Source apportionment modeling for multiple urban $PM_{2.5}$ chemical speciation sites within the Pacific Northwest.

Robert Kotchenruther
EPA Region 10
Kotchenruther.Robert@epa.gov
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1. Introduction and Summary

This report updates the source apportionment modeling reported in "Source apportionment modeling for multiple urban and rural $PM_{2.5}$ chemical speciation sites within the Pacific Northwest" dated March 6, 2012. The previous report discussed $PM_{2.5}$ source apportionment results for both the EPA Chemical Speciation Network (CSN) sites and IMPROVE network sites using available data through September 2011. This report updates that modeling only for the EPA CSN sites using available data through April 2013 and includes the Fairbanks Alaska site, which was not included in the previous report.

Source apportionment modeling was conducted using the Positive Matrix Factorization (PMF) model on chemically speciated $PM_{2.5}$ aerosol for 13 urban monitoring locations throughout the Pacific Northwest and Alaska. Data from EPA's mainly urban population-oriented Chemical Speciation Network (CSN) were used expect for 3 sites in central and Southern Oregon where winter 2012/2013 data were not yet available. Two sites were modeled that did not have winter 2012/2013 data, the Tacoma Washington Alexander Avenue site and the Seattle Washington Duwamish site. Monitoring at these sites was discontinued in April 2012, so no further data is expected. The time period modeled was roughly 2007 to the April 2013, but varied by site depending on the monitoring history of the site.

To allow for maximum site-to-site comparability of results, the raw data from each monitor was prepared using a consistent data preparation methodology. Following that, each site was modeled independently but followed a consistent modeling protocol. Model output consists of a number of 'factors' (between 8 and 11) that explain the majority of the data variability and a time series of these factors' mass impacts at the monitor.

The main goal of conducting this updated source apportionment modeling was to compare the mass impacts of factors/sources over the most recent 2012/2013 winter period with previous winter periods so as to better understand the reasons for any changes in the total observed mass in the most recent winter period.

There were multiple other goals in conducting this analysis as well as multiple lines of further investigation. Some of these are as follows:

- Some of the urban monitoring locations are currently designated $PM_{2.5}$ nonattainment areas (Utah sites, Tacoma WA), so this analysis provides additional information for understanding sources of $PM_{2.5}$ in those areas.
- For those monitoring locations currently in attainment of the $PM_{2.5}$ standard, some are likely to be designated as nonattainment areas after EPA's next review of the $PM_{2.5}$ standards (e.g., Yakima WA). So this analysis gives a jump start to understanding sources of elevated $PM_{2.5}$ in those areas.
- Modeling multiple sites using a consistent data preparation methodology and modeling protocol allows for better site-to-site comparability. For example, wood smoke factors can be compared between urban sites to examine the relative importance of wood smoke at each location and possibly also the effect of wood smoke controls between one location and another.

- An analysis of the site-to-site differences in chemical composition for a given factor can help distinguish the nature of sources contributing to that factor at each location.

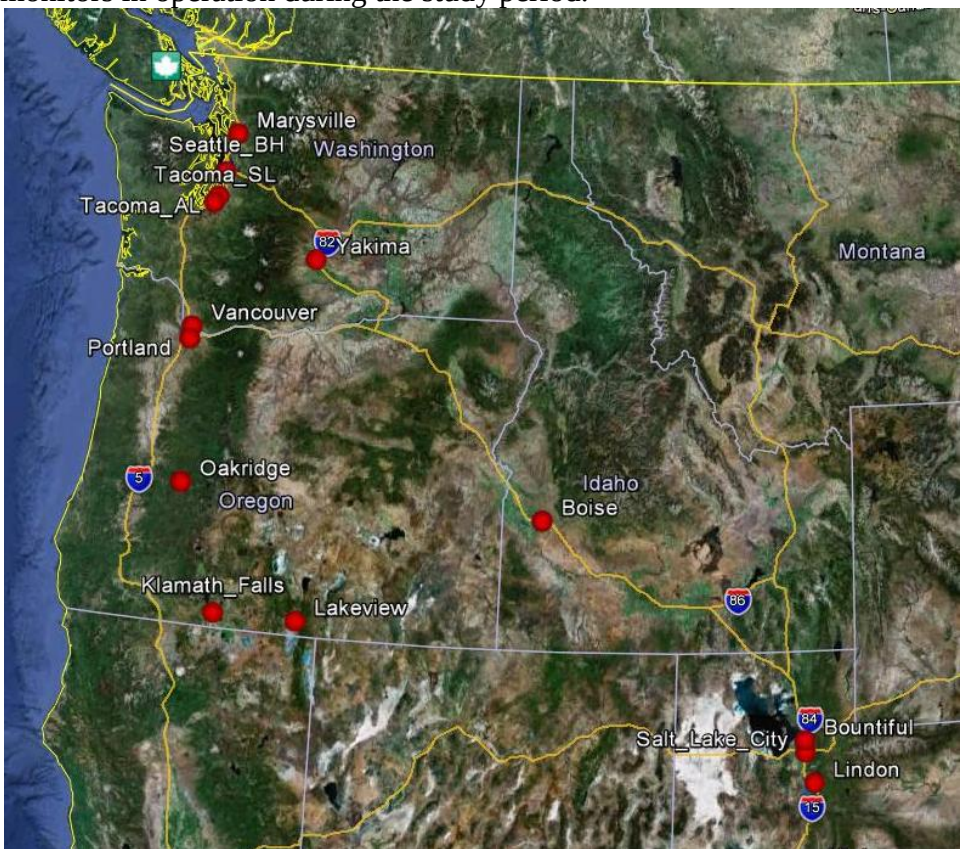
For this report, results have been aggregated into 2 time averages (winter and annual) and winter average contributions calculated for dates when total 24-hour $PM_{2.5}$ mass was greater than 12 ug/m3. Results are located in Tables 5.2 through 5.7 below. Most days with elevated $PM_{2.5}$ at the urban CSN sites occurred in winter. This is consistent with the general conceptual model that urban sites in the Pacific Northwest and Intermountain Northwest experience elevated $PM_{2.5}$ on wintertime days with air stagnation conditions, where primary $PM_{2.5}$ emissions can concentrate and/or wintertime nitrate aerosol formation is favored.

2. Monitoring Sites and Data.

2.1. Map of monitoring locations available for this analysis:

16 urban monitoring locations throughout the Pacific Northwest and Alaska were considered in this analysis. Figures 2.1 shows a map with the locations of 15 urban CSN monitors in the Pacific Northwest (the Fairbanks Alaska site is not shown). Of the 16 available sites, the three Oregon sites of Oakridge, Klamath Falls, and Lakeview were not modeled because data were not yet available for the 2012/2013 winter season at the time this analysis was performed. Table 2.1 provides more detailed monitoring site information for the modeled CSN monitors including; site identification information, the date range of data used, the number of samples used in this modeling analysis, and the latitude and longitude of the site.

Figure 2.1. Map of the Pacific Northwest showing the 15 Chemical Speciation Network (CSN) monitors in operation during the study period.



2.2. Metadata information:

Table 2.1. Site information for the CSN sites used in this modeling analysis.

Site Name	Date Range Modeled	Number of Measurements	AQS Number	State	County	Latitude	Longitude
Fairbanks	10/1/2009 - 4/7/2013	386	020900010	Alaska	Fairbanks North Star	64.840672	-147.722461
Boise	5/3/2007 - 4/7/2013	690	160010010	Idaho	Ada	43.600264	-116.347897
Portland	5/3/2007 - 4/7/2013	600	410510080	Oregon	Multnomah	45.49647386	-122.6034112
Bountiful	5/6/2007 - 4/4/2013	338	490110004	Utah	Davis	40.902967	-111.884467
Salt_Lake_City	5/9/2007 - 4/7/2013	637	490353006	Utah	Salt Lake	40.736389	-111.872222
Lindon	5/6/2007 - 4/4/2013	324	490494001	Utah	Utah	40.341389	-111.713611
Vancouver	4/7/2009 - 4/4/2013	235	530110013	Washington	Clark	45.648333	-122.586944
Seattle_DW (Duwamish)	11/8/2008 – 4/27/2012	198	530330057	Washington	King	47.5632	-122.3405
Seattle_BH (Beacon Hill)	5/3/2007 - 4/4/2013	525	530330080	Washington	King	47.568333	-122.308056

Tacoma_SL (South L St.)	5/12/2007 - 4/4/2013	302	530530029	Washington	Pierce	47.1864	-122.4517
Tacoma_AL (Alexander Ave.)	11/2/2008 - 4/27/2012	202	530530031	Washington	Pierce	47.2656	-122.3858
Marysville	4/7/2009 - 4/4/2013	225	530611007	Washington	Snohomish	48.054315	-122.171529
Yakima	11/8/2007 - 4/4/2013	257	530770009	Washington	Yakima	46.59678	-120.512215

- The source of CSN data was the EPA AQS database and data were downloaded on 7/16/2013. The data reports used were AMP350 raw data report and AMP503 sample blank report.
- The date range of data extracted for each site is shown in Table 2.1. The beginning of the date range was chosen to coincide with the installation of an updated carbon sampler (URG 3000N) at each location and the end date represents the latest data available at the time data were extracted.
- Final number of samples used in this modeling analysis at each site ranged from 198 to 690 and depended on sampling frequency, date of site installation, date of deactivation, and date of carbon sampling method change.
- Chemical species used in this modeling analysis depended on data quality at each monitoring location but generally included elements associated with soil such as Al, Fe, Ti, Ca, and Si, the elements associated with sea salt Na and Cl, major ions like NH4, NO3, and SO4, organic and elemental carbon species based on the thermal evolution method, and a range of other trace elements including Br, Cr, Cu, Pb, Ni, Mg, V, Zn, and K.

3. Data Preparation Methodology

The measured values, uncertainties, sample blank correction, and method detection limits for each chemical species were obtained and prepared as discussed in Kotchenruther (2013).

Other data processing choices included:

- To avoid double counting of chemical species, in the CSN datasets Na+, K+, S, and OP(TOT) were removed in favor of Na, K, $SO4^{2-}$, and OP(TOR) measurements. The chemical species that were retained were chosen based on either higher data completeness or higher S/N ratio. OP is reported by two measurement methods in the AQS database, TOT and TOR, here OP via TOR was retained to be consistent with the IMPROVE TOR measurement methodology.
- At all sites except for Utah, data from July 4 and 5 were removed to mitigate the influence of fireworks in the modeling result. For sites in Utah, a fireworks factor was obtained even after removing July 4 and 5 data, so for Utah July 4 and 5 data were retained to improve the fireworks factor identification.

4. Modeling

The PMF model version used in this analysis was EPA PMF v4.0 beta. The current publically available version at the time this modeling was conducted was v3.0

(http://www.epa.gov/heasd/research/pmf.html). EPA PMF v4.0 beta was used here because of its enhanced features related to setting model constraints. PMF modeling was conducted in two phases. The first phase, a 'baseline' model run, was performed to find the best solution using only model settings available in EPA PMF v3.0. Second, the results of the baseline solution were refined using modeling constraints available in EPA PMF v4.0 beta as discussed below.

User settings for the initial set of baseline model runs were as follows:

- $PM_{2.5}$ mass data was set as the 'Total Variable', which allows the model to estimate each factor contribution to total mass. Setting total $PM_{2.5}$ as the 'Total Variable' increases its uncertainty to down weight its influence in the model solution.
- For the base model run, 20 model runs were made to find the best least-squares minimum with the 'seed' variable set to 10 ('seed' determines where the model begins looking for a solution, setting a specific number rather than using the 'random' setting allows the user to reproduce the model results exactly).
- The optimum number of factors for each site was determined somewhat subjectively based on the interpretation of model results, but also from the quality of the least-squares fit (analysis of Q values) in the model output.

Upon inspection and analysis of the baseline model output for factor chemical composition and factor mass allocation, it was apparent that at many sites the factor representing gasoline vehicles was contaminated with mass from the factor representing aged wood smoke. This was most clearly identified in the results for Fairbanks and Boise where late summer wild fire impacts were identified through large 'spikes' in PM mass allocated to factors representing both fresh and aged wood smoke and PM mass for the factor representing gasoline vehicles also 'spiked' at these times. Less clear, but also troubling, was an observed pattern of gasoline vehicle factor mass increasing in winter along with fresh and aged wood smoke. However, there are plausible explanations as to why gasoline vehicle PM mass might increase in winter, so that is less conclusive. Both the factors identified as aged wood smoke and gasoline vehicles have large contributions from the organic carbon fractions OC1-OC4, so model difficulty separating these sources into their respective factors is understandable. Also, as additional evidence of aged wood smoke's contamination of the gasoline vehicle factor, the baseline factor chemical profile for gasoline vehicles in some cases showed elevated potassium (K) concentrations inconsistent with the low K concentrations reported in the EPA SPECIATE database for gasoline vehicles. K is a well known elemental tracer for wood smoke.

In order to better separate the factor representing gasoline vehicles from the factor representing aged wood smoke, the model constraint features of EPA PMF v4.0 beta were employed. Using the Fairbanks and Boise sites as test cases, the following model constraint settings allowed mass to shift from the factor representing gasoline vehicles to the factor representing aged wood smoke for those dates with clear late summer wild fire impacts.

Model constraint settings:

- The K allocation for the factor representing gasoline vehicles was forced to be zero,
- the K allocations for the factors representing both fresh and aged wood smoke were 'pulled up',
- and OC1-OC4 allocations for the factors representing aged wood smoke were 'pulled up'.

For most dates with clear summer wild fire impacts in Boise and Fairbanks, the above model constraints reduced the gasoline vehicles factor mass to be comparable to that when the site was not impacted by wild fires and increased the aged wood smoke factor mass by a comparable amount. Other factors were not significantly affected. When the above model constraints were applied to all sites, in many cases wintertime mass allocated to the gasoline vehicles factor was reduced, but usually remained at or above gasoline vehicles factor mass allocated at other times of the year. The mass allocated to the aged wood smoke factor increased a comparable amount and other factors were not significantly affected.

Despite the above model refinements, it is possible that there remains some level of gasoline vehicle contamination from aged wood smoke in the mass allocation results.

5. Modeling Results and Analysis

5.1. Resulting PMF Factors and Factor Classification

The PMF model uses a form of factor analysis where the underlying co-variability of many variables (e.g., sample to sample variation in particulate matter chemical species) is described by a smaller set of factors (e.g., particulate matter sources) to which the original variables are related. While PMF is often called a 'source apportionment' model, a more accurate description would be a 'factor apportionment' model. While this might seem to be an esoteric distinction, it is an important one to understand when viewing results in this report. PMF is a 'factor analysis' model, because it divides the full modeling dataset into a much simpler dataset that identifies the leading 'factors' causing day to day variability in the full dataset. A PMF factor could represent the impact on the monitor of a single source (e.g. industrial facility), a source category (e.g. a bunch of sources that have very similar chemical fingerprints and emissions patterns, so cars, trucks, wood burning sources could each be factors), or multiple sources or source categories grouped together (PMF may group multiple sources together because of insufficient variability in emissions, chemical composition, and/or temporal/spatial resolution).

The interpretation/classification of factors output by the PMF model is a subjective process. It can depend on the experience of the modeler, availability of current source profiles and literature references, availability of supplementary information about the airshed, and an understanding of the range of possible sources impacting the monitoring location.

It should be also noted that the factors determined in a PMF analysis are not necessarily mutually exclusive. For example, a factor identified as predominantly sulfate aerosol is possibly from a combination of multiple sources including diesel engines and industrial facilities, even though these sources may also have separately identified PMF factors. Hence, caution should be used in interpreting factor classifications too literally or with exclusivity.

The number of factors identified at each CSN monitor ranged between 8 and 11, with a total of 18 different factor types being identified between the 13 CSN sites modeled in this analysis. Tables 5.1 lists the names given to each factor type as well as a brief description of each factor's unique chemical characteristics. Tables 5.2 lists the number of factors found at each CSN

monitor, the multi-year annual average mass allocated to each factor by the PMF model, and the multi-year annual average $PM_{2.5}$ mass based on CSN observations.

Table 5.1. Factors found at Pacific Northwest CSN sites.

Factor Type Name	**Factor Description and Identifying Features**
Aged Wood Smoke, Organic Pyrolysis (OP) Rich	Factor mass dominated by the sum of OC fractions (often > 95% of mass). OC fractions are weighted to the higher temperature fractions (OC3, OC4, OP). OP is often the dominant mass of the OC fractions. This factor likely has a significant contribution from secondary organic aerosols. Higher factor masses that are coincident with late summer wood smoke factor impacts (likely wild fires) and wintertime wood smoke factor impacts (home heating) suggest this factor mostly represents aged wood smoke. OP rich factors have been previously identified by Hwang and Hopke, 2007; Kim et al., 2004; and Zhao and Hopke, 2006.
Ammonium Chloride	Factor mass dominated by the sum of Cl and NH_4.
Ammonium Nitrate	Factor mass dominated by the sum of NO_3 and NH_4.
Ammonium Sulfate	Factor mass dominated by the sum of SO_4 and NH_4.
Calcium Rich	Factor mass dominated by calcium and sulfate. Previous work has linked similar calcium rich factors to cement production (Kim et al., 2004; Shi et al., 2009).
Copper Rich	Factor mass dominated by Cu and EC. Factor possibly linked to metal processing industry (Amato and Hopke 2011).
Fireworks	Factor mass dominated by potassium and sulfate, with variable amounts of OC. Cu and Mg are trace metals but significant to factor identification. Factor identified by its chemical constituents and prevalence on days of the year known for significant fireworks activity.
Fugitive Dust	Factor mass dominated by the sum of Al, Ca, Fe, Ti, and Si.
Gasoline Vehicles	Factor mass dominated by the sum of OC2, OC3 and OC4 species with an approximate ratio of 1:2:1 between OC2:OC3:OC4. Usually no OC1 or OP component. EC component usually 15 – 20 % of total mass, and mostly EC1. Previous work has linked similar factors to gasoline vehicles (Hwang and Hopke, 2007; Kim et al., 2004; Maykut et al., 2003; Zhao and Hopke, 2004; Kim and Hopke, 2006; Kim and Hopke, 2008b; Zhao and Hopke, 2006).
Iron Rich	Fe is important in the identification of this factor with concentrations ranging between 5 - 50% of total factor mass. Factor also has variable amounts of OC, EC, SO_4, and/or NO_3. Factor identification is unclear but is thought likely to be diesel engine combustion. Previous work has linked similar Fe rich factors to a variety of sources including diesel vehicles, heavy-duty diesel, oil combustion, or industrial activity [Karanasiou et al., 2009 (Oil combustion); Kim et al., 2004 (Diesel); Maykut et al., 2003 (Diesel); Ramadan et al., 2000 (Heavy-duty diesel); Kim and Hopke, 2008a (Diesel); Kim and Hopke, 2008b (Metal processing); Lee et al., 2008 (Metal/Industrial processing); Wu et al., 2007 (Diesel); Zhao and Hopke, 2006 (Secondary Sulfate); Zhou et al., 2004 (Traffic); Amato and Hopke 2011 (Diesel); Kim et al., 2010 (Diesel); Lewis et al., 2003 (Diesel)].
Mixture, Ammonium Sulfate and Nitrate	Factor mass dominated by the sum of NO_3, SO_4, and NH_4.
Mixture, Iron Rich and Gasoline Vehicles	A roughly equal mixture of two other factors listed in this table, Iron Rich and Gasoline Vehicles.

Nitrate Rich	Factor mass dominated by NO_3.
Residual Fuel Oil Combustion	Factor mass dominated by SO4 and NH4 with Ni and V playing a significant role in factor identification at trace levels. Approximate ratio of V:Ni of 3:1. OC and EC also can contribute to factor mass. Previous work has linked similar factors to marine shipping fuel oil combustion due to Ni and V tracer species and high fuel sulfur content (Maykut et al., 2003; Kim and Hopke, 2008a; Kim and Hopke, 2008b).
Sea Salt	Factor mass dominated by the sum of Na and Cl.
Sulfate Rich	Factor mass dominated by SO_4.
Unidentified Urban	Factor dominated by a mixture of OC, EC, SO_4 and NO_3. EC2 is particularly important in the identification of this factor. Source identification is unclear, but it is likely that this factor is in some way connected to fuel combustion. Previous work has linked similar factors to a variety of sources including railroad traffic or diesel vehicles [Kim et al., 2004 (railroad traffic); Zhao and Hopke, 2004 (Diesel); Kim and Hopke, 2006 (secondary sulfate); Han et al., 2007 (Diesel)].
Wood Smoke	Factor dominated by the sum of OC and EC plus about 1% potassium. Usually OC dominated by the OC1 fraction, EC dominated by EC1, and no OP component. Majority of mass impacts in winter, but some sites have significant late summer impacts. EC can range between 10 – 30% of factor. Previous work has linked similar factors to wood smoke (Hwang and Hopke, 2007; Kim et al., 2004; Maykut et al., 2003; Kim and Hopke, 2008b; Zhao and Hopke, 2006).

Table 5.2. Number and type of PMF factors found, annual average $PM_{2.5}$ factor mass (ug/m3), and annual average total $PM_{2.5}$* mass (ug/m3) at each CSN site**.

Site	Number of Factors	Wood Smoke	Aged Wood Smoke (OP Rich)	Ammonium Nitrate	Nitrate Rich	Ammonium Sulfate	Sulfate Rich	Residual Fuel Oil Combustion	Mixture, Ammonium Sulfate and Nitrate	Ammonium Chloride	Gasoline Vehicles	Iron Rich	Mixture, Iron Rich and Gasoline Vehicles	Unidentified Urban	Sea Salt	Fugitive Dust	Calcium Rich	Copper Rich	Fireworks	$PM_{2.5}$ Mass
Fairbanks	9	2.2	3.1		0.5	1.3	1.8				1.4			0.4	0.2	0.4				11.6
Boise	8	0.8	1.8	1.3	0.5	0.8					0.9			0.2		0.9				7.4
Portland	8	1.8	1.3		1.0	0.9	1.1				1.5				0.2	0.4				8.4
Bountiful	11	0.6	1.4	2.2		0.6				0.3	0.8	0.1		0.3		0.6	0.5		0.2	7.8
Salt Lake City	10	0.6	1.3	2.6		0.8				0.3	1.1	0.6		0.1		1.3			0.2	9.1
Lindon	11	0.5	1.1	2.9		1.0				0.2	1.0	0.2	0.2	0.3		1.0			0.2	8.9
Vancouver	9	1.2	1.0		1.2		1.1		0.6		1.1			0.0	0.2	0.5				7.2
Seattle_DW	9	1.1	0.5	0.6			1.3	0.9			1.2	1.1			0.2		0.7			7.9
Seattle_BH	11	0.5	0.6		0.6	0.9		1.0			1.3	0.6		0.2	0.2	0.2		0.1		6.4
Tacoma_SL	9	2.3	1.1	1.2			0.9	0.7			1.2			0.5	0.3	0.5				9.0
Tacoma_AL	9	1.5	0.6	0.8				0.9			1.5	0.3		1.1	0.4	0.6				8.1
Marysville	8	2.4	0.8		1.2	0.5		1.3			1.0				0.3	0.6				8.5
Yakima	9	1.3	2.4	1.4	0.4	0.6					1.5			0.6	0.3	0.7				9.6

*$PM_{2.5}$ mass based on CSN monitoring data, not based on FRM data.
**Sum of factor masses may not equal total $PM_{2.5}$ mass because the PMF model does not necessarily attribute all measured mass to factors.

Because most days with elevated $PM_{2.5}$ occur in the winter months of November, December, January, and February, the average data from these months was computed. Table 5.3 shows the average $PM_{2.5}$ and PMF results only from winter month sample days when total $PM_{2.5}$ was elevated above 12 ug/m3. A focus was made in this report on days with elevated $PM_{2.5}$ because many wood smoke controls only come into effect when $PM_{2.5}$ is elevated (e.g., burn bans) and wood smoke is a dominant pollutant in the Region. A 12 ug/m3 threshold for elevated $PM_{2.5}$ was chosen because $PM_{2.5}$ on average days at all sites was less than this and the value is low enough to allow for statistics to be computed with sufficient sample size on elevated days. 12 ug/m3 is also the threshold of the new annual $PM_{2.5}$ standard.

Table 5.3. Winter* average $PM_{2.5}$ and average PMF model factor mass (ug/m3) when total $PM_{2.5}$ mass was greater than or equal to 12 ug/m^3. All available winter sample days from 2007 to most recent available data were used in these averages.

Site	Number of Samples	Wood Smoke	Aged Wood Smoke (OP Rich)	Ammonium Nitrate	Nitrate Rich	Ammonium Sulfate	Sulfate Rich	Residual Fuel Oil Combustion	Mixture, Ammonium Sulfate and Nitrate	Ammonium Chloride	Gasoline Vehicles	Iron Rich	Mixture, Iron Rich and Gasoline Vehicles	Unidentified Urban	Sea Salt	Fugitive Dust	Calcium Rich	Copper Rich	Fireworks	$PM_{2.5}$ Mass
Fairbanks	103	6.9	7.3		1.1	3.7	3.7				2.0			0.3	0.4	0.2				26.5
Boise	52	3.2	3.2	9.9	1.9	3.2					1.3			0.1		0.3				24.3
Portland	85	7.1	2.9		2.4	1.7	0.5				2.8			0.0	0.2	0.6				19.6
Bountiful	38	2.3	1.8	12.0		1.3				1.8	1.7	0.2		0.1		0.3	0.2		0.4	23.2
Salt Lake City	100	2.4	2.2	12.4		1.8				1.6	2.1	1.4		0.0		0.8			0.3	25.8
Lindon	41	2.1	1.4	15.4		1.7				1.4	1.4	0.5	0.7	0.1		0.3			0.1	25.9
Vancouver	24	6.3	3.6		3.3		0.4		1.5		2.0			0.0	0.2	0.5				18.9
Seattle_DW	21	4.7	1.5	1.7			1.2	0.9			1.8	3.3			0.2		0.9			17.6
Seattle_BH	15	3.0	1.5		2.1	2.2		0.3			3.5	1.8		0.3	0.2	0.3		0.1		15.9
Tacoma_SL	38	10.0	4.0	2.8			0.6	1.4			2.2			0.6	0.4	0.6				24.1
Tacoma_AL	23	7.6	2.5	2.4				1.3			2.5	0.7		1.1	0.4	0.7				19.8
Marysville	31	11.6	1.7		2.5	0.6		0.5			1.5				0.4	0.9				20.4
Yakima	73	4.3	5.1	4.8	0.9	0.9					2.6			0.5	0.8	0.6				21.1

*Winter months are defined as November, December, January and February.

One of the objectives of this analysis is to compare the most recent winter (2012/2013) factor impacts on elevated $PM_{2.5}$ days with that during previous winters. Table 5.4 shows the averages for a subset of the data used to make Table 5.3, excluding data from November 2012 through February 2013. Table 5.5 shows the averages for the remaining data, including only data from November 2012 through February 2013.

Table 5.4. Winter* average $PM_{2.5}$ and PMF model factor mass (ug/m3) when total $PM_{2.5}$ mass was greater than or equal to 12 ug/m^3. All available sample days from 2007 through February 2012.

Site	Number of Samples	Wood Smoke	Aged Wood Smoke (OP Rich)	Ammonium Nitrate	Nitrate Rich	Ammonium Sulfate	Sulfate Rich	Residual Fuel Oil Combustion	Mixture, Ammonium Sulfate and Nitrate	Ammonium Chloride	Gasoline Vehicles	Iron Rich	Mixture, Iron Rich and Gasoline Vehicles	Unidentified Urban	Sea Salt	Fugitive Dust	Calcium Rich	Copper Rich	Fireworks	$PM_{2.5}$ Mass
Fairbanks	72	7.0	7.5		1.2	3.9	3.0				1.9			0.3	0.5	0.2				26.7
Boise	43	2.9	3.0	6.9	1.6	1.7					1.4			0.1		0.3				18.6
Portland	72	7.2	3.1		2.3	1.8	0.5				2.7			0.0	0.3	0.6				19.8
Bountiful	32	2.1	2.0	11.9		1.2				1.6	1.5	0.2		0.1		0.4	0.2		0.4	22.6
Salt Lake City	89	2.4	2.2	12.2		1.5				1.2	2.0	1.4		0.0		0.9			0.3	24.9
Lindon	35	1.9	1.3	13.4		1.5				1.0	1.4	0.5	0.8	0.2		0.4			0.2	23.4
Vancouver	18	6.8	3.8		3.0		0.3		1.4		2.4			0.0	0.2	0.6				19.2
Seattle_DW	21	4.7	1.5	1.7			1.2	0.9			1.8	3.3			0.2		0.9			17.6
Seattle_BH	13	3.0	1.5		2.0	2.3		0.3			3.7	1.9		0.4	0.2	0.3		0.1		15.9
Tacoma_SL	33	10.5	4.5	2.8			0.5	1.6			2.0			0.6	0.4	0.7				24.9
Tacoma_AL	23	7.6	2.5	2.4				1.3			2.5	0.7		1.1	0.4	0.7				19.8
Marysville	26	11.5	1.9		2.4	0.7		0.5			1.4				0.4	0.9				20.4
Yakima	63	4.4	5.3	5.1	0.6	0.9					2.4			0.5	0.9	0.6				21.4

*Winter months are defined as November, December, January and February.

Table 5.5. Winter* average $PM_{2.5}$ and PMF model factor mass (ug/m3) when total $PM_{2.5}$ mass was greater than or equal to 12 ug/m^3. All available sample days from November 2012 through February 2013.

Site	Number of Samples	Wood Smoke	Aged Wood Smoke (OP Rich)	Ammonium Nitrate	Nitrate Rich	Ammonium Sulfate	Sulfate Rich	Residual Fuel Oil Combustion	Mixture, Ammonium Sulfate and Nitrate	Ammonium Chloride	Gasoline Vehicles	Iron Rich	Mixture, Iron Rich and Gasoline Vehicles	Unidentified Urban	Sea Salt	Fugitive Dust	Calcium Rich	Copper Rich	Fireworks	$PM_{2.5}$ Mass
Fairbanks	31	6.4	6.6		0.9	3.3	5.4				2.1			0.1	0.3	0.2				26.1
Boise	9	4.8	4.2	24.6	3.5	10.4					0.7			0.1		0.3				51.1
Portland	13	6.3	2.0		2.6	1.5	0.6				3.5			0.0	0.2	0.4				18.5
Bountiful	6	3.0	0.7	12.6		1.7				2.9	2.4	0.2		0.1		0.1	0.2		0.1	26.5
Salt Lake City	11	2.9	1.8	14.2		4.0				4.9	2.3	1.4		0.0		0.4			0.2	32.7
Lindon	6	3.0	2.4	26.8		2.8				3.3	1.3	0.5	0.4	0.0		0.2			0.0	40.6
Vancouver	6	4.8	2.8		4.0		0.7		2.0		1.1			0.0	0.2	0.2				18.2
Seattle_DW	NA																			
Seattle_BH	2	2.7	1.6		2.3	1.7		0.2			2.2	1.1		0.2	0.1	0.2		0.0		16.1
Tacoma_SL	5	6.4	0.6	2.6			0.6	0.6			3.5			0.5	0.5	0.3				19.0
Tacoma_AL	NA																			
Marysville	5	12.1	0.8		3.3	0.2		0.7			2.1				0.3	0.7				20.0
Yakima	10	3.7	4.0	3.0	2.7	0.6					3.7			0.5	0.5	0.4				19.5

*Winter months are defined as November, December, January and February.

Statistical hypothesis testing can be used to determine if there is a statistically significant difference between the most recent 2012/2013 winter's data and that from previous winters. This type of hypothesis testing attempts to disprove a 'null hypothesis', which in this case is that the two sets of data are sampled from an environment that is the same over the whole multiyear period. If the null hypothesis is disproven, in other words if a statistically significant difference is found between the 2012/2013 winter and previous winters, then there are a range of possible explanations. The two most obvious explanations would be that either emissions in the airshed have significantly changed or that there was a significant difference in seasonal meteorology (outside the range of meteorological conditions that occurred during the previous observations), which then affected observations.

The appropriate choice of hypothesis test depends on how the data is statistically distributed. Most hypothesis tests are divided into those applicable for normally distributed data and those applicable for data with non-normal distributions (nonparametric tests). Many environmentally sampled datasets are not normally distributed. A Shapiro Wilk test for normally distributed data performed on $PM_{2.5}$ values and factor mass allocations in this study confirmed that the data used

here is not normally distributed. The nonparametric hypothesis test used for this analysis was the Wilcoxon-Mann-Whitney test, which is a nonparametric test available in EPA's ProUCL software version 4.1.01 (http://www.epa.gov/osp/hstl/tsc/software.htm). A two-sided test was performed on the underlying data that make up the averages presented in Tables 5.4 and 5.5. As per the software guidance, to insure sufficient sample size the test was only performed when each data subset had at least 5 samples. This meant that the test was not performed for three sites, the Seattle Duwamish and Tacoma Alexander Ave sites, which had no data for the 2012/2013 winter period and the Seattle Beacon Hill site, which had only 2 CSN samples with total $PM_{2.5}$ greater than 12 ug/m3 for the 2012/2013 winter period. Table 5.6 shows the p-value result of the Wilcoxon-Mann-Whitney test, with values displayed in red where the two datasets were significantly different at or above the 95% confidence interval. For ease of reference, the difference in averages between data in Tables 5.5 and Table 5.4 are displayed in Table 5.7 (statistically significant changes in red, as per Table 5.6).

Table 5.6. Wilcoxon-Mann-Whitney significance test results (p-values) comparing elevated $PM_{2.5}$ data and PMF factor results* from the 2012/2013 winter (Table 5.5) with all available elevated $PM_{2.5}$ data and PMF factor results from previous years (Table 5.4). Statistically significant results at the 95% confidence interval are highlighted in red.

Site	Wood Smoke	Aged Wood Smoke (OP Rich)	Ammonium Nitrate	Nitrate Rich	Ammonium Sulfate	Sulfate Rich	Residual Fuel Oil Combustion	Mixture, Ammonium Sulfate and Nitrate	Ammonium Chloride	Gasoline Vehicles	Iron Rich	Mixture, Iron Rich and Gasoline Vehicles	Unidentified Urban	Sea Salt	Fugitive Dust	Calcium Rich	Copper Rich	Fireworks	$PM_{2.5}$ Mass
Fairbanks	0.47	0.37		0.55	0.32	**0.00**				0.66			**0.00**	0.58	0.33				0.89
Boise	**0.03**	0.09	0.12	**0.00**	**0.00**					**0.02**			0.13		0.40				**0.01**
Portland	0.79	0.20		0.59	0.14	0.40				**0.05**				0.44	0.07				0.66
Bountiful	**0.04**	0.06	0.59		0.51				**0.04**	0.16	0.89		0.10		**0.04**	0.80		0.72	0.14
Salt Lake City	0.09	0.66	0.31		0.19				**0.01**	0.73	0.65		0.55		0.12			0.76	0.13
Lindon	0.16	0.05	0.08		0.19				**0.01**	0.50	0.96	0.62	**0.02**		0.19			0.07	0.15
Vancouver	0.15	0.48		0.06		0.06		0.22		**0.04**			0.57	0.67	0.19				0.71
Seattle_DW																			
Seattle_BH																			
Tacoma_SL	0.34	**0.04**	0.86			0.55	**0.04**			**0.05**			0.44	0.80	0.23				0.41
Tacoma_AL																			
Marysville	0.77	0.13		0.07	**0.04**		0.44			0.14				0.69	0.50				0.81
Yakima	0.23	0.23	**0.04**	**0.00**	0.13					0.10			0.69	0.11	0.15				0.34

*Only from $PM_{2.5}$ samples greater than 12 ug/m3.

Table 5.7. The change in average elevated $PM_{2.5}$* (ug/m3) for the 2012/2013 winter period compared to previous winter periods (Table 5.5 data minus Table 5.4 data). Statistically significant changes in red as per Table 5.6.

Site	Wood Smoke	Aged Wood Smoke (OP Rich)	Ammonium Nitrate	Nitrate Rich	Ammonium Sulfate	Sulfate Rich	Residual Fuel Oil Combustion	Mixture, Ammonium Sulfate and Nitrate	Ammonium Chloride	Gasoline Vehicles	Iron Rich	Mixture, Iron Rich and Gasoline Vehicles	Unidentified Urban	Sea Salt	Fugitive Dust	Calcium Rich	Copper Rich	Fireworks	$PM_{2.5}$ Mass
Fairbanks	-0.6	-0.9		-0.2	-0.5	**2.3**				0.3			**-0.2**	-0.1	0.3				-0.7
Boise	**1.9**	1.2	17.7	**1.9**	**8.7**					**-0.7**			-0.5		-0.5				**32.4**
Portland	-0.8	-1.8		0.3	-0.3	0.9				**0.8**				-0.9	-0.2				-1.3
Bountiful	**0.9**	-1.3	0.7		0.5				**1.3**	0.9	0.9		-0.6		**-0.2**	-0.3		-0.3	3.8
Salt Lake City	0.6	-0.4	1.9		2.5				**3.6**	0.2	0.1		-0.8		-0.6			-0.2	7.8
Lindon	1.5	1.2	13.3		1.3				**2.3**	-0.6	0.5	-0.4	**-0.2**		-0.2			-0.1	17.2
Vancouver	-2.0	-1.0		1.8		0.4		0.6		**-1.3**			0.2	-0.4	-0.4				-0.9
Seattle_DW																			
Seattle_BH																			
Tacoma_SL	-4.8	**-3.9**	-0.2			0.8	**-1.0**			**1.5**			-0.1	0.1	-0.4				-5.9
Tacoma_AL																			
Marysville	0.6	-1.5		0.9	**-0.5**		0.2			0.7				-0.7	-0.2				-0.4
Yakima	-0.8	-1.3	**-2.1**	**2.1**	-0.4					1.3			-0.2	-0.4	-0.3				-1.9

*Average of samples greater than 12 ug/m3 $PM_{2.5}$ mass only.

5.2. Discussion of Significance Testing Results

5.2.1. Results for Boise

The only site where the average instances of elevated $PM_{2.5}$ (mass greater than 12 ug/m3) showed a significant difference between the 2012/2013 winter and previous winters was in Boise, where elevated $PM_{2.5}$ increased by 32.4 ug/m3 in the 2012/2013 winter period over previous winters. Based on the PMF factor results, this increase was largely from a significant increase in an ammonium sulfate factor (+8.7 ug/m3 between the two periods) and to a lesser extent from statistically significant increases in a wood smoke factor (+1.9 ug/m3) and a nitrate rich factor (+1.9 ug/m3). The ammonium nitrate factor also increased by 17.7 ug/m3, but was not statistically significant at the 95% confidence interval (significant at the 88% confidence interval). A meteorological analysis is beyond the current scope of this report, but anecdotal evidence suggest that the intermountain northwest had stronger and more frequent wintertime cold pool stagnation events in the 2012/2013 winter than in other recent winter periods. If accurate, this could at least partially explain the statistically significant increases.

5.2.2. Results for the Wasatch Front monitors; Bountiful, Salt Lake City, and Lindon

As discussed for the Boise site, anecdotal evidence suggest that the intermountain northwest had stronger and more frequent wintertime cold pool stagnation events in the 2012/2013 winter than in other recent winter periods. While not statistically significant at the 95% confidence interval, average instances of elevated $PM_{2.5}$ increased by 3.8, 7.8 and 17.2 ug/m3 at Bountiful, Salt Lake City, and Lindon sites, respectively. The three PMF factors representing secondary $PM_{2.5}$ (ammonium nitrate, ammonium sulfate, and ammonium chloride) all showed increases at each of the three monitoring sites for the 2012/2013 winter period. However, only the ammonium chloride factor increases were significant at the 95% confidence interval. Meteorological conditions play a significant role in ammonium chloride formation (Kelly et al., 2013). However, the source(s) of chlorine contributing to this factor are still uncertain in this airshed, so investigations into chlorine sources as well as meteorology would be needed further explain the significant increase PMF ammonium chloride factor mass.

5.2.3. Results for Tacoma Washington South L Street

Average instances of elevated total $PM_{2.5}$ mass (mass greater than 12 ug/m3) and most average PMF factor masses decreased in the 2012/2013 winter period over that of previous winter periods. However, statistically significant decreases occurred in only the aged wood smoke factor (-3.9 ug/m3) and residual fuel oil combustion factor (-1.0 ug/m3), while the gasoline vehicle factor showed a statistically significant increase (+1.5 ug/m3). The factor representing fresh wood smoke decreased by 4.8 ug/m3, but was not found to be statistically significant. There are several plausible explanations as to why decreases in the aged wood smoke factor would be more statistically significant than that of fresh wood smoke. Fewer prolonged (multi-day) stagnation events in the 2012/2013 winter period would lead to less aged wood smoke. Alternately, less wood combustion overall and better wood burning practices (more complete combustions) could lead to less semi-volatile organic carbon in the atmosphere, and it is possible that oxidation/condensation of this semi-volatile organic carbon contributes to the aged wood smoke factor.

5.2.4. Results for Yakima Washington

A statistically significant increases in PMF factor mass occurred for the nitrate rich factor (+2.1 ug/m3), but was offset by a statistically significant decrease in PMF factor mass for the ammonium nitrate factor (-2.1 ug/m3). This redistribution of nitrate mass between factors may represent a significant difference in the availability of ammonia in the airshed for the 2012/2013 winter compared to that of previous winters.

6.0 References

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