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Recent changes in winter PM_{2.5} contributions from wood smoke, motor vehicles, and other sources in the Northwest U.S.

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Abstract

In the Northwest U.S. elevated measurements of PM_{2.5} from anthropogenic sources occur most often in winter. Major contributors to winter PM_{2.5} are direct primary emissions of wood smoke from residential wood combustion, primary emissions from motor vehicles, gaseous NO_x emissions leading to particulate nitrate, and primary and secondary sources of particulate sulfate. A number of communities in the Northwest U.S. now have long data records of chemically speciated PM_{2.5} from which receptor-based source apportionment can be performed. This work uses receptor-based source apportionment on data from these monitoring sites to evaluate changes in the major contributors to winter PM_{2.5} over the available monitoring time span. Data from 9 sites are analyzed in this work using the Positive Matrix Factorization (PMF) source apportionment model. Each site was modeled individually rather than grouping the data from multiple sites. All sites had data through the summer of 2018, with most sites having 11 years of data and one site having 9 years of data. The number of PMF factors identified was between 5 to 10, depending on the site. Associations were made between PMF factors and PM_{2.5} sources based on comparison of PMF factor chemical profiles with published source test data and source profiles identified in other published studies. The most common factors found were: fresh wood smoke, aged wood smoke, soil dust, gas engines, mixed – gas engines and nitrate, ammonium sulfate, and ammonium nitrate. In this work, total wood smoke was identified as the combined contribution of fresh and aged wood smoke, and winter season data was defined as encompassing the last two months of a year and the first two months of the next year. To evaluate changes over time, average winter season PM_{2.5} measurements, major PM_{2.5} chemical components, and PMF factor results for the winter seasons of 2007 – 2009 were compared with the winter seasons of 2015 – 2017. The result for total 3-year average winter season PM_{2.5} was a decrease between 2% and 29% at the 9 sites, and the decreases were statistically significant at 3 sites. However, total winter season wood smoke contributions to PM_{2.5} decreased at every site between the two 3-year periods and the decreases were statistically significant at 8 of 9 sites, with decreases from 48% to 74% at those 8

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Declaration of competing interest

The author declares that he has no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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sites. All PMF factors associated with ammonium nitrate (identified at 5 of 9 sites) decreased a statistically significant 11% to 54% between the two 3-year winter season periods. All PMF factors associated with ammonium sulfate (identified at 7 of 9 sites) decreased a statistically significant 27% to 81% between the two 3-year winter season periods. In contrast to the significant reductions in PM_{2.5} from PMF factors related to wood smoke, ammonium nitrate and ammonium sulfate, PMF factors associated with gas engines increased from 6% to 226% between the two 3-year winter season periods. Increases in PM_{2.5} contributions from gas engine related factors explain why overall average winter season PM_{2.5} had more modest percent reductions compared to the percent reductions for wood smoke, ammonium nitrate, and ammonium sulfate factors between the two 3-year winter season periods.

Keywords

Positive matrix factorization; Residential wood combustion; Mobile sources; Source apportionment; PM_{2.5}

1. Introduction

In the Northwest U.S. elevated values of 24-hour average PM_{2.5} (particles with aerodynamic diameter <2.5 µm) from anthropogenic sources occur most often in winter. In this region of the U.S., 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), with percent contributions to total PM_{2.5} spanning from 11 – 93% depending on the community (Kotchenruther, 2016). Other common contributors to wintertime PM_{2.5} include primary emissions from motor vehicles, secondary ammonium nitrate, and secondary ammonium sulfate (Kotchenruther, 2016). 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 particular, 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.

The link between PM_{2.5} exposure and adverse health outcomes has led national, state and local governments in the U.S. in recent decades to target regulations that reduce emissions from directly emitting sources of PM_{2.5} as well as sources of gaseous precursors to PM_{2.5}, such as NO_x (the sum of NO and NO₂) and SO₂. Alongside these efforts to control PM_{2.5}, some communities now have multi-year monitoring records of chemically speciated PM_{2.5} that can be used to conduct source apportionment analysis. Results from such a source apportionment analysis can then be used to look at changes in source impacts over time and gauge the effectiveness of controlling PM_{2.5} on a source by source basis. A specific focus of this work is to evaluate the effectiveness of efforts to reduce winter wood smoke from residential wood combustion, since this is a significant source of direct PM_{2.5} pollution in

many communities in the Northwest U.S. and can be directly mitigated through local control measures. Additionally, winter wood smoke from residential wood combustion has for several decades been targeted for emissions reductions on the national, state, and local level because of its contribution to observed $PM_{2.5}$.

Previous work by this author has demonstrated that winter wood smoke from residential wood combustion can be identified and quantified using receptor-based source apportionment techniques, and that fresh wood combustion emissions can be identified separately from wood smoke that has undergone some degree of atmospheric aging (Kotchenruther, 2016). In a separate study, Kotchenruther (2017) demonstrated that source apportionment using ambient data can be used to evaluate the effects of regional emissions reduction efforts on $PM_{2.5}$. In the 2017 study, receptor-based source apportionment was conducted on multiple coastal $PM_{2.5}$ monitoring sites to quantify ambient $PM_{2.5}$ reductions from implementation of fuel sulfur regulations on commercial marine vessels.

This work builds on those previous publications by analyzing chemically speciated $PM_{2.5}$ from 9 urban locations in the Northwest U.S. that have long data records, conducts source apportionment on those datasets, identifies the $PM_{2.5}$ contributions from winter residential wood combustion as well as other major contributors to $PM_{2.5}$ such as motor vehicles, secondary nitrate and sulfate, and assesses the changes in these $PM_{2.5}$ sources in the context of ongoing $PM_{2.5}$ and $PM_{2.5}$ precursor emissions reduction efforts.

2. Methods

2.1. Chemically speciated $PM_{2.5}$ data and monitoring site selection

This study focuses on urban air quality in the Northwest U.S. and uses data collected from an EPA funded, and state and locally operated, network of monitors: The Chemical Speciation Network (CSN). CSN monitors collect 24-hour $PM_{2.5}$ on filter media that are analyzed at a laboratory to quantify the $PM_{2.5}$ chemical composition. Detailed information on the CSN and the analytical methods used can be found in Solomon et al. (2014). CSN data are available from the U.S. Environmental Protection Agency's (EPA's) Air Quality System (AQS) database.

Monitoring sites analyzed in this work are listed in Table 1 and depicted in Figure 1. The starting date for site data listed in Table 1 denotes the date that the CSN switched organic and elemental carbon (OC, EC) analytical methods to match that of the IMPROVE network, a separate network that collects chemically speciated $PM_{2.5}$. From 2007 – 2009 the EPA conducted an overhaul of CSN OC and EC measurement methodologies to harmonize both networks (U.S. EPA, 2009). OC and EC data before and after the methodology change are not easily comparable, hence data collected prior to the change are not used in this study. The ending date for site data listed in Table 1 represents the latest data available when data were extracted from the AQS database. Sites were excluded if monitoring was discontinued prior to 2018 or if data submissions were not up to date in the AQS database. All remaining sites were in operation for either 9 or 11 years and collected from 471 to 1306 24-hour samples.

2.2. Data Preparation and Treatment

The preparation and treatment of CSN data for input into the PMF model for this study is identical to that done in previous publications (Kotchenruther, 2013; Kotchenruther, 2016) and the reader is encouraged to consult those for detailed information. For this work, a synopsis of CSN data treatments is provided in the supplementary materials, including blank correction, handling of missing data, handling issues of chemical species double counting, data uncertainty estimation, the data quality metric used for data exclusion, and treatment of data with poor quality due to signal to noise ratio.

2.3. Source Apportionment

The EPA PMF 5.0 model was used for the receptor-based source apportionment analysis in this study (<https://www.epa.gov/air-research/positive-matrix-factorization-model-environmental-data-analyses>). Model documentation, including the underlying mathematical equations employed, can be found in Paatero & Hopke (2003) and Norris et al. (2014). Data from each site was modeled individually, PMF was run in the robust mode with 20 repeat runs, and the rotational F_{PEAK} variable held at 0.0. The final choice for the best number of PMF factors at each site was based on inspecting results after repeated runs where the number of factors modeled for each site was progressively increased. The results of the three error estimation methods available in this model version were compared across all runs at a given site to find the optimal solution. The three error estimation methods are: bootstrapping (BS), displacement (DISP), and bootstrapping with displacement (BS-DISP) (Norris et al., 2014; Paatero et al., 2014). For the solution chosen at each site, the scaled residuals generally fell into the recommended range of +3 to -3. 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. Association of PMF factors with specific sources or compositions of $\text{PM}_{2.5}$

Table 2 lists the 12 different factors that were identified throughout the 9 sites, and where numerical values are presented, indicates which factors were identified at which sites. The total number of PMF factors determined at each site ranged from 5 to 10. The factors listed in Table 2 were identified and labeled with descriptive names based on a combination of comparing the chemical composition of PMF factors with chemical profiles in EPA's SPECIATE database of source emissions test data (<https://www.epa.gov/air-emissions-modeling/speciate>), comparison with similar PMF factor chemical compositions identified in previous 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., soil dust, sea salt). The factors found and named in this work are consistent with those found in a previously published analysis (Kotchenruther, 2016). Kotchenruther (2016) provides a thorough description of how each factor was associated with a $\text{PM}_{2.5}$ source or composition and, for this work, that information is provided in the supplementary materials. Also provided in the supplementary materials are the PMF results for each site (the plotted factor mass time series and factor chemical profiles), and the average PMF factor chemical profiles

for those common factors found over multiple sites. The average PMF factor chemical profiles in this work are similar to those found in Kotchenruther (2016).

3.2. Winter season results

This work focuses on the results for winter $PM_{2.5}$, which for the Northwest U.S. is typically the season with the highest 24-hour average $PM_{2.5}$ from anthropogenic sources. For the purposes of this study, a winter season is defined as the last two months of the calendar year and the first two months of the following calendar year. So, for example, the 2007 winter season includes November and December of 2007 and January and February of 2008. This 4-month winter season was chosen to limit the potential influence of wild, prescribed, and agricultural fires on the observed data while at the same time including the months with highest anthropogenic influence. There is typically minimal fire activity during these four months in the Northwest U.S.

To evaluate changes in both PMF factor mass, measured $PM_{2.5}$, and measured major $PM_{2.5}$ chemical component concentrations over time, data from the 2007 through 2009 winter seasons are compared to data from the 2015 through 2017 winter seasons (note, for Butte MT only 2009 winter season data were available for the 2007 to 2009 period). A summary of the 3-year average winter season concentrations for these two time periods is provided in Table 2 and Table 3. The 3-year average winter season concentrations were calculated as the average of the monthly average concentrations. Table 4 presents the percent change in 3-year average factor mass, $PM_{2.5}$, and major $PM_{2.5}$ component concentrations between these periods. To determine if the change in 3-year averages was statistically significant, the distribution of data between the two time periods was compared using the non-parametric Wilcoxon-Mann-Whitney test, which does not require that the data distributions be normally distributed. Tables in the supplementary materials presents the p-value results of the Wilcoxon-Mann-Whitney tests, and where values were less than or equal to 0.05, indicate a statistically significant difference at or above the 95% confidence level. Percent changes in Table 4 are in bold font if the change was statistically significant at or above the 95% confidence level.

The evaluation of a 3-year time period was chosen to mitigate the effects of year-to-year meteorological fluctuations and, for the same reason, is the number of averaging years used by the U.S. EPA in its regulatory evaluation of $PM_{2.5}$.

There are a range of circumstances that influence observed winter season 24-hour average $PM_{2.5}$ mass measurements in the Northwest U.S. High winter $PM_{2.5}$ concentrations in this region of the U.S. are typically associated with air stagnation events and are exacerbated if coincident with temperature inversions and low surface temperatures, which can lead to increased emissions from some sources of direct $PM_{2.5}$ (e.g., residential wood combustion and motor vehicles). A detailed meteorological analysis of air stagnation events at all 9 monitoring sites over the time span covered by this work is beyond the scope of this study. However, some meteorological analysis regarding surface temperature and wind speed was performed.

The results for the Sea Salt factor presented in Table 4 suggest that there were some meteorological differences between the two 3-year periods at the three monitoring sites where this factor was identified: Seattle WA, Tacoma WA, and Portland OR. Table 4 shows that impacts to $PM_{2.5}$ from Sea Salt declined a statistically significant amount, between 43% and 71%, at these 3 sites over the two 3-year periods. Of the 3 sites, the Tacoma WA site had sonic anemometer measurements throughout the study period. Analysis of Tacoma WA wind data showed a decrease in average wind speed and a decrease in the frequency of days with higher daily average wind speeds. The average winter wind speed over the two 3-year periods decreased from 3.1 to 2.8 mph and, as an example of the decrease in days with higher daily average wind speeds, the frequency of days with 24-hour average wind speeds greater than 6 mph decreased by 66%, from 35 days in 2007–2009 to 12 days in 2015–2017. The lower average wind speed and lower frequency of higher wind speed days in the latter 3-year period likely explains the reduced influence of sea salt to total $PM_{2.5}$ in the latter 3-year period. However, for the concentrations of anthropogenic $PM_{2.5}$ sources that are the focus of this study, it is likely the frequency of air stagnation events, rather than the frequency of higher wind speed events that most effect observed anthropogenic concentrations. High winter concentrations of $PM_{2.5}$ in the Northwest U.S. are most often linked to air stagnation events, where horizontal transport of $PM_{2.5}$ is limited and fresh $PM_{2.5}$ emissions or secondary formation can add to existing concentrations.

Average winter season daily temperatures at meteorological sites in close proximity to the $PM_{2.5}$ monitors were evaluated to determine if there were any consistent warming or cooling trends that may have influenced anthropogenic emissions that are temperature dependent. Average winter season temperatures showed no consistent pattern of increases or decreases across the 9 sites studied, and temperature data for the two 3-year periods is provided in the supplemental materials. However, as with the discussion of wind speed, it is likely the frequency of air stagnation events that most effect observed concentrations of anthropogenic $PM_{2.5}$ sources. As mentioned above, the choice to compare 3-year time periods was used to mitigate the effects of year-to-year changes in meteorology.

3.3. Winter season $PM_{2.5}$

When inspecting the source apportionment mass attribution results in Tables 2 and 3, the majority of winter season mass on the filters at the 9 sites evaluated in this study, in both 3-year time periods, can be explained by a combination of wood smoke, sulfate and nitrate dominant factors, and gasoline and diesel engine related factors.

All sites observed a decrease in 3-year average winter season $PM_{2.5}$ between the two periods, with decreases in the range of 2% to 29% (Boise, ID and Salt Lake City, UT, respectively). However, at only 3 of the 9 sites were decreases in 3-year average winter season $PM_{2.5}$ statistically significant at the 95% confidence level (Tacoma WA, Portland OR, and Salt Lake City UT).

3.4. Results for winter season wood smoke factors

In contrast to winter season $PM_{2.5}$, all sites except one saw a large and statistically significant reduction in winter season total wood smoke $PM_{2.5}$ over the two 3-year periods.

Total wood smoke is defined as the sum of fresh and aged wood smoke factors. Table 4 shows that, with the exception of Boise ID (a 5% reduction), 3-year average winter season total wood smoke $PM_{2.5}$ diminished between 48% and 74%. Figure 2 shows the yearly winter season average total wood smoke $PM_{2.5}$ for the 9 monitoring sites.

3.5. National, state, and local efforts to reduce winter residential wood smoke pollution

Nationally, the U.S. EPA has a multi-decade history of developing and implementing New Source Performance Standards (NSPS) for residential wood heaters. These standards govern the manufacture and sale of new devices and impose emissions limits on those devices. EPA issued the first NSPS for residential wood room heaters and certain wood burning fireplace inserts in 1988. The 1988 NSPS created a device certification process and mandated that adjustable burn rate wood stoves built after 1988 emit not more than a weighted average of 4.1 g/hr or 7.5 g/hr of particulate matter (PM), depending on whether the stove was equipped with a catalytic combustor or not, respectively. Prior to the 1988 NSPS, adjustable burn rate wood stoves typically emitted in the range of 15 to 30 g/hr of PM.

In 2015 EPA issued revisions and additions to the 1988 residential wood heater NSPS. These changes strengthened the emissions standards for new wood heaters and broadened the types of devices requiring emissions certification. The 2015 NSPS for wood burning room heaters included all residential wood heaters not specifically exempt in the rule, which now included both adjustable and fixed burn rate stoves and defined a single PM emissions limit for both catalytic and noncatalytic wood heaters. The emissions limits for wood burning room heaters are phased in via a two-step compliance approach, with the step 1 limit being not more than 4.5 g/hr of PM for devices sold on or after December 31, 2015, and the step 2 limit being not more than 2.0 g/hr, effective in 2020 (2.5 g/hr for an optional cord wood test). In addition to new standards for wood burning room heaters, the 2015 NSPS set PM emissions standards for two specific types of wood heaters not previously covered in the 1988 NSPS, hydronic heaters and wood fired forced-air furnaces. The 2015 NSPS for these devices is also phased in via a two-step compliance approach, with various timelines and emissions thresholds for step 1 compliance, depending on the device, and step 2 compliance and emissions thresholds mandated by 2020.

In 1991 Washington State enacted wood burning device standards that were more stringent than the EPA 1988 NSPS, and are in some respects more stringent than the 2015 NSPS. Under these Washington State standards, catalytic wood burning devices are limited to 2.5 g/hr $PM_{2.5}$ and all other solid fuel burning devices are limited to 4.5 g/hr $PM_{2.5}$ (with the exception of factory-built fireplaces and masonry heaters, having a 7.3 g/kg limit).

On a State and Local level in the U.S. Pacific Northwest and Intermountain West, there are a range of additional measures that have been taken to reduce residential wood smoke PM emissions. Both the 1988 and 2015 NSPS only effected the manufacture and sale of new devices, not existing devices. Hence, their effects on emissions reductions for current wood burners is expected to be slow due to the long lifespan of these devices. One common measure to speed up the emissions reduction benefits of newer devices is to offer incentives to replace older higher-emitting devices with cleaner burning devices. State tax incentives are one example of this. Idaho offers a tax deduction for replacing a wood stove that no

longer meets EPA standards, Montana offers a tax credit for installing a low-emissions wood burning device, and Oregon provides a tax credit for installing a high efficiency wood and pellet stoves.

Some local communities also periodically offer partial or full device exchange grants to home owners willing to remove old wood stoves and replace them with cleaner options. These local programs are often of limited duration and are dependent on the availability of funding. For example, under such programs from 2007 – 2019 the City of Yakima WA reported a combined 1071 woodstove removals or changeouts of uncertified woodstoves, to cleaner burning devices (233 removals, 838 changeouts). The Yakima WA countywide estimate of total woodstoves is 30,000 – 40,000 (YRCAA, 2019). From 2012 – 2019, the City of Tacoma WA reported more than 3,000 changeouts of uncertified wood burning devices to cleaner burning devices out of a total of approximately 40,000 wood burning (fireplaces excluded) home heating devices (PSCAA, 2019). In Boise ID, from 2007 – 2017 incentive programs resulted in a total of 135 uncertified wood stoves being changed out to cleaner burning options, out of a total estimated woodstove population of approximately 30,000 in Ada county (IDEQ, 2019). In the greater Salt Lake City region of Utah, from 2014 – 2017 incentive programs resulted in a total of 80 wood stoves being changed out to natural gas options, out of a total estimated woodstove population of 33,000 in the 3 Utah counties of Davis, Salt Lake and Utah containing the monitors in this study (UDEQ, 2019).

Some communities couple changeout or removal incentives with even stronger measures. For example, Oregon requires the removal of uncertified wood stoves upon sale of a property, and in Tacoma WA, as of October 1, 2015, it was made illegal to operate an uncertified wood stove. This measure in Tacoma WA was credited for a large number of wood stove replacements prior to 2015.

Another measure to reduce winter wood smoke is to curtail wood burning on days measured or forecast to have poor air quality; these are commonly referred to as ‘burn bans’. Where burn bans occur, the specific triggering thresholds and burning restrictions are specific to state and local jurisdictions. Some examples of this are as follows. In the Washington State cities of Seattle, Tacoma and Yakima, a ‘Stage 1’ burn ban prohibiting outdoor burning and indoor burning except for EPA certified woodstoves or pellet stoves can be called when $PM_{2.5}$ is forecast to exceed $30 \mu\text{g}/\text{m}^3$ (Tacoma and Yakima) or $35 \mu\text{g}/\text{m}^3$ (Seattle). A ‘Stage 2’ burn ban prohibiting all outdoor and indoor burning can be called when 24-hour $PM_{2.5}$ is measured above $25 \mu\text{g}/\text{m}^3$. In the Salt Lake City region of Utah, burn bans effect communities surrounding all three Utah monitors in this study. Voluntary burn bans can be called when $PM_{2.5}$ is forecast to exceed $12 \mu\text{g}/\text{m}^3$ and mandatory burn bans can be called when $PM_{2.5}$ is forecast to exceed $25 \mu\text{g}/\text{m}^3$. In the Idaho municipality of Meridian (which contains the city of Boise), progressively more stringent wood burning curtailments are called when 24-hour average $PM_{2.5}$ exceeds certain thresholds. When 24-hour average $PM_{2.5}$ exceeds $16.3 \mu\text{g}/\text{m}^3$ (EPA Air Quality Index (AQI) ≥ 60) all outdoor burning is prohibited. When 24-hour average $PM_{2.5}$ exceeds $23 \mu\text{g}/\text{m}^3$ (AQI ≥ 74) indoor burning in all fireplaces and woodstoves is prohibited. It should be noted that the effectiveness of burn bans is largely dependent on the amount of local resources dedicated to informing the public about the bans and local methods and resources used to enforce the bans.

Nationally and on the State and Local level, governments also promote websites with educational materials on best burning practices that can reduce PM_{2.5} emissions in existing devices.

The reductions in 3-year average winter season PM_{2.5} contributions from wood smoke, from 48% to 74% at 8 of 9 sites in this study, suggest that the suite of combined wood smoke control measures in these communities have been effective.

3.6. Results for winter season ammonium sulfate and ammonium nitrate factors

Those sites with factors associated with ammonium sulfate (7 sites) and ammonium nitrate (5 sites) also all had statistically significant reductions in 3-year average winter season mass for those factors, with reductions between 27% to 81% for ammonium sulfate and 11% to 54% for ammonium nitrate over the two 3-year periods. Additionally, table 4 indicates that total measured sulfate decreased at every site, from 9% to 75%, and total measured nitrate decreased at every site, from 2% to 39%, over the two 3-year winter season periods.

Gaseous SO₂ and NO_x are precursors to particulate sulfate and nitrate, formed through atmospheric reactions. The largest sources of both SO₂ and NO_x are fossil fuel combustion for power generation, in industrial facilities, and in transportation. Recent efforts to reduce SO₂ emissions have focused on industrial emissions controls and reducing the sulfur content of transportation fuels. Efforts to reduce NO_x have focused on implementing more stringent emissions control technologies and improving engine designs. Yearly emissions inventory data (U.S. EPA, 2019) for the 5 states with monitoring sites covered in this analysis, after removing the emissions from wildfires, show a 5-state average reduction in NO_x emissions of 24% between the same two 3-year periods that were evaluated above for PM (range, 11% to 31% reduction depending on the State), a 5-state average reduction in SO₂ emissions of 45% (range, 21% to 69% depending on the State), and a 5-state average reduction in NH₃ emissions of 39% (range, 19% to 57% depending on the State). SO₂, NO_x, and NH₃ emissions inventory data for the 5 states with monitoring sites covered in this analysis is provided in the supplemental materials. While it is hard to compare changes in yearly state-wide PM_{2.5} gaseous precursor emissions to winter observations at specific monitoring locations, the decreases in ammonium nitrate and ammonium sulfate factor mass at every site where they are identified, and decreases in total measured sulfate and nitrate at every site, are consistent with the across the board 5 state reductions in PM_{2.5} gaseous precursor emissions.

3.7. Results for winter season residual oil combustion factors

The 2 sites with sulfate dominated factors associated with residual oil combustion both had a statistically significant 85% reduction in 3-year average winter season mass over this study time period. These sites are in the major port cities of Seattle and Tacoma, WA, and the main source using residual fuel oils has been large ocean-going vessels. The 85% reduction in this factor is consistent with previous published work investigating the impacts on ambient PM_{2.5} before and after implementing fuel sulfur standards on ocean going vessels in 2012 and 2015 (Kotchenruther, 2017) as part of the International Maritime Organization North American emissions control area.

3.8. Results for winter season gas engine factors

There is an apparent incongruity when comparing the measured changes in 3-year average winter season $PM_{2.5}$, which are smaller and less significant than the decreases in specific $PM_{2.5}$ components identified as factors by PMF and associated with wood smoke, ammonium nitrate, ammonium sulfate, and residual oil combustion. To account for this discrepancy, there must be other factors that are major contributors to $PM_{2.5}$ that are not significantly decreasing, or that are increasing. This is the case for factors associated with gas engines and to a lesser extent also for diesel engines. The two factors with the highest attributed mass in these categories were those identified as Gas Engines and Mixed – Gas Engines & Nitrate. One or the other of these two factors was identified at 8 of the 9 sites (4 sites for each factor) and the average chemical profiles for these two factors are nearly identical except for the presence and absence of nitrate (see average factor profiles in the supplementary materials). At the 4 sites where the Gas Engine factor was identified, 3-year average winter season $PM_{2.5}$ associated with this factor ranged from 2.21 to 3.05 $\mu\text{g}/\text{m}^3$ in the 2007–2009 period and 2.48 to 3.66 $\mu\text{g}/\text{m}^3$ in the 2015–2017 period. At the 4 sites where the Mixed – Gas Engine & Nitrate factor was identified, 3-year average winter season $PM_{2.5}$ associated with this factor ranged from 1.85 to 2.61 $\mu\text{g}/\text{m}^3$ in the 2007–2009 period and 2.44 to 6.04 $\mu\text{g}/\text{m}^3$ in the 2015–2017 period. While the 3-year average winter season mass increased for these factors at all 8 sites between the two 3-year time periods, the increases were statistically significant at only 3 of the 8 sites. The Gas Engine factor increased between 6% and 66% at the 4 sites where found, and the Mixed – Gas Engines & Nitrate factor increased between 31% and 226% at the 4 sites where found. Figure 3 shows the yearly average winter season mass attributed to these two factors for the 8 monitoring sites where found. It should be noted that these two factors associated with Gas Engines only account for a portion of $PM_{2.5}$ that should be attributable to gas engine emissions. Gas engines also emit gaseous NO_x , volatile organic compounds, SO_2 , and NH_3 , that can also form $PM_{2.5}$ through atmospheric chemical reactions. Similar results of increasing Gas Engine factor mass were also found by Masiol et al. (2019), where 8 sites in New York State were analyzed from 2005 to 2017, and increases of up to 0.2 $\mu\text{g}/\text{m}^3$ per year were identified in a similar PMF factor (identified as spark-ignition vehicles by Masiol et al.).

For the remaining site where neither the Gas Engine nor the Mixed – Gas Engine & Nitrate factors were found, in Boise ID, Gas Engine $PM_{2.5}$ was associated with the factor Mixed – Gas Engines and Soil / Dust. $PM_{2.5}$ mass associated with this factor increased by 24% between the two 3-year winter season periods, but the increase was not statistically significant.

The largest percent increase in winter season mass between the two 3-year periods, 226%, was in the Mixed – Gas Engines & Nitrate factor identified in the Yakima WA data. Inspecting table 4, PMF results for Yakima also show a statistically significant 48% reduction in the ammonium nitrate factor, but also show that observed winter season nitrate only decreased by 10% between the two 3-year periods. This suggests that at least part of the 226% increase in the Mixed – Gas Engines & Nitrate factor resulted from a shift in the atmospheric chemistry of particulate nitrate production in Yakima, with less ammonium nitrate production and more particulate nitrate association with the gas engine factor.

The 3 Factors associated with diesel engines were Diesel Engines, Mixed – Gas and Diesel Engines, and Mixed – Diesel Engines and Nitrate. One of these 3 factors was identified at 6 of the 9 monitoring sites in this study. The 3-year average winter season mass attributed to diesel engine factors was much less than that attributed to gas engines, $0.53 \mu\text{g}/\text{m}^3$ or less for diesel engine factors for the 2015–2017 3-year winter season period. The change in 3-year average winter season $\text{PM}_{2.5}$ associated with these factors ranged from –16% to 80%, with half of the sites showing decreases and half increases. Only 2 of the sites showed statistically significant changes between the two 3-year periods, and these were both increases (23% and 80% at Boise ID and Salt Lake City UT, respectively).

Excluding meteorological effects addressed earlier, changes in gas and diesel engine $\text{PM}_{2.5}$ impacts over time are influenced by changes in vehicle miles traveled (VMT), congestion, and average vehicle emissions due to fleet turnover. Of these influences, only VMT data were readily available for analysis. Yearly total urban VMT data was analyzed for the 5 states with monitoring sites covered in this analysis. On average for the 5-state area, 3-year average VMT increased by 22% (range, increases of 10% to 46%, depending on State) between the two 3-year periods. VMT data is provided in the supplemental materials.

The increases shown here, in 3-year average winter season $\text{PM}_{2.5}$ from gas and diesel engines, largely explain why reductions in total measured winter season $\text{PM}_{2.5}$ is muted compared to the large significant reductions in wood smoke, ammonium nitrate, and ammonium sulfate related factors over the period of this study.

4. Conclusions

This work uses PMF source apportionment modeling to identify factors contributing to $\text{PM}_{2.5}$ pollution in the Northwest U.S. at monitoring locations in 9 cities. $\text{PM}_{2.5}$, the chemical components of $\text{PM}_{2.5}$, and PMF factor contributions to $\text{PM}_{2.5}$ were tracked for 9 or 11 winter seasons, depending on the site. The PMF model allocated the majority of $\text{PM}_{2.5}$ mass to factors associated with fresh wood smoke, aged wood smoke, soil dust, gas engines, mixed – gas engines and nitrate, ammonium sulfate, and ammonium nitrate. Results for two 3-year winter season periods were compared, 2007–2009 and 2015–2017, to assess changes in average concentrations and determine if those changes were statistically significant.

Total $\text{PM}_{2.5}$ mass decreased between 2% and 29% at the 9 sites between the two 3-year winter season periods, and the decreases were statistically significant at 3 sites. On the state and local level, as well as nationally, a range of efforts have been employed to try and reduce the contributions of winter residential wood smoke to measured concentrations of $\text{PM}_{2.5}$. The source apportionment results for winter season wood smoke demonstrate that these efforts have been successful at 8 of 9 sites analyzed in this study, with total wood smoke $\text{PM}_{2.5}$ decreasing a statistically significant 48% to 74% between those 8 sites.

Recent national and state efforts to reduce SO_2 emissions have focused on industrial emissions controls and reducing the sulfur content of transportation fuels. Efforts to reduce NO_x have focused on implementing more stringent emissions control technologies and improving engine designs. Analysis of yearly emissions inventories for SO_2 and NO_x for the

5 states with monitors used in this study demonstrate success in this regard and indicate a 5-state average decrease of 45% and 24%, respectively, for SO₂ and NO_x for the 3-year average emissions between the two 3-year periods. Additionally, each individual state showed an emissions decrease in these precursors as well as the 5-state average. While it is hard to compare changes in yearly state-wide PM_{2.5} gaseous precursor emissions to winter PM_{2.5} observations at specific monitoring locations, the decreases in ammonium nitrate and ammonium sulfate PMF factor mass at every site where they are identified are consistent with the across the board state reductions in PM_{2.5} gaseous precursor emissions. Factors identified and associated with ammonium nitrate (5 of 9 sites) decreased a statistically significant 11% to 54% between the two 3-year winter season periods. Factors identified and associated with ammonium sulfate (7 of 9 sites) decreased a statistically significant 27% to 81% between the two 3-year winter season periods. At every site, measured sulfate and nitrate decreased, with measured sulfate decreasing from 9% to 75% and measured nitrate decreasing from 2% to 39% over the two 3-year winter season periods.

In contrast to the reduction in PM_{2.5} from factors associated with wood smoke, ammonium nitrate, and ammonium sulfate, factors associated with gas engine PM_{2.5} increased from 6% to 226% between the two 3-year winter season averages at the 9 sites in this study. Increases in PM_{2.5} from gas engine related factors explain why overall average winter season PM_{2.5} had more modest percent reductions compared to the percent reductions for wood smoke, ammonium nitrate, and ammonium sulfate factors between the two 3-year periods. Analysis of yearly total urban VMT data for the 5 states with monitoring sites covered in this analysis showed that for the 5-state area, 3-year average VMT increased by 22% (range, increases of 10% to 46%, depending on State) between the two 3-year periods. However, VMT is only one variable influencing observed gas engine Factor PM_{2.5}, with congestion and average vehicle emissions due to fleet turnover also influencing results.

It is hoped that these source apportionment results will help guide policy-makers in formulating future policies aimed at improving particulate air quality by identifying the specific sources and pollutants where success has been achieved as well as areas where further progress is possible.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Figure 1.
CSN monitoring sites analyzed in this study.

Figure 2.
Yearly winter season average total wood smoke $PM_{2.5}$ mass (the sum of fresh and aged wood smoke factors).

Figure 3.
Yearly winter season average gas engine and mixed gas engine combined with nitrate $PM_{2.5}$ factor mass.

Table 1.

CSN monitoring sites analyzed in this study.

City	State	Start date	End date	Number of samples	EPA AQS Number	Latitude	Longitude
Seattle	WA	5/3/2007	9/29/2018	1143	530330080	47.5683	-122.3081
Tacoma	WA	5/12/2007	9/29/2018	614	530530029	47.1864	-122.4517
Yakima	WA	11/8/2007	9/29/2018	539	530770009	46.5968	-120.5122
Portland	OR	5/3/2007	9/29/2018	1185	410510080	45.4965	-122.6034
Butte	MT	10/4/2009	9/29/2018	471	300930005	46.0026	-112.5012
Boise	ID	5/3/2007	9/29/2018	1306	160010010	43.6003	-116.3479
Bountiful	UT	5/6/2007	9/29/2018	630	490110004	40.903	-111.8845
Salt Lake City	UT	5/9/2007	9/29/2018	1213	490353006	40.7364	-111.8722
Lindon	UT	5/6/2007	9/29/2018	605	490494001	40.3414	-111.7136

Table 2.

Average winter season PM_{2.5} mass (µg/m³) for PMF factors, PM_{2.5}, and major chemical components from 2007 – 2009*.

<i>PMF Factor Names</i>	Seattle, WA	Tacoma, WA	Yakima, WA	Portland, OR	Butte, MT	Boise, ID	Bountiful, UT	Salt Lake City, UT	Lindon, UT
Fresh Wood Smoke	0.91	5.73	3.15	2.63	7.08	2.47	0.44	0.88	
Aged Wood Smoke	0.88	1.95	4.38	1.78	5.56	1.66	1.97	1.87	
Soil / Dust	0.28		0.46	0.13	0.76		0.49	0.49	0.87
Gas Engines	2.25			3.05	2.21			2.35	
Ammonium Sulfate			0.51	0.62	0.65	1.07	0.86	1.60	0.50
Ammonium Nitrate			4.77			3.29	6.61	9.08	7.85
Diesel Engines					0.21	0.38	0.22		
Sulfate Dominant	0.34	0.38		0.38					
Sea Salt	0.11	0.56		0.16					
Residual Oil Combustion	0.80	1.37							
Nitrate Dominant	0.56			1.51	2.35				
Industrial	0.11								
Mixed - Gas Engines & Nitrate		2.61	1.85				1.87		2.48
Mixed - Gas & Diesel Engines	0.52			0.57		0.24			
Mixed - Gas Engines & Soil / Dust		0.87						0.29	
Mixed - Diesel Engines & Nitrate									2.51
Mixed - Fresh & Aged Wood Smoke									
Wood Smoke Summary									
Total Wood Smoke	1.79	7.68	7.52	4.40	12.64	4.14	2.41	2.75	2.51
Measured PM_{2.5} and Major Components									
PM _{2.5}	6.68	13.49	14.93	10.42	18.42	8.88	12.57	16.52	13.04
Organic Mass (OC*1.4)	3.15	7.58	7.42	6.10	12.21	3.01	2.67	3.79	3.15
Elemental Carbon	0.93	1.44	1.40	1.23	1.90	0.71	0.91	1.17	0.83
Soil (IMPROVE Formula)	0.26	0.35	0.35	0.27	0.41	0.24	0.46	0.75	0.65
Sulfate	0.61	0.69	0.61	0.77	0.48	0.55	0.84	0.91	0.60
Nitrate	0.87	1.00	2.75	1.16	1.29	2.53	4.77	5.99	5.17
Ammonium	0.35	0.44	1.01	0.51	0.44	0.88	1.79	2.20	1.71

* for Butte, MT, only 2009 available.

Table 3.

Average winter season $PM_{2.5}$ mass ($\mu g/m^3$) for PMF factors, $PM_{2.5}$, and major chemical components from 2015 – 2017.

<i>PMF Factor Names</i>	Seattle, WA	Tacoma, WA	Yakima, WA	Portland, OR	Butte, MT	Boise, ID	Bountiful, UT	Salt Lake City, UT	Lindon, UT
Fresh Wood Smoke	0.43	3.53	1.09	0.88	3.43	2.45	0.05	0.37	
Aged Wood Smoke	0.31	0.34	1.22	0.57	3.16	1.48	0.74	0.50	
Soil / Dust	0.23		0.32	0.11	0.99		0.61	0.47	0.67
Gas Engines	2.78			3.36	3.66			2.48	
Ammonium Sulfate			0.20	0.12	0.13	0.32	0.63	0.75	0.11
Ammonium Nitrate			2.47			2.92	4.80	5.26	3.64
Diesel Engines					0.18	0.46	0.24		
Sulfate Dominant	0.24	0.46		0.34					
Sea Salt	0.06	0.16		0.09					
Residual Oil Combustion	0.12	0.21							
Nitrate Dominant	0.42			1.17	2.36				
Industrial	0.04								
Mixed - Gas Engines & Nitrate		3.79	6.04				2.44		3.12
Mixed - Gas & Diesel Engines	0.46			0.50		0.30		0.53	
Mixed - Gas Engines & Soil / Dust		0.78							0.65
Mixed - Diesel Engines & Nitrate									
Mixed - Fresh & Aged Wood Smoke									
Wood Smoke Summary									
Total Wood Smoke	0.74	3.88	2.31	1.45	6.59	3.93	0.80	0.87	0.65
Measured $PM_{2.5}$ and Major Components									
$PM_{2.5}$	5.40	9.91	12.40	7.62	14.66	8.69	10.88	11.72	9.83
Organic Mass (OC*1.4)	2.56	5.46	5.14	3.95	8.15	2.66	1.79	2.32	2.02
Elemental Carbon	0.60	1.07	0.90	0.69	1.79	0.62	0.56	0.66	0.58
Soil (IMPROVE Formula)	0.18	0.21	0.28	0.19	0.46	0.27	0.49	0.64	0.49
Sulfate	0.22	0.33	0.35	0.30	0.12	0.33	0.76	0.62	0.31
Nitrate	0.54	0.61	2.48	0.77	1.24	2.48	4.25	4.16	3.41
Ammonium	0.05	0.06	0.46	0.10	0.16	0.57	1.15	1.07	0.70

Table 4.

Percent change in winter season PM_{2.5} mass (µg/m³) for PMF factors, PM_{2.5}, and major chemical components from 2007–2009* to 2015–2017. Values in bold text indicate a statistically significant change at the 95% confidence level using the non-parametric Wilcoxon-Mann-Whitney test.

<i>PMF Factor Names</i>	Seattle, WA	Tacoma, WA	Yakima, WA	Portland, OR	Butte, MT	Boise, ID	Bountiful, UT	Salt Lake City, UT	Lindon, UT
Fresh Wood Smoke	-53	-38	-65	-66	-52	-1	-88	-58	
Aged Wood Smoke	-65	-82	-72	-68	-43	-11	-62	-73	
Soil / Dust	-18		-31	-19	30		25	-5	-22
Gas Engines	24			10	66			6	
Ammonium Sulfate			-60	-81	-80	-71	-27	-53	-79
Ammonium Nitrate			-48			-11	-27	-42	-54
Diesel Engines					-16	23	9		
Sulfate Dominant	-28	20		-11					
Sea Salt	-49	-71		-43					
Residual Oil Combustion	-85	-85							
Nitrate Dominant	-26			-23	0				
Industrial	-62								
Mixed - Gas Engines & Nitrate		45	226				31		26
Mixed - Gas & Diesel Engines	-11			-12		24			
Mixed - Gas Engines & Soil / Dust		-10						80	-74
Mixed - Diesel Engines & Nitrate									
Mixed - Fresh & Aged Wood Smoke									
Wood Smoke Summary									
Total Wood Smoke	-59	-50	-69	-67	-48	-5	-67	-68	-74
Measured PM_{2.5} and Major Components									
PM _{2.5}	-19	-27	-17	-27	-20	-2	-13	-29	-25
Organic Mass (OC*1.4)	-19	-28	-31	-35	-33	-12	-33	-39	-36
Elemental Carbon	-36	-26	-36	-44	-6	-13	-39	-43	-31
Soil (IMPROVE Formula)	-29	-40	-20	-31	13	13	6	-14	-24
Sulfate	-64	-52	-43	-61	-75	-39	-9	-32	-47
Nitrate	-39	-39	-10	-34	-4	-2	-11	-31	-34
Ammonium	-85	-86	-55	-81	-63	-35	-36	-51	-59

*
for Butte, MT, only 2009 available.

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