Source apportionment of PM$_{2.5}$ at multiple Northwest U.S. sites: Assessing regional winter wood smoke impacts from residential wood combustion

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HIGHLIGHTS

- Source apportionment of PM$_{2.5}$ was performed on 19 Northwest U.S. monitoring sites.
- Common sources of PM$_{2.5}$ were identified across multiple sites.
- Winter wood smoke PM$_{2.5}$ impacts from residential wood combustion were quantified.
- Residential wood combustion contributions to PM$_{2.5}$ spanned a wide range.
- Source apportionment at multiple sites has advantages over single site analysis.

ARTICLE INFO

ABSTRACT

Wood smoke from residential wood combustion is a significant source of elevated PM$_{2.5}$ in many communities across the Northwest U.S. Accurate representation of residential wood combustion in source-oriented regional scale air quality models is challenging because of multiple uncertainties. As an alternative to source-oriented source apportionment, this work provides, through receptor-oriented source apportionment, an assessment of winter residential wood combustion impacts at multiple Northwest U.S. locations. Source apportionment was performed on chemically speciated PM$_{2.5}$ from 19 monitoring sites using the Positive Matrix Factorization (PMF) receptor model. Each site was modeled independently, but a common data preparation and modeling protocol was used so that results were as comparable as possible across sites. Model solutions had from 4 to 8 PMF factors, depending on the site. PMF factors at each site were associated with a source classification (e.g., primary wood smoke), a dominant chemical composition (e.g., ammonium nitrate), or were some mixture. 15 different sources or chemical compositions were identified as contributing to PM$_{2.5}$ across the 19 sites. The 6 most common were; aged wood smoke and secondary organic carbon, motor vehicles, primary wood smoke, ammonium nitrate, ammonium sulfate, and fugitive dust. Wood smoke was identified at every site, with both aged and primary wood smoke identified at most sites. Wood smoke contributions to PM$_{2.5}$ were averaged for the two winter months of December and January, the months when wood smoke in the Northwest U.S. is mainly from residential wood combustion. The total contribution of residential wood combustion, that from primary plus aged smoke, ranged from 11.4% to 92.7% of average December and January PM$_{2.5}$ depending on the site, with the highest percent contributions occurring in smaller towns that have fewer expected sources of winter PM$_{2.5}$. Receptor modeling at multiple sites, such as that conducted in this work, provided some significant advantages over modeling a single or small number of sites. Analysis at multiple sites allowed common factor chemical compositions to be identified, making it easier to evaluate when a PMF factor at a particular site represents a mix of sources versus a single source. The identification of similar PMF factors across multiple sites also allowed average chemical profiles to be established for the 6 the most commonly identified PM$_{2.5}$ sources or compositions in this study. These average profiles have the potential to be used as source profile inputs in future Chemical Mass Balance receptor modeling, when a limited number of samples may restrict the ability to conduct PMF receptor modeling, or when the availability of local source profiles is limited. Receptor modeling results spanning a range of community sizes and source compositions, as in this study, could be used to

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evaluate and improve the representation of wood smoke and other specific sources in source-oriented regional scale air quality models by providing an independent source impact assessment.

1. Introduction

Wood smoke is a common source of ambient $PM_{2.5}$ (particles with aerodynamic diameter <2.5 μm) worldwide. In developing countries, wood burning has widespread use as a fuel for cooking and heating. In more developed countries like the U.S., wood burning is most often used as a source of supplemental home heating and for esthetic purposes, but can also be a primary source of home heating. Human exposure to $PM_{2.5}$ has been linked to cardiovascular and pulmonary disease (Künzli et al., 2005), and lung cancer and premature mortality (Pope and Dockery, 2006). Wood burning, in addition to being a source of $PM_{2.5}$, is also a source of carcinogenic organic compounds such as benzene and formaldehyde, and respiratory irritants like phenols and acetaldehyde (Naeher et al., 2007). Recently, Noonan et al. (2015) have suggested that the number of vulnerable people in the U.S. exposed to residential wood smoke has been significantly underestimated. In the Northwest U.S., exceedances of the 24-h National Ambient Air Quality Standards for $PM_{2.5}$ occur most often in winter. In communities ranging from small mountain towns to large metropolitan areas, wood smoke from residential wood combustion frequently contributes a significant fraction of wintertime $PM_{2.5}$ (Jeong et al., 2008; Kim and Hopke, 2008; Ward and Lange, 2010; Wang and Hopke, 2014). Identifying the proportional contribution of wood smoke, and other sources, to wintertime $PM_{2.5}$ is a key step to developing targeted and cost effective $PM_{2.5}$ reduction strategies.

Regional scale efforts to assess source impacts to ambient $PM_{2.5}$ are often addressed in the U.S. using source apportionment tools within source-oriented photochemical grid models like CMAQ and CAMx. These models predict source impacts from emissions inventories, emissions modeling, meteorological simulations, and chemical transport modeling (Wagstrom et al., 2008). Source-oriented models also have the benefit of being able to explore the impact of emissions control scenarios on predicted $PM_{2.5}$. However, evaluating the contribution of residential wood combustion to observed $PM_{2.5}$ with these models can be challenging for a number of reasons. Grid models can be overly dispersive under the low wind speed conditions that often lead to high winter $PM_{2.5}$ (Holtslag et al., 2013) and these models can have difficulty replicating multiday wintertime temperature inversions and air stagnation episodes (Baker et al., 2011). For small mountain valley towns with high residential wood combustion impacts, even the finest horizontal grid resolutions that are typically used can be too course. Also, developing accurate residential wood combustion emissions inventories can be challenging because of the large variety of wood burning devices in use, difficulties in obtaining an accurate count and spatial representation of each device type, and differences between standard wood burning device emissions tests in a laboratory and emissions from these devices when they are used in the real world.

While source-oriented source apportionment methods have their challenges, receptor-based methods also have their limitations. Results in receptor-based source apportionment studies can be dependent on the chemical species measured, quality and amount of measured data, choice of chemical source markers to identify sources, and the QA/QC modeling protocol used. Despite these limitations, this work demonstrates that a regional assessment of $PM_{2.5}$ using receptor-based source apportionment methods like Positive Matrix Factorization (PMF) can provide a complementary approach to source-oriented techniques and could be used as an independent means of evaluating them.

While there are numerous published receptor-based source apportionment studies, most published studies report results for only a few monitoring sites, cover differing time periods, and use differing data preparation and modeling protocols. These differences make it hard to compare results between studies or use them to compile a regional assessment. Recently, there have been several regional assessments published using receptor-based methods focusing on marine vessel impacts in the western U.S. (Kotchenruther, 2013, 2015). This work uses a similar approach as Kotchenruther (2013, 2015) to assess regional impacts of winter wood smoke from residential wood combustion in the Northwest U.S. Source apportionment is performed using the PMF model on chemically speciated $PM_{2.5}$ from 19 sites. As in the previous work, the approach taken is to model each site independently and to treat data from all sites with a common data preparation and modeling protocol. The benefits of this approach are that results between sites are as comparable as possible since site-to-site data and modeling have undergone the same treatments. An additional benefit of receptor modeling at multiple sites is that common factors across sites can be identified, making it easier to evaluate when a PMF factor at a particular site represents a mix of sources versus a single source.

2. Methods

2.1. Chemically speciated $PM_{2.5}$ data

The Chemical Speciation Network (CSN) is one of several urban and suburban monitoring networks funded by the U.S. Environmental Protection Agency (EPA) and operated by state and local agencies. CSN monitors collect 24-h integrated $PM_{2.5}$ mass on filters that are sent to a laboratory for chemical analyses. Laboratory analyses includes quantification of total $PM_{2.5}$ mass, elemental composition by energy dispersive X-ray fluorescence, organic and elemental carbon (OC, EC) by thermal evolution in 8 temperature fractions, and anions and cations by ion chromatography. Detailed information about the CSN network is provided by Solomon et al. (2014). CSN monitors are typically operated on a daily, every third day, or every sixth day schedule depending on the site. Quality assured CSN data are housed in EPA's Air Quality System (AQS) database.

Monitoring sites analyzed in this work are listed in Table 1 and depicted in Fig. 1. From 2007 to 2009 EPA conducted a systematic replacement of all CSN carbon samplers to match those of the IMPROVE program (a chemically speciated $PM_{2.5}$ monitoring network of mostly rural and remote sites) and switched to IMPROVE-based carbon analytical measurement protocols (U.S. EPA, 2009). Consequently, EC and OC data before and after the change are not easily comparable in the CSN network. For this reason, the start date for data used from each site in this study is

Table 1
CSN monitoring sites analyzed in this study.

City	State	Data start date	Data end date	Number of samples	EPA AQS number	Latitude	Longitude
Fairbanks	AK	10/1/2009	9/29/2014	573	20900010	64.8407	−147.7225
Fresno	CA	1/1/2012	9/29/2014	304	60190011	36.7853	−119.7742
Bakersfield	CA	1/2/2010	9/7/2013	310	60290014	35.3561	−119.0412
Sacramento	CA	1/2/2010	9/29/2014	565	60670006	38.6138	−121.3680
Boise	ID	5/3/2007	9/26/2014	851	160010010	43.6003	−116.3479
Klamath Falls	OR	7/12/2009	6/28/2014	180	410350004	42.1889	−121.7225
Lakeview	OR	10/16/2009	8/3/2014	177	410370001	42.1889	−120.3519
Oakridge	OR	7/6/2009	9/30/2012	177	410392013	43.7444	−122.4805
Portland	OR	5/3/2007	9/29/2014	736	410510080	45.4965	−122.6034
Bountiful	UT	5/6/2007	9/26/2014	417	490110014	40.9030	−111.8845
Salt Lake City	UT	5/9/2007	9/29/2014	777	490353006	40.7364	−111.8722
Lindon	UT	5/6/2007	9/26/2014	404	490494001	40.3414	−111.7136
Vancouver	WA	4/7/2009	8/26/2013	259	530110013	45.6483	−122.5869
Seattle (Duwamish)	WA	11/8/2008	4/27/2012	198	530330057	47.5632	−122.3405
Seattle (Beacon Hill)	WA	5/3/2007	9/29/2014	678	530330080	47.5683	−122.3081
Tacoma (South L St.)	WA	5/12/2007	9/26/2014	376	530530029	47.1864	−122.4517
Tacoma (Alexander Ave.)	WA	11/2/2008	4/27/2012	203	530530031	47.2656	−122.3858
Marysville	WA	4/7/2009	9/26/2014	305	530611007	48.0543	−122.1715
Yakima	WA	11/8/2007	9/26/2014	335	530770009	46.5968	−120.5122

Fig. 1. CSN monitoring sites analyzed in this study.

based on when it converted to IMPROVE-based carbon sampling methods, if the site was in existence during the transition. The end date for data represents what was available in AQS at the time data were extracted. All sites were in operation for over two years and collected from 177 to 851 24-h samples.

2.2. Data preparation and treatment

A detailed discussion of CSN data preparation and treatment is provided in a previous publication (Kotchenruther, 2013) and briefly summarized here. Prior to source apportionment analyses the data were processed to correct for field blanks. Chemical species were omitted in PMF modeling if more than 40% of samples had missing data. Missing values were replaced with median concentrations and the uncertainty set to a very high value compared to measured data, typically four times the species median concentration, to minimize the influence of the replaced data on the model solution. Any negative concentrations were reset to zero. The uncertainty of each measurement was estimated based on the measured analytical uncertainty plus 1/3 of the method detection limit. The signal-to-noise (S/N) ratio was also used to evaluate whether chemical species should be included in the PMF modeling, and was used to adjust the data uncertainty. Chemical species were omitted in PMF modeling if the S/N ratio was less than 0.2. For chemical species with S/N between 0.2 and 1.0, data uncertainties were multiplied by a factor of 3 to down-weight the influence of these species in the model solution. For chemical species measured by both elemental and ion analyses, such as sodium (Na) and potassium (K), Na ion and elemental K were used because these species had better S/N ratios, and elemental Na and K ion were not used to avoid double counting. In addition to these treatments, sulfate was retained in the dataset and non-sulfate sulfur (NSS) was calculated by subtracting the sulfur component of measured sulfate from the measured sulfur concentration. Also, the reported lowest temperature fraction of EC, EC1, is actually the sum of pyrolyzed organic carbon (OP) and low temperature combusting EC. Hence, EC1 was recalculated as EC1-OP and the measured OP value was used, so as not to double count measured OP.

2.3. Source apportionment

Source apportionment modeling was performed using EPA PMF 5.0 (http://www.epa.gov/heasd/research/pmf.html). A discussion of the mathematical equations underlying EPA PMF can be found in Paatero and Hopke (2003) and Norris et al. (2014). Data from each monitoring site was modeled independently. In each case, the model was run in the robust mode with 20 repeat runs to insure the

model least-squares solution represented a global rather than local minimum and the rotational F_{PEAK} variable was held at the default value of 0.0. The model solution with the optimum number of factors was determined somewhat subjectively and was based on inspection of the factors in each solution, the quality of the least-squares fit (analysis of Q_{Robust} and Q_{True} values), and the results from three error estimation methods available in PMF 5.0; bootstrapping (BS), displacement (DISP), and bootstrapping with displacement (BS-DISP) (Norris et al., 2014; Paatero et al., 2014). The scaled residuals for final model solutions were generally normally distributed, falling into the recommended range of $+3$ to -3.

PMF factors can represent a single source or source category (e.g., cement manufacture, wood burning), a chemical composition (e.g., ammonium nitrate, sea salt), or mixtures of sources and compositions. During modeling of each of the 19 sites in this work, it was sometimes the case that the number of factors that appeared to present the best delineation of sources and compositions, were in fact shown to have too much solution instability after analysis with DISP, BS, and BS-DISP (e.g., factor swaps; Brown et al., 2015). In these cases, reducing the number of factors often led to improved solution stability, but also caused some factors to combine and become mixtures of sources, or sources and compositions. Preference in PMF solutions was given to the number of factors with improved solution stability, even if that lead to reduced source delineation. Further information on how the model solution with the optimal number of factors was selected is provided in the Supplemental Materials.

3. Results and discussion

3.1. Identified $PM_{2.5}$ sources and compositions

The number of PMF factors ranged from 4 to 8, depending on the site. Table 2 lists the 15 different sources and compositions identified over all time periods at the 19 sites, and how often they were identified in a factor by themselves versus in a factor mixed with other sources or compositions. A table in the Supplemental Materials lists each site, the number of factors found, and the factor attributions using the source or composition identifiers listed in Table 2. All PMF factor mass impacts and factor chemical profiles for each site are also provided in the Supplementary Materials. The chemical profiles presented are those after the factor chemical composition from each site was normalized. A factor chemical composition was normalized by assuming an organic mass (OMC)

to OC ratio of 1.4 (i.e., multiplying all OC fraction by 1.4), summing all of the measured chemical constituents using the assumed OMC instead of OC, and dividing each chemical component by the summed constituents. Additionally, for factors associated with fugitive dust, a metal oxide to metal ratio was assumed for aluminum (Al, ratio of 2.2), calcium (Ca, 1.63), iron (Fe, 2.42), titanium (Ti, 1.94), and silicon (Si, 2.49) based on the ratios used in the IMPROVE network (Solomon et al., 2014).

The sources and compositions listed in Table 2 were identified by comparing the chemical composition of PMF factors with chemical profiles in EPA's SPECIATE database of source emissions test data (https://www3.epa.gov/ttnchie1/software/speciate/), comparison with similar PMF factor chemical compositions identified in existing published studies, knowledge of existing sources in the airsheds and their seasonal emissions patterns, and composition of aerosols found in the natural environment (e.g., fugitive dust, sea salt). The sections below describe how each source or composition was identified, and for those most commonly found, a figure is provided depicting the average PMF factor chemical profile from those factors that were determined not to be a mixture. Data tables for the average profiles are provided in the Supplemental Materials. The average PMF factor chemical profile was taken after the PMF factor chemical profile from each site was normalized as described above.

3.1.1. PMF factors associated with aged wood smoke and secondary OC

These factors were dominated by OC and EC, with higher temperature OC fractions more abundant than that found in Primary Wood Smoke (see section 3.1.3) and almost none of the lowest temperature OC1 fraction. K was a significant trace constituent, but not chlorine (Cl). The average chemical profile from PMF factors at 11 sites where this source was not mixed with other sources is shown in Fig. 2a.

The seasonal pattern of mass impacts depended on the site. Two illustrative examples are provided here, Fig. 3 shows the time series of mass impacts for this factor in Fairbanks, AK and Fig. 4 for Lindon, UT. These figures also show the time series of mass impacts for the PMF factor associated with primary wood smoke at these sites. At sites like Fairbanks, AK, with significant winter primary wood smoke impacts, the aged wood smoke and secondary OC factor had both elevated winter impacts and high summer impacts on those years corresponding to high wildfire activity. Summers with low wildfire activity had small, but not zero, summer impacts. At sites

Table 2
Sources and chemical compositions identified and the number of sites where appearing as a single PMF factor, or in a factor mixed with other listed sources or chemical compositions.

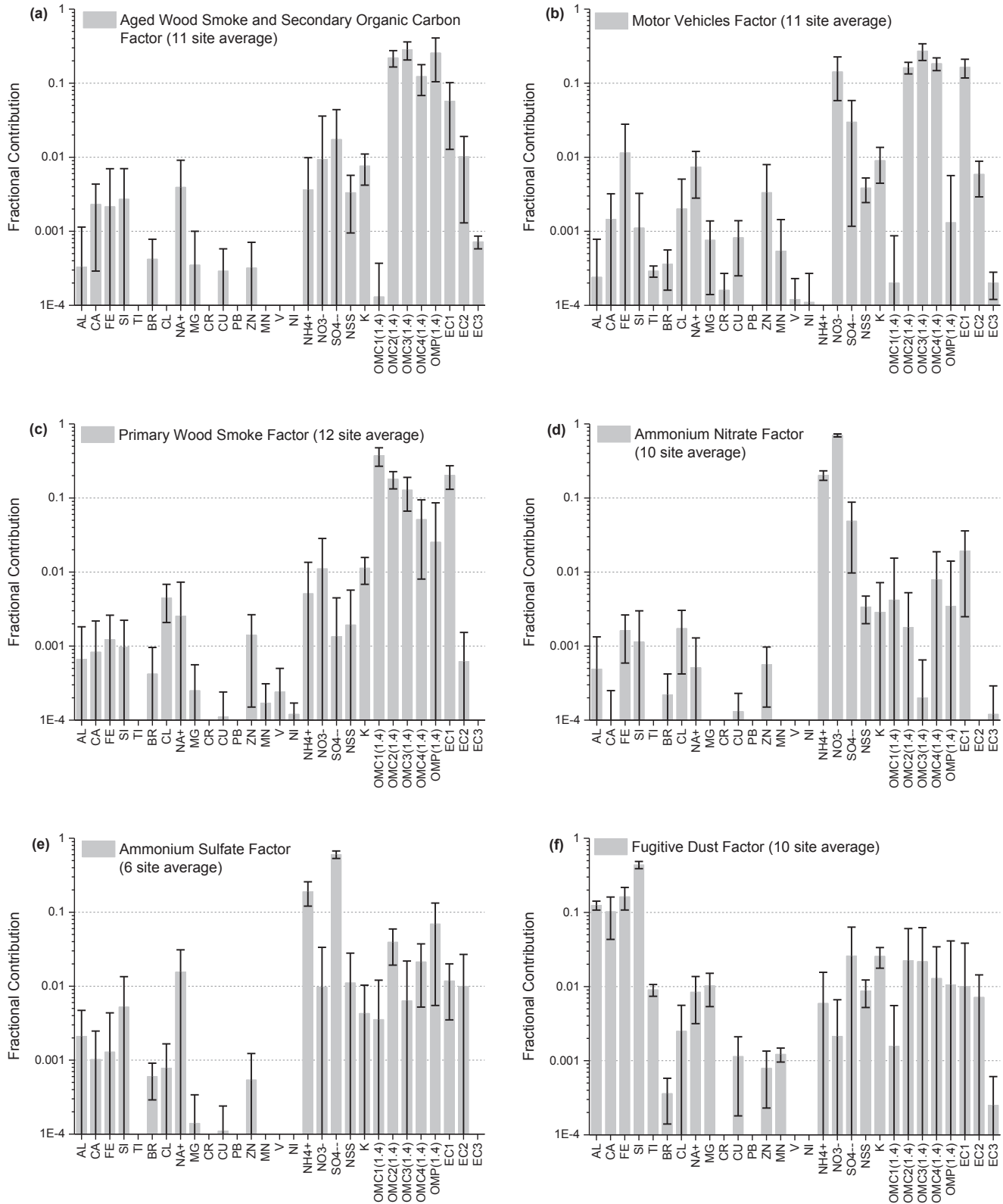| Source or composition identifier	Identified source or composition	Number of sites where appears	Number of sites where a single factor	Number of sites where in a mixed factor
1	Aged Wood Smoke and Secondary Organic Carbon	19	11	8
2	Motor Vehicles	18	11	7
3	Primary Wood Smoke	17	12	5
4	Ammonium Nitrate	16	10	6
5	Ammonium Sulfate	16	6	10
6	Fugitive Dust	16	10	6
7	Sea Salt	7	6	1
8	Sulfate Rich	6	1	5
9	Iron Rich	4	2	2
10	Aged Sea Salt	4	3	1
11	Undetermined	4	0	4
12	Elemental Carbon and Sulfate Rich	3	0	3
13	Residual Fuel Oil Combustion	3	2	1
14	Nitrate Rich	2	0	2
15	Cement Kiln	1	1	0

Fig. 2. Average and standard deviation of chemical profiles from PMF factors from multiple sites associated with (a) aged wood smoke and secondary OC, (b) motor vehicles, (c) primary word smoke, (d) ammonium nitrate, (e) ammonium sulfate, and (f) fugitive dust.

like Lindon, UT, where primary wood smoke plays a relatively minor role in winter $PM_{2.5}$, the aged wood smoke and secondary OC

factor had high mass impacts in summer months on those years corresponding to high wildfire activity, with low impacts at most
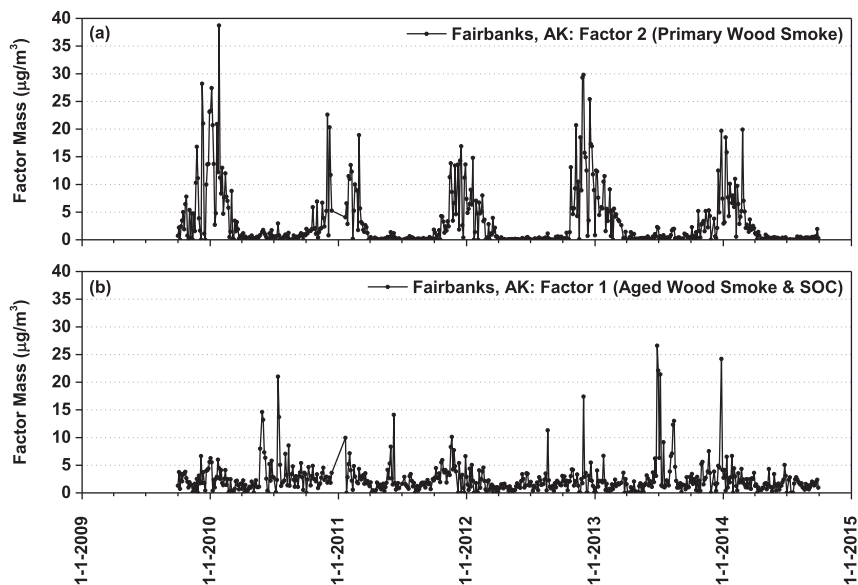
Fig. 3. Time series of PM$_{2.5}$ mass impacts in Fairbanks, AK for PMF factors associated with (a) primary wood smoke and (b) aged wood smoke and secondary OC.
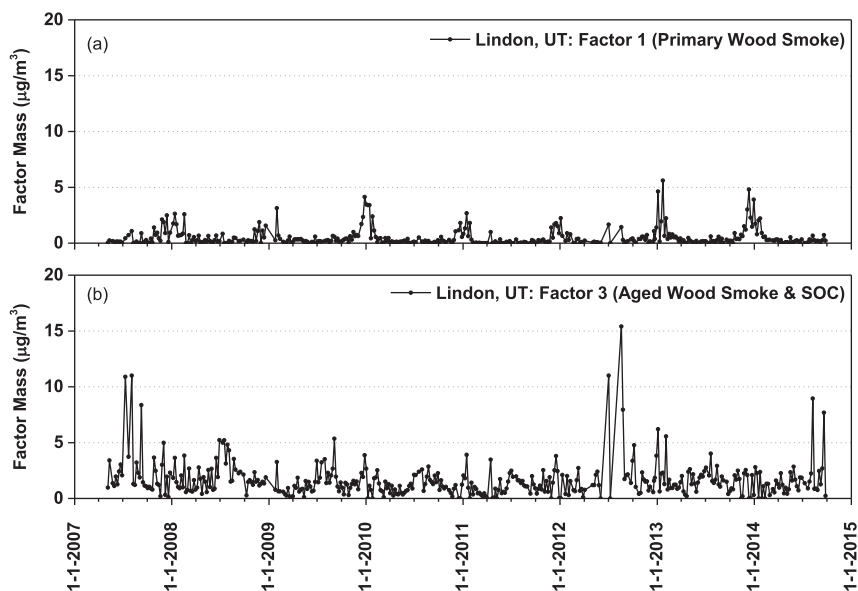

Fig. 4. Time series of PM$_{2.5}$ mass impacts in Lindon, UT for PMF factors associated with (a) primary wood smoke and (b) aged wood smoke and secondary OC.

other times. Yearly totals of state wildfire acres burned from 2007 to 2014 were obtained from the National Interagency Fire Center (https://www.nifc.gov/fireInfo/fireInfo_statistics.html). Yearly wildfire data for all states with monitoring sites in this study is provided in the Supplementary Materials. Further pointing to the impact of wildfire in this factor, the ratio of char-EC to soot-EC (measured (EC1-OP)/(EC2+EC3) in the CSN datasets) in the average profile was 5.2, much lower than the ratio found in the average profile for primary wood smoke (ratio = 283). This lower char-EC to soot-EC ratio is consistent with differences found by Han et al. (2010) between forest fire emissions and biomass combustion for home heating.

The rational for associating this factor with wood smoke comes from the correspondence of elevated summer impacts with high wildfire activity, the correspondence of elevated winter impacts with those areas also having significant winter Primary Wood Smoke impacts, the dominance of OC and EC in the chemical profile, and the presence of the wood smoke marker K. The determination that the wood smoke is aged comes from the observation that OC fractions in this factors' profile are shifted to higher temperature fractions compared with primary wood smoke, which is consistent with oxidative aging of organic carbon. Also, the OC to EC ratio in the average profile was 9.3, higher than that from Primary Wood Smoke (2.7) and the K to OC ratio, 0.012, was lower than that from Primary Wood Smoke (0.021), both of which are consistent with organic gases from wood fires undergo gas to particle conversion and adding organic mass to the aerosol during aging. The absence of Cl in the chemical profile, compared to that in Primary Wood Smoke, is also an indication of aging, similar to the Cl replacement chemistry that occurs when sea salt aerosol ages (Adachi and Buseck, 2015). Lastly, the rational for also associating this factor with secondary organic carbon comes from the small

elevation in summertime mass at most sites, even during years with low wildfire activity.

3.1.2. PMF factors associated with motor vehicles

The principal chemical constituents in this factor were EC1, OC2, OC3, OC4, and nitrate. Significant trace constituents were zinc (Zn), copper (Cu), and Fe. The average chemical profile from PMF factors at 11 sites where this source was not mixed with other sources is shown in Fig. 2b. The dominant chemical constituents are similar to those found for motor vehicles in previous publications (Zhao and Hopke, 2004; Kim and Hopke, 2006; Hwang and Hopke, 2007). The significant trace metal constituents match those commonly found in PM$_{2.5}$ associated with motor vehicles (Song and Gao, 2011; Pant and Harrison, 2013). The near ubiquity of this source at the sites in this study matches the conceptual understanding of motor vehicles as a common source of particulate pollution in urban areas. Separate factors for gasoline and diesel vehicles were not found in this study, and this factor likely represents a combination of these sources.

3.1.3. PMF factors associated with primary wood smoke

This factor was dominated by OC and EC, with the lower temperature OC and EC fractions having the highest abundance. K and Cl were significant trace constituents. The seasonal pattern of mass impacts showed high winter and low summer impacts. The average chemical profile from PMF factors at 12 sites where this source was not mixed with other sources is shown in Fig. 2c. The K to OC ratio in the average profile, 0.021, was consistent with K to OC ratios in PM$_{2.5}$ from Northwestern U.S. forest biomass combustion (Munchak et al., 2011). The OC to EC ratio was 2.7. The pattern of OC and EC temperature fractions in the average profile and the presence of K and Cl are similar to that found in many wood smoke profiles in EPA's SPECIATE database. Examples of similar profiles include SPECIATE profile 3921 representing PM$_{2.5}$ from pine wood burning in a fireplace and profile 3937 from oak burning in a woodstove.

3.1.4. PMF factors associated with ammonium nitrate

The main chemical constituents in this factor were ammonium and nitrate. The typical seasonal pattern of mass impacts showed high winter and very low summer impacts, which is indicative of secondary formation, and likely from multiple sources of NO$_x$ and ammonia. The average chemical profile from PMF factors at 10 sites where this source was not mixed with other sources is shown in Fig. 2d. The fractional contribution of nitrate and ammonium in the average profile was 0.70 and 0.20, respectively, which is the ratio expected when nitrate is fully neutralized by ammonium.

3.1.5. PMF factors associated with ammonium sulfate

The main chemical constituents in this factor were ammonium and sulfate. The typical seasonal pattern of mass impacts showed higher summer impacts, but also some sites like Fairbanks, AK with higher winter impacts. This factor likely arises from multiple sources of SO$_2$ and ammonia. The average chemical profile from PMF factors at 6 sites where this source was not mixed with other sources is shown in Fig. 2e. The fractional contribution of sulfate and ammonium in the average profile was 0.60 and 0.19, respectively, which demonstrates near full neutralization of sulfate by ammonium.

3.1.6. PMF factors associated with fugitive dust

The principal chemical constituents in this factor were Al, Ca, Fe, and Si. Significant trace constituents were Ti and K. The typical seasonal pattern of mass impacts is higher in late summer and lower in winter and spring and corresponds to the typical seasons

with less and more precipitation, respectively. The average chemical profile from PMF factors at 10 sites where this source was not mixed with other sources is shown in Fig. 2f. The fractional contributions of the principal and trace chemical constituents in the average profile is similar to that of numerous soil dust profiles in EPA's SPECIATE database.

3.1.7. PMF factors associated with sea salt

This factor was dominated by Na and Cl. Significant trace constituents were magnesium (Mg) and Ca. Mass impacts for this factor had no discernable seasonal pattern and was only found at cities near salt water bodies. The site locations and lack of seasonal pattern suggest these factors are from natural sources rather than winter road salting.

3.1.8. PMF factors identified as Sulfate Rich

The main identifying feature of this factor was a significant presence of sulfate in the absence of ammonium. This most often occurred when sulfate was mixed with another factor that typically did not contain sulfate (e.g., mixed with fugitive dust in results for Vancouver, WA). The source of the sulfate is likely multiple sources of SO$_2$.

3.1.9. PMF factors identified as iron rich

This factor was dominated by Fe. Significant trace constituents were chromium, Cu, Zn, manganese, and Ni. This factor was only found in the large cities of Seattle, Portland, and Tacoma. It is likely this factor is related to metal fabrication or other industrial activity.

3.1.10. PMF factors associated with aged sea salt

This factor had the same identifying features as Sea Salt, but with little or no Cl and the addition of a significant contribution from nitrate. The replacement of Cl with nitrate is typical of sea salt after aging (Adachi and Buseck, 2015).

3.1.11. PMF factors identified as undetermined

This classification was given to factors that contained a significant amount of organic mass that could not otherwise be identified. 4 of the 19 sites had factors like this.

3.1.12. PMF factors identified as elemental carbon and Sulfate Rich

The main identifying feature of this factor was a significant presence of sulfate and the EC2 fraction. While always appearing as mixed with other sources in this study, previous studies have linked elevated EC2 and sulfate to fuel combustion sources (Kim et al., 2004; Han et al., 2010).

3.1.13. PMF factors associated with residual fuel oil combustion

The main chemical constituents in this factor were ammonium and sulfate. Significant trace constituents were vanadium (V) and Ni. The V to Ni ratio for this factor was close to 3, typical of residual fuel oil combustion (Kotchenruther, 2015). Because this factor was found only in the major port cities of Seattle and Tacoma, the main source is likely marine vessels.

3.1.14. PMF factors identified as nitrate rich

The main identifying feature of this factor was a significant presence of nitrate in the absence of ammonium. The source of the nitrate is likely multiple sources NO$_x$.

3.1.15. PMF factors associated with cement kiln emissions

The main chemical constituents in this factor were sulfate and Ca, similar to high Ca and sulfate profiles for cement kilns in EPA's SPECIATE database. There is a cement plant in the vicinity of the Seattle monitor where this factor was identified.

3.2. Winter residential wood combustion impacts

For the winter months of December and January, there is typically no wild or prescribed fire activity that would impact the urban monitors in this study. Therefore it is assumed here that both the Primary Wood Smoke and Aged Wood Smoke and Secondary OC factors for these months come predominantly from residential wood combustion. Table 3 lists the December and January two-month average mass and percent contribution for each wood smoke factor, when not mixed with other sources or compositions. For those sites where both factors were identified and quantified, total average wood smoke impacts are calculated as the sum of these two factors. At the Bakersfield site the two wood smoke factors were mixed with each other, therefore, while separate wood smoke factors are not quantified, the total average wood smoke impact is indicated. Average winter and annual PM$_{2.5}$ results for all sites and factors are provided in the Supplementary Materials.

Table 3 and Fig. 5 show the average winter residential wood combustion results found in this study. Available results encompass small towns to large cities and show a wide range of total wood smoke percent contributions to average winter PM$_{2.5}$, from 11.4% in Bakersfield, CA to 92.7% in Lakeview, OR. The highest winter wood smoke percent contributions occur in small towns where, in addition to residential wood combustion, there are fewer potential sources of primary PM$_{2.5}$ compared to larger urban areas. It would be difficult to make a consistent link between town size and winter percent contribution of residential wood combustion because secondary inorganic PM$_{2.5}$ (e.g., ammonium nitrate) in some areas contributes significantly to total winter PM$_{2.5}$.

The winter wood smoke results presented in this study could be used for performance evaluation and improvement of source-oriented models. For example, exploring the effect of uncertainties in meteorological modeling, emissions inventories, and the effects of grid resolution on wood smoke predictions over the wide range of city sizes and complexity of PM$_{2.5}$ sources presented here.
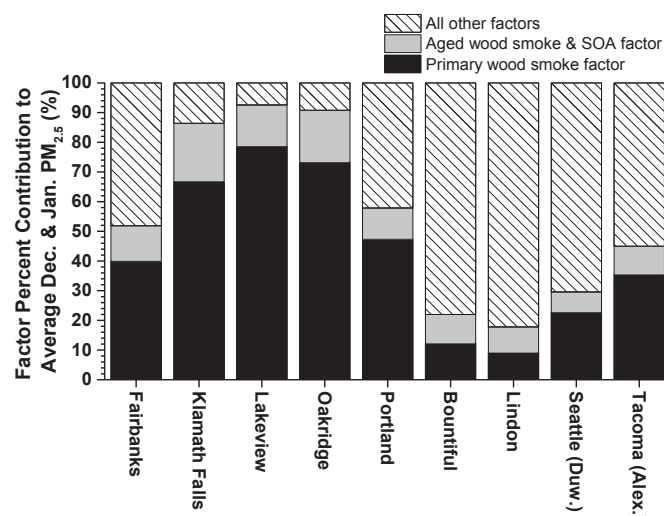

Fig. 5. Factor percent contribution to average December and January PM$_{2.5}$ for sites where two wood smoke factors were identified.

3.3. Comparison of residential wood combustion impacts with emissions inventories and previous studies

Every three years EPA, working with States and Tribes, develops a comprehensive and detailed estimate of air pollution emissions and publishes them in a National Emissions Inventory (NEI, https://www.epa.gov/air-emissions-inventories). The 2011 NEI provides annual estimates of PM$_{2.5}$ and PM$_{2.5}$ precursor emissions from numerous source categories at both state and county levels and includes residential wood combustion. To compare PMF results with the NEI, annual average PMF results for residential wood combustion were computed for 2011 for those sites in Table 3 where total primary + aged wood smoke impacts could be computed. A table is provided in the Supplemental Materials

Table 3
December and January average wood smoke PM$_{2.5}$ mass and percent of total PM$_{2.5}$, from PMF factor results.

Monitor location	Primary wood smoke factor mass ($\mu g/m^3$)	Primary wood smoke factor mass (%)	Aged wood smoke & SOA factor mass ($\mu g/m^3$)	Aged wood smoke & SOA factor mass (%)	Total primary + aged wood smoke ($\mu g/m^3$)	Total primary + aged wood smoke (%)
Fairbanks	9.8	39.9	3.0	12.0	12.8	51.8
Fresno	Not found	Not found	Mixed[c]	Mixed[c]		
Bakersfield	Mixed[a]	Mixed[a]	Mixed[d]	Mixed[d]	3.5	11.4
Sacramento	Mixed[b]	Mixed[b]	Mixed[c]	Mixed[c]		
Boise	Not found	Not found	Mixed[b]	Mixed[b]		
Klamath Falls	15.6	66.7	4.6	19.7	20.2	86.4
Lakeview	19.0	78.5	3.4	14.1	22.4	92.7
Oakridge	12.9	73.1	3.1	17.7	16.0	90.7
Portland	5.8	47.3	1.3	10.6	7.1	57.9
Bountiful	1.8	12.1	1.5	9.9	3.3	22.0
Salt Lake City	Mixed[b]	Mixed[b]	Mixed[c]	Mixed[c]		
Lindon	1.6	9.0	1.6	8.8	3.3	17.8
Vancouver	Mixed[b]	Mixed[b]	0.9	7.6		
Seattle (Duwamish)	2.3	22.6	0.7	7.0	3.0	29.5
Seattle (Beacon Hill)	2.0	30.4	Mixed[e]	Mixed[e]		
Tacoma (South L St.)	8.1	59.6	Mixed[f]	Mixed[f]		
Tacoma (Alexander Ave.)	3.9	35.3	1.1	9.7	5.0	45.0
Marysville	Mixed[b]	Mixed[b]	0.8	6.5		
Yakima	5.4	31.7	Mixed[c]	Mixed[c]		

[a] Mixed with Aged Wood Smoke & SOA.
[b] Mixed with Motor Vehicles.
[c] Mixed with Ammonium Sulfate.
[d] Mixed with Primary Wood Smoke.
[e] Mixed with Residual Fuel Oil Combustion.
[f] Mixed with Sulfate Rich.

containing the county level 2011 NEI emissions data for residential wood combustion, for those counties containing the monitoring sites in this study, and the 2011 PMF results for residential wood combustion. Percent contribution from residential wood combustion in the NEI and PMF data compared poorly ($r^2 = 0.0$). This is not surprising given the many factors influencing source contributions at a monitor, including the relative contribution of primary versus secondary $PM_{2.5}$ at a location and differences between the composition of county level emissions sources and those in the local airshed impacting the monitor. Counties in the Western U.S. are typically much larger than the size of the airsheds impacting the monitors in this study.

The winter wood smoke impacts presented in this work are somewhat higher, but are generally consistent with, those of other recent published studies conducted for monitoring sites in the western northern hemisphere.

Kim and Hopke (2008) found that annual average wood smoke impacts at 5 sites in the Seattle area ranged from 7% to 31% of total $PM_{2.5}$. These results are generally lower than those found in this work for Seattle (29.5%), but the Kim and Hopke results are an annual average, which may dilute the impact of winter wood smoke compared to this study.

Wang and Hopke (2014) found that winter wood smoke contributed 40% to total $PM_{2.5}$ in Fairbanks Alaska, which is also lower than the results found in this work for Fairbanks (51.8%). However, the Wang and Hopke results represent a 6-month winter average, whereas the results in this work are only for the highest impacted months of December and January.

Jeong et al. (2008) found that wood smoke contributed 74% to winter $PM_{2.5}$ in the small community of Golden, British Columbia, Canada and Ward and Lange (2010) analyzed $PM_{2.5}$ in five western Montana valley communities and determined that winter wood smoke contributed between 56% and 77% of total $PM_{2.5}$. These results are also lower than those found in this work for the smaller Oregon communities of Klamath Falls, Lakeview, and Oakridge (86.4%–92.7%). As with the Fairbanks results, this may represent the difference in temporal averaging in this work compared to the previous studies, but also may also represent different source compositions in these communities.

4. Conclusions

The source apportionment analyses reported in this work, at multiple monitoring sites, allowed for improved PMF factor identification over that of receptor modeling at a single site or a small number of sites. By comparing chemically similar PMF factors found at multiple monitoring sites, factors representing a single source or chemical composition could be delineated from those constituting some mixture. From the 19 monitoring sites analyzed in this work, PMF receptor model solutions had from 4 to 8 factors, depending on the site, and a total of 15 different sources or chemical compositions were identified as contributing to $PM_{2.5}$ across the 19 sites. The 6 most commonly identified sources or chemical compositions making up PMF factors were; aged wood smoke and secondary OC, motor vehicles, primary wood smoke, ammonium nitrate, ammonium sulfate, and fugitive dust. For these 6, average chemical profiles were established based on PMF factors at sites where the factor was determined not to be from a mixture. These average profiles could be useful as source profile inputs for Chemical Mass Balance receptor modeling, and also helpful with factor identification in other PMF modeling studies.

$PM_{2.5}$ from wood smoke was identified at every site, with both primary and aged wood smoke identified at most sites. The contribution of residential wood combustion to average winter $PM_{2.5}$ was calculated by summing the average contributions of

primary and aged wood smoke during December and January, months when other wood smoke sources such as wild and prescribed fire are minimal. 10 of the 19 sites analyzed had PMF results where primary and aged wood smoke were in factors well delineated from other sources or chemical compositions. At these 10 sites, the average December and January contribution of residential wood combustion to $PM_{2.5}$ ranged from 11.4% to 92.7% depending on the site, with the highest percent contributions occurring in smaller towns that have fewer expected sources of winter primary $PM_{2.5}$. The breadth of these results, spanning a wide range of community sizes and source compositions, could be useful to improve evaluations of source-oriented regional scale air quality models, where the impacts of grid size and emissions inventory quality on model performance are typical concerns.

Appendix A. Supplementary data

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