indoor real-time PM<sub>2.5</sub> averaged over the same periods as the corresponding  $C_o$ . The linear regression algorithm was applied to Eq (1) to calculate coefficient  $b_1$ . The obtained  $b_1$  coefficient is a modified  $F_{\text{inf}}$  or particle attenuation efficiency ( $A$ ), which represents a combined effect of  $F_{\text{inf}}$  from home outdoor to home indoor environments and PM<sub>2.5</sub> spatial variations between the home and the central sites. Thus,  $A$  is not bounded by 1. It was calculated for each subject's home from an average of 44 paired 12-hr observations (range: 14-61  $\mu\text{g}/\text{m}^3$ ).

We also modeled the  $F_{inf}$  using a recursive model (RM) as described in detail by Allen et al. (2003) and Wu et al. (2004):

$$(b_{sp})_t^{in} = a_1(b_{sp})_t^{out} + a_2(b_{sp})_{t-1}^{in} + S_t^{in} \quad (2)$$

where  $(b_{sp})_t^{in}$  is the indoor light scattering value at time  $t$  (i.e. hourly indoor  $PM_{2.5}$  at residence measured with either nephelometer or pDR),  $(b_{sp})_t^{out}$  is the outdoor light scattering value at time  $t$  (i.e. hourly outdoor  $PM_{2.5}$  at residence estimated from the GLM spatial model),  $(b_{sp})_{t-1}^{in}$  is the indoor light scattering value at time  $t-1$ , and  $S_t^{in}$  is the contribution from indoor sources. The  $a_1$  and  $a_2$  parameters in Eq (2) were obtained through linear regression with the influence from  $S_t^{in}$  minimized by censoring the indoor data for indoor sources. The  $F_{inf}$  was then calculated as  $a_1/(1-a_2)$ . The RM was performed from the best 10 monitoring days at each residence, based on the following criteria (in order of priority): (1) Nephelometer was used and its nighttime median hourly ratio of indoor to outdoor  $PM_{2.5} < 1$  (N=8 residences); (2) pDR was used and its nighttime median hourly ratio of indoor to outdoor  $PM_{2.5} < 1$  (N=9); (3) Median hourly ratio of indoor to outdoor  $PM_{2.5}$  during the best 10 days  $< 1$  (N=2); and (4) Median nighttime hourly ratio of indoor to central-site  $PM_{2.5}$  during the best 10 days  $< 1$  (n=4). Both the RCS model and RM rely on good correlation between the  $PM_{2.5}$  at the central site and the home sites. Site B32 was excluded in this analysis, as it was located outside the main Pullman area.

*Estimate the personal exposures due to ambient-generated PM.* A total exposure model was used to calculate personal exposures to ambient-generated PM ( $E_{ag}$ ) on a 12-hr basis:

$$E_{ag} = \alpha * C_c = [f_i * A + (1-f_i)] * C_c \quad (3)$$

where  $\alpha$  is the ambient contribution fraction (Allen et al., 2004), and  $f_i$  is the time fraction spent indoors at home. Because during the school day all subjects went to the WSU campus where the central-site was located, we made a strong assumption that  $A$  at school indoors or other indoor microenvironments was 1. Since our main interest is to estimate  $E_{ab}$  and most of the PM excursions from Ag burning occurred during the nighttime when subjects were at home (Jimenez et al., 2005), it is not as critical to obtain an accurate  $A$  for the school and other indoor microenvironments during the day.

Estimate the personal exposures due to agriculture-burning PM ( $E_{ab}$ ). The  $E_{ab}$  at any 12-h sampling period  $t$  was calculated as:

$$E_{ab,t} = (\text{CMB}_{\text{VEG},t}/C_{c,t}) * E_{ag,t} \quad (4)$$

where  $\text{CMB}_{\text{VEG},t}$  is the vegetative burning mass concentration estimated from the CMB model (Jimenez et al., 2005). Eq (4) assumes that the proportion of Ag burning PM in personal exposure to ambient-generated PM is identical to the proportion of Ag burning PM in total outdoor  $\text{PM}_{2.5}$  measured at the central site in any corresponding 12 hour period, regardless of the location of subject residences. Therefore this proportion is fixed for all subjects, but the actual level varies depending on subject specific  $E_{ag}$ . The  $E_{ag}$  estimates were evaluated by comparing them with those estimated from the sulfur tracer method (Allen et al., 2003), while the  $E_{ab}$  was evaluated by comparison with the LG tracer method.

## **2.4 Results and discussion**

### **2.4.1 Data quality control**

A total of 118  $\text{HI}_{2.5}$  samples with Teflon filters were collected at the central site, and 74 and 44  $\text{HPEM}_{2.5}$  samples were collected on the subjects and outside the subjects' home, respectively. Five  $\text{HI}_{2.5}$  samples with Teflon filters at the central site were removed from the dataset due to either filter (e.g. filter mishandled or damaged) or air flow problems (e.g. not within 10% of the designated flow rate). Five personal and 9 home outdoor samples were removed due to similar fatal errors. All filter samples were blank corrected ( $1.5 \pm 4.1 \mu\text{g}$  and  $7.7 \pm 5.2 \mu\text{g}$  for  $\text{HI}_{2.5}$  and  $\text{HPEM}_{2.5}$  samples, respectively). The nephelometer at the central site was calibrated against the collocated  $\text{HI}_{2.5}$  measurements. The pDRs were collocated with and calibrated against the central-site nephelometer over 4 days at the end of the study period. The calibration curve developed for each pDR had an  $r^2$  ranging between 0.986 and 0.992. The histogram of LG revealed that there was one potential outlier of  $0.17 \mu\text{g}/\text{m}^3$  and the TAD confirmed that this subject was exposed to smoke from a woodstove fire for 160 min during that sampling period. Since we were using LG as a tracer for Ag burning related smoke, this sample was removed from the dataset.

## 2.4.2 PM<sub>2.5</sub> spatial analysis

During the second half of the study, the average PM<sub>2.5</sub> concentrations at these monitoring sites (i.e. residential outdoor and DOE) and the central site were  $11.7\pm 6.1$  and  $14.8\pm 4.6$   $\mu\text{g}/\text{m}^3$ , respectively. The average elevation of these monitoring sites was  $757\pm 19$  m (range: 732~790 m) with 3 sites located above the central site. The average distance from the home outdoor to the central site was  $1.2\pm 0.6$  km (range: 0.4~2.1 km). The average temperature was  $42.4\pm 6.7^\circ\text{F}$  (range: 28.8~51.9°F). The high PM<sub>2.5</sub> concentrations generally occurred in the valley and decreased with higher elevations based on data collected during the second half of the study. Site B16, located on the northwest hill facing away from the central Pullman valley, was most likely in a different airshed as it had the lowest correlation with the central site measurements ( $r=0.25$ ) and the lowest monitoring to central sites PM ratio ( $0.34\pm 0.08$ , Figure 1). The ratios of home outdoor to central site (BC on Figure 1) PM<sub>2.5</sub> measurements ranged between 0.27 and 1.81, with the lowest mean ratios observed for the hill site B16 (mean $\pm$ SD= $0.34\pm 0.08$ ), median for B33 ( $0.74\pm 0.09$ ), and the highest for valley sites B02 ( $1.05\pm 0.25$ ) and BJJ ( $1.09\pm 0.38$ ) (Figure 1). The nighttime drainage flow (or mountain wind) that brought down upper-layer PM remaining from the daytime Ag burning to the ground level and possible accumulation of residential wood burning account for the observed vertical distributions in concentrations. Based on the 2000 census data, there were 30 (0.3%) housing units in Pullman using wood as the heating source. Four sites (i.e. B02, B03, B28, and BJJ) had a relatively larger variation (SD>0.2) in the ratio over the monitoring period while the other three sites had small variation (SD<0.2), suggesting a possible temporal-spatial interaction among these sites.

Results from the stepwise regression analysis for spatial variation showed that the significant predicting factors, in the order of importance, are the central site PM measurements, home elevation, and outdoor ambient temperature ( $R^2=0.63$  without B16 and 0.49 with B16) (Table 1). As none of the other home sites were located near B16 (Figure 1), B16 was thus excluded in the final spatial model. The distance from the home to the central site was the least and insignificant contributor to the total  $R^2$  (partial  $R^2<0.01$ ,  $p=0.81$ ). This is partially due to the high correlation ( $r=0.88$ ) between elevation and distance. The PM<sub>2.5</sub> at sites above the central site was on average  $4.7$   $\mu\text{g}/\text{m}^3$  lower

than those below the central sites. The  $PM_{2.5}$  at the outdoor sites was positively associated with the central-site  $PM_{2.5}$  and temperature. Vukovich and Sherwell (2002) also found positive correlation between  $PM_{2.5}$  and temperature within a season at an urban site (Washington, DC) and a remote site (Shenandoah National Park, VA) based on Principal Component Analysis. It was argued that temperature influences secondary particle chemistry, producing greater rates of reaction at higher temperature.

### **2.4.3 Characterization of personal exposure**

Subjects spent an average of 19% of their time at school or work and 61% of their time at home indoors (Table 2). We examined the Hawthorn effect (Franke and Kaul, 1978), i.e., how subjects might change their activities simply due to the participation in the study or the declaration of episodes. We examined the overall time spent in each microenvironment during declared episode and non-episode days and found significant differences in time spent at the home indoors, in transit, and indoors in other microenvironments using a GLM (Table 2). During declared episodes (i.e. real plus sham episodes) subjects spent less time indoors at home and more time in transit or indoors away from home than during non-declared episode periods. The differences were sustained even when data were limited to weekdays only. We also examined whether subjects have different activities during *real* episode vs. non-episode days (regardless of whether the episodes were declared or not) and obtained similar results as to those analyzed by declaration status (Table 2). We attributed these differences to the fact that subjects had to come to our lab for respiratory health tests during episodes, thus increased the time spent in transit or indoors away from home. The time spent outdoors, however, was not different between the episode and non-episode periods. In addition, results from the GLM on activity patterns with subject ID and the status of real episode as the fixed effects show that there is a significant between-subject variation in the fraction of time spend indoors at home, indoors away from home, and in transportation. This between-subject variation might bias the health effect assessment if the analysis was performed based on binary coding of episode status and on using the central-site measurements to represent individual exposures. Applying personal exposure measurements or activity-specific exposure estimates can avoid this problem since they take into account the subjects' activity patterns.

The mean personal exposure to PM<sub>2.5</sub> among the 16 monitored subjects was 13.8±11.1 µg/m<sup>3</sup> (N=68, median=11.4 µg/m<sup>3</sup>). Results from GLM showed that there was no significant difference in PM exposure among subjects (p=0.12). Thus we pooled all subjects to test for episode effects. The personal exposure was on average 7.9 µg/m<sup>3</sup> higher during the episodes (N=23, mean=19.1±12.1 µg/m<sup>3</sup>) than non-episodes (N=45, mean=11.1±9.7 µg/m<sup>3</sup>). The mean personal exposure to EC was 0.4±0.5 µg/m<sup>3</sup> and 8.5±2.7 µg/m<sup>3</sup> to OC (N=64). Personal OC exposures were higher during episodes than during other times (10.1±2.9 vs. 7.8±2.2 µg/m<sup>3</sup>). However, there was also a significant subject effect (p<0.01) on OC exposure. Due to the small sample size, we could not separate episode effect from the subject effect.

LG in PM samples is regarded as a unique tracer for biomass combustion (Simpson et al., 2004). It has been used previously in source apportionment analyses to quantify biomass smoke (Zheng et al., 2002; Cass, Larson et al., 2004; Jiminez et al., 2004). In the current study, the mean personal LG exposure was 0.018±0.024 µg/m<sup>3</sup> (N=47, median=0.012 µg/m<sup>3</sup>) after correcting for the analytical recovery (mean=82±8%). This level is lower than the central-site LG (mean=0.068±0.082 µg/m<sup>3</sup>) and the LG measured at fixed sites in southeastern US (0.166 µg/m<sup>3</sup>, Zheng et al., 2004) and Seattle, WA (0.204 µg/m<sup>3</sup>, Larson et al., 2004). Personal LG exposures were higher during episodes than during other times (0.024±0.029 vs. 0.013±0.018 µg/m<sup>3</sup>). However, similar to the personal OC exposures, we could not separate the episode effect from the subject effect on the LG measurements (p<0.01).

The cross-sectional Pearson correlation coefficient between central-site and personal PM<sub>2.5</sub> was 0.29, identical to the 0.29 reported in Seattle, WA (Liu et al., 2003) and higher than the 0.15 in Vancouver, BC (Ebelt et al., 2000).. From the scatter plot, 5 personal samples (> 22 µg/m<sup>3</sup>, denoted as open circles in Figure 2) were identified as heavily influenced by indoor sources or personal activities. Four of these five samples occurred when the subjects spent more than 2 hours at social events during the corresponding sampling period, while the 5<sup>th</sup> sample occurred when the subject spent 5 hours at home with unknown activities. After removing these 5 observations, the cross-sectional correlation increased to 0.65 (Figure 2), similar to the 0.67 in Baltimore, MD

(Sarnat et al., 2000) Previous studies showed that the median longitudinal correlation usually was higher than the cross-sectional correlation because the interpersonal variability was adjusted for in the former correlation (Ebelt et al., 2000; Sarnat et al., 2000; Wallace, 2000; Liu et al., 2003). In this study, the longitudinal Pearson correlation coefficient between central site and personal PM<sub>2.5</sub> for individual subjects (N=11) who had more than 3 daily observations ranged between -0.67 and 0.98, with a median of 0.52. The negative longitudinal correlations are likely due to the limited number of samples per subject (N ranged from 3 to 5) (Lumley and Liu, 2004).

#### 2.4.4 Personal exposure modeling

The particle attenuation efficiency ( $A$ ) that represents a combined effect of the outdoor spatial variation and infiltration estimated from the RCS model for each home is shown in Table 3.  $A$  ranged from -0.12 to 2.21; the negative  $A$  had one of the poorest model fits ( $r^2=0.04$ ). Eleven of the 28 residences had low  $r^2$  (<0.2), suggesting that central site PM<sub>2.5</sub> was a poor predictor for home outdoor PM<sub>2.5</sub> in the RCS model (Eq 1) for some subjects. Thus, residences with  $R^2$  lower than 0.2 were removed from the subsequent analyses. Table 3 also shows the  $F_{inf}$  estimated from the RM (Eq 2). Five sites had no qualified 10-day periods, resulting in a unity  $F_{inf}$  estimate. Considering the fact that  $A$  included the spatial variation effect while  $F_{inf}$  did not, they correlated reasonably well ( $r=0.27$ ,  $N=12$ ) excluding those residences of  $F_{inf}=1$ . Removing the residence that was 2 km (B21, not shown in Figure 1) away from the central site improved the correlation to 0.35 ( $N=11$ ). The  $A$  estimates were chosen to be used in the subsequent analyses because of the limitations associated with the RM. Namely, this model relied on hourly home outdoor concentrations which were estimated using the spatial model built on 24-hr to 1-week data (Table 1). errors in the outdoor PM<sub>2.5</sub> estimates could affect the censoring algorithm and the RM itself. Furthermore, there was no indicator for the goodness of model fit for the RM. On the other hand, the RCS model comes with an  $R^2$  value that can readily be used to judge the goodness of the model fit.

The estimated 12-h exposures to ambient-generated PM ( $E_{ag}$ ) and Ag-burning PM ( $E_{ab}$ ) are shown in Table 4. Due to incomplete TAD information, two subjects (i.e. B06 and B15) had relatively small sample size. The mean  $E_{ag}$  ranged from 6.1 to 21.6  $\mu\text{g}/\text{m}^3$  and the mean  $E_{ab}$  ranged from 2.0 to 7.1  $\mu\text{g}/\text{m}^3$ . The  $E_{ab}$  was on average 33% of the  $E_{ag}$

(SD=3%, range=26%~36%). The GLM results show that after controlling for the significant subject effects ( $p < 0.01$ ), the  $E_{ab}$  was on average  $1.4 \mu\text{g}/\text{m}^3$  higher during the episodes than non-episodes. The fact that the subject effect on  $E_{ab}$  was significant ( $p < 0.01$ ) suggests that one should be cautious when assigning the same central-site monitoring data as the exposure index for all subjects in the health data analysis.

Conventionally, the ratio of personal sulfur to central-site sulfur is used as the “true” ambient contribution fraction of  $\text{PM}_{2.5}$  ( $\alpha_{\text{sulfur}}$ ) from outdoor to personal environment, assuming that sulfur is a tracer of outdoor  $\text{PM}_{2.5}$  (Sarnat et al., 2002; Allen et al., 2003; Ebelt et al., 2004). We evaluated our estimated ambient contribution fraction ( $\alpha_{\text{model}}$ ) against the true  $\alpha_{\text{sulfur}}$ , thus indirectly verifying  $A$ . The  $\alpha_{\text{model}}$  is the sum of the fraction of time spent outdoors ( $1-f_i$ ) and the fraction of time spent indoors ( $f_i$ )\* $A$ , as shown in Eq (3). Due to the observed spatial variation of outdoor  $\text{PM}_{2.5}$ , the ambient contribution fractions based on the central-site measurements are not bounded by 1. For example, subjects residing at locations with  $\text{PM}_{2.5}$  concentration higher than that at central site may experience higher  $\text{PM}_{2.5}$  exposures than the central-site measurements, resulting in a contribution fraction greater than 1. The correlation between  $\alpha_{\text{model}}$  and  $\alpha_{\text{sulfur}}$  ( $r=0.86$ , Figure 3) was high, even with the strong assumption about  $A=1$  for the school and indoor microenvironments other than home. We attributed this good agreement to the close proximity of most subjects to the central-site monitors and the well ventilated campus microenvironments where the subjects spent on average 19% time during the day.

The  $E_{ab}$  estimates calculated from Eq (4) were evaluated against the personal LG measurements ( $r=0.53$ ,  $p < 0.01$ , Figure 4a). For the same samples, the correlation between the central site and personal LG was 0.75 (Figure 4b), indicating that personal LG exposure predominantly originates from common outdoor sources. Thus, 24-h central site LG may be representative of 24-h average personal exposure to LG in this study population. The lower correlation between the measured personal LG and the modeled  $E_{ab}$  is expected as we assumed a uniform Ag burning contribution across all subjects regardless of the location of their residences. In reality, residents in the valley may receive higher Ag burning exposure than those residing on hill. The uncertainties associated with the CBM estimates of biomass smoke (Eq 4) (Jimenez et al. 2005) also

partially account for this lower correlation. Nevertheless, as it was cost prohibitive to measure daily personal exposure to LG on every subject, the estimated  $E_{ab}$  serves as an informative estimate of exposure to agricultural burning derived PM. The mean ratio of personal to central site LG (or  $\alpha_{LG}$ ) was  $0.46 \pm 0.79$  (median=0.27). Its correlation with  $\alpha_{sulfur}$  was 0.79, and decreased to 0.23 after removing the influential point of  $\alpha_{LG}=3.1$  (Figure 5). Both regression slopes were also significantly different from 1. The low correlation and absence of a 1:1 relationship indicate that total  $PM_{2.5}$  from all sources (using sulfur as the tracer) and  $PM_{2.5}$  from only biomass burning (using LG as the tracer) may have different spatial distribution, as well as different chemical and physical characteristics. Long & Sarnat (Aerosol Science and Technology, 38(S2):91–104, 2004) analyzed infiltration efficiencies for various elemental constituents in  $PM_{2.5}$  and reported various infiltration behaviors among the elements. Thus, the  $A$  estimated using the total  $PM_{2.5}$  measurements may not represent the true  $A$  for the  $PM_{2.5}$  constituents originated from biomass burning smoke. To improve the  $E_{ab}$  estimates, future studies should consider obtaining a better estimate of the infiltration efficiency for biomass related  $PM_{2.5}$  by collecting LG samples at the subjects' home and at the indoor and outdoor microenvironments. The National Research Council also suggests future studies should focus on characterizing contribution fraction for specific PM components (NRC, 2004).

## **2.5 Conclusions**

We found that during the Ag burning season in Pullman, outdoor  $PM_{2.5}$  exhibited significant spatial variation, with the highest  $PM_{2.5}$  concentrations occurring in valley areas. The best predictors for such a spatial variation included elevation and outdoor temperature. The mean personal  $PM_{2.5}$  exposure was  $13.8 \mu\text{g}/\text{m}^3$  and was higher during the episodes than non-episodes. The personal LG and OC exposure also was higher during episodes than during non-episodes, although there also existed a significant between-subject variation. The cross-sectional correlation between the central-site and personal  $PM_{2.5}$  concentrations was higher than those reported previously excluding outliers, probably due to the close proximity of subject work places and residences to the central site. However, since it was found that subjects had different activity patterns

during episodes, one should be cautious when applying central-site measurements as estimates of individual exposures in a cohort health study. Using the RCS and total exposure models, we estimated that the personal exposure to PM<sub>2.5</sub> from agriculture burning smoke ranged from 2.0 to 7.1 µg/m<sup>3</sup>. These E<sub>ab</sub> estimates correlated with the personal LG measurements (r =0.53), although a higher correlation (r =0.75) between the personal and central-site LG was found. The slightly lower correlation between E<sub>ab</sub> and personal LG was likely due to the differences in infiltration between total PM<sub>2.5</sub> and vegetative burning related PM, as well as the uncertainties in the CMB modeling results. On the other hand, the observed between-subject variation discussed above may bias the health analysis when using the central-site measurements as the exposure index. Therefore, it is advisable to collect either more personal samples to measure the E<sub>ab</sub> or more microenvironmental samples to model the E<sub>ab</sub>.

## 2.6 References

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## **2.7 Figure Captions**

Figure 1. The locations of the subjects' residences. The red ID labels indicate sites with outdoor PM<sub>2.5</sub> measurements. The mean ratio and the standard deviation of the outdoor to central-site PM<sub>2.5</sub> were in parenthesis.

Figure 2. Correlation between personal and central-site PM<sub>2.5</sub> measurements. The blank circle represents the five potential outliers.

Figure 3. Comparison between the ambient contribution fraction ( $\alpha$ ) estimated from the sulfur tracer and total exposure model. Each point represents one subject-day observation (from 4 subjects).

Figure 4. (a) Comparison between  $E_{ab}$  and Personal LG. (b) Comparison between central-site and Personal LG.

Figure 5. Comparison between the ambient contribution fraction ( $\alpha$ ) estimated from the sulfur tracer and LG. Each point represents one subject-day observation (from X subjects).

Table 1. The spatial modeling results. The dependent variable is the HPEM samples at home outdoor locations and nephelometer data at the DOE site (N=37).

Parameter	GLM				Stepwise regression			
	Estimate	SE	t Value	Pr >  t	Partial R-Square	Model R-Square	F value	Pr>F
Intercept	-6.46	4.72	-1.37	0.18				
Neph PM <sub>2.5</sub> (μg/m <sup>3</sup> )	0.89	0.14	6.59	<.001	0.43	0.43	26.04	<.001
Elevation								
above central site	-4.74	1.26	-3.78	<.001	0.16	0.59	13.02	<.001
below central site	0.00	.	.	.				
Temperature at the central site (°F)	0.19	0.10	2.00	0.05	0.04	0.63	4.00	0.05

Table 2. Percent of time spent in each microenvironment.

microenvironment	All Mean(SD) N=1419	Declared as Episode		Real Episode	
		No Mean(SD) N=1147	Yes Mean(SD) N=272	No Mean(SD) N=1098	Yes Mean(SD) N=321
Indoors at home	61.1(20.6)	62.4(19.8)**	55.3(22.7)	62.3(20.2)**	56.9(21.2)
Outdoors near home	0.5(2.3)	0.5(2.3)	0.5(2.0)	0.5(2.4)	0.5(1.9)
Transportation	6.5(5.6)	6.2(5.6)**	8.0(5.4)	6.3(5.7)**	7.3(5.2)
Work	18.9(15.8)	18.8(15.6)	19.4(16.7)	18.7(15.6)	19.9(16.2)
Outdoors away home	2.5(6.5)	2.3(6.2)	3.1(7.8)	2.4(6.2)	3.0(7.4)
Indoors away home	10.5(17.7)	9.7(17.0)**	13.6(20.3)	9.9(17.2)*	12.4(19.2)

\*\* significantly different between episodes and non-episodes,  $p < 0.01$

\* significantly different between episodes and non-episodes,  $p < 0.05$

Table 3. The particle attenuation efficiency ( $A$ ) estimated from the Random Component Superposition model and particle infiltration efficiency ( $F_{inf}$ ) estimated from the recursive model (sorted by RCS model's  $r^2$  values).

ID	RCS model			RM
	$A$	$r^2$	N	$F_{inf}$
B01	1.16	0.60	61	0.73
B21	1.01	0.58	54	0.63
B02	1.18	0.57	54	0.79
B14	0.75	0.55	20	1*
B10	0.75	0.51	41	0.9
B19	0.74	0.40	46	0.7
B09	2.21	0.34	29	1*
B20	0.46	0.31	54	0.55
B15	1.27	0.29	54	1*
B03	0.90	0.28	50	1*
B24	0.58	0.28	56	0.94
B30	0.53	0.28	56	0.78
B06	0.50	0.27	46	0.49
B13	0.28	0.26	54	0.71
B16	0.48	0.25	54	0.58
B29	0.53	0.21	48	0.57
B27	1.77	0.20	30	1*
B22	0.32	0.14	53	0.56
B17	0.40	0.13	16	0.45
B18	0.48	0.12	33	0.41
B33	0.71	0.10	44	0.54
B26	0.21	0.09	50	0.61
B23	0.15	0.07	26	0.25
B07	0.39	0.07	51	0.69
B04	0.41	0.06	54	0.66
B28	-0.12	0.04	14	0.49
B08	0.11	0.02	41	0.62
B31	0.11	0.01	54	0.51

\* No qualified 10-day periods in the recursive model.

Table 4. Summary of the estimated personal exposures to PM<sub>2.5</sub> from outdoor sources (E<sub>ag</sub>) and from agriculture burning smoke (E<sub>ab</sub>)

ID	N	E <sub>ag</sub> (µg/m <sup>3</sup> )			E <sub>ab</sub> (µg/m <sup>3</sup> )		
		Mean	Median	SD	Mean	Median	SD
B01	75	13.0	11.4	6.5	4.3	3.9	2.9
B02	82	13.1	11.8	6.6	4.4	3.9	2.9
B03	82	11.0	10.1	5.0	3.6	3.1	2.3
B06	18	8.0	7.9	3.6	2.1	2.2	1.1
B09	81	21.6	18.1	13.5	7.1	5.6	5.8
B10	84	9.0	8.3	4.4	2.9	2.5	2.0
B13	93	7.7	7.5	4.3	2.5	2.2	1.7
B14	60	9.0	8.9	4.2	2.7	2.7	1.5
B15	18	11.7	10.1	6.9	3.3	2.9	2.3
B16	93	7.4	7.3	3.7	2.4	2.3	1.5
B19	70	9.7	8.8	4.4	3.5	3.2	2.1
B20	69	6.1	5.6	2.9	2.0	1.9	1.2
B21	74	11.6	10.8	5.5	4.1	3.8	2.6
B24	91	8.1	7.8	3.8	2.7	2.5	1.6
B27	83	17.0	16.8	7.7	5.8	5.7	3.3
B29	66	8.2	7.4	4.1	2.9	2.5	1.8
B30	46	8.6	8.8	3.5	3.1	3.1	1.5

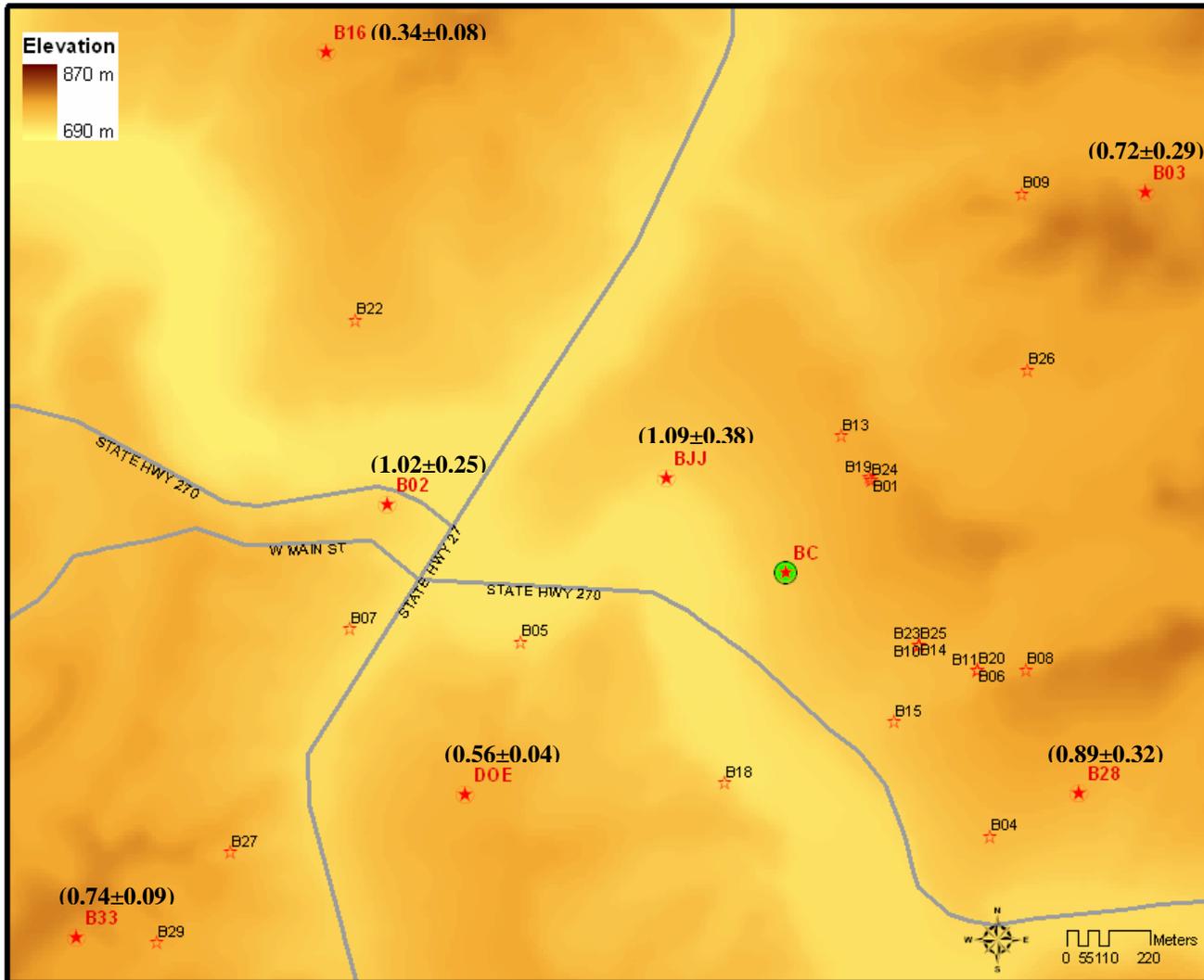


Figure 1.

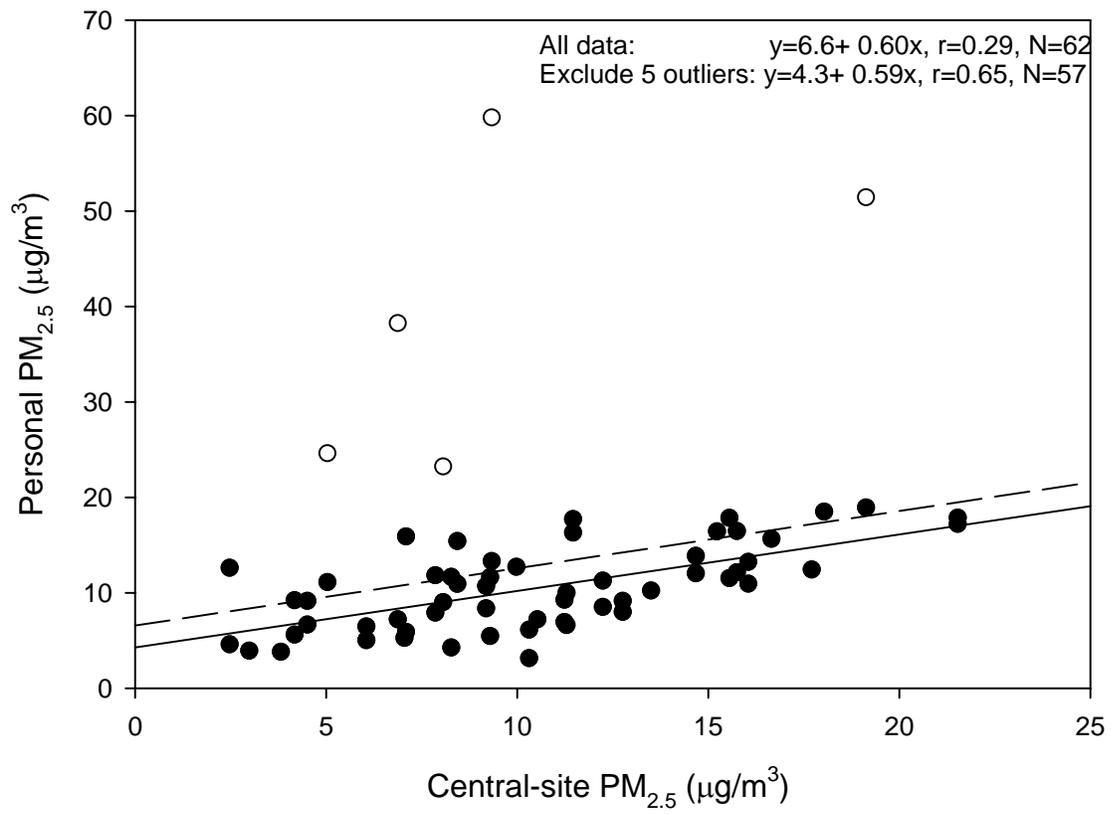


Figure 2.

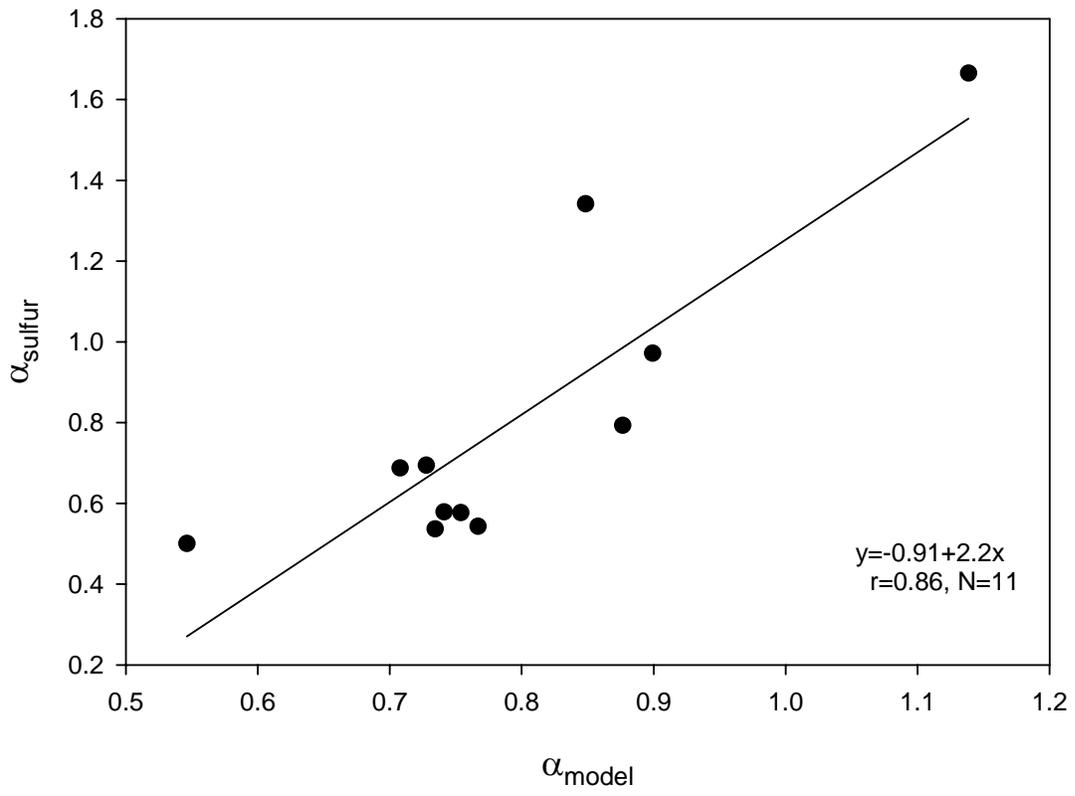


Figure 3.

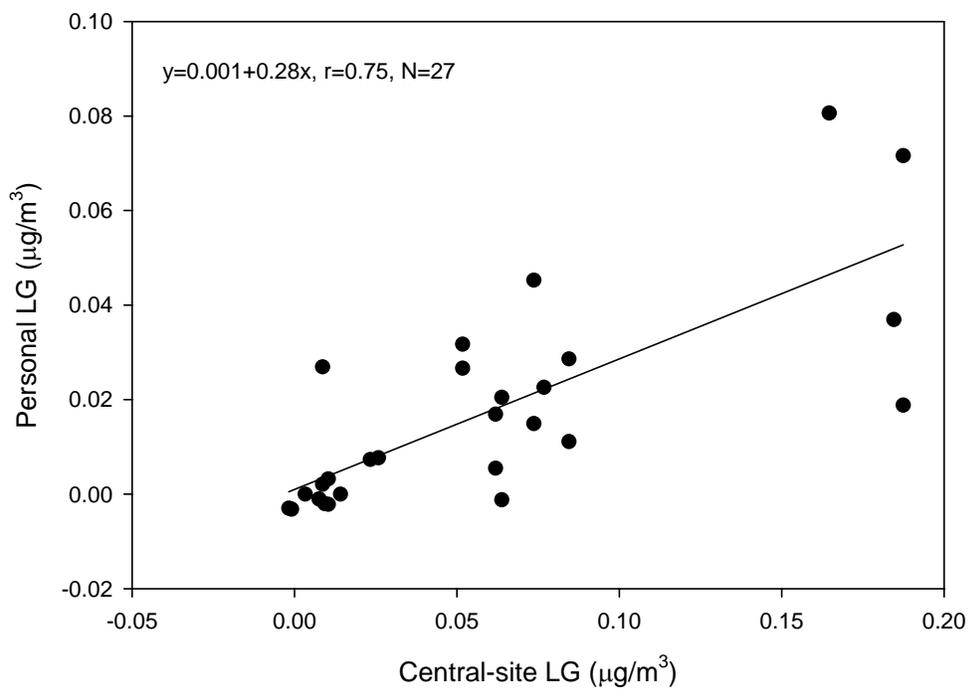
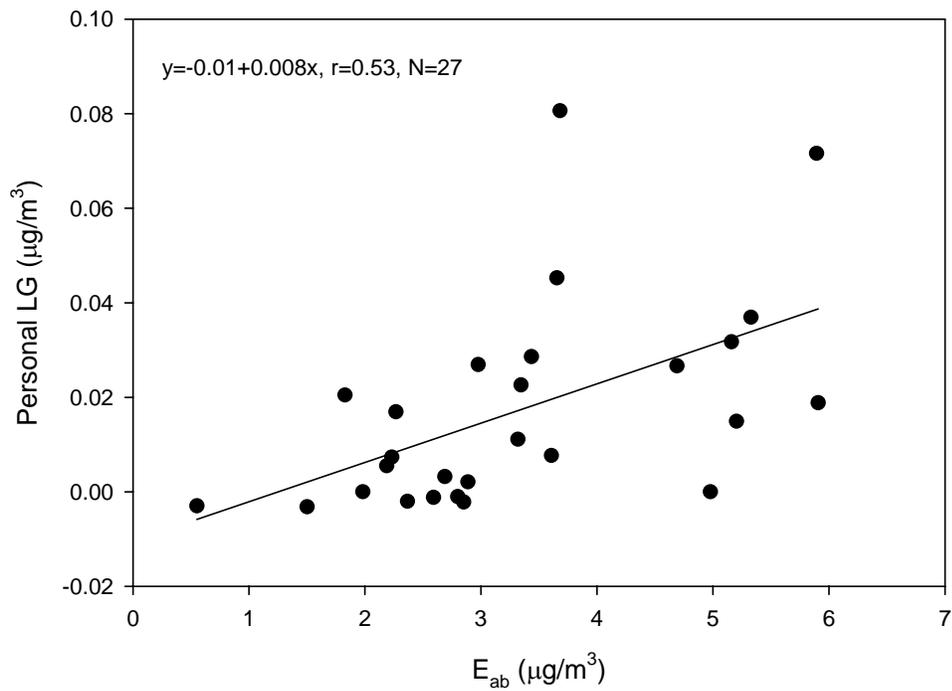


Figure 4.

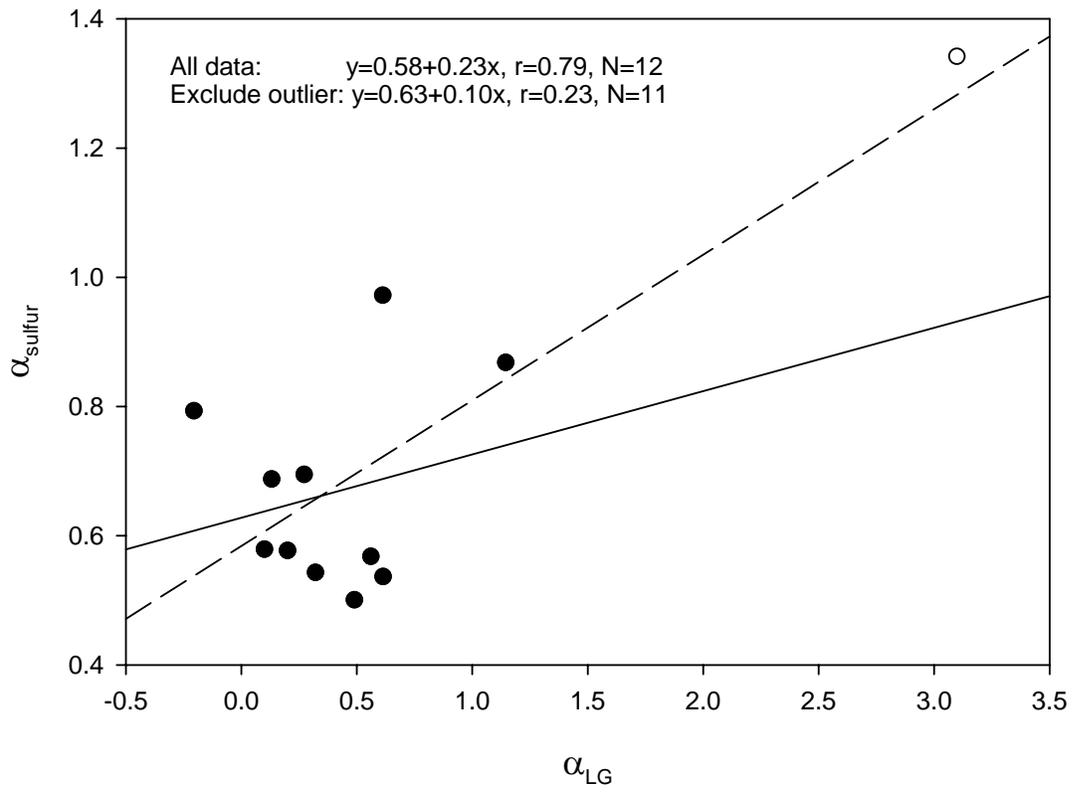


Figure 5.

## Chapter 3. Health Assessment

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### **3.1 Abstract**

To determine whether wheat field burning has adverse pulmonary effects in adults with mild to moderate asthma, we performed repeated measures of on-line exhaled nitric oxide (eNO)(594 measures) and coached spirometry (591 measures) on 32 individuals with asthma during field burning season (September-October 2002) in Pullman, WA. These pulmonary measures were assessed against measurements of fine particulate air pollution (PM<sub>2.5</sub>) at a central location and to agricultural (Ag) burning related exposure estimates for each individual. We hypothesized that participants who were not using anti-inflammatory medication would show a positive association of exhaled nitric oxide (eNO) and negative association of forced expiratory volume in one second (FEV<sub>1</sub>) and maximal mid-expiratory flow (MMEF) with the peak 1-h average of PM<sub>2.5</sub> during the previous 24 hours. Pulmonary effects of PM<sub>2.5</sub> were assessed with a generalized estimating equation model that included fixed covariates for gender, age, BMI, time of day, an interaction term between medication use and exposure and adjusted for temperature and relative humidity. The 32 participants ranged in age from 18 –52 years (median 24y), and 66% were female. 11 individuals were prescribed asthma controller medications and 3 individuals had baseline FEV<sub>1</sub> < 80% of predicted. The observed 1-h average PM<sub>2.5</sub> concentrations ranged between 0.3 and 59.6 µg/m<sup>3</sup>, averaging 13.0±9.2 µg/m<sup>3</sup> during the study period. There was no significant effect of peak 1-h PM<sub>2.5</sub> on measures of eNO among those not prescribed anti-inflammatory medications: -0.35 ppb (95% CI: -1.70, 1.01) or those prescribed controller medications: 1.68 ppb (95% CI: -1.51, 4.87) per 10 µg/m<sup>3</sup> increase of PM<sub>2.5</sub>. Similar null effects of peak PM<sub>2.5</sub> exposure were noted for spirometric measures of MMEF and FEV<sub>1</sub>. Sensitivity analyses with refined Ag burning specific exposure measures did not change these null results. In conclusion, at the observed range of PM<sub>2.5</sub> concentrations, we did not find an association between peak PM<sub>2.5</sub> episodes from field burning and decrements in pulmonary function or increases in on-line eNO measures in adults with mild to moderate asthma.

### **3.2 Introduction**

Despite increasing recognition of the deleterious effects of ambient generated PM and co-pollutants from industrial, motor vehicle and indoor cooking sources on human health, surprisingly little is known regards the health effects of agricultural (Ag) field burning on individuals with chronic respiratory or cardiac conditions living in rural communities.

Field burning represents a considerable potential air pollution exposure in rural areas. Wheat growers in eastern Washington and northern Idaho burn wheat stubble that remains in the fields after the wheat is harvested. In fall 2000 and spring 2001, a total of 177,346 acres of cereal grain were burned in eastern Washington (DOE, 2001). This field burning results in the production of various air pollutants with known health effects, including particulate matter (PM), nitrogen dioxide (NO<sub>2</sub>), CO, polycyclic aromatic hydrocarbons (PAH), and a series of semi-volatile and volatile organic compounds (Bouble et al. 1969, Kakareka et al. 2003; Ryu et al. 2004; Chun et al. 2004). Kim et al. (2003) reported a 44% contribution of PM<sub>2.5</sub> from biomass burning in Spokane, Washington. Oppenheimer and coworkers (2004) measured significant elevations in NO<sub>2</sub> levels [40 ppb] during the burning of cereal crop residue in Brazil.

Prior studies have examined either asthma hospitalizations or symptom reports as the primary health effect of agricultural field burning smoke on neighboring communities. Jacobs and coworkers (1997) found links between acreage of rice stubble burned and asthma hospitalizations in Butte County, CA. However, peak burning acreage was not correlated with O<sub>3</sub>, CO, and PM<sub>10</sub>. Torigoe and coworkers (2000) reported a correlation between 24-hour averaged PM<sub>10</sub> and the number of children admitted to ER or hospital with asthma exacerbation, with the increase in PM<sub>10</sub> most likely due to the influence of emission from rice straw burning. Although these administrative database studies suggest potential for adverse health effects, they may underestimate the total burden of effect as they only capture individuals who experienced significant decrements in asthma control. A survey of 428 individuals with COPD living in Manitoba during a period of elevated PM from wheat field burning found that 42% of the respondents reported worsening of respiratory symptoms including cough, wheezing, chest tightness, and shortness of breath.

Multiple epidemiological studies have found associations between particulate matter air pollution and increased clinic visits, hospitalizations and sub-clinical decrements in lung function among individuals with asthma. More recently, Delfino et al. (2004) underscored the importance of longer-term health effects by showing stronger inverse associations of FEV<sub>1</sub> with increasing PM<sub>2.5</sub> exposure using multi-day moving averages of PM for both personal and stationary-site PM during the 24 hr before the FEV<sub>1</sub> maneuver. Koenig et al. (2003) has found positive associations between exhaled NO levels in asthmatic children not on controller medications and increases in resident level PM<sub>2.5</sub>, that may contain a high portion of wood burning related PM.

In light of these findings, we hypothesized that adults with mild-moderate asthma who were not using anti-inflammatory medication (inhaled corticosteroids, leukotriene inhibitors) would show a significant positive association of eNO with the peak 1-hour avg. of PM<sub>2.5</sub> during the previous 24 hours from the time of measurement. We also postulated that these same individuals would demonstrate a significant negative association of maximal mid-expiratory flow (MMEF) and FEV<sub>1</sub> with the peak 1-hour average of PM<sub>2.5</sub> during the previous 24 hours from the time of measurement. We further refined exposure by measuring central site levels of levoglucosan (LG) to reflect agricultural sources. In addition, we estimated individual exposure to agricultural burning smoke by using resident-specific filtration estimates and examined the association between agricultural field burns and health effects.

### **3.3 Methods**

#### **3.3.1 Study Design**

We conducted this longitudinal study in Pullman from September 3 to November 1, 2002. Pullman, located in eastern Washington (population ~ 25,000), 80 miles from Spokane, WA and 10 miles from Moscow, ID, was selected due to its relatively large population and its proximity to many regional Ag burning activities in Eastern Washington and North Idaho. This study consisted of 32 young adults with asthma and 2 randomly assigned monitoring sessions for each subject, including an active session and an on-call session. The active session required 16 participants to perform in-lab measures of on-line eNO (Sievers, Boulder CO), coached

spirometry (microDL) and complete symptom questionnaires at the same time of day every Monday, Wednesday and Friday during a 30-day period. The on-call session occurred during the remaining 30-days of the 60-day monitoring session for these 16 individuals. During a declared episode (as described below), the on-call subjects would be paged in to have all health measures performed with the active subjects over the 3 successive days from the initial called episode. During our study, there were four real episodes and a sham episode.

Episodes were declared based on the combination of TEOM measurements at the central site, visual observations, and burn calls from the Washington State Department of Ecology (DOE) and Idaho Department of Environmental Quality (Jimenez et al., 2004). There were four real episodes, among which two were declared during the field campaign and the other two were identified later during the data analysis phase. One “sham” episode was declared during the field campaign and was used as a control (Jimenez et al., 2005).

### **3.3.2 Study Population**

Individuals were considered eligible for participation if they had a clinical diagnosis of mild-intermittent to moderate asthma as defined by National Academy of Early Childhood Programs (NAECP) 1994 guidelines. All participants were non-smokers, living in non-smoking residences. Individuals were excluded if they had an  $FEV_1 < 70\%$  of predicted on baseline spirometry or were chronically prescribed prednisone for asthma control. They were also excluded if they contracted an upper respiratory tract infection or had been prescribed prednisone or increased their controller medication use within the 30 days prior to study. Individuals were recruited by the posting of fliers at sites in Pullman and at residential halls and a student health clinic at the University of Washington State University. The study protocol was approved by the University of Washington and Washington State University Human Subjects Review Boards.

### **3.3.3 Health outcomes**

The primary health outcome described in this article is eNO, which potentially represents the degree of airway inflammation in individuals with asthma. Collection of exhaled NO preceded spirometry. The gas for assessment of the fractional exhaled NO concentration was collected into

a Sievers model 280i on-line collection system (Boulder, CO) at a constant flow rate of 50 ml/s without a breath hold. We followed manufacturer's recommendation to scrub inhalational air. Methods adhered to ATS standards.<sup>94</sup> The analyzer has sensitivity from 0.4-500,000 ppb of NO, and accuracy  $\pm 0.2$  ppb. Ambient NO levels were recorded prior to each measurement. The analyzer was calibrated daily using a certified concentration of NO in nitrogen. For quality control, eNO measurements with technical problems (n=4 events or 0.6% of all measures) were excluded from analysis. Health models were performed on all remaining eNO measurements; however, a sensitivity analysis was also performed excluding subjects with baseline eNO greater than 100 ppb.

Secondary outcomes include FEV<sub>1</sub>, and MMEF, representing the degree of airway obstruction, which is a key phenotype of asthma. Analyses focus on the percentage of predicted normal FEV<sub>1</sub> and MMEF (Hankinson et al. 1999) for a given height, age, sex, and race/ethnicity. This standardizes measurements between subjects and gives clinically meaningful overall estimates of association for the study population. The participants performed 3-coached spirometric maneuvers using a portable MicroDL spirometer (MicroDL, Florida), without nose clips. Subjects were coached by 2 trained staff members who adhered to strict study protocol. Measures were considered reproducible if the 3-maneuvers' values were within 5%, up to a maximum of 6 maneuvers. All maneuvers were performed at the same time of day for each participant. Participants were to refrain from albuterol use within 4-hours of the maneuver. Before the study, calibration of spirometers was performed with a 3-L syringe. Spirometry measurements with technical problems or operation errors were excluded from analysis in case of the following criteria.

### **3.3.4 Symptom Questionnaires**

Symptom questionnaires were completed at each lab visit during the active session and at each declared episode. The questionnaire was modified from the symptom questionnaire used in the Children's Asthma Control Program Study (Slaughter et al. 2003). It included questions on symptoms, rescue medication use, clinic visits and work/ school absence. Asthma episodes were summarized as a continuous response, rescue medications were categorized as 0=no use, 1=1-3 times over 24-hours and 2=4 or more inhalations per 24-hours.

### 3.3.5 Exposure Assessment

Detailed atmospheric characterization and exposure assessment methodologies have been described elsewhere (Jimenez et al. 2005; Wu et al. 2005). Briefly, a central site was set up at the rooftop of the WSU Engineering building to collect measurements of PM<sub>2.5</sub> from a Tapered Element Oscillating Microbalance (TEOM, Series 1400a, Rupprecht & Patashnick Co., Inc) and a nephelometer (Radiance Research, Seattle, WA), and integrated 12-hr (starting at 8 AM and 8 PM) PM<sub>2.5</sub> samples from two pairs of collocated Harvard Impactors (HI<sub>2.5</sub>) (Air Diagnostics and Engineering, Inc., Naples, ME). These HI<sub>2.5</sub> samples were used for gravimetric analysis, trace element analysis via X-ray fluorescence, LG analysis via GC-MS as the biomass burning marker, and EC/OC analysis via thermal optical transmittance (Jimenez et al. 2005). The Nephelometer measurements were calibrated against the HI<sub>2.5</sub> measurements and were only used for health effect assessment when the TEOM<sub>2.5</sub> measurements were not available due to instrument malfunction or less than 0.

Home indoor PM was monitored at most, except for 4 subjects' residences using either the Radiance nephelometer or the personal DataRAM (pDR, Thermo-Andersen, Smyrna, GA) (Liu et al., 2002). Daily personal monitoring was performed on 2 subjects per week for 5 consecutive days using two collocated Harvard Personal Environmental Monitor (HPEM<sub>2.5</sub>, Harvard School of Public Health, Boston, MA). The HPEM<sub>2.5</sub> samplers were subject to the same series of analyses as for the Central site HI<sub>2.5</sub> samples. Each subject kept a 10-min time-activity diary.

Individual exposure were estimated based on 1) the central site 1-hr peak PM<sub>2.5</sub> measurements as the primary crude non-specific exposure index; 2) declared episodes as a crude index for Ag-burning specific exposure; 3) the central site LG measurements as a more targeted exposure indices related to biomass burning exposure; 4) home indoor nighttime PM measurements as a more relevant exposure indices as episodes occurred mostly during the nighttime when subjects were asleep at home (Wu et al. 2005); and 5) ambient originated Ag-burning exposures estimated for each subject as the most refined exposure index (described briefly below and by Wu et al.. 2005). Using the central site and home indoor PM measurements, we applied the random component superposition model to estimate the attenuation of central-site outdoor PM to

home outdoor and indoor environments (Wu et al. 2005). A total exposure model and a chemical mass balance model were used to calculate personal exposures to ambient-generated PM and exposure to biomass burning related PM.

### **3.3.6 Statistical Methods**

The data was analyzed using SAS software (version 9.2, Carey, NC). Prior to initiating the study, we performed power simulations to assess sample size based findings in our previous health assessment studies (Koenig et al. 1993). Our simulations were based on an estimated FEV<sub>1</sub> effect of -30 ml per 10 µg/m<sup>3</sup> increase in PM<sub>2.5</sub>, an unexplained variation in FEV<sub>1</sub> of 0.18, 10% probability of episodes, 14 monitoring days on 24 subjects. The simulation results indicated a greater than 90% power for detection of a 3 ml decline in FEV<sub>1</sub> (Slaughter et al. 2005)

Primary Analyses. We modeled the association between the peak 1-hour central-site PM<sub>2.5</sub> concentrations (Exposure Index # 1) measured over the 24-hours prior to measures of exhaled NO, MMEF and FEV<sub>1</sub> using Generalized Estimating Equation (GEE) models that controlled for sex, age, BMI, time of day, and included linear and quadratic terms to control for temperature and relative humidity. Our alternative models also included an interaction term between medication use and PM exposure to distinguish effects between subjects on and not on medication. These same models were repeated to measure the association of 1-day lagged peaked 1-hour PM<sub>2.5</sub>, i.e., measured 24-48 hours prior to measures of exhaled NO, MMEF and FEV<sub>1</sub>. To focus our exposure estimates to Ag burning related PM, we performed models that assessed health effects vs. true episodes (Exposure Index #2) whether declared or not. These models included the 2 declared real episodes that included 195 eNO and spirometric measures on 32 participants and 2 undeclared (missed) real episode with 30 health measures on the 16 active participants.

Secondary Analyses. We performed several sensitivity analyses to further refine our exposure metric. These included health effect models that assessed effects of eNO, FEV<sub>1</sub> and MMEF from LG (Exposure Index #3), the impact of nocturnal inversions of field burning PM (Exposure Index #4), and individually-based estimates on ambient originated exposure from Ag burning smoke (Exposure Index #5).

## **3.4 Results**

### **3.4.1 Subject Characteristics**

The 32 subjects ranged in age from 18 –52 years (median 24y), with 66% female. Eleven individuals were prescribed asthma controller medications and 3 individuals had baseline FEV<sub>1</sub> < 80% of predicted (Table 1). The median baseline eNO level was 42 ppb (range 10-229 ppb). There are no significant differences in the baseline eNO and spirometry measures between subjects with or without medication use. After adjusting for missing or unacceptable health or exposure data, we collected 594 laboratory measures of on-line eNO and 591 coached spirometry measures from these subjects (Table 1).

Results from our asthma symptom questionnaire indicated that 16 of the 32 participants had at least one mild or severe asthma symptom on days with corresponding in-lab measurements. These 16 individuals responded that they had increased symptoms on 87 (14.6%) subject-days (Table 2). However, only one respondent described a severe episode of wheezing during the 60-day study period.. No respondents contacted their physician for asthma symptoms. Regarding the use of rescue medicine, 87% of the responses described no rescue inhaler use during the study, while only 2.2% responses (from 4 participants) described using a rescue inhaler greater than 3 times a day during the study (Table 2).

### **3.4.2 Exposure Results**

The average ( $\pm$ SD) TEOM PM<sub>10</sub> and PM<sub>2.5</sub> levels observed during the study were 45.0 $\pm$ 42.3 and 13.0 $\pm$ 9.4  $\mu$ g/m<sup>3</sup>, respectively. The mean value of the nighttime samples observed higher PM<sub>2.5</sub> and LG concentrations (15.8 and 0.10  $\mu$ g/m<sup>3</sup>) as compared with the daytime samples (10.13 and 0.05  $\mu$ g/m<sup>3</sup>) due to nighttime inversions. Our source apportionment analysis (Jimenez et al. 2005) indicated four major sources accounting for more than 95% of the PM<sub>2.5</sub> mass, including soil (4.5  $\mu$ g/m<sup>3</sup> or 38%) and vegetative burning (4.0  $\mu$ g/m<sup>3</sup> or 35%) as the two largest contributors and sulfate aerosol (2.2  $\mu$ g/m<sup>3</sup> or 20%) and vehicular traffic (0.2  $\mu$ g/m<sup>3</sup> or 2%).

The mean personal exposures to PM<sub>2.5</sub> was 13.8 $\pm$ 11.1  $\mu$ g/m<sup>3</sup>, which was on average 7.9  $\mu$ g/m<sup>3</sup> higher during the episodes (19.1 $\pm$ 12.1  $\mu$ g/m<sup>3</sup>) than non-episodes (11.1 $\pm$ 9.7  $\mu$ g/m<sup>3</sup>) (Wu et al.

2005). The personal LG also was higher during the episodes than non-episodes. The estimated exposure to ambient originated Ag burning related PM<sub>2.5</sub> ( $E_{ab}$ ) ranged between 2.0 and 7.1  $\mu\text{g}/\text{m}^3$  (mean=3.5±1.3  $\mu\text{g}/\text{m}^3$ ) and correlated with personal LG measurements with an r of 0.53. The correlation between the central-site LG and personal LG was r=0.75. A significant inter-subject variation between episodes and non-episodes was found for both the  $E_{ab}$  estimates and subjects' activity patterns.

### 3.4.3 Primary Hypotheses Testing

Results of our primary hypotheses testing are presented in Table 3 and Figure 1. We found no significant effect resulted from a peak 1-hour increase in central-site PM during the preceding 24-hours on eNO (0.36 ppb; 95% CI: -1.19, 1.90 ppb), MMEF (0.41liters/min; 95% CI: -0.86, 1.68), or FEV<sub>1</sub> (3.96 ml; 95% CI: -4.93, 12.85). Moreover, there was no evidence for effect in the subset of individuals not on asthma controller medications on measures of eNO (0.35 ppb; 95% CI: -1.70, 1.01 ppb), MMEF (0.41liters/min; 95% CI: -1.18, 1.99), or FEV<sub>1</sub> (5.16 ml; 95% CI: -6.80, 17.11). Similar null results were found at 1-day lagged peak 1-hour PM<sub>2.5</sub> exposures (Figure 2). Models that adjusted for ambient NO did not change the null association between eNO levels and peak 1-hour PM<sub>2.5</sub> measurements at the same day or 1-day lagged measure (data not shown).

We analyzed the respiratory health effects between baseline and agricultural field burn episodes (real vs. sham) for the study participants (Figure 3). These models find no association between the real field burn episodes (declared or not, shown as filled black dots and error bars) and estimated changes in measures of eNO (2 ppb; 95% CI: -1.5, 5.5), FEV<sub>1</sub> (18.4 ml; 95% CI: -16.4, 53.2), or MMEF (1.9 liters/min; 95% CI: -2.5, 6.2).

### 3.4.4 Sensitivity Analyses

Our exposure analyses showed that the cross-sectional Pearson correlation coefficient between central-site and personal PM<sub>2.5</sub> was only 0.29. On the other hand, the correlation between the central site and personal LG was 0.75, indicating that personal LG exposure was predominantly originated from outdoor sources and that central-site LG was more representative of personal

exposure to Ag burning smoke than central-site  $PM_{2.5}$ . Thus, the exposure indices used in our primary hypotheses testing were crude proxies of exposure at best. Central site 12-h average LG measurements were thus tested in the health effect models. MMEF and eNO showed similar null results as observed in the primary tests, although  $FEV_1$  seemed to increase to an average of 28.8 ml (95% CI: -0.1,57.6) for all subjects and 40 ml (95% CI:0.11,79.9) for the non-medicated group in response to a  $0.2 \mu\text{g}/\text{m}^3$  increase in LG (an approximate equivalent of  $10 \mu\text{g}/\text{m}^3$  of  $PM_{2.5}$ ) (Table 4 and Figure 4) .

Because Ag burning episodes occurred often at night when subjects were likely asleep at home and not close to the central site (Wu et al. 2005), exposure to nocturnal  $PM_{2.5}$  peaks due to nocturnal inversions after field burns might not be captured by the central  $PM_{2.5}$  measurements. We performed similar GEE models that reflected respiratory effects of 12-hour average indoor PM both 24 hours and 48 hours prior to the measures of eNO, MMEF and  $FEV_1$  . Similar null results for were observed for all three health measures, regardless of the use of medication (Figure 5). Null results were also noted in 1-day lagged analyses (data not shown).

Our exposure analyses demonstrated a substantial spatial variation of  $PM_{2.5}$  in the Pullman area due to the complex topography (Wu et al. 2005). Thus in place of the central-site peak concentrations in the GEE models, we used the estimates of exposure from ambient originated particles ( $E_{ag}$ ). The  $E_{ag}$  was estimated based on subjects' time-activity information and the particle attenuation estimates from the random component superposition model that take into account both spatial variation and infiltration efficiency (Wu et al. 2005). These effect models using the  $E_{ag}$  estimates demonstrated no effect of the same day  $E_{ag}$  on eNO: 4.69 ppb (95% CI: -5.41, 14.79 ppb), MMEF: -1.08 l/min (95% CI: -4.29, 2.13), or  $FEV_1$ : -6.26 ml (95% CI: -29.6, 17.04) (Figure 6, black filled circles and bars). Similar null associations were obtained in models that stratified by medication use (Figure 6, blue filled circles and bars) and that used a 1-day lagged exposure (data not shown).

The final sensitivity analyses were performed for estimated exposures to ambient originated  $PM_{2.5}$  from Ag burning sources ( $E_{ab}$ ) based on the previous  $E_{ag}$  estimates and the estimated ratio of Ag burning related mass from our source apportionment results (Jimenez et al. 2005). We

performed GEE analyses of the association between these modeled  $E_{ab}$  on individual measures of eNO, MMEF and FEV<sub>1</sub>. Again, similar null associations were observed between  $E_{ab}$  and eNO, MMEF, and FEV<sub>1</sub> (Figure 7) regardless of the use of medication. The broader confidence intervals reflected the multiple models and assumptions made to generate  $E_{ab}$ .

The primary analysis was repeated with and without inclusion of subjects with baseline eNO greater than 100 ppb (from 6 subjects, 3 in each medication category). When subjects with high baseline eNO were excluded from the analysis a significant association was found between eNO and same day peak 1-hr max: -0.95 ppb (95% CI: -1.84, -0.06), among subjects not taking controller medications.

### **3.5 Discussion**

To our knowledge this is the first community level study that examined the potential for sub-clinical effects of agricultural field burning related PM exposure on individuals with asthma. Our study did not find an association between peak PM<sub>2.5</sub> concentrations from Ag burning and decrements in pulmonary function or increases in pulmonary inflammation measured by eNO in the 32 individuals with mild to moderate asthma. The null results from the primary hypotheses testing with central-site based exposure estimates were further validated with the multiple sensitivity analyses using refined exposure estimates. Although the null results may be true, the following factors could have contributed to the absence of effect in our study: the selection of a relatively non-susceptible adult population, non-linear effects of agricultural burning related PM on eNO and spirometric measures, timing of the health assessments, uncertainties in estimating the spatial and infiltration efficiencies of PM, and the imprecision in estimating the agricultural combustion contributions to the PM mass.

Our study population consisted of young adults with asthma. They may have been less sensitive to the effects of biomass burning than children or elderly individuals with pre-existing pulmonary or cardiovascular disease. Prior studies that find association between agricultural or wood smoke related PM and asthma exacerbations reported effects primarily in children with asthma (Koenig et al. 1993; Jacobs et al. 1997; Sheppard et al. 1999; Torigoe et al. 2000; Golshan et al. 2002). Moreover, our restriction to a study population that had stable asthma

without routine prednisone or recent increase in controller medication use may also have diminished our ability to find effect. However, the small population of young children and elderly individuals with pre-existing disease made it logistically difficult for recruiting and monitoring in Pullman. Adults with asthma were chosen because of the potential of using a centrally located monitoring station to capture Ag burning smoke exposure of a population residing at or near the campus, ability of these adults to comply with the scheduled lab visits, flexibility of these adults' schedule to allow for measures on episode days, and ability to perform reproducible measures of pulmonary function.

The absence of Ag burn effects on pulmonary measures could be a result of a non-linear relation between the pulmonary effects end-points and PM exposure. However, our results are similar to those of controlled human exposure studies for concentrated ambient particles, ultrafine particulate, and diesel exhaust that found no effects on spirometric or eNO measures even at much higher levels of PM (Gong et al. 2003; Ghio & Huang 2004; Pietropaoli et al. 2004; Gong et al. 2005). At present, there are no controlled exposure studies that have assessed dose-response relationship between biomass burning smoke and measures of eNO or spirometry in humans.

The timing of our eNO measure could have been too far away from the peak Ag burn exposure to capture short-lived changes in eNO due to increased PM. Most peaks occurred during the night. The nocturnal inversions may have generated transient increases in eNO during the night that returned toward baseline levels at time of measure later in the following day. Although we modeled for effect of nocturnal changes in PM levels on eNO measures, we had both insufficient air measurements to accurately quantify the nighttime spatial variation of outdoor PM<sub>2.5</sub> or Ag burning related PM<sub>2.5</sub> and inadequate lab measures of eNO occurring within 1-4 hours of the PM peaks to assess for an acute response. Thus, wider confidence intervals were observed for the GEE models that used modeled exposures (Table 4).

The exposure assessment was complicated by the increased percentage of PM from wind blown dust during the dry Ag burning season. Our source apportionment analysis indicated that the largest PM<sub>2.5</sub> mass contribution came from soil, even during the declared Ag burning episodes.

It is possible that the dust related PM biased a true association between Ag burning and pulmonary effects towards the null. Furthermore, it was difficult to accurately assess Ag burning related PM<sub>2.5</sub> due to the lack of a validated source profile for wheat straw burning. LG levels reflect both Ag burning and wood burning; therefore, our study could not detect potential differences in biologic effect between these two common sources of vegetative burning in Pullman.

Our study had several strengths that add to the validity of the results. These included repeated in-lab measures of sub-clinical effect (eNO and spirometry); inclusion of a sham Ag burn episode to control for non-agricultural PM related changes in pulmonary measures and symptoms measures; and detailed exposure measures that included residential, indoor and personal measures in a community where agricultural burning represented a relatively high fraction of total PM<sub>2.5</sub>. Although the frequency and peak levels of Ag burn related PM<sub>2.5</sub> were low, they were representative of the recent Ag burning related PM (Jimenez et al. 2005). Since the 1999 Memorandum of Understanding between the wheat growers and the WA Department of Ecology, and the subsequent implementation of the Ag burning control strategies, acreage burned and Ag related PM emission have been decreasing annually. The low and infrequent exposures observed in our study were comparable to the exposures in the preceding two years.

### **3.6 Conclusions**

Although we did not find an association between peak PM<sub>2.5</sub> from field burning and decrements in pulmonary function or increases in eNO in young adults with asthma, we cannot rule-out health effects from field burning in more susceptible populations or at higher PM concentrations. We recommend future studies that measure sub-clinical effects on children with asthma, older individuals with cardiac disease, or farm workers exposed to potentially greater agricultural PM concentrations.

### 3.7 References

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Table 1. Demographics and Baseline Values of eNO and Spirometry in the 32 study Participants.

	Overall	Medication Use	
		No	Yes
<b>Subject-Specific Characteristics</b>			
Number of subjects	32	21	11
Number of Females (%)	21 (66)	14 (67)	7 (64)
Age in Years	23.5 (18, 52)	23 (18, 47)	25 (18, 52)
Body Mass index in kg/m <sup>2</sup>	24 (18, 55)	24 (18, 55)	26 (19, 44)
<b>Baseline Measures</b>			
eNO	42 (10, 229)	39 (16, 229)	52 (10, 196)
FEV <sub>1</sub> , liters	3.3 (2.1, 5.7)	3.3 (2.6, 5.3)	3.5 (2.1, 5.7)
MEF, liters/min	196 (118, 346)	198 (118, 322)	194 (122, 346)
<b>Baseline % Predicted Values</b>			
FEV <sub>1</sub> % predicted	96 (72, 119)	94 (72, 117)	101 (75, 119)
MEF % predicted	80 (47, 138)	82 (47, 138)	75 (58, 122)
<b>Subject-Days Characteristics</b>			
Number of subject-days, overall	595	389	206
Number of subject-days with eNO	594	388	206
Number of subject-days with LF	591	388	203

\*Unless otherwise specified, values listed are median (range)

Table 2. Frequency of Asthma Symptom Reporting and Medication Use in the 32 Study Participants for days summarized only for days with corresponding lab measurements.

Symptom	Level	Overall	Medication Use	
			No	Yes
Asthma episode code	missing	41 (6.9%)	25 (6.4%)	16 (7.8%)
	no episodes	467 (78.5%)	326 (83.8%)	141 (68.4%)
	1-3 mild episodes	69 (11.6%)	31 (8%)	38 (18.4%)
	4 or more mild episodes	15 (2.5%)	7 (1.8%)	8 (3.9%)
	1 or more severe episode	3 (0.5%)	. (%)	3 (1.5%)
Contacted Dr. for asthma	missing	41 (6.9%)	25 (6.4%)	16 (7.8%)
	0	554 (93.1%)	364 (93.6%)	190 (92.2%)
Missed class/work because of asthma	missing	41 (6.9%)	25 (6.4%)	16 (7.8%)
	0	554 (93.1%)	364 (93.6%)	190 (92.2%)
Rescue inhaler (puffs/day)	missing	41 (6.9%)	25 (6.4%)	16 (7.8%)
	no use	518 (87.1%)	350 (90%)	168 (81.6%)
	1-3 puffs/day	23 (3.9%)	12 (3.1%)	11 (5.3%)
	4 or more puffs/day	13 (2.2%)	2 (0.5%)	11 (5.3%)

Rescue Inhaler use categorized as 0=no use; 1=1-3 puffs/ day; 2>3/puffs/day

Table 3. Effects of Peak 1-hour PM<sub>2.5</sub>\* within the 24-hours Preceding Measure on mean eNO levels, and Spirometric Measures in 32 Individuals with Asthma.

Exposure Time	Pulmonary Measure	Medication Use Subgroup	Change in Measure	Confidence Interval
Same day	eNO	All participants	0.36 ppb	(-1.19, 1.90)
	eNO	On Meds	1.68 ppb	(-1.51, 4.87)
	eNO	No Meds	-0.35 ppb	(-1.70, 1.01)
	FEV <sub>1</sub>	All Participants	3.96 ml	(-4.93,12.85)
	FEV <sub>1</sub>	On Meds	1.59 ml	(-8.90,12.09)
	FEV <sub>1</sub>	No Meds	5.16 ml	(-6.80,17.11)
	MMEF	All Participants	0.41 liters/ min	(-0.86, 1.68)
	MMEF	On Meds	0.42 liters	(-1.22, 2.06)
	MMEF	No Meds	0.41 liters	(-1.18, 1.99)
1-day lagged	eNO	All Participants	-0.12 ppb	(-1.56, 1.32)
	eNO	On Meds	1.76 ppb	(-1.25, 4.77)
	eNO	No Meds	-1.17 ppb	(-2.60, 0.25)
	FEV <sub>1</sub>	All Participants	1.63 ml	(-13.3,16.57)
	FEV <sub>1</sub>	On Meds	-5.10 ml	(-23.8,13.57)
	FEV <sub>1</sub>	No Meds	5.42 ml	(-12.7,23.56)
	MMEF	All Participants	-0.29 liters/ min	(-2.03, 1.45)
	MMEF	On Meds	-2.09 liters/ min	(-4.66, 0.48)
	MMEF	No Meds	0.73 liters/min	(-0.91, 2.38)

\*The peak 1-hr. PM value changes are reported for a 10- $\mu\text{g}/\text{m}^3$  increase in the central-site PM<sub>2.5</sub>.

Table 4. The Effects of Refined Exposure Metric for Ag Burn PM on the Change in mean value of eNO and Spirometric Measures in the 32 individuals with Asthma.

Exposure Metric	Change in Exposure	Pulmonary Measure	Medication Use Subgroup	Change in Measure	Confidence Interval
Central-site levoglucosan	0.2 $\mu\text{g}/\text{m}^3$	eNO	All Participants	-0.75 ppb	(-3.62, 2.12)
		eNO	On Meds	-3.41 ppb	(-9.52, 2.70)
		eNO	No Meds	0.71 ppb	(-4.89, 6.31)
		FEV <sub>1</sub>	All Participants	28.76 ml	(-0.10,57.63)
		FEV <sub>1</sub>	On Meds	8.15 ml	(-17.9,34.17)
		FEV <sub>1</sub>	No Meds	40.00 ml	(-0.11,79.89)
		MMEF	All Participants	1.82 liters/min	(-2.18, 5.82)
		MMEF	On Meds	2.45 liters/ min	(-3.68, 8.59)
		MMEF	No Meds	1.48 liters/ min	(-3.54, 6.51)
Indoor exposure to PM <sub>2.5</sub>	10- $\mu\text{g}/\text{m}^3$	eNO	All Participants	-0.40 ppb	(-1.11, 0.32)
		eNO	On Meds	-1.17 ppb	(-3.06, 0.73)
		eNO	No Meds	-0.16 ppb	(-0.97, 0.64)
		FEV <sub>1</sub>	All Participants	1.61 ml	(-3.78, 7.01)
		FEV <sub>1</sub>	On Meds	3.47 ml	(-12.7,19.69)
		FEV <sub>1</sub>	No Meds	1.08 ml	(-4.28, 6.44)
		MMEF	All Participants	0.03 liters/ min	(-1.06, 1.12)
		MMEF	On Meds	2.06 liters/ min	(-0.70, 4.82)
		MMEF	No Meds	-0.58 liters/ min	(-1.45, 0.29)
Exposure to PM <sub>2.5</sub> of biomass burning origin (E <sub>ab</sub> )	10- $\mu\text{g}/\text{m}^3$	eNO	All Participants	2.56 ppb	(-10.2,15.26)
		eNO	On Meds	-3.67 ppb	(-14.9, 7.53)
		eNO	No Meds	4.86 ppb	(-14.1,23.85)
		FEV <sub>1</sub>	All Participants	-40.4 ml	( -128,46.72)
		FEV <sub>1</sub>	On Meds	2.23 ml	(-67.8,72.22)
		FEV <sub>1</sub>	No Meds	-56.0 ml	(-170,57.82)
		MMEF	All Participants	-5.11 liters/ min	(-17.8, 7.57)
		MMEF	On Meds	-5.78 liters/ min	(-18.2, 6.62)
		MMEF	No Meds	-4.85 liters/ min	(-21.0,11.30)

Health responses to peak TEOM2.5 at central site, lag 0

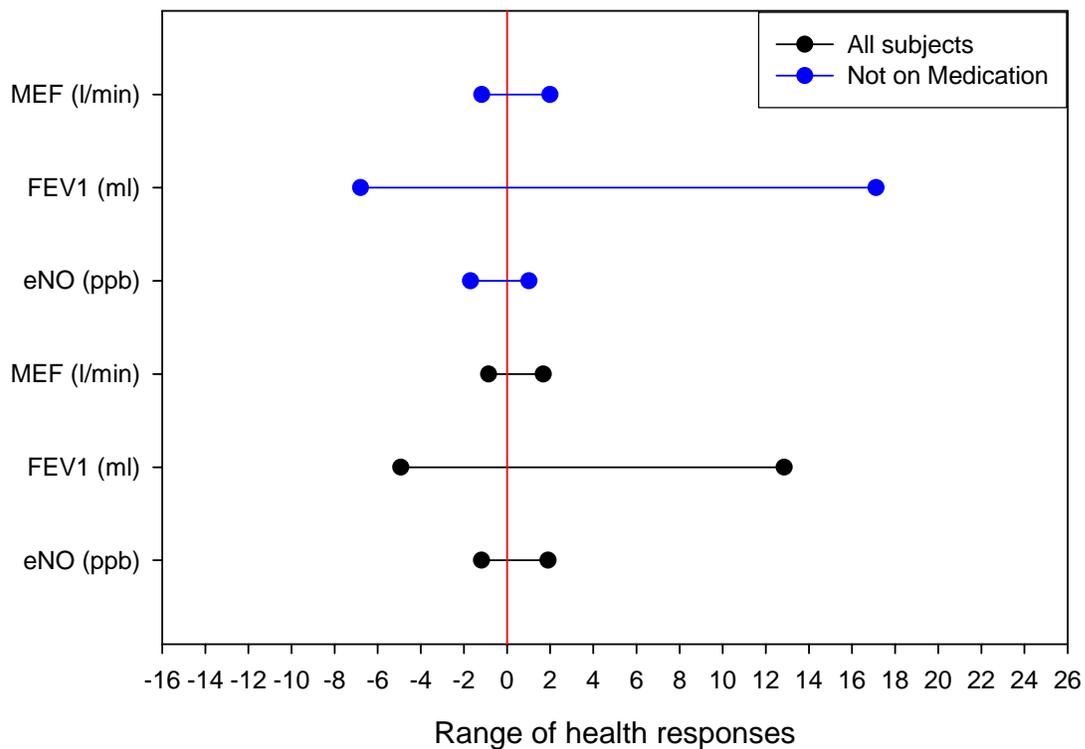


Figure 1: Health Effects of Peak 1-hour Agricultural PM within the 24-hours Preceding Health Measure in 32 Individuals with Asthma.

### Health responses to peak TEOM2.5 at central site, lag 1

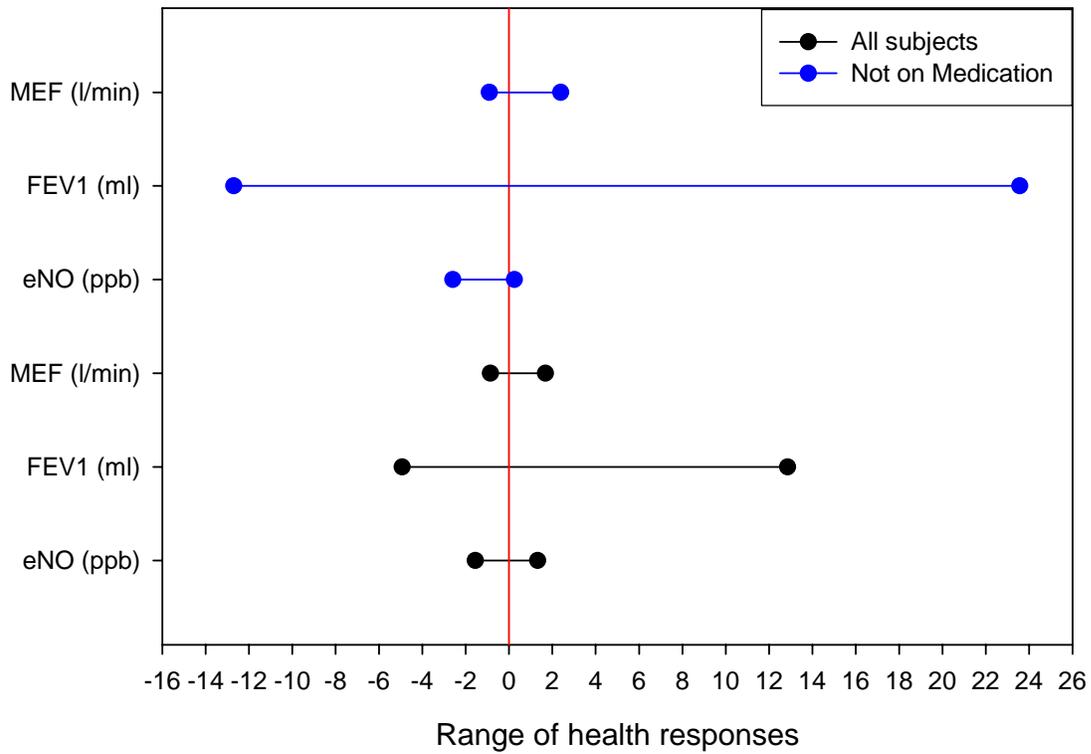


Figure 2: Health Effects of Peak 1-hour Agricultural PM within the 24-48 hours Preceding Health Measure in 32 Individuals with Asthma.

### Health responses during episodes, non-episodes, and sham episode

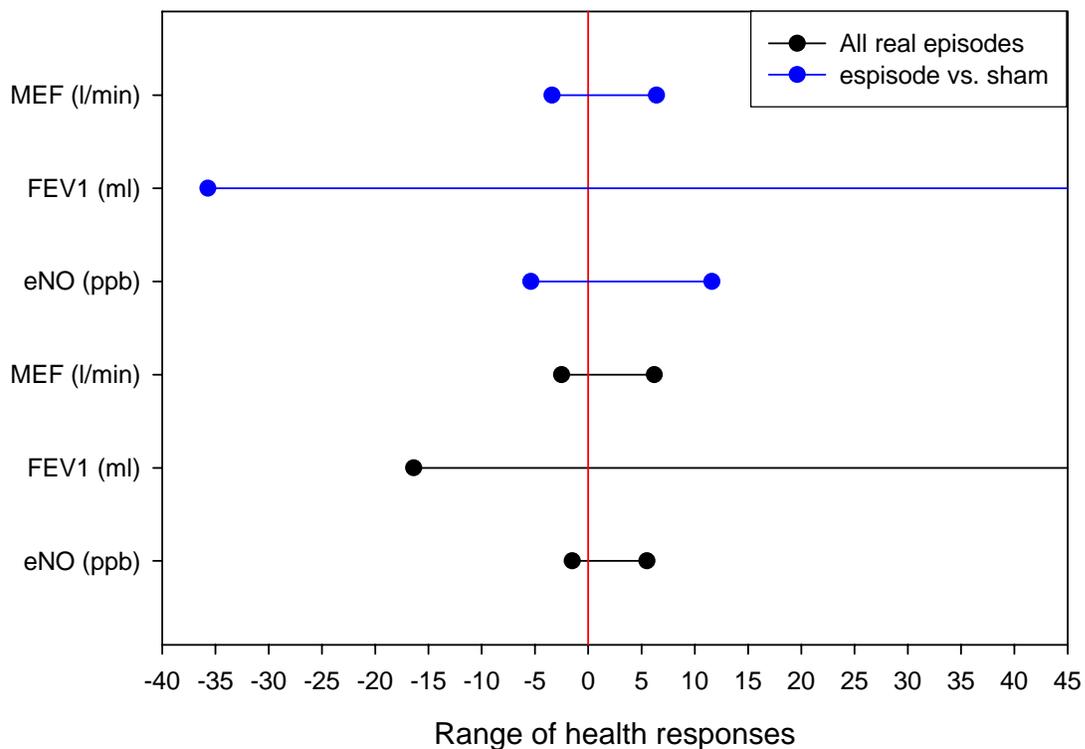


Figure 3: Health Effects of agricultural PM Episodes compared with non-episode measures in 32 individuals with Asthma.

Health responses to exposure to central site levoglucosan ( $\mu\text{g}/\text{m}^3$ ), per  $0.2 \mu\text{g}/\text{m}^3$  increase in LG level

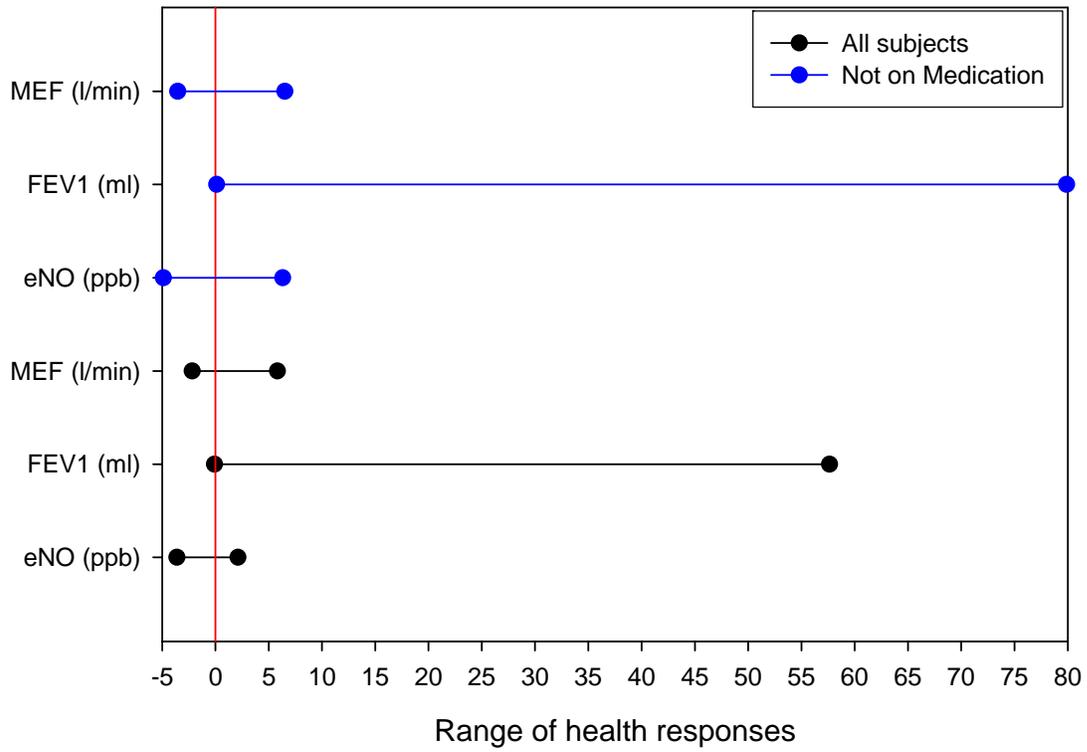


Figure 4. Health responses to exposure to  $0.2 \mu\text{g}/\text{m}^3$  increase in central-site levoglucosan, a biomass burning marker.

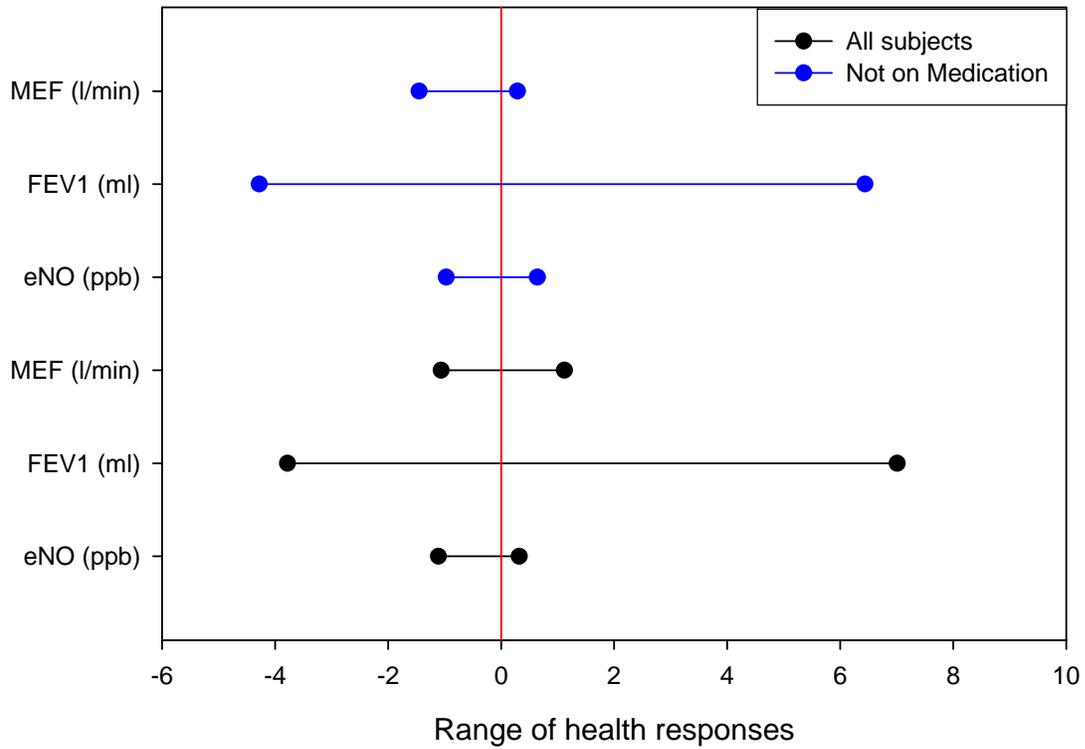


Figure 5. Health responses to 12-h average home indoor PM measurements that represent exposure to peak Ag burn PM due to nocturnal inversions.

### Health responses to exposure to ambient generated PM<sub>2.5</sub> (E<sub>ag</sub>)

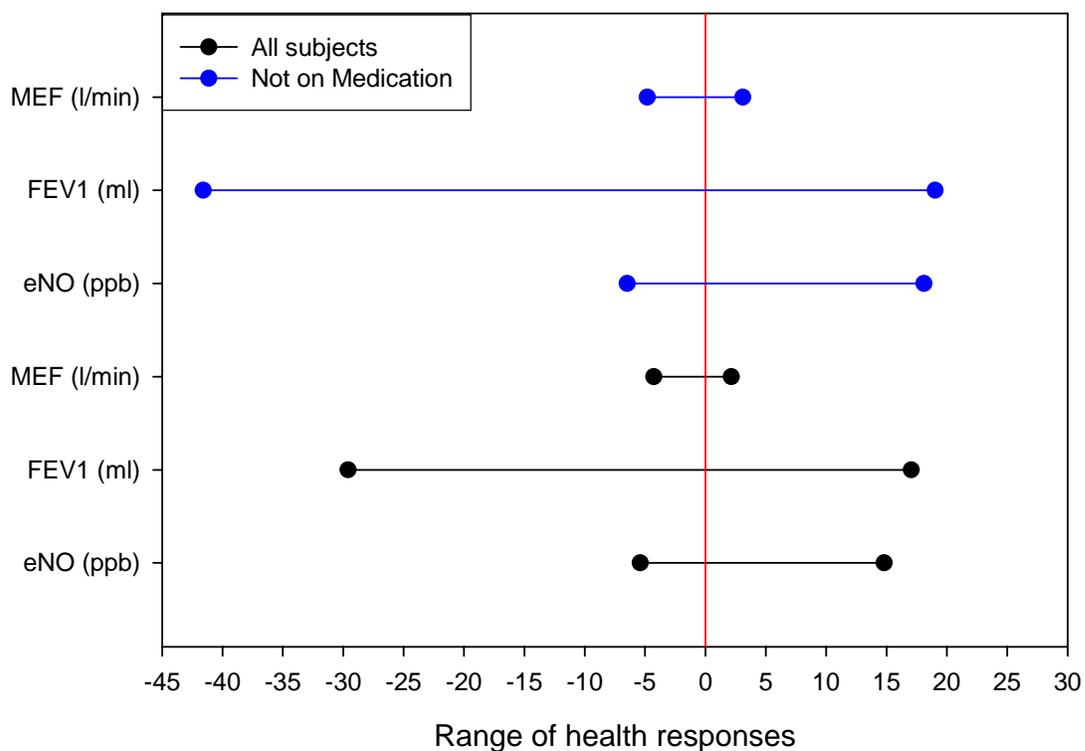


Figure 6. Effect of Ambient Generated PM<sub>2.5</sub> on levels of eNO and spirometry in the 32 Study Participants.

Figure 7. The Effect of Agricultural Burn Related PM<sub>2.5</sub> on levels of eNO and spirometry in the 32 Study Participants.

