



## Is Alaska Truly the Great Escape from Air Pollution? – Long Term Source Apportionment of Fine Particulate Matter in Fairbanks, Alaska

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### ABSTRACT

Alaska is generally considered to be a place that is one of the last great escapes from air pollution. However, they have not spent a winter in Fairbanks or the nearby village of North Pole, where the daily average PM<sub>2.5</sub> concentration was 170 µg/m<sup>3</sup> in December 2012 according to the Alaska Department of Environmental Conservation (DEC) Air Monitoring Network. In this study, source apportionment using Positive Factorization Matrix (EPA PMF 5.0) has been conducted based on the 2005 to 2012 Fairbanks PM<sub>2.5</sub> compositional data including elements, sulfate, nitrate, ammonia, elemental carbon (EC), and organic carbon (OC) from the U.S. Environmental Protection Agency speciation network. Seven sources were identified: soil, gasoline, sulfate, diesel, wood smoke, road salt, and nitrate. The average contributions to PM<sub>2.5</sub> of these seven sources were 3.4%, 16.3%, 19.5%, 14.3%, 40.5%, 1.5%, and 4.5%, respectively. Wood smoke provided the highest contributions to PM<sub>2.5</sub>. Its contributions were the lowest in 2007 (3.5 µg/m<sup>3</sup>) and peaked in 2009 (5.7 µg/m<sup>3</sup>). The winter contributions of sulfate, nitrate, diesel, road salt, and wood smoke were all substantially higher compared to other seasons. Wood smoke is the only source with weekend's contribution greater than the weekdays' corresponding to the times when residential wood combustion is more likely to occur. The contributions of diesel, wood smoke, and sulfate were approximately doubled on violation days (daily average PM<sub>2.5</sub> higher than 35 µg/m<sup>3</sup>) compared to all days. This finding indicates that winter heating is the most important factor affecting the air quality in Fairbanks. In the future, additional source apportionment using other receptor models and tracers will need to be conducted to confirm these results.

**Keywords:** Air pollution; Fine particular matter; Source apportionment; Positive matrix factorization; Wood combustion; Alaska.

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### INTRODUCTION

Alaska's pristine, mountainous landscapes suggest crystal clean air. Most people including ourselves think of Alaska as one of the last great escapes from air pollution. But we have not spent a winter in Fairbanks or the nearby town of North Pole, where the daily average fine particulate matter (PM<sub>2.5</sub>) concentration was 182 µg/m<sup>3</sup> on December 18, 2012 (Alaska DEC, 2013). PM<sub>2.5</sub> presents high risks to human health. Numerous studies have suggested links between long-term exposure to PM<sub>2.5</sub> and a wide range of serious health effects including premature death (especially related to heart disease), cardiovascular effects (such as heart attacks and strokes), reduced lung development, and chronic respiratory diseases (such as asthma) (EPA, 2012a).

Previously, source characterization has been conducted

for the PM<sub>2.5</sub> chemical speciation data collected at Alaskan National Park sites from 1986 to 1995 by Polissar *et al.* (1996; 1998a, b). Four major sources were identified at seven monitoring sites in Alaska: arctic haze, sea salt, soil dust, and forest fires. Ambient PM<sub>2.5</sub> speciation samples collected at one monitoring site located in Anchorage, Alaska between 2002 and 2003 were characterized through the application of the source apportionment method of positive matrix factorization (PMF). Gasoline emission was the largest source of PM<sub>2.5</sub>, but it was likely originated from a parking lot close to the monitoring site (Kim and Hopke, 2008).

Ward *et al.* (2012) performed PM<sub>2.5</sub> source apportionment using Chemical Mass Balance (CMB) model at four locations in Fairbanks, Alaska over a three-winter period (2008/2009, 2009/2010, and 2010/2011). The daily average PM<sub>2.5</sub> concentrations frequently exceeded the 24-hour National Ambient Air Quality Standard (NAAQS) of 35 µg/m<sup>3</sup>. The CMB model identified five sources including sulfate, ammonium nitrate, diesel exhaust, automobiles, and wood smoke. Wood combustion (including residential wood stoves, biomass open burning, outdoor boilers, and small industrial sources) was the major source of PM<sub>2.5</sub>

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throughout the winter months, contributing between 63% and 80% of the measured PM<sub>2.5</sub> at the four sites.

The effects of exchanging noncertified with certified wood-burning devices on the 24h-average PM<sub>2.5</sub> concentrations in Fairbanks, Alaska, in a cold season (October to March) were investigated using the Weather Research and Forecasting model inline coupled with a chemistry package (Tran and Mölders, 2012). Changing out 2930 uncertified woodstoves and 90 outdoor wood boilers reduced the 24 h-average PM<sub>2.5</sub> concentrations on average by 0.6 µg/m<sup>3</sup> and avoided seven of 55 simulated exceedance days during this half-a-year. The highest estimated reductions on any exceedance day ranged between 1.7 and 2.8 µg/m<sup>3</sup> (Tran and Mölders, 2012).

In this study, the major sources of PM<sub>2.5</sub> in Fairbanks, Alaska were identified and their contributions to PM<sub>2.5</sub> concentrations were estimated based on data collected between 2005 and 2012 using the PMF model. This study provides the record of the longest PM<sub>2.5</sub> chemical composition data in Alaska.

## MATERIALS AND METHODS

### *Description of the Study Area and the Sampling Site*

Fairbanks is the largest city in the interior region of Alaska, and second largest in the state after Anchorage. It has a total area of 85 km<sup>2</sup>, and is located in the central Tanana Valley. Fairbanks is the northernmost Metropolitan Statistical Area in the United States, lying less than 190 km south of the Arctic Circle. According to 2012 estimates, the population of the city was 32,070. Fairbanks' climate is classified as subarctic, with long, cold winters, and short, warm summers. Winter lasts from late September/early October until late April/early May. Snow is limited from February to May. Average winter low temperatures range from −26 to −32°C, extremes can range from −51 to −59°C. In winter, Fairbanks' location at the bottom of the Tanana Valley causes cold air to accumulate in and around the city. Warmer air rises to the tops of the hills north of Fairbanks, while the city itself experiences substantial temperature inversions. In the summer, temperatures typically range between 21 and 10°C. The dominant wind direction is northerly.

Twenty-four hour PM<sub>2.5</sub> samples were collected every three days at the State Office Building (675 7<sup>th</sup> Ave) Speciation Trend Network (STN) site (site ID: 02-090-0010, 64.84°N, 147.72°W) between March 2005 and September 2012. The site is 132 m above the sea level.

### *Sample Collection and Analytical Method*

The PM<sub>2.5</sub> samples were collected on Teflon, nylon, and quartz filters with a spiral aerosol speciation sampler (Met One Instruments) (EPA, 2012b). The Teflon filters were used for the analysis of mass concentrations of up to 48 elements by the energy-dispersive x-ray fluorescence (EDXRF). The nylon filters were used for the analysis of cations and anions by ion chromatography (IC). The quartz filters were used for organic carbon (OC) and elemental carbon (EC) by the thermal optical reflectance/thermal optical transmittance (TOR/TOT) method (EPA, 2012b). A

total of 850 samples were collected.

### *Receptor Modeling*

PMF is a least-squares variant of factor analysis with non-negative factor elements. It takes into account the uncertainties in the observed data values (Polissar *et al.*, 1998b). EPA PMF version 5.0 was used in this study. The input data matrix includes 850 rows and 27 columns. The measured concentrations below method detection limit (MDL) values were replaced by half of the MDL value, and their uncertainties were set at five sixths of the MDL values. Missing concentrations were replaced by the geometric mean of the concentrations, and their accompanying uncertainties were set at four times this geometric mean concentration (Polissar *et al.*, 1998b). An additional uncertainty of 25% was added. The signal to noise ratio (S/N) was greater than 2.0 for the following 12 species including Al, Ca, Cu, Cl, Fe, Si, Zn, S, K, NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup>, and OC. The species with S/N value between 0.9 and 2.0 were marked as “weak” species, including As, Br, Cr, Pb, Mn, Ni, Mg, Se, Ti, V, Sr, Na, EC, and PM<sub>2.5</sub>. All of the weak variables were down-weighted by a factor of 3. PM<sub>2.5</sub> was set as the total variable. Both sulfur and sulfate are strong tracers of the sulfate factor, and they both are available from the STN site. In this study, sulfur has a greater signal to noise ratio value than sulfate and is less likely to suffer from positive artifacts resulting from adsorption of SO<sub>2</sub>. Therefore, we chose sulfur as the input variable for the PMF analysis.

Between June and August 2009, a number of forest fires occurred over interior Alaska. The image from the Moderate Resolution Imaging Spectroradiometer (MODIS) on NASA's Aqua satellite shows several lightning-triggered fires southwest of Fairbanks, Alaska, on July 7 (Fig. S1). Hourly PM<sub>2.5</sub> concentrations greater than 60 µg/m<sup>3</sup> were frequently observed. These data were excluded from the PMF modeling since such high contributions will distort the analysis.

## RESULTS AND DISCUSSION

### *Data Overview*

Table 1 provides the arithmetic mean, standard deviation, geometric mean, MDL values, and prevalence of missing and below detection limits (BDL) observations for the measured species at the sampling site. A comparison between the reconstructed and measured PM<sub>2.5</sub> mass concentrations (Fig. 1) shows that the reconstructed PM<sub>2.5</sub> effectively reproduced the measured values (slope = 1.09, R<sup>2</sup> = 0.93). The equation for calculating the reconstructed PM<sub>2.5</sub> mass concentration is obtained from Malm and Hand (2007):

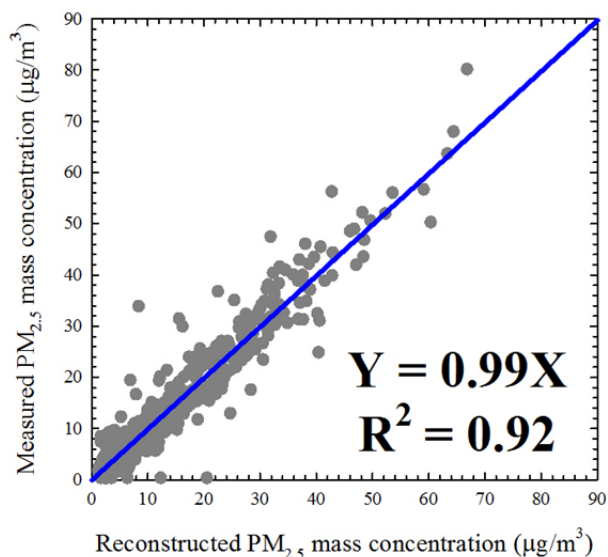
$$\begin{aligned} \text{PM}_{2.5}(\text{reconstructed}) = & 1.37[\text{SO}_4^{2-}] + 1.29[\text{NO}_3^-] + \\ & 1.4[\text{OC}] + [\text{EC}] + 2.2[\text{Al}] + 2.49[\text{Si}] + 1.94[\text{Ti}] + 1.63[\text{Ca}] \\ & + 2.42[\text{Fe}] + 1.8[\text{Cl}] \end{aligned} \quad (1)$$

### *Source Identification and Source Profiles*

A range of factor numbers from 6 to 9 was examined. On the basis of the quality of fit as indicated by the scaled residuals distributions and the interpretability of the resulting

**Table 1.** Summary statistics for the PM<sub>2.5</sub> and species concentration (ng/m<sup>3</sup>) at Fairbanks, Alaska during March 2005 to September 2012.

Species	Arithmetic Mean	Standard Deviation	Geometric Mean	Percent BDL <sup>a</sup>	MDL <sup>b</sup>	Percent Missing
As	0.5	0.9	0.8	82.2%	1.6	4.3%
Al	30.5	40.9	23.3	45.2%	16.4	4.3%
Br	2.5	3.0	1.8	44.9%	1.6	4.3%
Ca	32.0	29.2	22.7	9.0%	6.3	4.3%
Cr	2.5	14.6	1.3	76.4%	2.3	4.3%
Cu	2.3	3.8	1.6	57.4%	1.9	4.3%
Cl	38.2	87.5	15.1	52.7%	7.6	4.3%
Fe	60.5	71.0	43.2	0.1%	2.0	4.3%
Pb	2.3	3.9	2.4	75.7%	3.8	4.3%
Mn	1.2	1.9	1.0	75.5%	1.9	4.3%
Ni	0.8	4.4	0.6	83.4%	1.4	4.3%
Mg	5.9	17.3	9.3	84.6%	15.2	4.3%
Se	0.2	0.6	0.5	93.7%	2.1	4.3%
Ti	2.8	5.5	2.8	77.1%	4.7	4.3%
V	0.5	1.0	0.9	92.7%	3.2	4.3%
Si	97.0	126.2	56.3	8.9%	12.2	4.3%
Zn	21.7	30.1	9.4	20.6%	2.4	2.3%
Sr	1.9	8.3	1.4	80.3%	2.3	4.3%
S	673.4	858.2	389.4	0.1%	8.2	4.3%
K	61.1	104.4	37.1	4.4%	7.0	4.3%
Na	36.8	78.5	32.0	70.6%	44.0	4.3%
NH <sub>4</sub> <sup>+</sup>	792.5	1146.2	358.0	5.7%	16.2	3.9%
NO <sub>3</sub> <sup>-</sup>	557.0	631.8	318.4	0.1%	8.8	3.9%
OC	6969.8	9939.2	4845.1	0.0%	240.1	42.1%
EC	914.2	882.2	567.8	9.9%	240.1	42.1%
PM <sub>2.5</sub>	12,770.5	15,998.2	8,490.1	0.8%	743.4	4.7%

<sup>a</sup>Below detection limit.<sup>b</sup>Method detection limit.**Fig. 1.** Comparison of reconstructed and measured PM<sub>2.5</sub> concentrations.

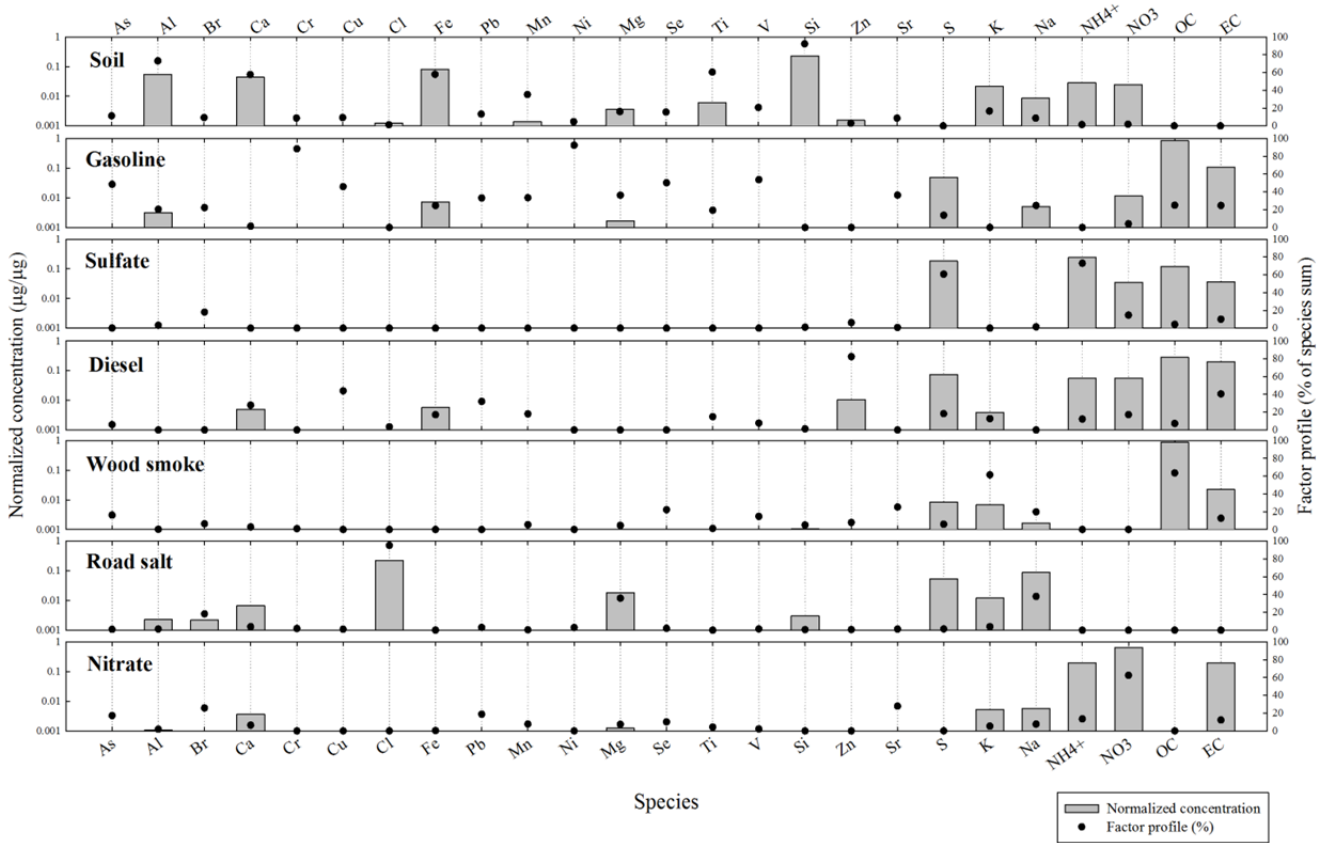
profiles, a seven-factor solution was identified. Each of these seven factors has a distinctive grouping of species that can be associated with a specific source class. The scaled residuals were approximately symmetrically distributed

between  $-3$  and  $+3$ . Rotations were introduced to the solutions by adjusting the FPEAK value (Paatero *et al.*, 2002). The non-rotated solutions (FPEAK = 0.0) were judged most interpretable. Seven sources include soil, gasoline, sulfate, diesel, wood smoke, road salt, and nitrate. The average contributions to PM<sub>2.5</sub> of these seven sources were 3.4%, 16.3%, 19.5%, 14.3%, 40.5%, 1.5%, and 4.5% respectively during March 2005 to September 2012.

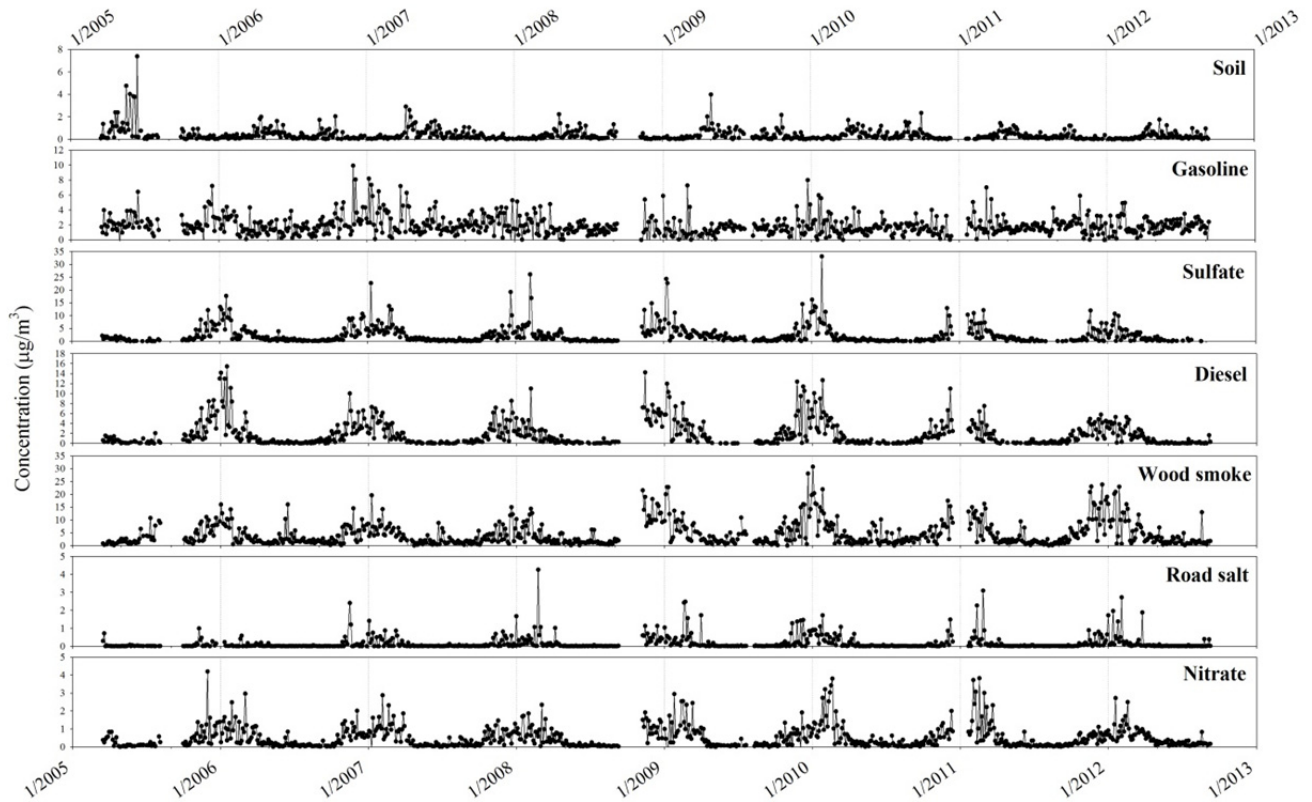
Fig. 2 shows the source profiles while Fig. 3 presents the time series of contributions from each source. The sum of the PM<sub>2.5</sub> contributions derived from the PMF model and measured PM<sub>2.5</sub> values are compared in Fig. 4. The coefficient of determination (0.95) and slope (1.04) suggest good agreement between modelled and measured results.

The species associated with the first source included Al, Ca, Fe, and Si, and this profile was classified as soil. In the summer when there is no snow cover, contributions were higher than other seasons. The major marker species contributing to the second source included S, OC, and EC, and this profile was classified as gasoline. Prior studies found that the in-use gasoline available in the Fairbanks area had sulfur contents in the range of 160 to 215 ppm (e.g., Alaska DEC, 2006). No distinct seasonal variation was observed for this source.

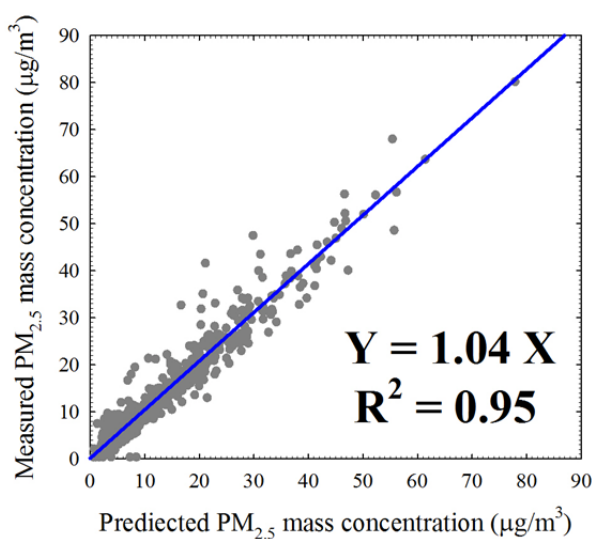
The species contributing to the third source included S and NH<sub>4</sub><sup>+</sup>. This source was identified as sulfate. The seasonal



**Fig. 2.** Source profiles (bars represent normalized mass contribution, y-axis on the left, black dots represent contribution in percentage, y-axis on the right).



**Fig. 3.** Temporal variation of source contributions to total  $PM_{2.5}$ .



**Fig. 4.** Comparison of PMF-predicted and measured  $PM_{2.5}$  concentrations.

average mass contributions show peaks in the winter suggesting the impact of sulfur in fuels that are burned for building heating. The majority of buildings in Fairbanks are heated with oil (52%) (Carlson *et al.*, 2010). The presence of OC in the sulfate profiles supports the suggestion of the sulfur being primary in nature arising from the combustion of S-containing fuel.

The fourth source was classified as diesel. The main species in this source are OC, EC, S,  $NO_3^-$ , Fe, Zn, and Ca. For diesel emission, EC was higher compared to the gasoline vehicle profile. Zn and Ca are additives in motor oil, and Fe is released from brake pads and muffler ablation (Garg *et al.*, 2000; Maykut *et al.*, 2003; Alander *et al.*, 2005; Wahlin *et al.*, 2006; Zhao and Hopke, 2006). The winter contributions were higher than other seasons. Because of the access to Prudhoe Bay during the winter when the ice road is open, there are of the order of 250 heavy duty diesel trucks per day operating from Fairbanks along the James W. Dalton Highway compared to 160 per day during the summer (Alaska DOT, 2010). The lower mixing-layer height also contributes to the high winter values due to the reduced dispersion of these ground level emissions.

The validation of the gasoline and diesel factors was conducted by reviewing the G-Space plots for combinations of gasoline, diesel and sulfate factors (Fig. S2). Both the gasoline vs. diesel and gasoline vs. sulfate contribution values completely fill the scatter plot space showing that there is no correlation between them. This result suggests that these three factors are independent of one another.

The major species contributing to the fifth source included K, OC and EC. This source was assigned to wood smoke. As shown in Fig. 3, the highest level of wood combustion occurred in the winter. Wood combustion is used in 23% of the homes in Fairbanks (Carlson *et al.*, 2010). This factor may include limited contributions from meat cooking, because the combination of OC, EC, and K would also be the tracers for this source.

The major species contributing to the sixth source included

Cl, S, Na, Mg, Si, and Ca. This factor was assigned to be road salt. The peak seasonal mass contributions were in the winter. The seventh source was interpreted as nitrate.  $NO_3^-$ ,  $NH_4^+$ , and EC were major species contributing to this source. The mass contributions of nitrate show peaks in the winter when conditions favor the formation of particulate nitrate from gas-phase nitric acid and ammonia.

#### Temporal Variations of Sources

Fig. 5 shows the annual variations of contributions to total  $PM_{2.5}$  mass of each source. Wood smoke was the dominant source during the entire period of 2005 to 2012. Its contributions were the lowest in 2007 ( $3.5 \mu\text{g}/\text{m}^3$ ) and reached the maximum in 2009 ( $5.7 \mu\text{g}/\text{m}^3$ ). The contributions of gasoline, diesel and sulfate had values ranging between  $1.0 \mu\text{g}/\text{m}^3$  and  $2.8 \mu\text{g}/\text{m}^3$ . The gasoline contributions gradually increased during 2009 to 2012. Conversely, diesel and sulfate experienced a downward trend. According to Alaska DOT, the number of loaded trucks ( $> 27,200 \text{ kg}$ ) crossing the Fox Weigh Station (north of Fairbanks) substantially decreased in 2009 (Fig. S3). The annual contributions of soil, road salt, and nitrate were all below  $0.9 \mu\text{g}/\text{m}^3$  with no distinct annual variation.

Table 2 summarizes the seasonal contributions for each source and shows the average source contributions for the whole sampling period. The winter contributions of sulfate, nitrate, diesel, road salt, and wood smoke were all substantially higher compared to other seasons. Wood smoke is the dominant source of  $PM_{2.5}$  for all four seasons. Fig. 6 illustrates weekday and weekend comparison results. Wood smoke is the only source with its weekend contribution greater than the weekdays. These times correspond to the traditional at-home weekend hours when residential wood combustion is more likely to occur. Fig. 7 compares source contributions on violation days and all days. Violations days are defined as the days with daily  $PM_{2.5}$  mass concentration greater than  $35 \mu\text{g}/\text{m}^3$  and 36 of the 39 violation days detected were during the wintertime. The contributions of diesel, wood smoke, and sulfate were approximately double on violation days compared to all days. The ambient temperatures were significantly lower on violations days compared to all winter days ( $-26.5 \pm 7.6^\circ\text{C}$  vs.  $-18.4 \pm 9.0^\circ\text{C}$ ,  $p < 0.01$ ). These findings indicate that the combination of heating and strong inversion on cold winter days plays an important role in affecting the air quality in Fairbanks. No distinct difference was observed for other sources.

#### Wood Smoke Source

Fig. 8 shows the wood smoke contributions only during winter season (Dec–Feb). There was a 30% increase between 2008 and 2009. After 2009, the contributions became very steady with the lowest average value ( $10.6 \mu\text{g}/\text{m}^3$ ) observed in 2012 and the highest average value ( $11.3 \mu\text{g}/\text{m}^3$ ) in 2010. The Fairbanks North Star Borough (FNSB) Wood Stove Change-Out program started in 2010. It offers a reimbursement incentive for replacing, removing, repairing or upgrading solid fuel burning appliances. Our analysis results show that this program made weak impact in reducing wood smoke contributions to  $PM_{2.5}$  during the winter.

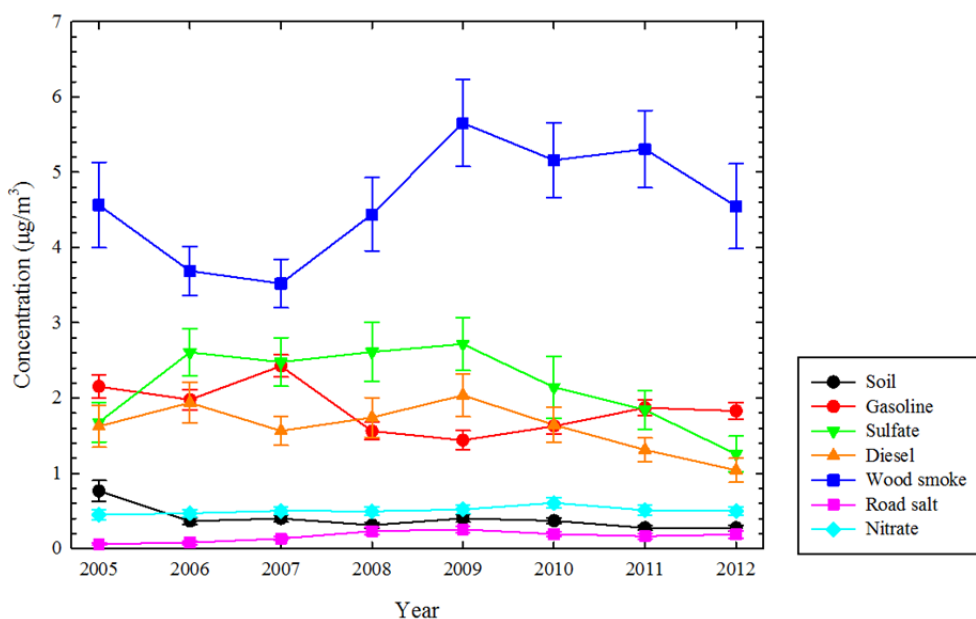


Fig. 5. Annual variation of source contributions to total  $PM_{2.5}$  in mass. Error bars represent one standard error.

Table 2. Average seasonal source contributions.

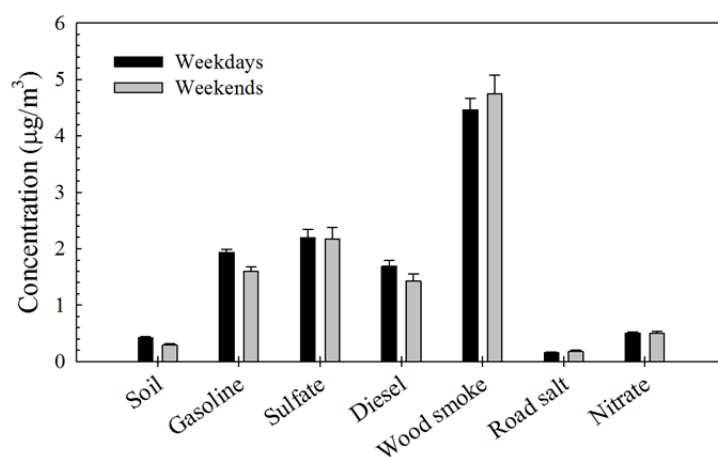
Source name	Winter (Dec–Feb)		Spring (Mar–May)		Summer (Jun–Aug)		Fall (Sep–Nov)		Average	
	Mean $\pm$ SE <sup>a</sup> ( $\mu\text{g}/\text{m}^3$ )	%	Mean $\pm$ SE <sup>a</sup> ( $\mu\text{g}/\text{m}^3$ )	%	Mean $\pm$ SE <sup>a</sup> ( $\mu\text{g}/\text{m}^3$ )	%	Mean $\pm$ SE <sup>a</sup> ( $\mu\text{g}/\text{m}^3$ )	%	Mean $\pm$ SE <sup>a</sup> ( $\mu\text{g}/\text{m}^3$ )	%
Soil	0.086 $\pm$ 0.008	0.4	0.651 $\pm$ 0.043	8.3	0.412 $\pm$ 0.04	7.1	0.342 $\pm$ 0.030	3.0	0.385 $\pm$ 0.020	3.4
Gasoline	2.219 $\pm$ 0.129	9.7	1.648 $\pm$ 0.080	20.9	1.717 $\pm$ 0.06	29.7	1.925 $\pm$ 0.096	17.6	1.857 $\pm$ 0.046	16.3
Sulfate	5.761 $\pm$ 0.371	25.2	1.698 $\pm$ 0.106	21.5	0.312 $\pm$ 0.03	5.4	1.579 $\pm$ 0.164	14.5	2.215 $\pm$ 0.119	19.5
Diesel	4.244 $\pm$ 0.221	18.6	0.705 $\pm$ 0.070	8.9	0.123 $\pm$ 0.02	2.1	2.000 $\pm$ 0.162	18.3	1.631 $\pm$ 0.084	14.3
Wood smoke	9.061 $\pm$ 0.424	39.6	2.621 $\pm$ 0.159	33.3	3.035 $\pm$ 0.28	52.5	4.487 $\pm$ 0.311	41.1	4.607 $\pm$ 0.171	40.5
Road salt	0.436 $\pm$ 0.044	1.9	0.107 $\pm$ 0.016	1.4	0.021 $\pm$ 0.002	0.4	0.140 $\pm$ 0.022	1.3	0.166 $\pm$ 0.013	1.5
Nitrate	1.051 $\pm$ 0.056	4.6	0.450 $\pm$ 0.033	5.7	0.166 $\pm$ 0.033	2.9	0.455 $\pm$ 0.037	4.2	0.510 $\pm$ 0.021	4.5
Sum	22.858 $\pm$ 0.929	100.0	7.879 $\pm$ 0.320	100.0	5.786 $\pm$ 0.308	100.0	10.910 $\pm$ 0.621	100.0	11.371 $\pm$ 0.358	100.0

<sup>a</sup> SE: Standard error.

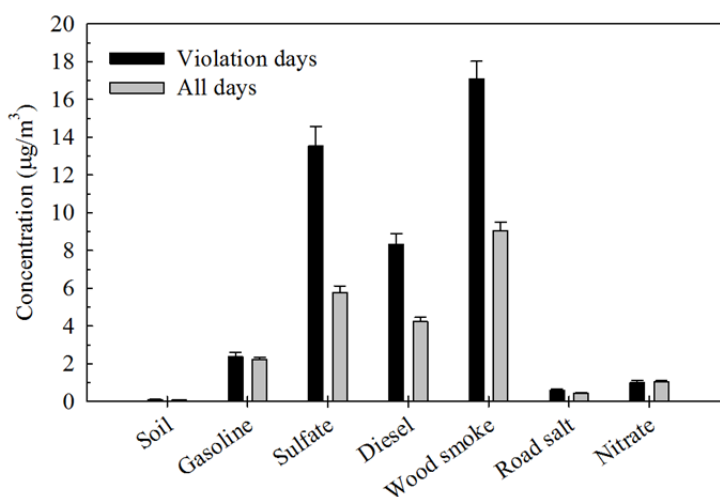
Ward *et al.* (2012) calculated the wintertime source contributions between 2008 and 2010 (Fig. S4). The CMB results show that wood smoke is the dominant  $PM_{2.5}$  source with the lowest and highest contributions observed in 2008 and 2009, respectively. This trend was consistent with our annual variation results shown in Fig. 5, but different from our wintertime results in Fig. 8. The 2010 winter  $PM_{2.5}$  chemical composition data Ward *et al.* (2012) used for their CMB modeling were collected between November 2010 and February 2011. In our study, the 2010 winter started in December 2010 and ended in February 2011. The exclusion of November data could probably contribute to the discrepancy.

## CONCLUSIONS

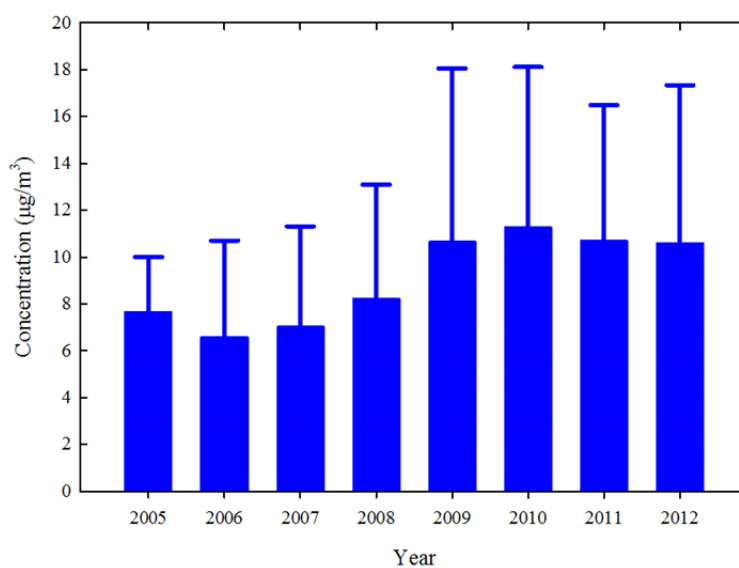
In this study, source apportionment using PMF has been conducted based on the March 2005 to September 2012  $PM_{2.5}$  compositional data collected in Fairbanks, Alaska. This study holds the record of the longest  $PM_{2.5}$  chemical composition data in Alaska. Seven sources were identified including soil, gasoline, sulfate, diesel, wood smoke, road salt, and nitrate. Their average contributions to  $PM_{2.5}$  were 3.4%, 16.3%, 19.5%, 14.3%, 40.5%, 1.5%, and 4.5%, respectively. Wood smoke contributed the most to  $PM_{2.5}$  throughout the entire study period. Sulfate and diesel together with wood smoke were the major sources on violation days



**Fig. 6.** Weekday and weekend comparison of source contributions to total PM<sub>2.5</sub> in mass. Error bars represent one standard error.



**Fig. 7.** Violation day and all day comparison of source contributions to total PM<sub>2.5</sub> in mass. Error bars represent one standard error.



**Fig. 8.** Annual variation of wintertime wood smoke contributions to total PM<sub>2.5</sub> in mass. Error bars represent one standard deviation.

indicating a strong impact of winter heating on the local air quality.

In the future, source apportionment using other receptor models and tracers will need to be conducted to confirm these results. For example, aethalometer delta-C, a simple, cheap, readily accessible, and of high time resolution marker of wood smoke, could help further apportion the wood stove contributions to PM<sub>2.5</sub> (Wang *et al.*, 2011).

## ACKNOWLEDGMENTS

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## SUPPLEMENTARY MATERIALS

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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Fig. S4. 2008-2010 annual variation of wintertime source contributions to total PM<sub>2.5</sub> mass  
(Data source: Ward et al., 2012).