

Ambient Woodsmoke and Associated Respiratory Emergency Department Visits in Spokane, Washington

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Three multivariate receptor algorithms were applied to seven years of chemical speciation data to apportion fine particulate matter to various sources in Spokane, Washington. Source marker compounds were used to assess the associations between atmospheric concentration of these compounds and daily cardiac hospital admissions and/or respiratory emergency department visits. Total carbon and arsenic had high correlations with two different vegetative burning sources and were selected as vegetative burning markers, while zinc and silicon were selected as markers for the motor vehicle and airborne soil sources, respectively. The rate of respiratory emergency department visits increased 2% for a 3.0 $\mu\text{g}/\text{m}^3$ interquartile range change in a vegetative burning source marker (1.023, 95% CI 1.009–1.038) at a lag of one day. The other source markers studied were not associated with the health outcomes investigated. Results suggest vegetative burning is associated with acute respiratory events. *Key words:* air pollution; generalized additive models; multivariate receptor models; source apportionment; source health effects analysis.

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Airborne particulate matter (PM) mass has been associated with adverse health outcomes.^{1–3} However, chemical composition and average particle size vary by source. Since one of the research priorities of the National Research Council (NRC, 1998) is the identification of specific constituents and/or sources of PM as contributors to adverse health outcomes, we used a long time series of speciated PM

data to learn more about the association between PM sources and health.

Previous time-series analyses have examined directly the relationships between estimated source contributions to fine particle mass and specific health outcomes in given communities.^{1,4,5,6} These studies use time series of estimated sources from factor-analysis-based algorithms, such as positive matrix factorization (PMF) or principal-component analysis, in time-series regression analysis. Health-effect estimates from these studies are potentially biased because they use estimated sources as predictors in the regression models rather than using measured variables with known error. Using these imputed measures in a regression model results in regression-attenuation bias.⁷ The resulting bias is attributable both to measurement error in the species and to rotational uncertainty in the receptor-model solutions. Due to rotational uncertainty, the bias can result in either an overestimate or an underestimate of the association; however, statistical methods have not advanced sufficiently to eliminate this bias.⁸ Alternatively, we can identify measured individual species that are highly correlated with the estimated source contributions derived from the multivariate receptor models. This approach potentially allows a limited set of independent variables with known laboratory analytical measurement errors to be identified and incorporated in the health-effect analysis. We apply this approach to a seven-year data set of speciated PM from Spokane, Washington.

Spokane's location in a semi-arid Eastern Washington valley makes it subject to frequent dust storms and pollutant-trapping temperature inversions. This location also has low concentrations of secondary aerosol and potentially confounding gaseous air pollutants such as SO_2 and O_3 . There is strong seasonal variability of the primary particulate sources, and several potential particulate metals sources are present.¹⁰ A previous PMF analysis of a subset of the measurements in Spokane identified several major sources of fine particles, including vegetative burning, sulfate aerosol, motor vehicle, nitrate aerosol, airborne soil, chlorine-rich source, and metal processing.¹⁰ The primary combustion sources were the dominant contributors to $\text{PM}_{2.5}$.

Of these combustion sources, previous studies have shown that daily variations in fine PM derived from vegetative burning are associated with respiratory emer-

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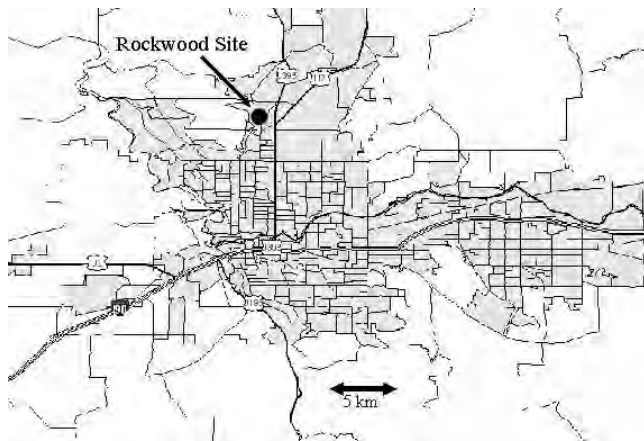


Figure 1—Map of the Rockwood sampling site located in a residential area of Spokane, Washington.

gencies,^{11,12} whereas variations in fine PM from mobile source emissions are associated with both cardiac^{6,9,13–16} and respiratory^{6,9,13,17} outcomes. Previous studies in Spokane have shown that emergency department (ED) visits for asthma are associated with combustion-derived PM, including fine particulate Zn, but not with fine particle soil.¹⁸ In addition, no significant associations were observed in Spokane between acute mortality and the coarse fraction of PM,^{19,20} although studies in other locations have observed associations between acute adverse health outcomes and this PM fraction.^{4,21}

METHODS

Our data consist of daily measures taken over a seven-year period from 1 September 1995 to 15 May 2002 in Spokane, Washington.

Chemical Speciation Data

Daily fine PM filter samples were collected at a residential monitoring site between 1995 and 2002. Figure 1 shows this site's (Rockwood) location 8 km north of the central business district. These filters were analyzed for nitrate, sulfate, ammonium, elemental (EC) and organic carbon (OC), and elemental analyses that included the following trace elements: Al, As, Br, Ca, Cu, Fe, K, Mn, Na, Pb, S, Si, Ti, and Zn. Details of the mass estimation and uncertainty methods are given elsewhere.^{22,23} During the first four years of this study, we used the thermal manganese oxidation method (TMO) to quantify particulate OC and EC. An overestimate of the ratio of EC to OC was expected due to this method's failure to correct for pyrolytically formed char.^{10,24} Therefore, only total carbon (TC) was available for the entire study period. TC, the sum of OC and EC, was obtained from TMO between 1 September 1995 and 20 September 1999, and from thermal optical transmission (TOT) between 24 September 1999 and 15 May 2002.

Health Data

Hospital admissions for Spokane were aggregated from the Comprehensive Hospital Admissions Reporting System (CHARS) as daily counts of cardiac hospital admissions (International Classification of Disease, Ninth Revision World Health Organization, Geneva (ICD9) codes 390-459). Emergency department visit records were obtained directly from four Spokane-area hospitals (Deaconess, Valley, Sacred Heart, and Holy Family). Daily respiratory ED visits (all respiratory causes, ICD9 codes 460-519) were aggregated. Any events from subjects who did not have Spokane-area zip codes as their residences were removed. Readmissions within two weeks of the first admission were also excluded.

Meteorologic Data

The Spokane County Air Pollution Control Authority (SCAPCA) supplied temperature and relative humidity data aggregated into daily averages.

Source Apportionment

We used 13 species (K, Fe, S, Zn, Ca, Si, Cu, Mn, Br, Na, As, NO₃⁻, TC) with good data in each of 2,925 daily samples for the source-apportionment analyses. These species were chosen because: they were available for use by all three multivariate receptor-model algorithms; they were consistently above their respective detection limits; and they did not exhibit any systematic changes in concentration due to a corresponding change in analytical method that might have occurred during the study period. See Table 1 for a summary of the fine-particle mass and species concentrations. We had a maximum of 38% of days with data below the limit of detection (LOD) and 29% missing (Br, Na, As; see Table 1). These daily values were used in the source apportionment analysis described below.

The source contributions were estimated using three different multivariate receptor algorithms: Unmix,^{25,26} positive matrix factorization (PMF),²⁷ and table-driven least squares as implemented in the multilinear engine version 2 (ME2).²⁸ The latter two least-squares algorithms weight the observed concentrations by their individual uncertainty estimates that vary for each species, for each observation. Less weight is given to more uncertain measurements, including those that are reported as either below the LOD and/or missing. Following Polissar et al., we replaced values below the LOD by half of the LOD and set the corresponding uncertainty to 5/6 of the LOD values.²⁹ Missing values were replaced by the geometric mean of all the measured values and the corresponding uncertainty was set to four times the geometric mean value. For the PMF analysis, we used the same criteria and approach

TABLE 1 Summary of fine Particle Mass (ng/m³) and 13 Species Concentrations Used for Unmix/PMF/ME2

Species	Arithmetic Mean	Geometric Mean*	Minimum	Maximum	BDL† No. (%)	Missing No. (%)
PM _{2.5}	10,580	8,790	930	43,230	—	698 (23.9)
K	78	61	2	698	127 (4.3)	98 (3.4)
Fe	125	89	2	1,970	31 (1.1)	98 (3.4)
S	288	244	2	1,270	125 (4.3)	98 (3.4)
Zn	12	8	1	280	269 (9.2)	98 (3.4)
Ca	49	31	3	957	686 (23.5)	98 (3.4)
Si	312	189	6	8,616	752 (25.7)	98 (3.4)
Cu	15	7	0.6	501	833 (28.5)	99 (3.4)
Mn	3.5	2.6	0.5	69	1,146 (38.2)	98 (3.4)
Br	1.1	0.9	0.01	13	0 (0)	848 (29.0)
Na	63	49	0.01	6,995	4 (0.1)	848 (29.0)
As	0.4	0.3	0.05	9.3	0 (0)	848 (29.0)
NO ₃ ⁻	579	455	20	5,923	450 (15.4)	224 (7.7)
TC	4,540	3,900	130	23,600	—	220 (7.5)

*Data below the limit of detection were replaced by half of reported detection limit values for the geometric mean calculations.
†Below detection limit.

detailed by Kim et al.¹⁰ Robust, individual markers of a particular source’s total mass contributions were identified using the correlations between the source contribution estimates from all three algorithms and the concentrations of individual species.

Health-effects Analysis

All statistical analyses were conducted using S-Plus 6.1 (Insightful Inc., Seattle, WA). We regressed daily cardiac hospital admissions and respiratory ED visits against the measured concentrations of selected source markers using Poisson regression with a generalized additive model (GAM)³⁰ and an exact GAM standard-error estimate.³¹ We controlled for season, temperature, relative humidity, and day of the week. The general form for the regression of event counts Y_t on daily levels of exposures X_t was: $E(Y_t) = \exp\{\beta_0 + \beta_1 X_{t-l} + S(\text{time}_t, \lambda_1) + S(\text{temp}_t, \lambda_2) + \gamma * \text{DOW} + \alpha \text{rhum}_t\}$, where temp is the daily average temperature; DOW are indicator variables for day of week, rhum is daily average relative humidity and l is the lag of the exposure. Our hypotheses used $l = 1$ day lag in the models. The smooth function, S , is constructed using smoothing splines with a parameter, λ . Six degrees of freedom per

year were used in this investigation ($\lambda_1 = 6 * 7 = 42$ df). Temperature was smoothed with $\lambda_2 = 2$ df. A sensitivity analysis of potentially influential points was conducted by repeating the analyses with these points removed, which did not change the results.

RESULTS

Source Apportionment and Source Marker Compounds

Eight sources were identified using three different multivariate receptor algorithms (PMF, ME2, Unmix). Their estimated average source contributions to fine PM mass are shown in Table 2. Both the source contributions and the source compositional features are consistent with the previous source-apportionment analysis at this site.¹⁰ The three algorithms (PMF, ME2, and Unmix) could not account for averages of 2.0, 1.4, and 0.3 $\mu\text{g}/\text{m}^3$, respectively, of the overall mean PM_{2.5} mass concentration at this site. The magnitudes of the top-ranking source contribution, vegetative burning, are similar across the three receptor-modeling methods. Due to the larger number of samples in this analysis, we were able to discern an additional As-rich source that was not evident in the earlier apportionment.¹⁰

TABLE 2 Estimated Average Source Contributions to Fine-particle Mass Concentrations in $\mu\text{g}/\text{m}^3$

	PMF	ME2	Unmix	PMF†
Vegetative burning	3.6 (2.3)*	4.1 (2.6)	4.3 (3.8)	5.3 (4.3)
As-rich	1.2 (1.3)	0.9 (1.0)	0.5 (0.7)	—
Motor vehicle	0.9 (0.9)	1.0 (1.0)	0.6 (0.8)	1.3 (0.9)
Sulfate aerosol	1.1 (1.0)	1.5 (1.2)	2.3 (1.7)	2.3 (1.2)
Nitrate aerosol	0.7 (0.8)	0.6 (0.7)	1.4 (1.3)	1.1 (1.5)
Airborne soil	1.0 (1.2)	1.0 (1.2)	0.6 (0.8)	1.0 (1.2)
Cu-rich	0.08 (0.15)	0.06 (0.12)	0.3 (0.6)	0.3 (0.3)
Marine	0.06 (0.12)	0.07 (0.14)	0.3 (0.3)	0.7 (0.9)

*() = standard deviation of daily estimates.

†Previous analysis by Kim et al. (2003) of a subset of 945 samples taken between January 1995 and December 1997.

TABLE 3 Estimated Percentages of Species Contributed by Each Source Feature, Unmix/PMF/ME2

Sources/ Species	Vegetative Burning	As-rich	Motor Vehicle	Sulfate Aerosol	Nitrate Aerosol	Airborne Soil	Cu-rich	Marine
K	37/13/19	1/14/—*	6/19/22	11/5/9	3/2/2	37/46/46	—	4/—/—
Fe	15/4/4	—	7/12/12	3/—/—	—	68/80/80	—	7/3/4
S	14/27/31	—/1/—	2/—/—	65/34/38	14/19/17	1/18/15	2/—/—	2/—/—
Zn	7/3/2	2/3/—	74/90/90	4/—/4	5/3/3	5/—/—	2/1/1	—
Si	3/—/—	—	3/3/3	—	1/—/—	91/97/97	—	3/—/—
Cu	—/—/2	—	—	—	—	—	99/96/97	—
Mn	—3/—	—/6/—	12/18/16	20/—/—	—/3/2	68/74/75	—	—/2/2
Na	8/—/—	—	4/—/—	6/—/—	—	14/5/—	—	67/95/100
As	1/—/—	97/97/88	—	—/—/7	—	—	—	—
NO ₃ ⁻	—	—	—	—	99/99/99	—	—	—
TC	55/96/85	5/—/11	7/2/4	12/—/—	14/—/—	—	4/—/—	3/1/—

*— indicates <1%.

The As-rich source profile is similar to our derived vegetative-burning profile (with the exception of more As) and therefore may be due to the burning of As-treated wood.³² Burning treated wood was prohibited by the Spokane County Air Pollution Agency in late 1994, which is consistent with the fact that the contribution from this study's As-rich source is relatively small compared with the more general vegetative-burning source, and that it also declined over the time period of this study. Table 3 summarizes the average percentage of each species mass concentration associated with each source. As expected, the nitrate, sulfate, As-rich, Cu-rich, and airborne soil sources contributed most of the NO₃⁻, S, As, Cu, and Si, respectively (our "Cu-rich" feature was previously named "metals"¹⁰). In addition, the motor vehicle source contributed most of the Zn and the vegetative-burning source contributed most of the TC.

Table 4 shows correlations for potential source-marker compounds with the source-contribution estimates. To facilitate interpretation, only correlations > 0.3 are shown. From Table 4, TC, As, and Zn are most highly correlated with the vegetative-burning source,

As-rich source, and motor-vehicle source, respectively. Finally, as expected, Si is highly correlated with the airborne-soil source. Based on these results, we treat Si, TC, and Zn as markers of the fine particle mass contributed by three different source-related features: airborne soil, vegetative burning, and motor-vehicle exhaust, respectively. For completeness, we also examined the association between As and respiratory ED visits, hypothesizing that As is a tracer for an additional, smaller source of wood combustion.

Health Effects Analysis

Table 5 gives summary statistics for the variables used in the analyses. This data set is a slightly smaller subset of that used in the source-apportionment analysis and therefore the species means differ somewhat in Tables 1 and 5. Table 6 gives overall estimated relative risks (RRs) for an interquartile (IQR: 75th–25th percentile) range increase in the identified source-marker compounds. There was no association of Si, As, or Zn with either of the health outcomes. In contrast, we estimated a 2% increase in respiratory ED visits for an interquar-

TABLE 4 Pearson Correlation Coefficients for Source Features vs Species, Unmix/PMF/ME2

Sources/ Possible Markers	Vegetative Burning	As-rich	Motor Vehicle	Sulfate Aerosol	Nitrate Aerosol	Airborne Soil	Cu-rich	Marine
K	0.57/0.42/0.43	—/0.37/—	—/0.44/0.45	—/—/0.36	—	0.71/0.73/0.73	—	—
Fe	—	—	—	—	—	0.94/0.94/0.95	—	—
S	—/0.35/0.36	—/0.35/—	—	0.87/0.50/0.52	—/0.46/0.47	—	—	—
Zn	0.35/0.46/0.44	—	0.92/0.94/0.93	—	0.32/—/—	—	—	—
Ca	—	—	—	—	—	0.85/0.93/0.94	—	—
Si	—	—	—	—	—	0.97/0.92/0.93	—	—
Cu	—	—	—	—	—	—	0.99/0.98/0.99	—
Mn	—	—	—	—	—	0.91/0.80/0.81	—	—
Br	0.50/0.45/0.42	0.31/0.41/0.35	—/0.38/0.37	—/0.91/0.94	—	—	—	—
Na	—	—	—	—	—	—	—	—/0.73/0.73
As	0.35/0.52/0.38	0.96/0.95/0.85	—/0.33/0.38	—/0.31/0.40	—	—	—	—
NO ₃ ⁻	—/0.34/0.34	—	—/0.30/0.30	—	0.99/0.94/0.95	—	—	—
TC	0.86/0.92/0.91	—/0.50/0.35	0.41/0.54/0.56	—	0.41/0.33/0.33	—	—	—

*— indicates a correlation <0.30.

TABLE 5 Daily Summary Statistics for Model Variables

Outcome	Mean	SD	Percentile				
			5th	25th	50th	75th	95th
Daily cardiac hospital admissions	5.3	1.9	2	4	5	7	9
Daily respiratory ED visits	16.3	6.9	7	11	15	20	29
TC	4.6 µg/m ³	2.6 µg/m ³	1.4 µg/m ³	2.8 µg/m ³	4.0 µg/m ³	5.8 µg/m ³	9.4 µg/m ³
As	0.45 ng/m ³	0.47 ng/m ³	0.08 ng/m ³	0.18 ng/m ³	0.31 ng/m ³	0.57 ng/m ³	1.1 ng/m ³
Zn	12.0 ng/m ³	11.5 ng/m ³	2 ng/m ³	4 ng/m ³	8 ng/m ³	16 ng/m ³	34 ng/m ³
Si	310 ng/m ³	410 ng/m ³	4 ng/m ³	85 ng/m ³	190 ng/m ³	410 ng/m ³	930 ng/m ³
Relative humidity	69%	19%	38%	53%	70%	86%	95%
Temperature	47.4° F	15.5° F	25.3° F	35.4° F	45.4° F	60.0° F	73.4° F
PM _{2.5}	10.6 µg/m ³	6.8 µg/m ³	2.9 µg/m ³	5.8 µg/m ³	8.9 µg/m ³	13.5 µg/m ³	25.1 µg/m ³

tile range change in TC (vegetative-burning marker) (IQR 3.0 µg/m³, 1.023, 95% CI 1.009–1.038) lagged one day. Table 7 shows the RRs lagged one day by heating (October–February) versus non-heating (March–September) season. The non-heating season diluted the association, while the relationship was larger (1.051, 95% CI 1.010–1.094) in the heating season.

DISCUSSION

Our source apportionment results are consistent with those of a previous source apportionment in Spokane.¹⁰ They used PMF to deduce the sources of PM_{2.5} at the same residential site in Spokane for 1995–1997 using a subset of our final dataset. Using the same species with the exception of S rather than SO₄⁼ and Cl rather than Na, they found seven of the eight source features reported here. We found not only source-contribution estimates (see Table 2), but also source compositional

features similar to those reported by Kim et al.¹⁰ Our substantially larger data set over the entire study period (2,925 vs 945 samples) allowed us to also resolve an additional, albeit minor, As-rich feature that they could not discern. It is noteworthy that in this airshed total particulate carbon and zinc were excellent individual tracers of motor-vehicle source and vegetative-burning contributions, respectively. The motor-vehicle source is enriched in Zn, presumably because of the use of the anti-wear additive zinc dialkyldithiophosphate.³³ The total carbon association is due to the fact that vegetative-burning particles are enriched in total carbon relative to other species and that this vegetative burning source is a dominant contributor to PM_{2.5} in this airshed, especially in the winter heating season.

We found evidence for adverse health outcomes associated with TC, our source marker for general vegetative-burning. The vegetative burning feature derived from the receptor model predicts that wood-smoke par-

TABLE 6 Relative Risks (RRs) and 95% CIs for the Entire Study Period of Hypothesized Health Outcomes for a Given IQR Increase

	IQR Increase	Cardiac Hospital RR (95% CI)	All Respiratory ED RR (95% CI)
TC (vegetative burning)	3.0 µg/m ³		
lag 0		1.008 (0.983–1.032)	1.012 (0.997–1.027)
lag 1		0.999 (0.975–1.024)	1.023 (1.009–1.088)*
As (As-rich)	0.39 ng/m ³		
lag 0		0.994 (0.976–1.012)	1.008 (0.998–1.019)
lag 1		0.997 (0.980–1.015)	1.002 (0.992–1.013)
Zn (motor vehicle)	11.6 ng/m ³		
lag 0		1.007 (0.987,1.028)	1.012 (0.999–1.024)
lag 1		1.003 (0.983,1.024)	1.006 (0.994–1.019)
Si (airborne soil)	324 ng/m ³		
lag 0		1.011 (0.994–1.028)	0.999 (0.990–1.009)
lag 1		1.005 (0.989–1.021)	0.999 (0.989–1.009)
PM _{2.5}	7.7 µg/m ³		
lag 0		1.008 (0.985–1.032)	1.011 (0.997–1.025)
lag 1		1.000 (0.978–1.023)	1.013 (0.999–1.027)

*p = 0.002.

TABLE 7 Relative Risks (RRs) and 95% CIs by Season of Hypothesized Health Outcomes Lagged One Day

	IQR Increase	Cardiac Hospital RR (95% CI)	All Respiratory ED RR (95% CI)
TC (vegetative burning)			
Heating season*	3.5 µg/m ³	1.024 (0.968–1.084)	1.051 (1.010–1.094)**
Non-heating	2.2 µg/m ³	0.992 (0.966–1.018)	1.013 (0.998–1.028)
As (As-rich)			
Heating	0.53 ng/m ³	0.992 (0.958–1.026)	1.013 (0.992–1.036)
Non-heating	0.30 ng/m ³	1.000 (0.981–1.021)	0.998 (0.986–1.011)
Zn (Motor vehicle)			
Heating	14.9 ng/m ³	1.002 (0.969–1.037)	1.014 (0.994–1.036)
Non-heating	7.9 ng/m ³	1.004 (0.980–1.029)	1.000 (0.986–1.014)
Si (Airborne soil)			
Heating	162 ng/m ³	1.002 (0.986–1.019)	1.001 (0.992–1.010)
Non-Heating	376 ng/m ³	1.024 (0.973–1.078)	1.002 (0.972–1.033)
PM _{2.5}			
Heating	10.1 µg/m ³	1.015 (0.968–1.063)	1.018 (0.985–1.052)
Non-heating	5.5 µg/m ³	0.995 (0.969–1.021)	1.009 (0.994–1.025)

*October–February.
 †p = 0.01.

ticles (not total PM_{2.5}) contain about 80% TC by mass. Therefore, this epidemiologic association translates to a 2.3/3.0 * 0.8 = 0.6% increase in respiratory ED visits per µg/m³ of woodsmoke particle mass. This is reasonably consistent with the results of McGowan et al.,¹¹ who found a 0.2% (3.37/14.8) increase in all respiratory admissions per µg/m³ increase in PM₁₀ in Christchurch, New Zealand, a region that was significantly impacted by woodsmoke during the heating season. This result is also consistent with the reported associations between outdoor fine PM and measures of acute respiratory response in woodsmoke-impacted neighborhoods,^{34–37} and the fact that ambient wood-smoke particles readily penetrate indoors.³⁸

In this study, the other combustion-source marker compounds, As and Zn, did not show associations with the health outcomes studied. In contrast, previous health analyses in Spokane found an increased risk of ED visits for asthma with higher meteorologic stagnation index and an increased risk of all respiratory and asthma-only ED visits for increases in both fine particle Zn and CO, the latter being a general indicator of stagnant air conditions and increased levels of surface combustion sources.^{23,18,39} In addition, Si, a marker of fine-particle soil, also did not show associations with any of the health outcomes, consistent with previous observations in Spokane.¹⁸ However the inability to detect association with the other source markers is not surprising, given their relatively low PM_{2.5} source contributions compared with vegetative burning at this location, as summarized in Table 2.

Our results suggest vegetative burning is associated with acute respiratory events. This conclusion is qualified by the relatively small community population and

the relatively low pollution levels in Spokane. While we compensated for these features by using a seven-year time series, the low pollution level and our need to use TC rather than the more traditional temperature-resolved carbon fractions results in less-than-optimal resolving power in the source-apportionment analysis. Another important limitation is potential misclassification of exposure resulting from: 1) the use ambient air-pollution measurements at a single site as a proxy for ambient-source personal exposure, and 2) increased potential for indoor exposures to woodsmoke from increased use of wood stoves during cold, stable conditions that are associated with increased outdoor woodsmoke levels. These limitations are offset by some important additional strengths: we had a large number of chemically resolved filter samples over the seven-year time period, the study population lives within 8 km of the monitor, and in Spokane vegetative burning is an important source of PM_{2.5} during the heating season.

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