Trends in Source Impacts at Long-Running $PM_{2.5}$ CSN Monitoring Sites in the Pacific Northwest and Intermountain West

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Motivation:

- Some PM2.5 monitors have a long record of chemically speciated data.
- These data can be used for source apportionment modeling (e.g., PMF)
- We can use source apportionment results to look at changes in source impacts over time.
- Some sources have a long history of being targeted for emissions reductions (e.g., residential wood combustion, sulfur in fossil fuels)
- ??? How successful have we been?
- ??? Are there any unexpected results?

This Approach:

Data:

1) Review currently active sites that collect chemically speciated PM2.5 data.
2) Select sites that are up to date in data submissions to EPA's AQS system (data through summer 2018)
3) Select sites that have a data record going back to when EPA switched carbon measurement methods to match the IMPROVE methodology (2007-2009).

Modeling:

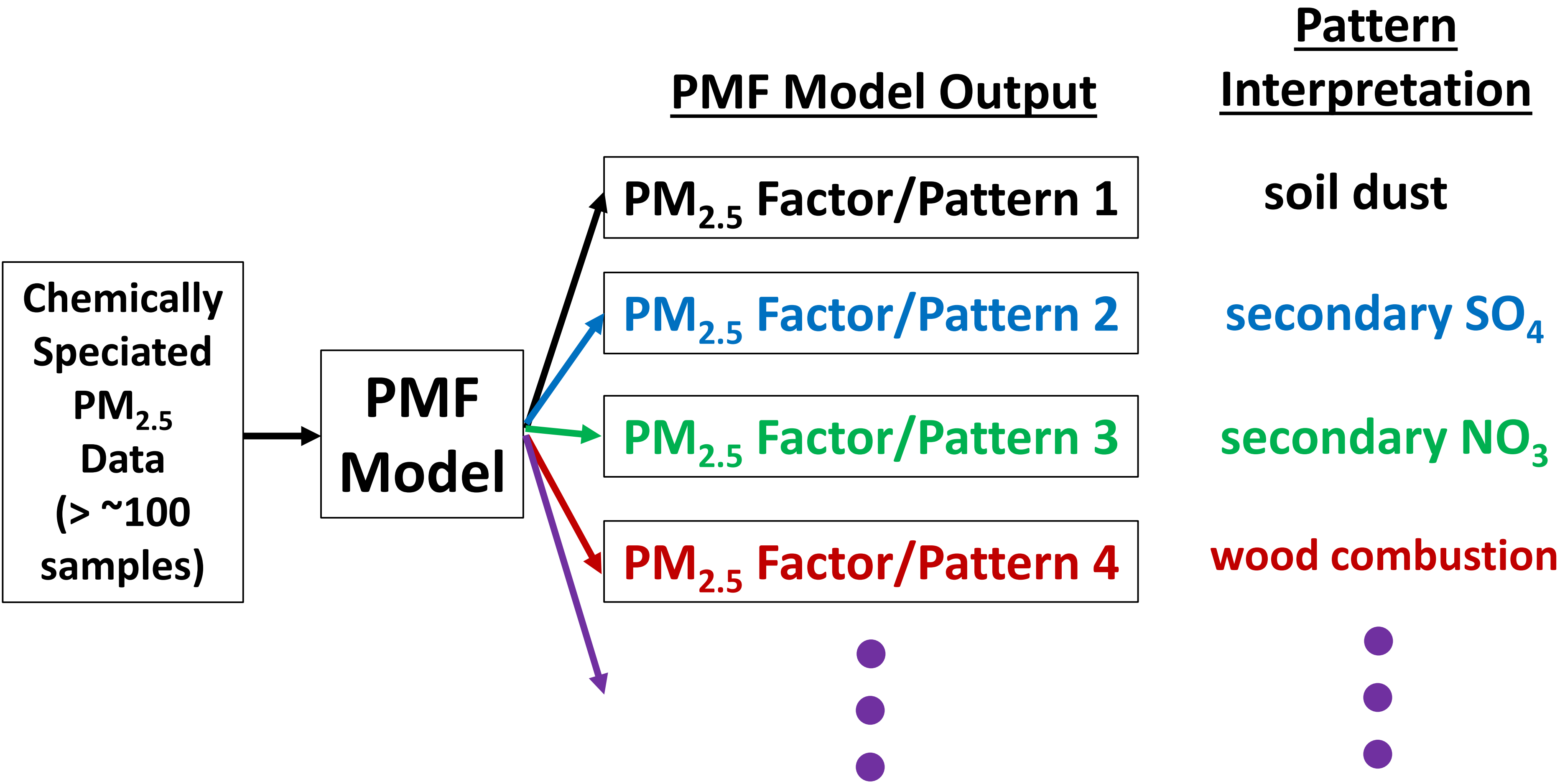
1) Run sites' data in PMF and get source apportionment results (each site run independently)
2) Analyze year to year changes in resulting sources.

Note:

- For this presentation I focus on year to year results for winter months Nov – Feb (but all data was used in modeling)
- A 'winter year' is Nov – Feb. So, for example, '2015 winter' is Nov-Dec 2015 and Jan-Feb 2016

How does the PMF source apportionment model work?

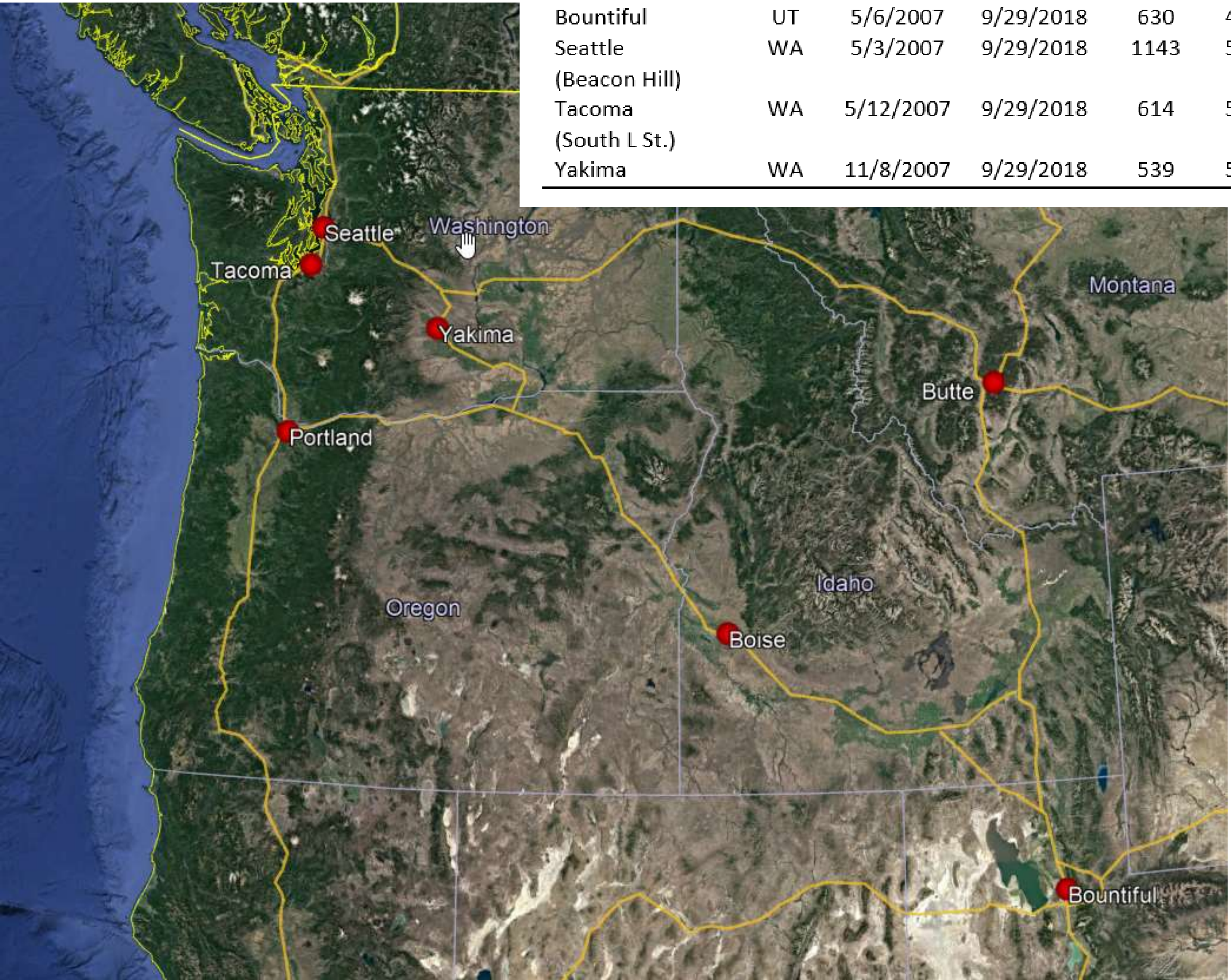
The model looks for systematic patterns in the day-to-day chemical variations and quantifies a smaller set of 'factors' that can explain the overall data variability.



7 Sites Analyzed

CSN monitoring sites analyzed in this study.

City	State	Start date	End date	Number of samples	EPA AQS Number	Latitude	Longitude
Boise	ID	5/3/2007	9/29/2018	1306	160010010	43.6003	-116.3479
Butte	MT	10/04/2009	9/29/2018	471	300930005	46.0026	-112.5012
Portland	OR	5/3/2007	9/29/2018	1185	410510080	45.4965	-122.6034
Bountiful	UT	5/6/2007	9/29/2018	630	490110004	40.9030	-111.8845
Seattle (Beacon Hill)	WA	5/3/2007	9/29/2018	1143	530330080	47.5683	-122.3081
Tacoma (South L St.)	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



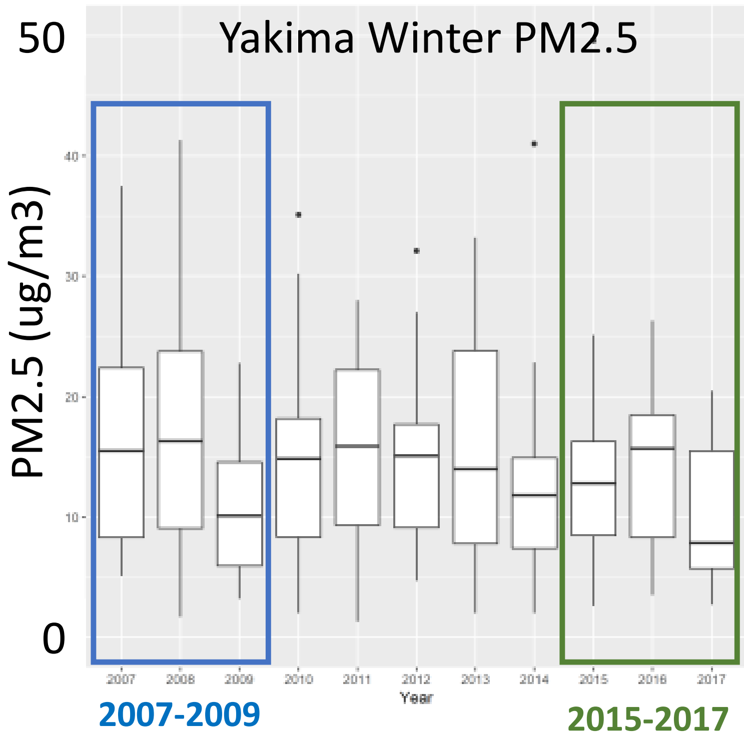
What were the common types of chemical patterns / factors / sources found from PMF analysis?

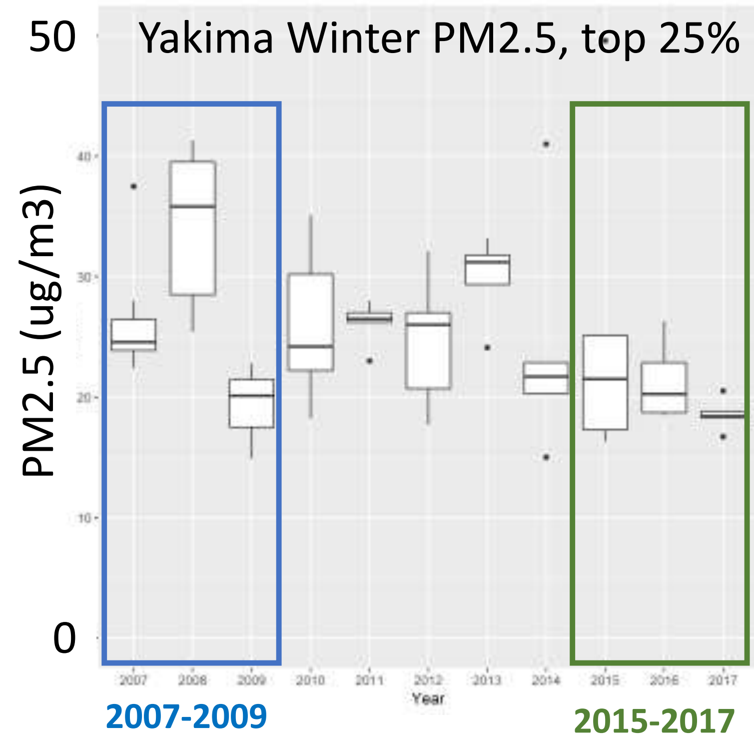
Factor / Pattern	Boise, ID	Butte, MT	Portland, OR	Bountiful, UT	Seattle, WA (Beason Hill)	Tacoma, WA (South L St.)	Yakima, WA
Fresh Wood Smoke	✓	✓	✓	✓	✓	✓	✓
Aged Wood Smoke	✓	✓	✓	✓	✓	✓	✓
Soil / Dust	✓	✓	✓	✓	✓		✓
Gas Engines		✓	✓	✓	✓	✓	✓
Ammonium Sulfate	✓	✓	✓	✓			✓
Ammonium Nitrate	✓	✓		✓			✓
Diesel Engines	✓	✓		✓	✓		
Sulfate Dominant			✓		✓	✓	
Sea Salt			✓		✓	✓	
Residual Oil Combustion					✓	✓	
Nitrate Dominant			✓		✓		
Industrial (Sulfate & Metals)					✓		
Mixed			✓			✓	

Format of results analysis - Example of Yakima WA

Changes in winter (Nov-Feb) $PM_{2.5}$* from 2007-2017.

*only using PM2.5 data coincident with CSN monitoring





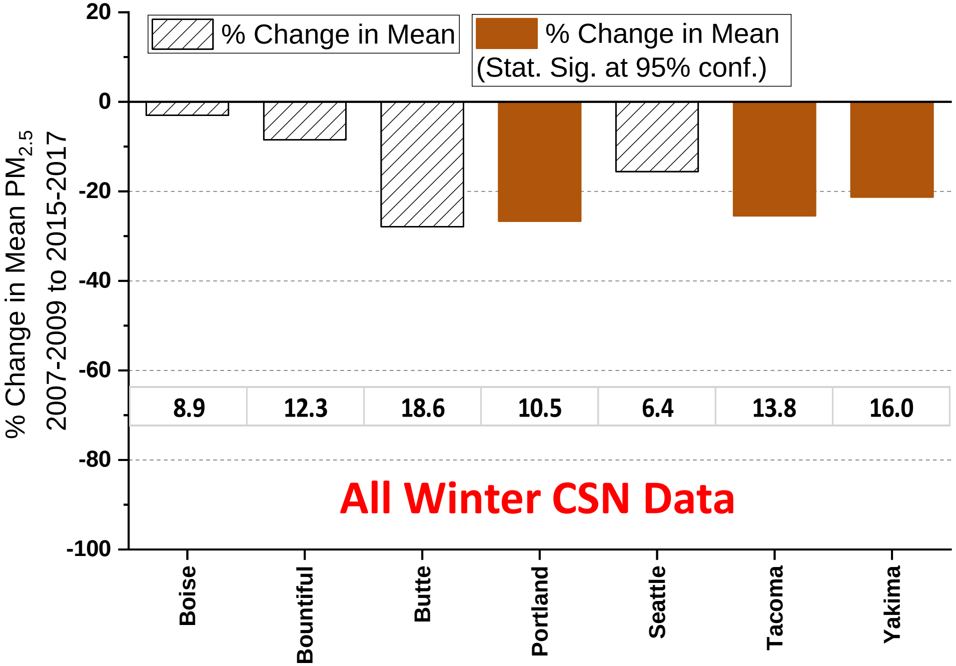
Comparing 2007-2009 with 2015-2017

		2007-2009			2015-2017				
		Mean (ug/m3)	Stdev (ug/m3)	Median (ug/m3)	Mean (ug/m3)	Stdev (ug/m3)	Median (ug/m3)	WMW# p-value (Δ significant?)	% Change in Mean
Yakima	Winter PM2.5	16.0	9.8	14.5	12.6	8.1	10.9	*0.050*	*-21.3*
Yakima	Winter PM2.5 (top 25%)	28.6	7.6	25.9	22.0	8.5	18.8	*0.003*	*-23.0*

#WMW = Wilcoxon-Mann-Whitney significance test (non-parametric)

Changes in winter (Nov – Feb) $PM_{2.5}$

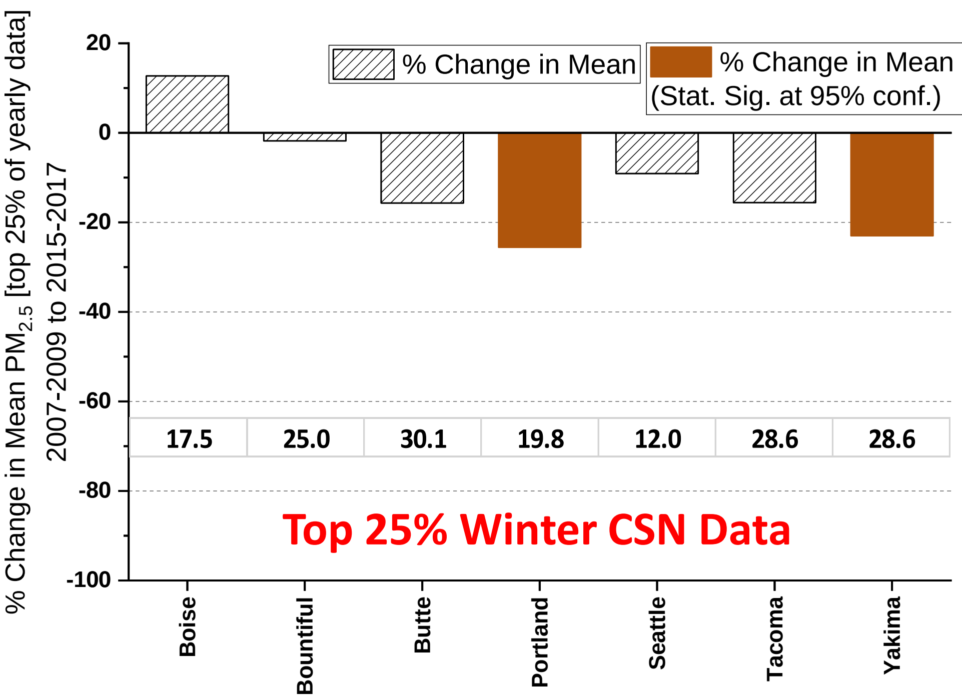
2007-2009 to 2015 - 2017



Change in 3-year average winter $PM_{2.5}$* [2007-2009 to 2015-2017]

*only using PM2.5 data coincident with CSN monitoring

Beginning 2007-2009 average winter PM2.5 mass (ug/m3)



Change in 3-year average winter $PM_{2.5}$* [2007-2009 to 2015-2017] for top 25% of yearly data.

*only using PM2.5 data coincident with CSN monitoring

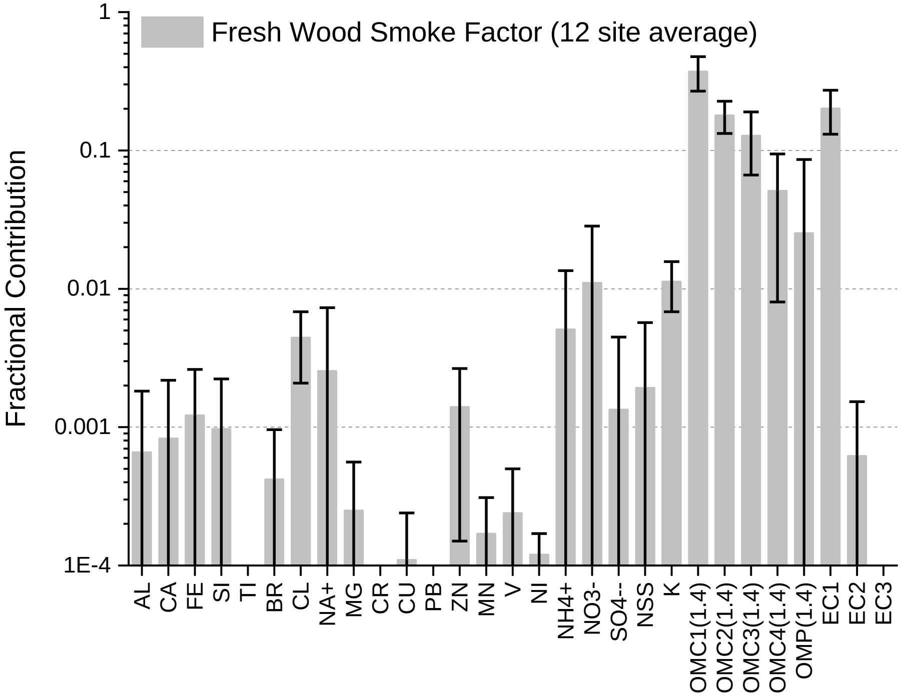
Beginning 2007-2009 average winter PM2.5 mass (ug/m3) for top 25% of yearly data.

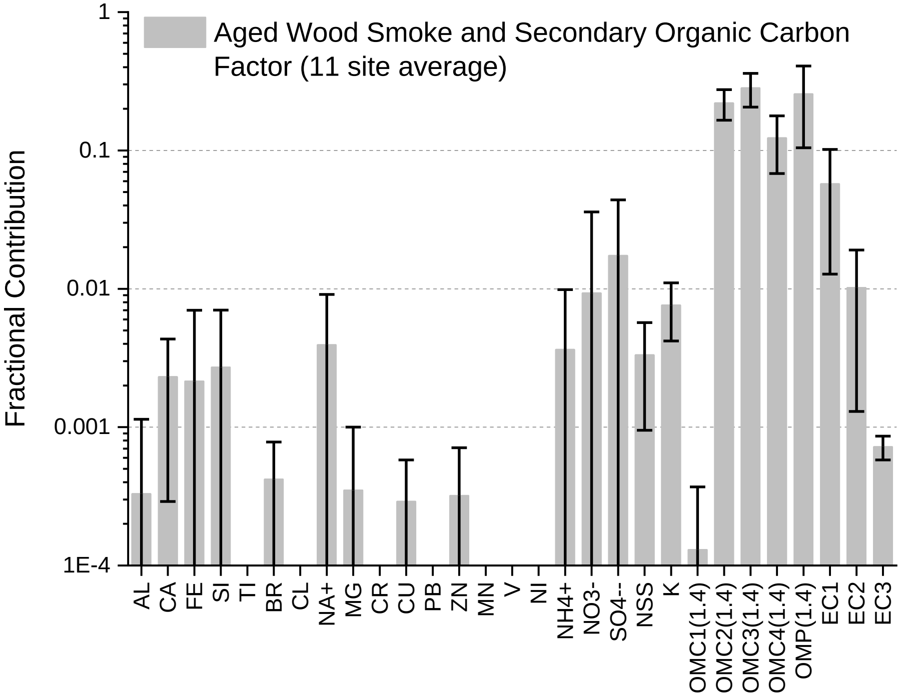
Results for Winter Wood Smoke $PM_{2.5}$

(2007-2009 to 2015 – 2017)

How was Wood Smoke Identified in the Chemical Data?

(Figures from R.A. Kotchenruther / Atmospheric Environment 142 (2016) 210-219)





Fresh wood smoke

- OC and EC dominate
- OC components shifted to lower boiling fractions
- K contribution
- Cl contribution

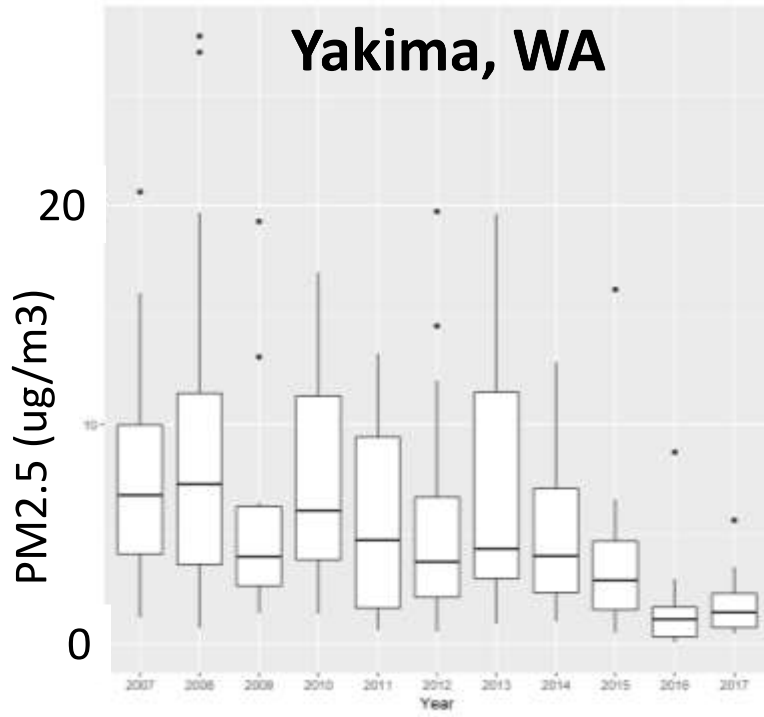
Aged wood smoke

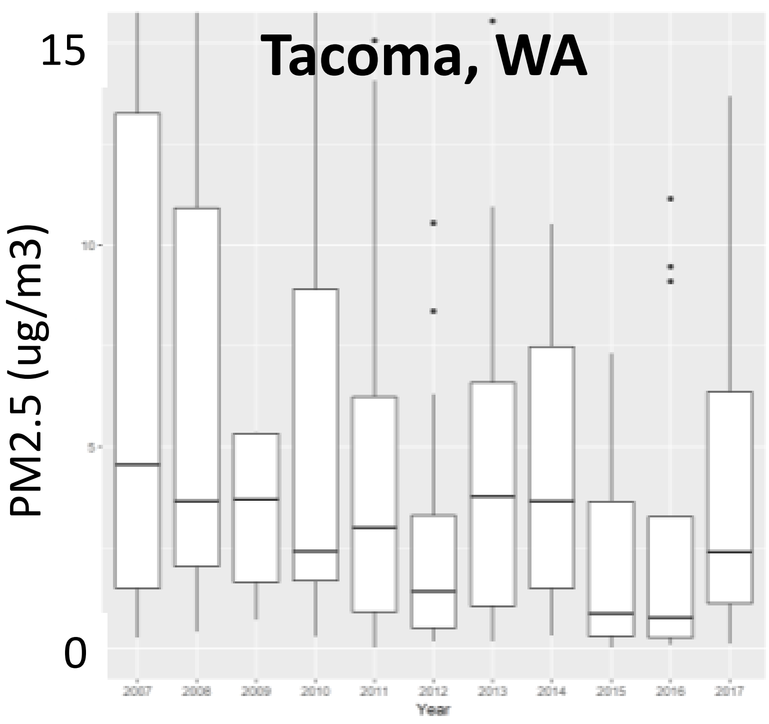
- OC and EC dominate
- OC components shifted to higher boiling fractions (more oxidative processing)
- Higher OC/EC ratio then fresh smoke
- K contribution
- No Cl contribution

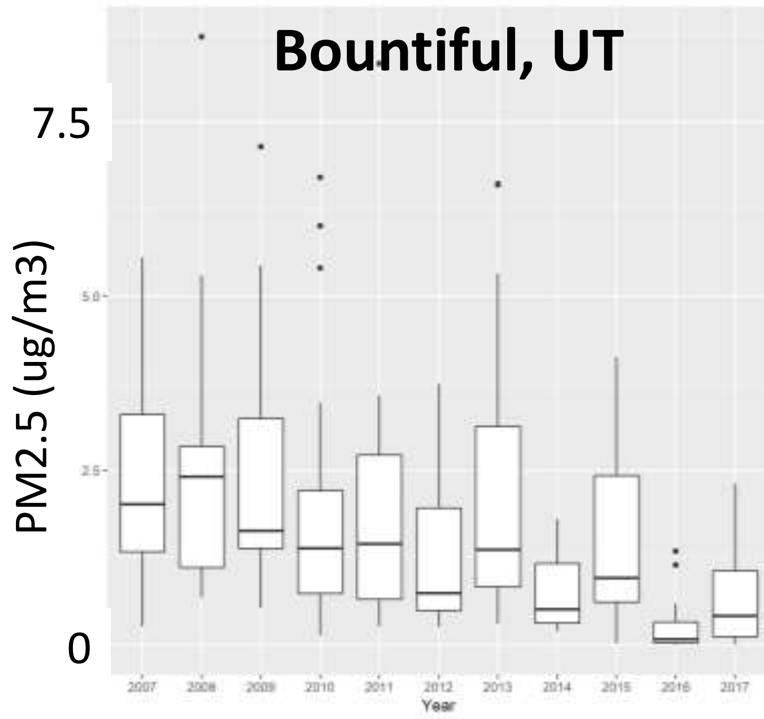
<u>In winter</u>, fresh + aged = RWC

<u>In summer</u>, fresh + aged = wild, Rx, Ag fire & SOA

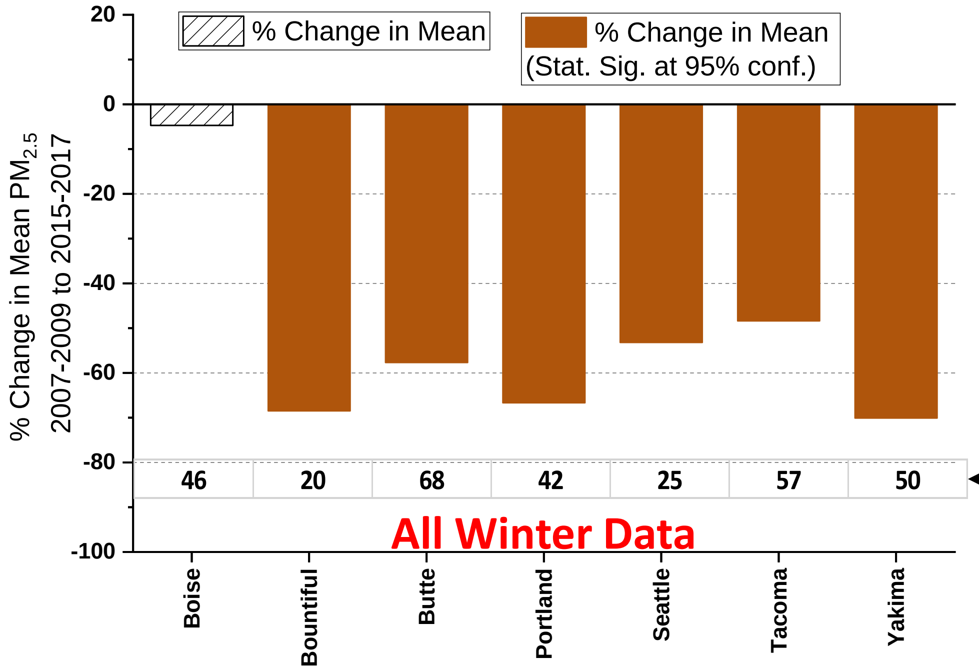
Boxplot examples, winter wood smoke $PM_{2.5}$ (fresh + aged)





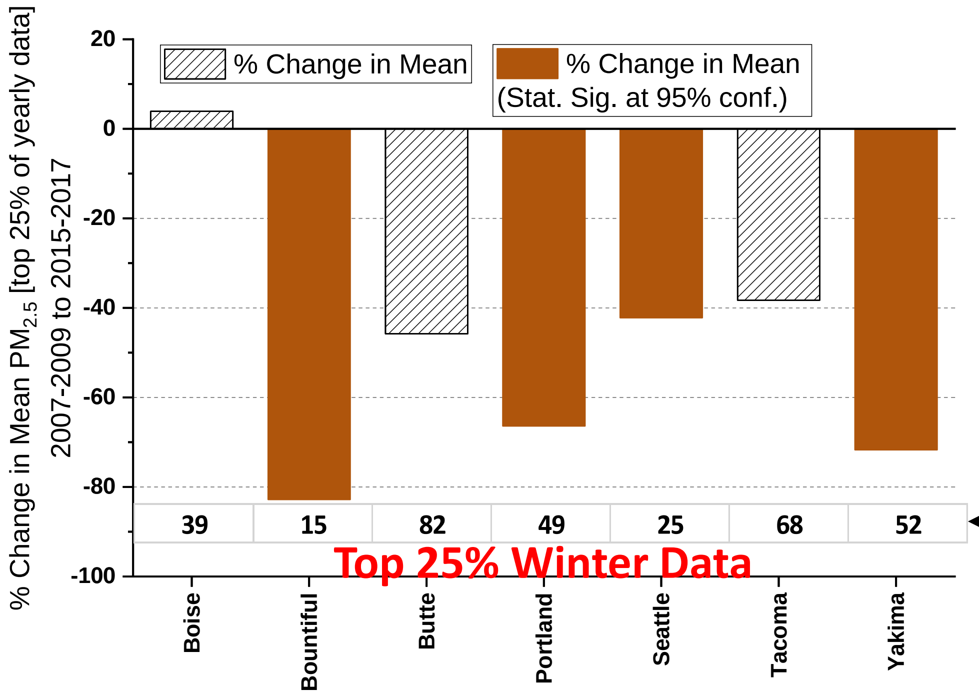


Change in 3-year average winter $PM_{2.5}$ from wood smoke (fresh + aged) [2007-2009 to 2015-2017]



Percent contribution of beginning 2007-2009 winter PM2.5 mass

Change in 3-year average winter $PM_{2.5}$ from wood smoke (fresh + aged) [2007-2009 to 2015-2017] for top 25% of yearly data



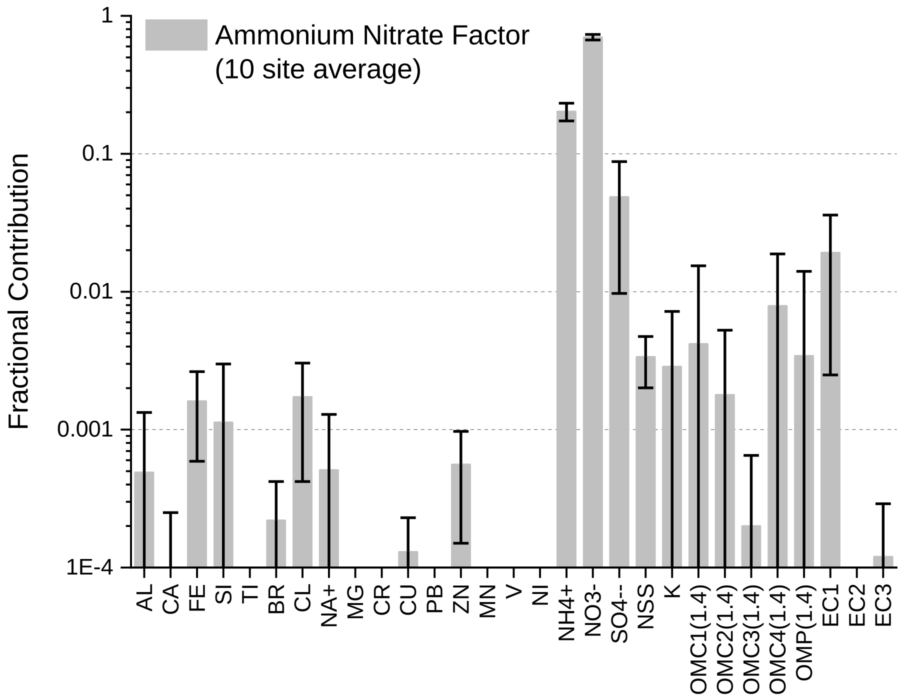
Percent contribution of beginning 2007-2009 winter PM2.5 mass (top 25% of yearly data)

Results for Winter Ammonium Nitrate $PM_{2.5}$

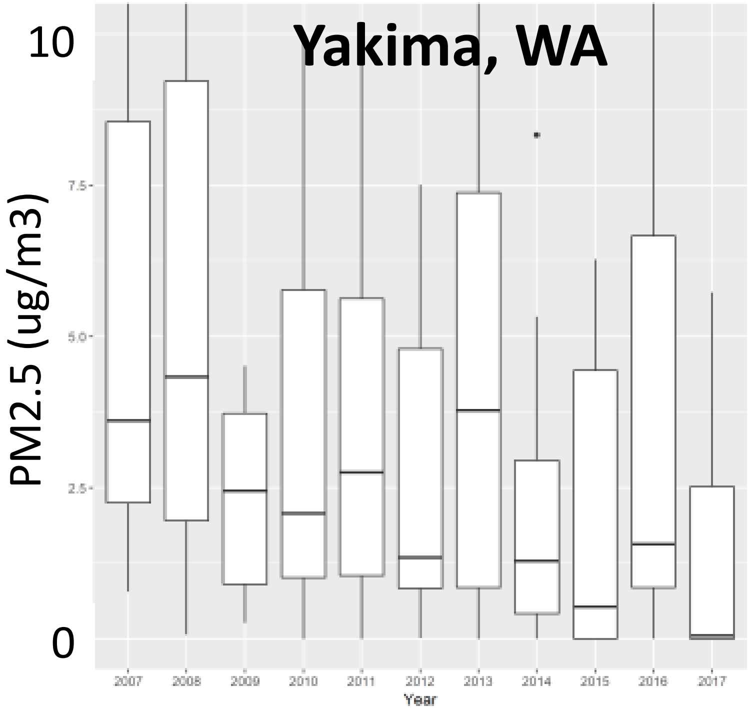
(2007-2009 to 2015 – 2017)

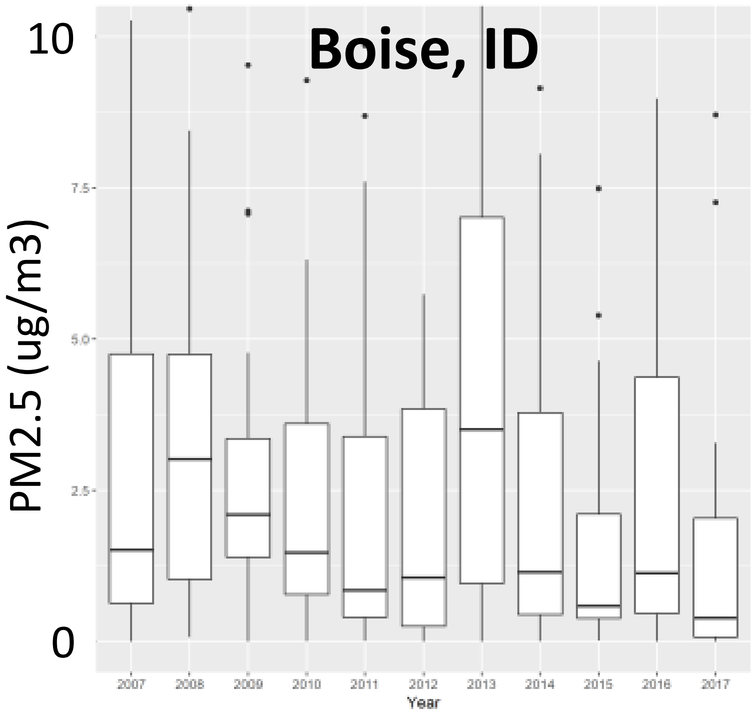
How was Ammonium Nitrate Identified in the Chemical Data?

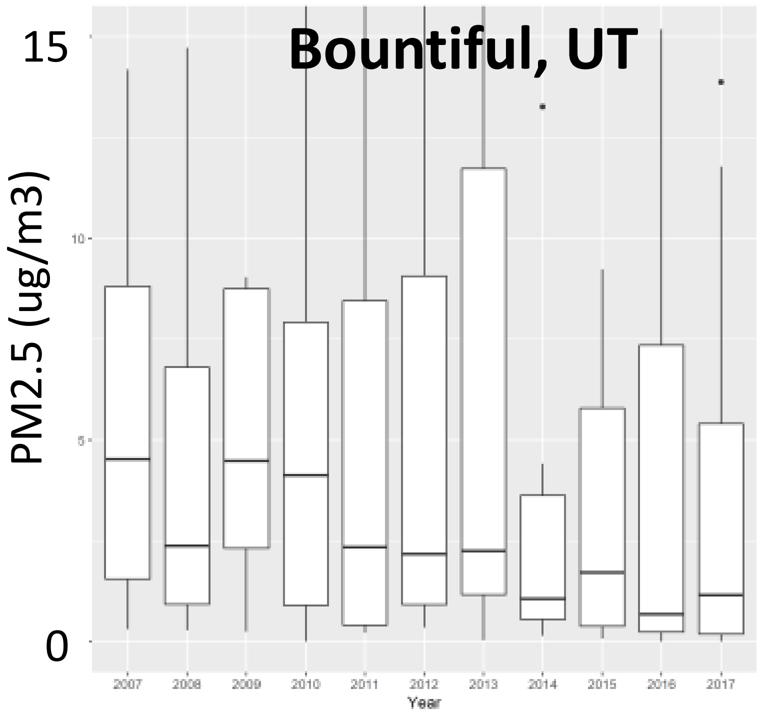
(Figure from R.A. Kotchenruther / Atmospheric Environment 142 (2016) 210-219)



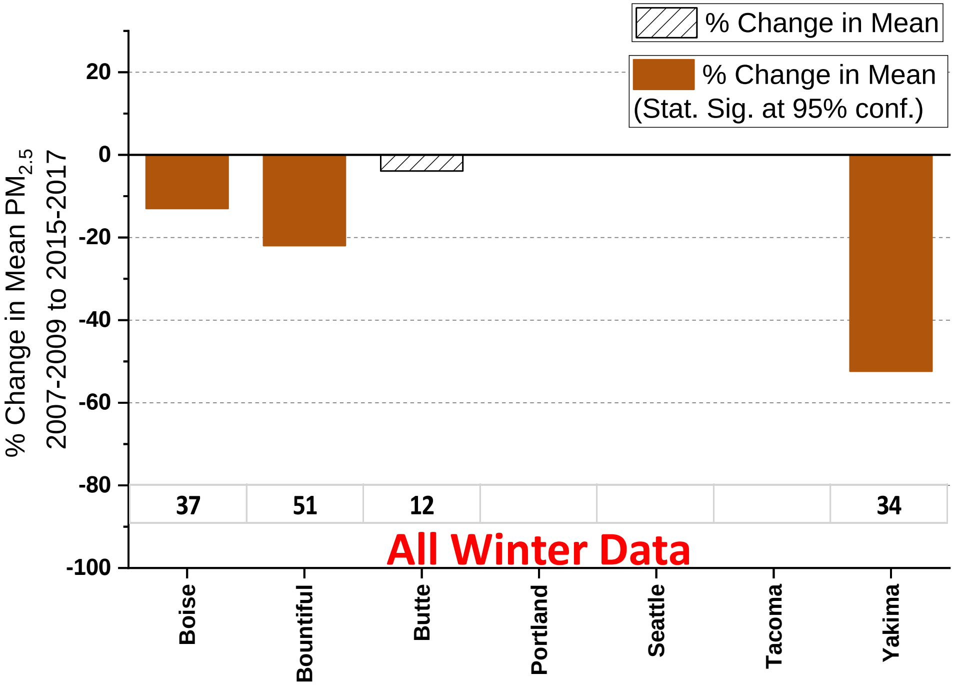
Boxplot examples, winter ammonium nitrate PM2.5





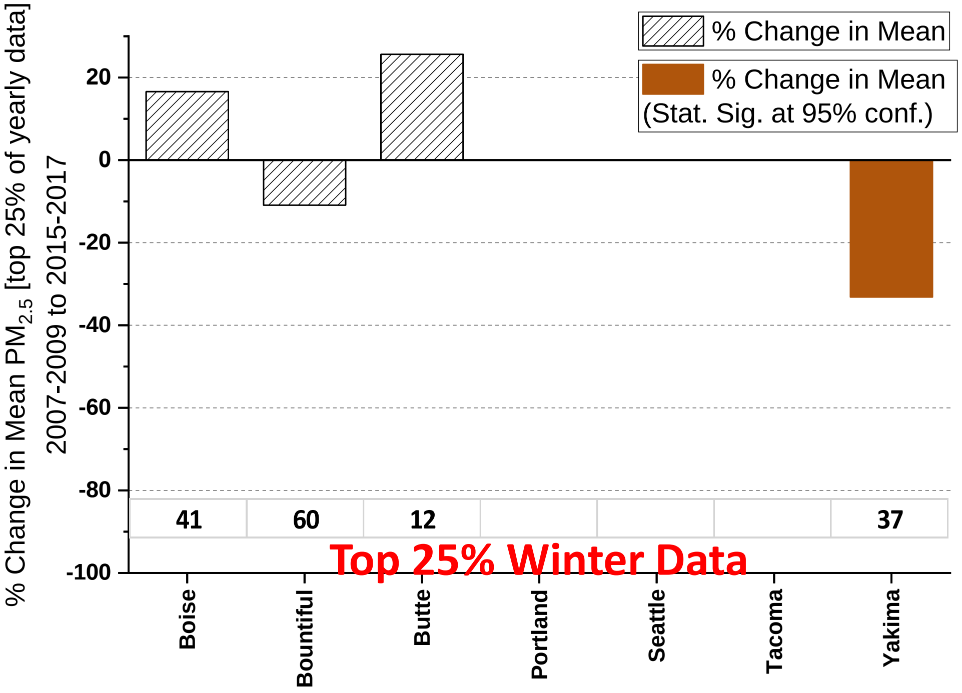


Change in 3-year average winter $PM_{2.5}$ from ammonium nitrate [2007-2009 to 2015-2017]



Percent contribution of beginning 2007-2009 winter PM2.5 mass

Change in 3-year average winter $PM_{2.5}$ from ammonium nitrate [2007-2009 to 2015-2017] for top 25% of yearly data



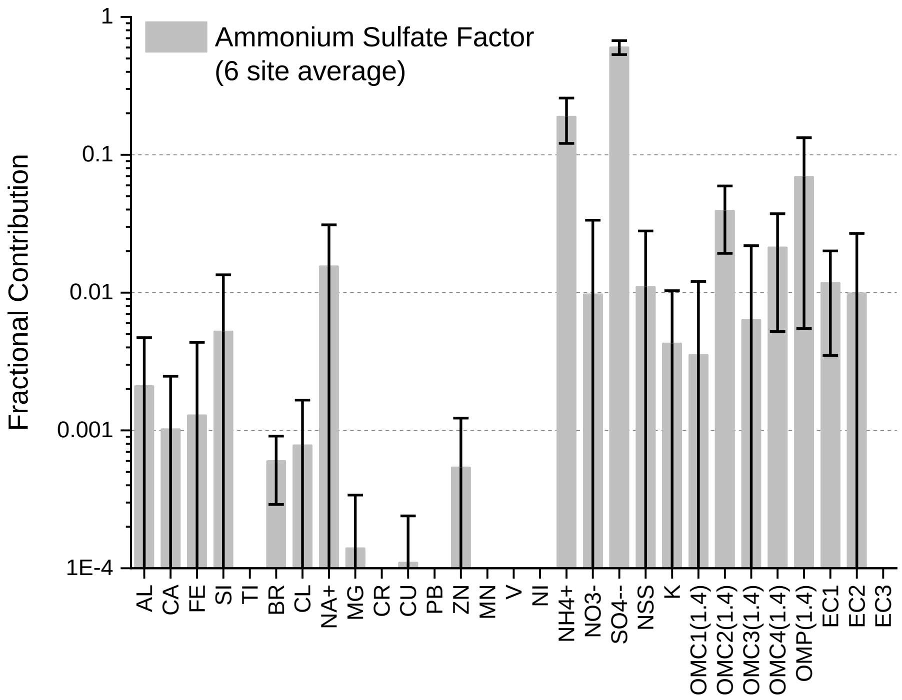
Percent contribution of beginning 2007-2009 winter PM2.5 mass (top 25% of yearly data)

Results for Winter Ammonium Sulfate $PM_{2.5}$

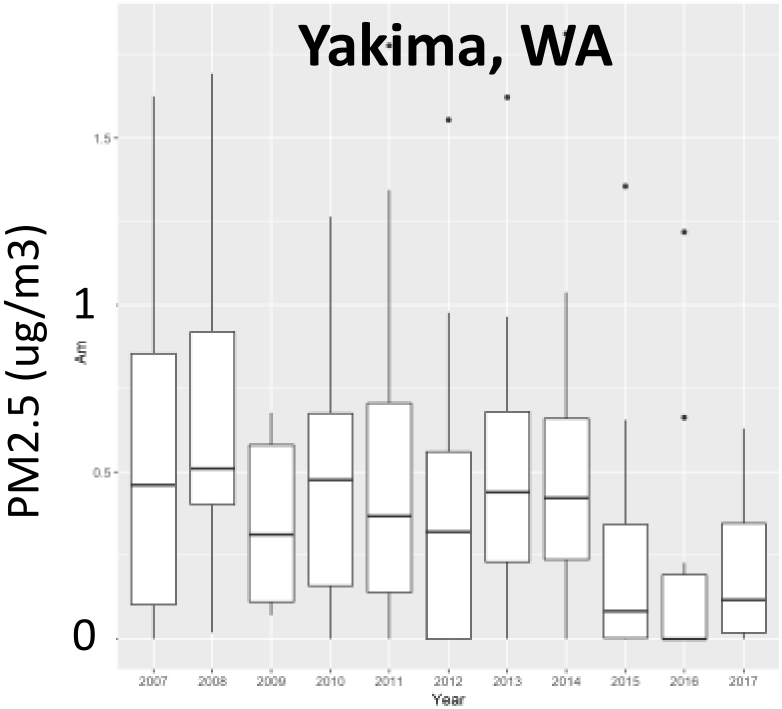
(2007-2009 to 2015 – 2017)

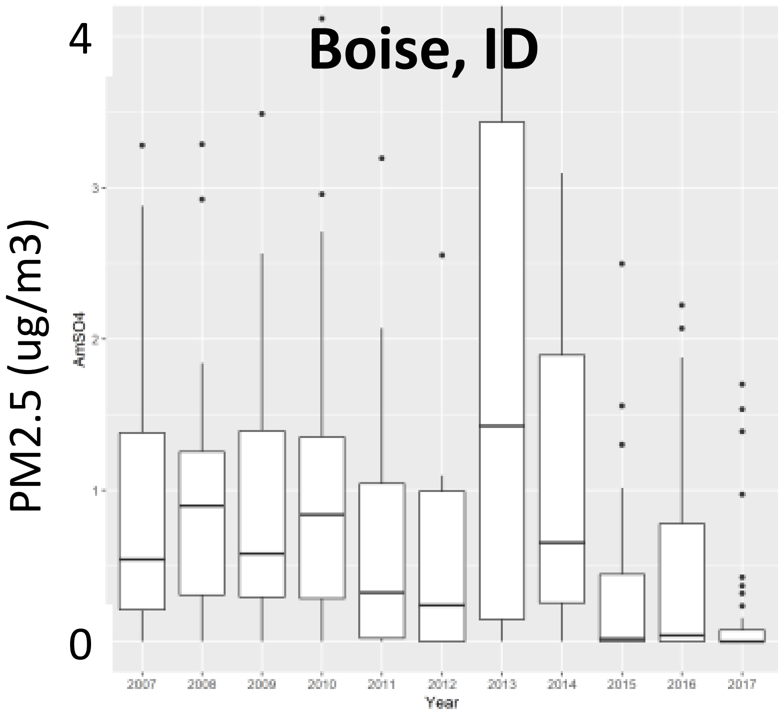
How was Ammonium Sulfate Identified in the Chemical Data?

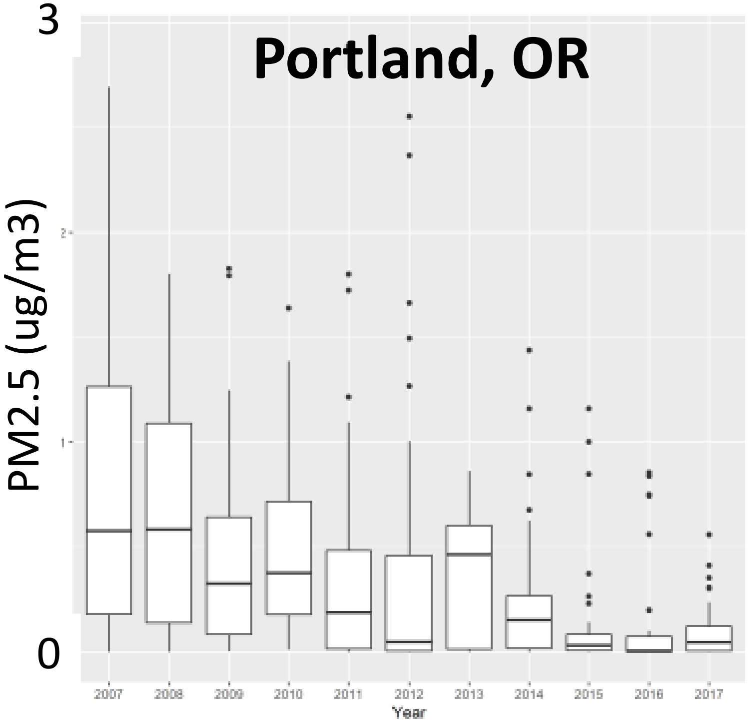
(Figure from R.A. Kotchenruther / Atmospheric Environment 142 (2016) 210-219)

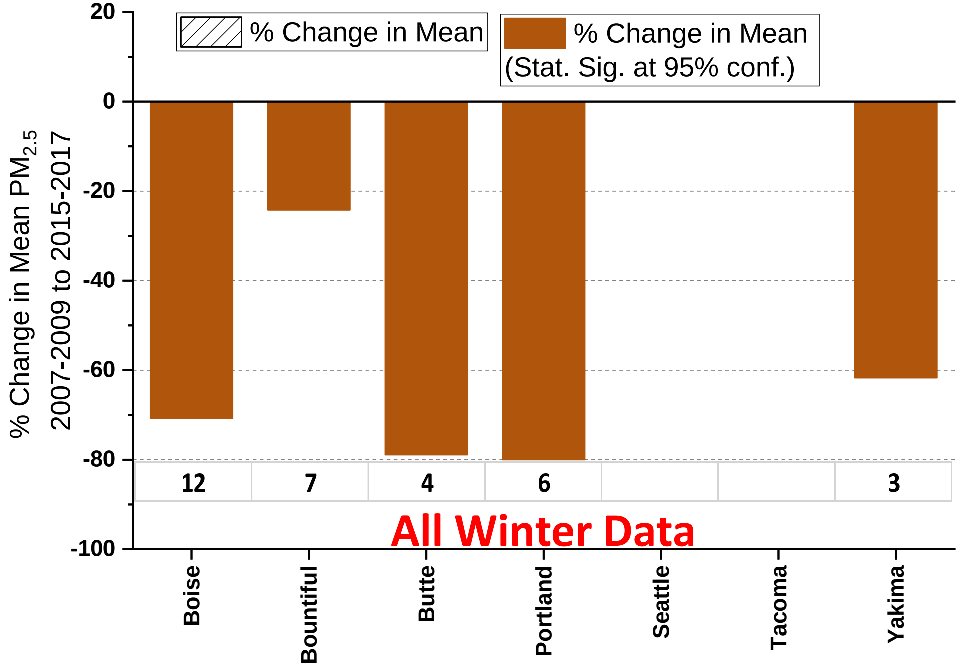


Boxplot examples, winter ammonium sulfate PM2.5



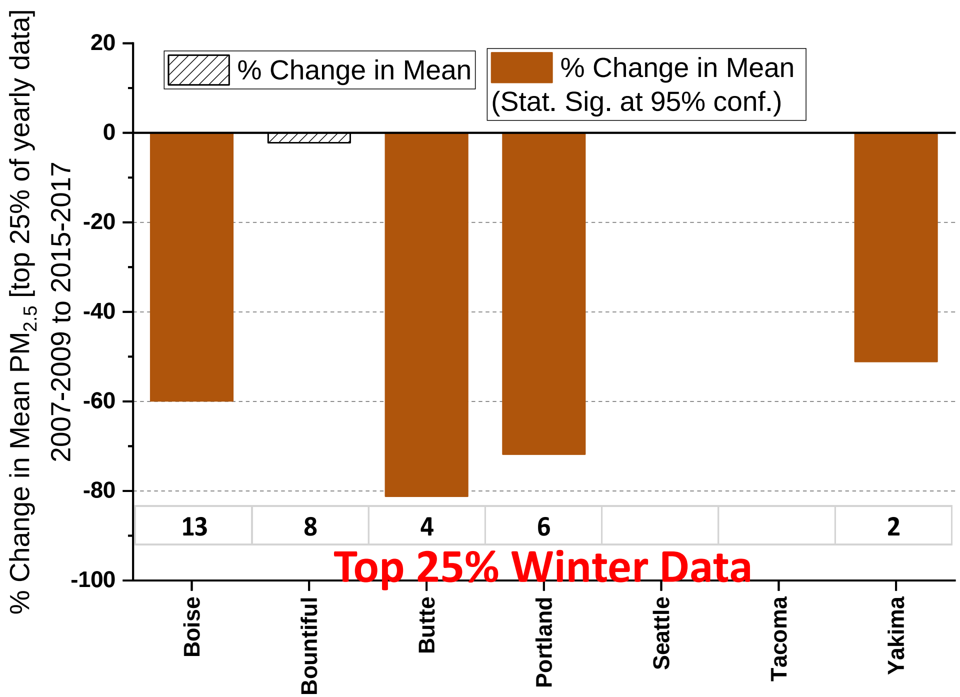






Change in 3-year average winter $PM_{2.5}$ from ammonium sulfate [2007-2009 to 2015-2017]

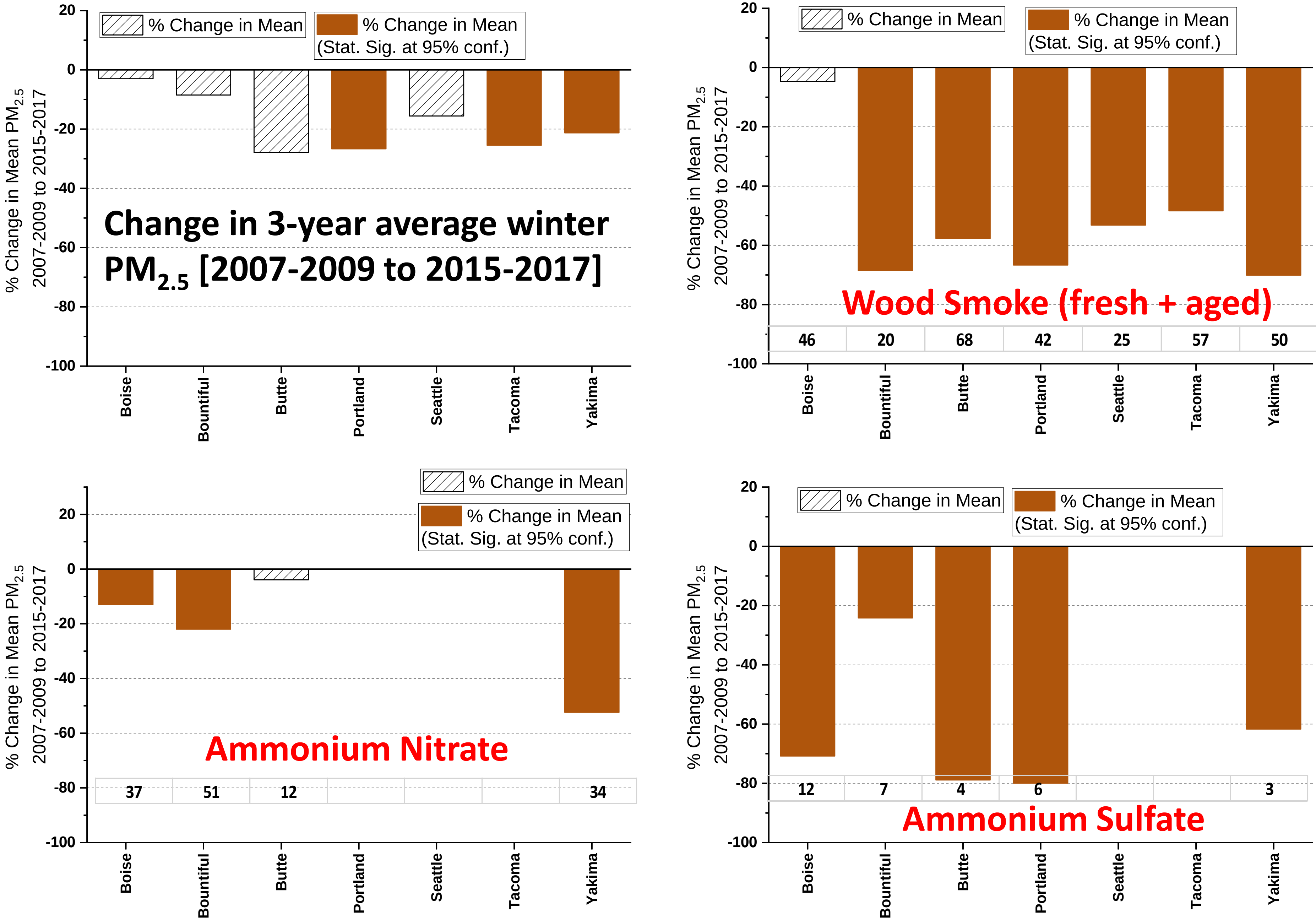
Percent contribution of beginning 2007-2009 winter PM2.5 mass



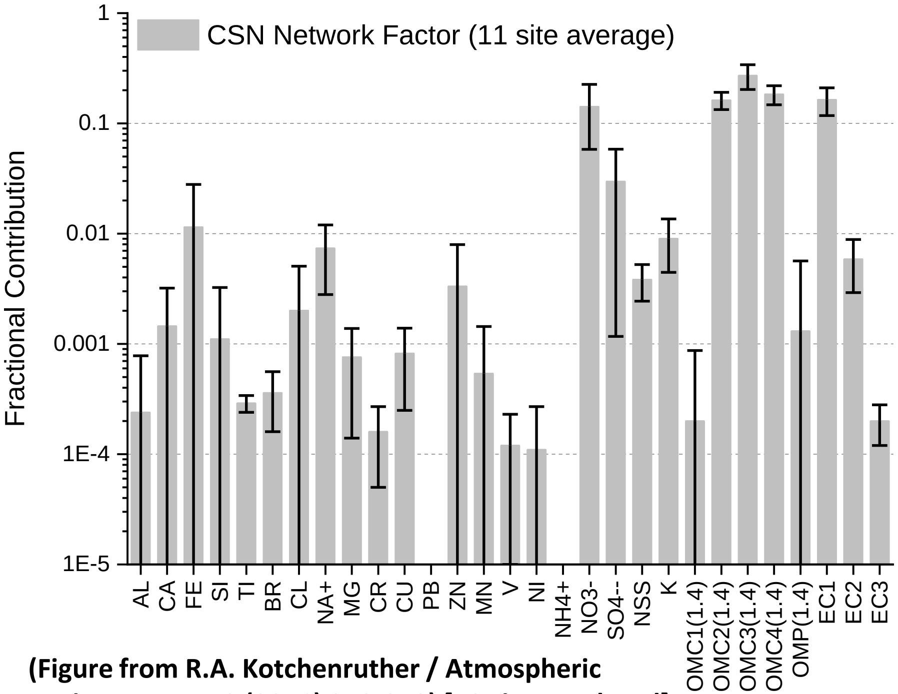
Change in 3-year average winter $PM_{2.5}$ from ammonium sulfate [2007-2009 to 2015-2017] for top 25% of yearly data

Percent contribution of beginning 2007-2009 winter PM2.5 mass (top 25% of yearly data)

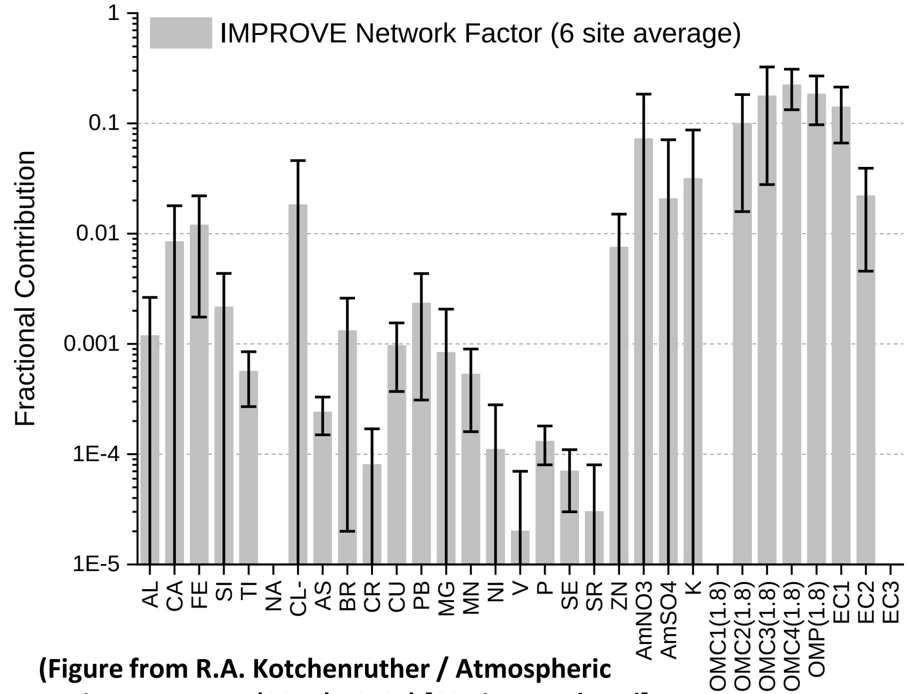
Many important contributors to $PM_{2.5}$ going down, why not total $PM_{2.5}$ so much?



One chemical pattern in the $PM_{2.5}$ data has been increasing. What is it?



(Figure from R.A. Kotchenruther / Atmospheric Environment 142 (2016) 210-219) [19 sites analyzed]



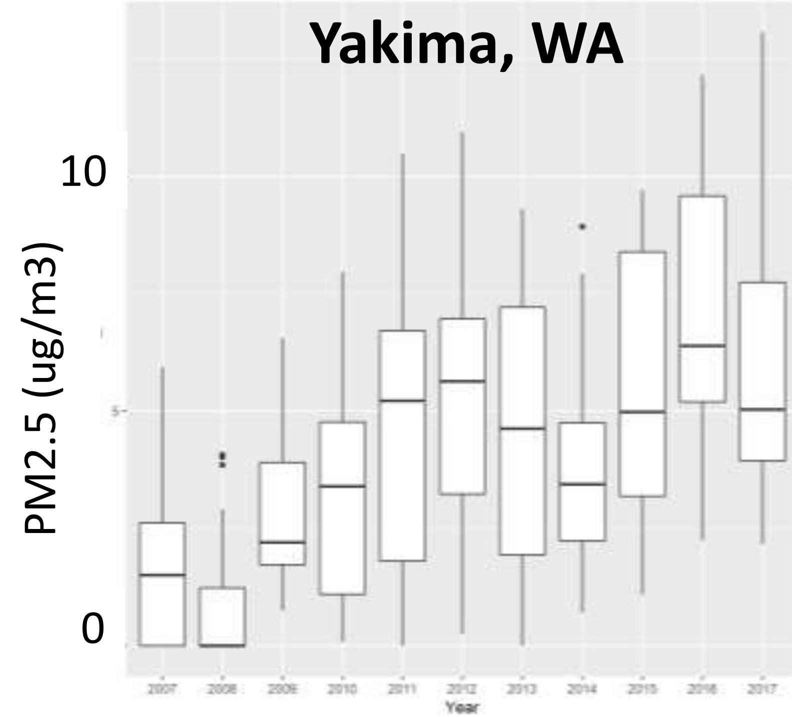
(Figure from R.A. Kotchenruther / Atmospheric Environment 151 (2017) 52-61) [22 sites analyzed]

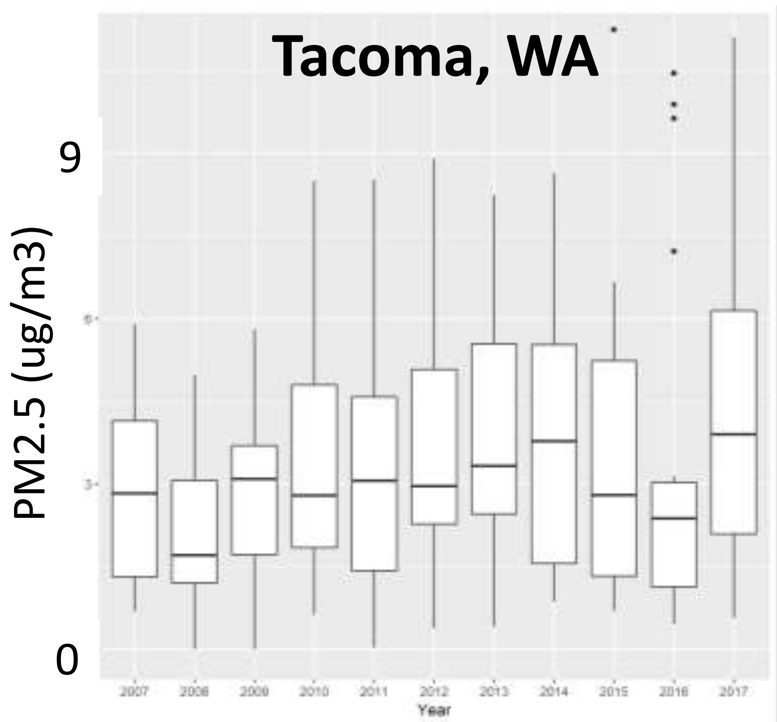
<u>What do we know?</u>

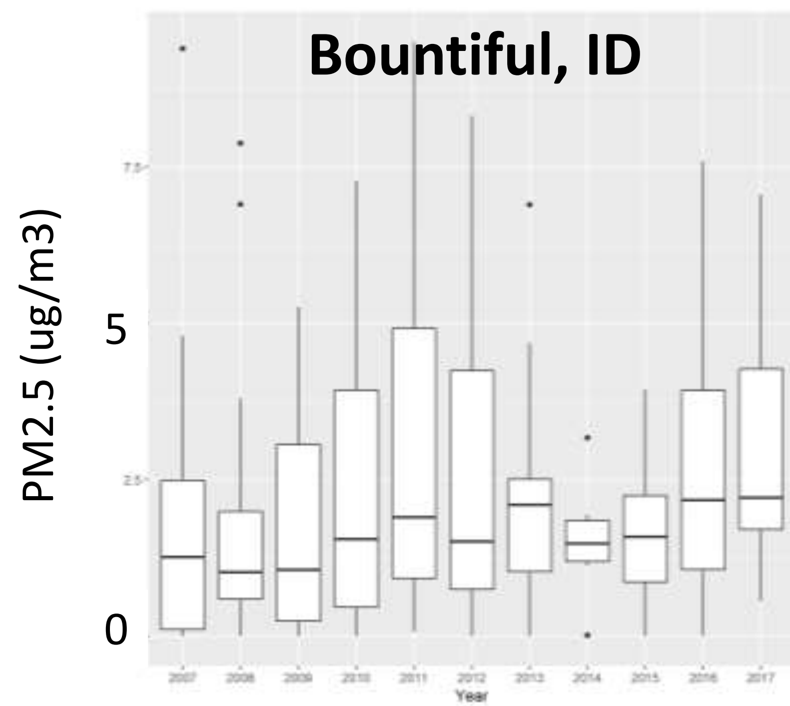
- **Ubiquitous**, appears in PMF analysis at every CSN and IMPROVE site (either on own or mixed with other patterns/sources)
- **OC & EC dominate**, **->** some kind of fuel combustion.
- **Not associated with wood smoke factors.**
- **Trace metals**, Fe, Ti, Cu, Zn.
- **IMPROVE factor,** OC shifted to higher thermal evolution components (OC4 & OP) -> Likely more atmospheric oxidative processing (more aged from source)
- **IMPROVE factor**, fewer sites where factor clearly resolved -> further form source
- **My take on what source fits this pattern …**

Gas Vehicles

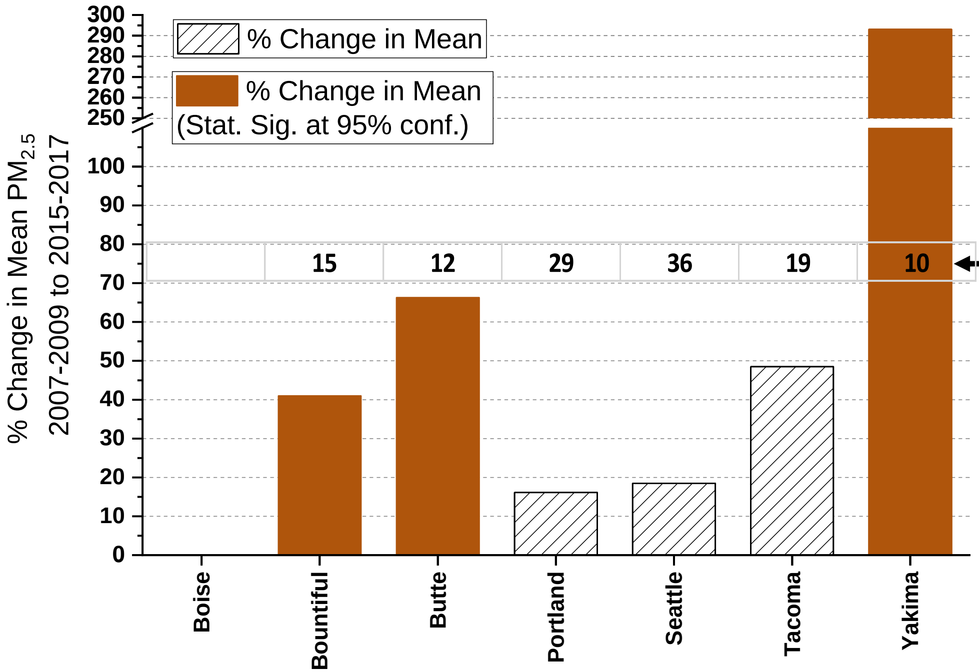
Boxplot examples, winter gas vehicles PM2.5





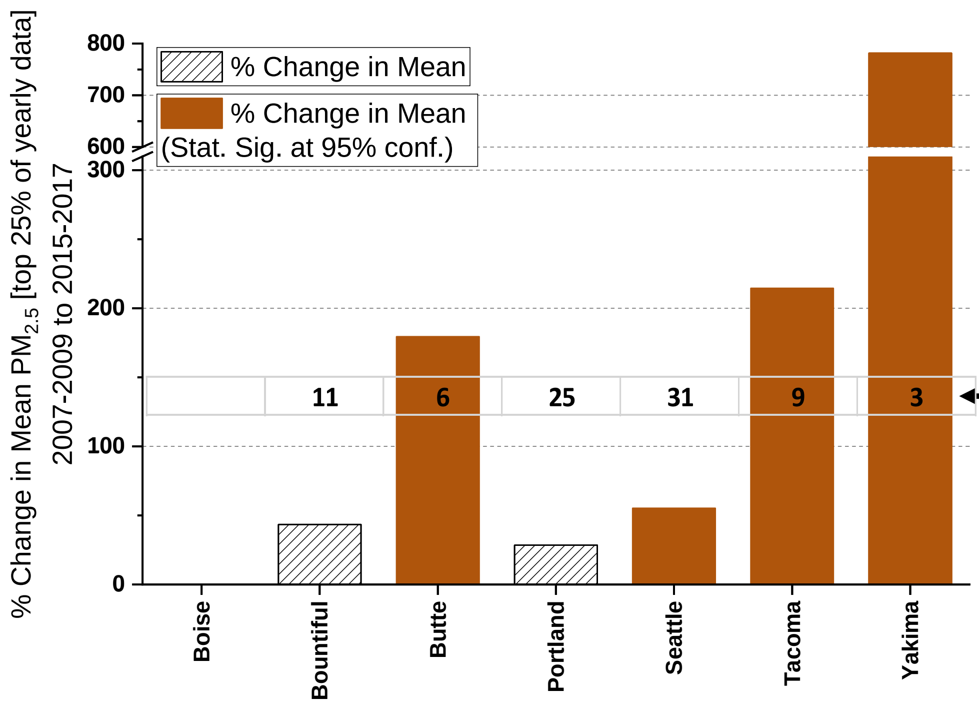


Change in 3-year average winter $PM_{2.5}$ from gas vehicles [2007-2009 to 2015-2017]



Percent contribution of beginning 2007-2009 winter PM2.5 mass

Change in 3-year average winter $PM_{2.5}$ from gas vehicles [2007-2009 to 2015-2017] for top 25% of yearly data



Percent contribution of beginning 2007-2009 winter PM2.5 mass (top 25% of yearly data)

What have other source apportionment efforts found?

Multi-site PMF analysis of New York State sites from 2005 – 2016

Squizzato et al., **A long-term source apportionment of PM2.5 in New York State during 2005–2016** Atmospheric Environment 192 (2018) 35–47
Masiol et al., **Long-term trends (2005–2016) of source apportioned PM2.5 across New York State** Atmospheric Environment 201 (2019) 110–120

Similar story

Masiol et al., 2019 [from abstract]
"Spark-ignition vehicles were the only source type experiencing upward annual trends at all urban sites with slopes ranging from 0.02 µg/m3/y to ~0.2 µg/m3/y …"

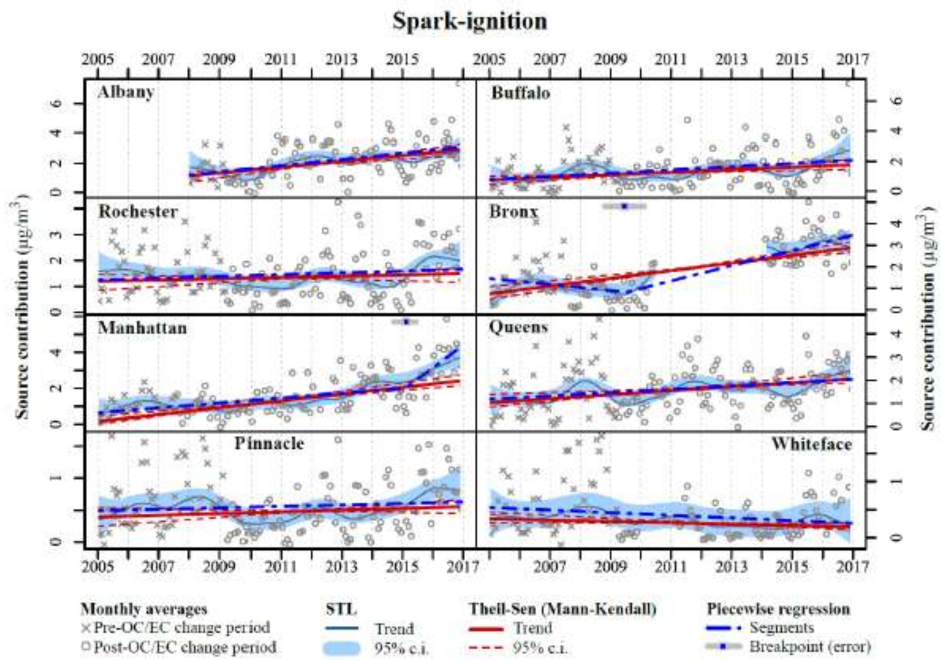
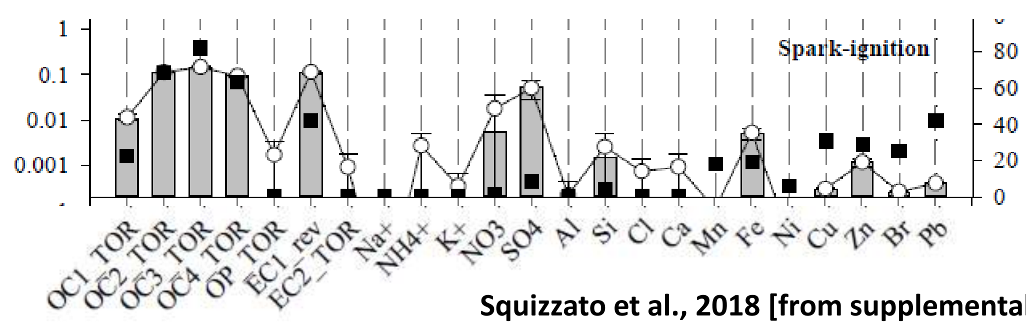


Figure S15. Results of trend analyses for the spark-ignition source computed over all the year (STL in light blue; Theil-Sen nonparametric estimator of slope in red; piecewise linear regression model with breakpoints estimation in dark blue). The confidence intervals (c.i. at 0.05 level) for STL and Theil-Sen slopes are computed by bootstrap estimation. **Masiol et al., 2019 [from supplemental]**

PMF chemical pattern / factor for 'Spark–ignition'

Same factor / pattern as identified in this work



Squizzato et al., 2018 [from supplemental]

Why is this chemical pattern / source increasing?

Masiol et al., 2019 attribute increases in this factor to:

- **a similar patter in increases in registered vehicles**
- **gas vehicle emissions are a significant source of secondary organic aerosol precursors**

Some questions about the gas vehicle pattern …

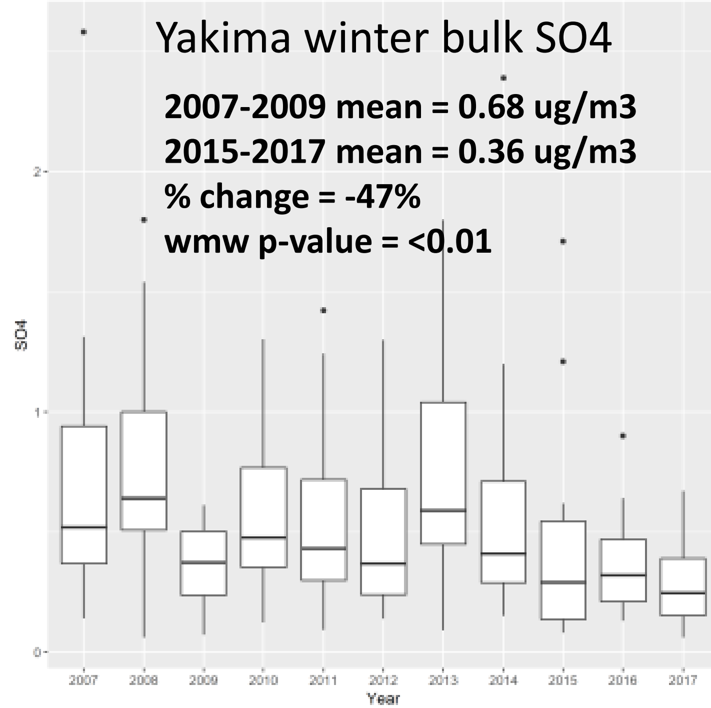
- **Why such a large increase in Yakima?**
- **Can it really all be attributed to increasing vehicle population?**
- **Why is ammonium nitrate going down in Yakima (52% reduction in mean from 2007-2009 to 2015-2017), if this pattern is going up? (i.e., if most of NOx is from vehicles)**
  (note: 2007-2009 to 2015-2017 mean monitored NO3 down only 17%, mean monitored NH4 down 58%, mean SO4 down 47%)
- **Maybe there is a shift in winter nitrate chemistry, away from ammonium nitrate production, and towards organic nitrate production?**

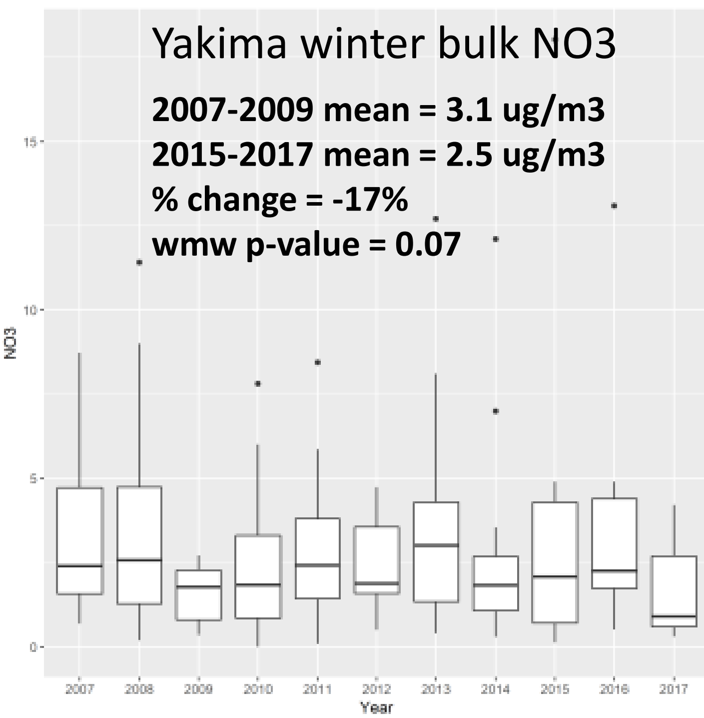
Your thoughts?

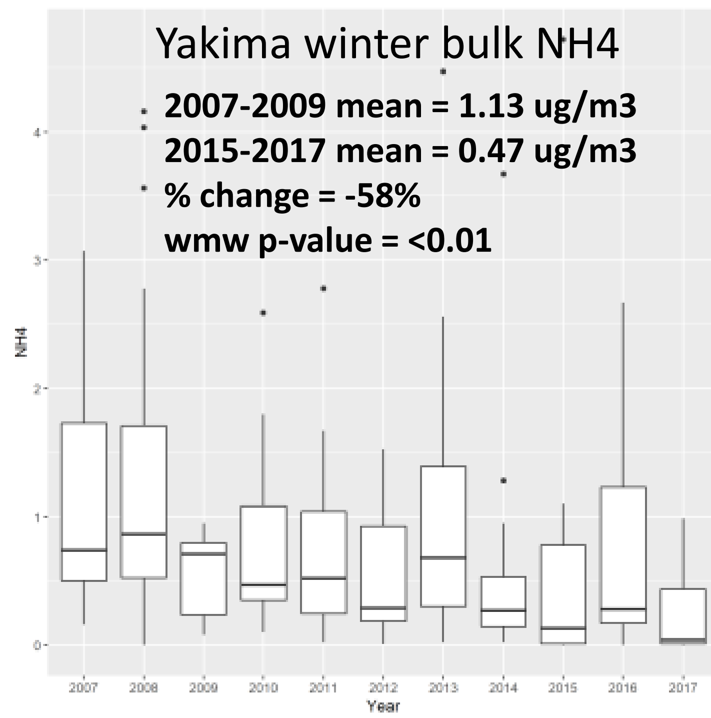

Thank You!
Questions?

Supplementary Slides

Change in Yakima winter monitored NO3, NH4, and SO4





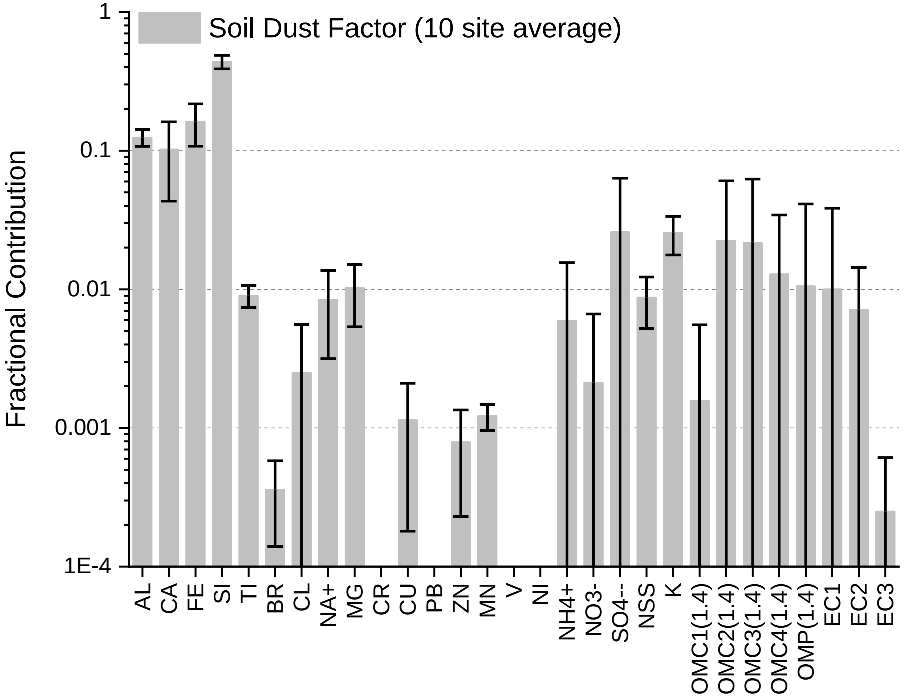


Results for Winter
Soil Dust $PM_{2.5}$

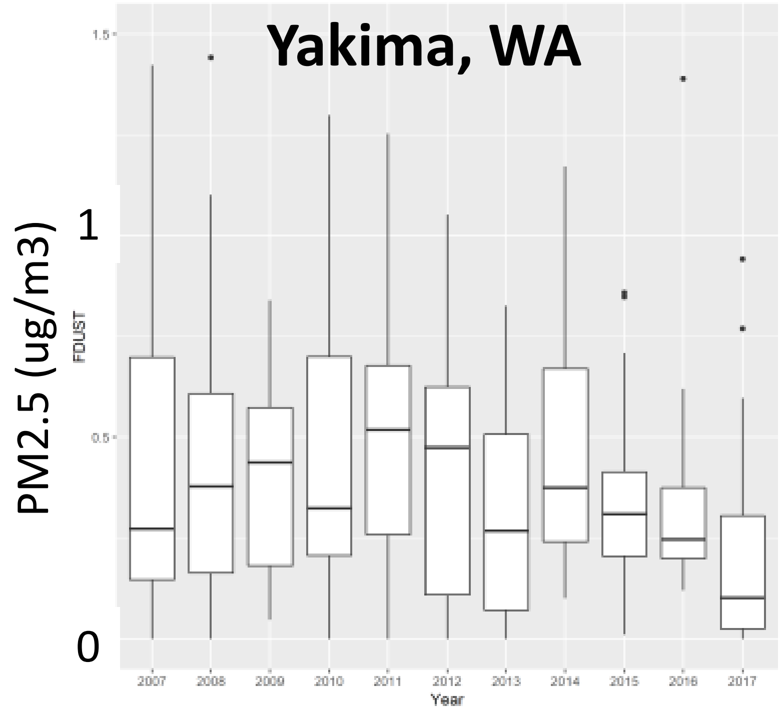
(2007-2009 to 2015 – 2017)

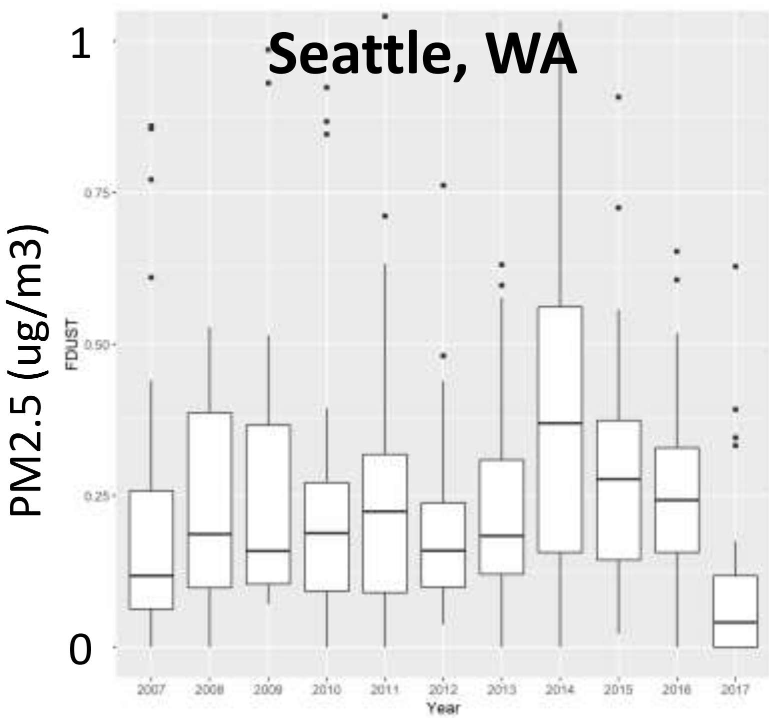
How was Soil Dust Identified in the Chemical Data?

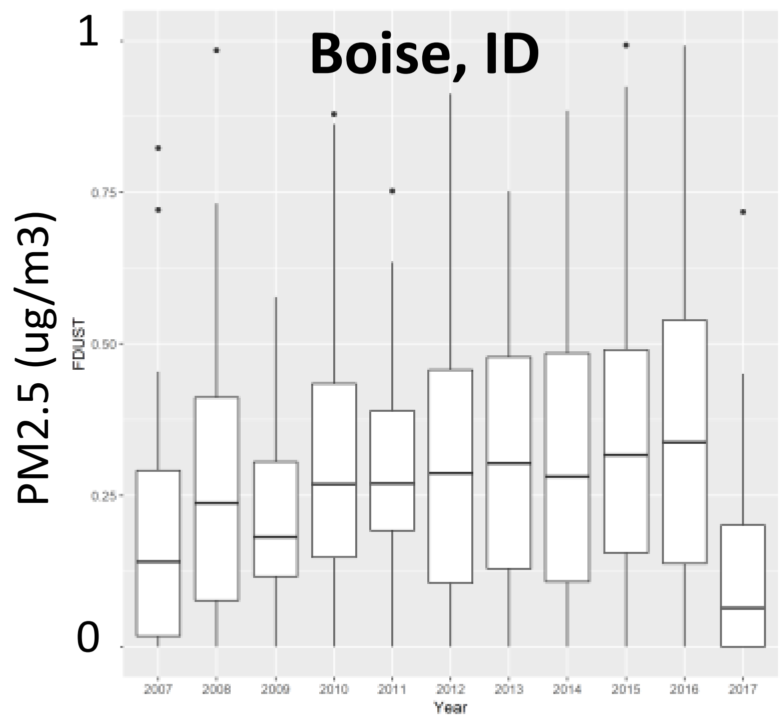
(Figure from R.A. Kotchenruther / Atmospheric Environment 142 (2016) 210-219)



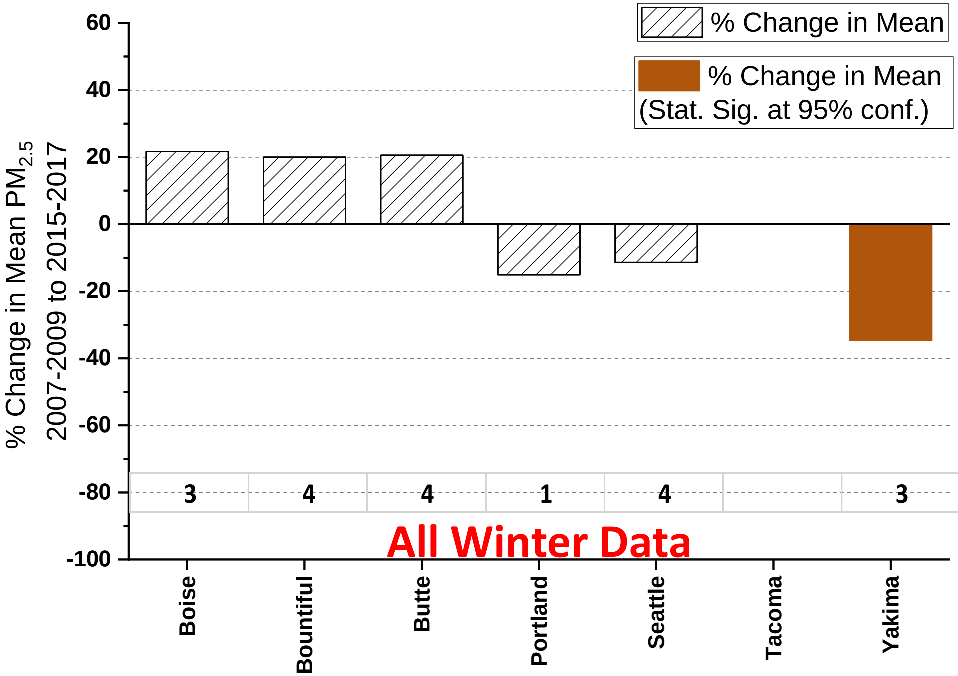
Boxplot examples, winter soil dust PM2.5





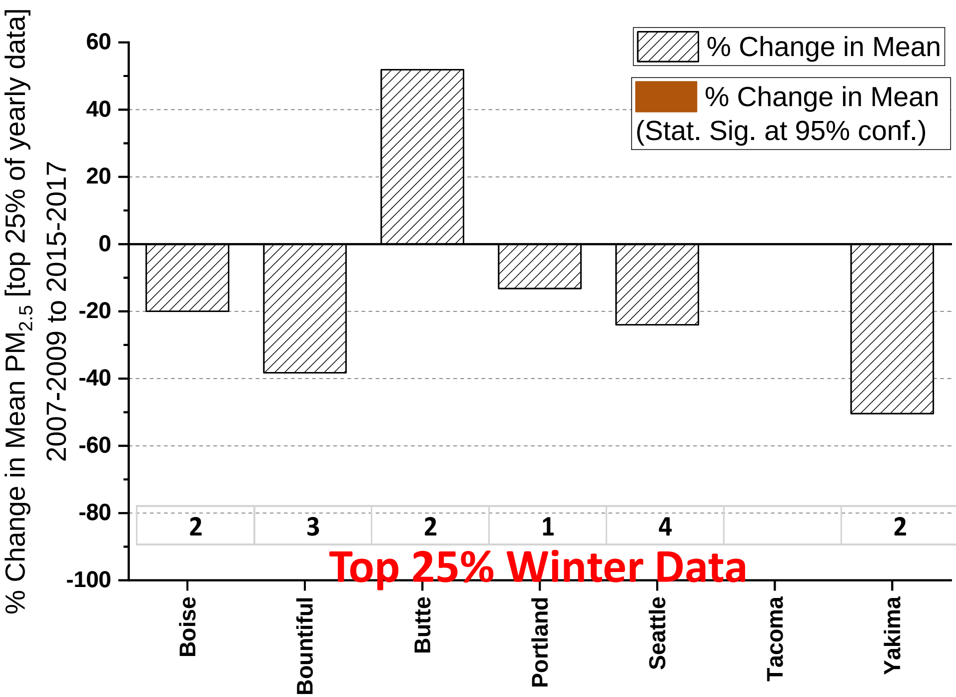


Change in 3-year average winter $PM_{2.5}$ from soil dust [2007-2009 to 2015-2017]



Percent contribution of beginning 2007-2009 winter PM2.5 mass

Change in 3-year average winter $PM_{2.5}$ from soil dust [2007-2009 to 2015-2017] for top 25% of yearly data



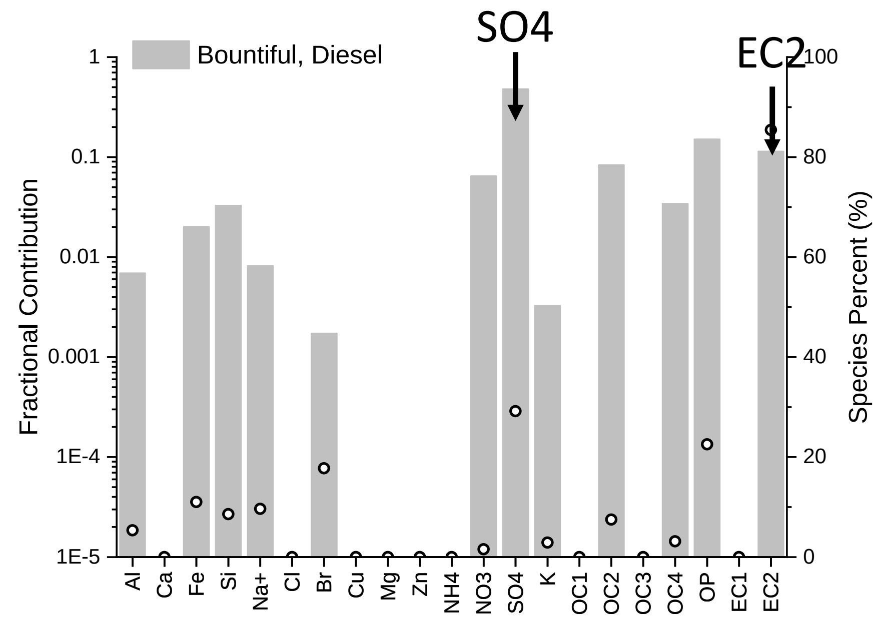
Percent contribution of beginning 2007-2009 winter PM2.5 mass (top 25% of yearly data)

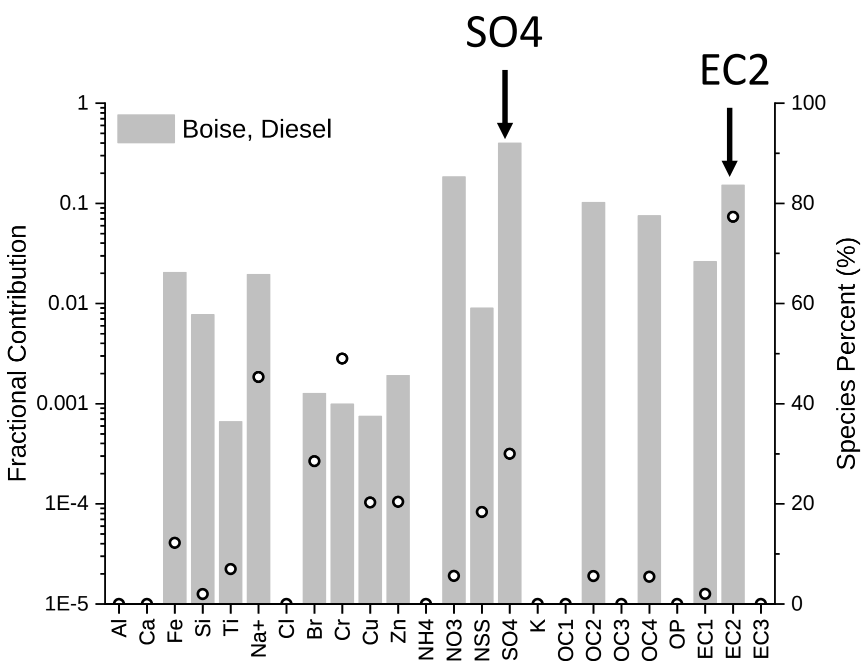
Results for Winter Diesel Engine $PM_{2.5}$

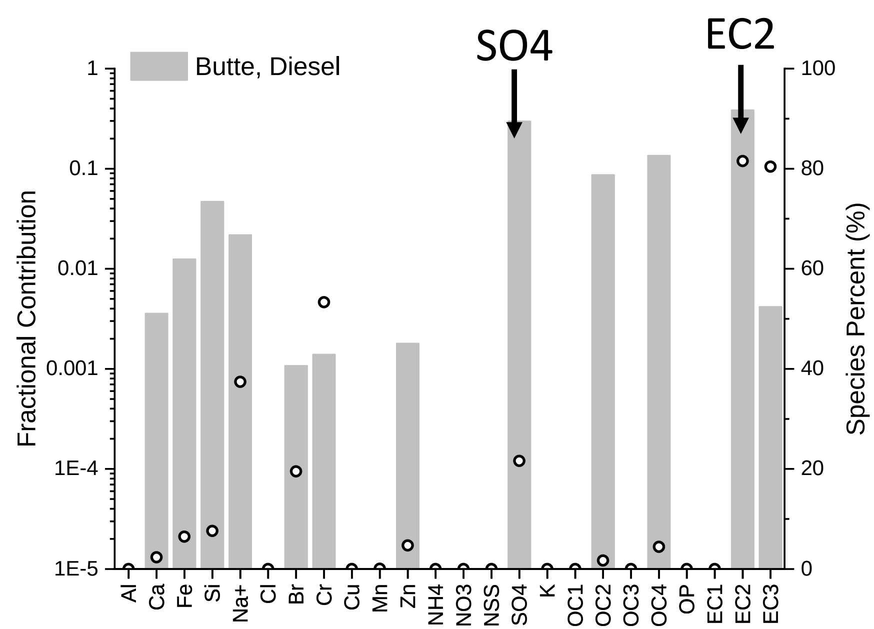
(2007-2009 to 2015 – 2017)

How were Diesel Engines Identified in the Chemical Data?

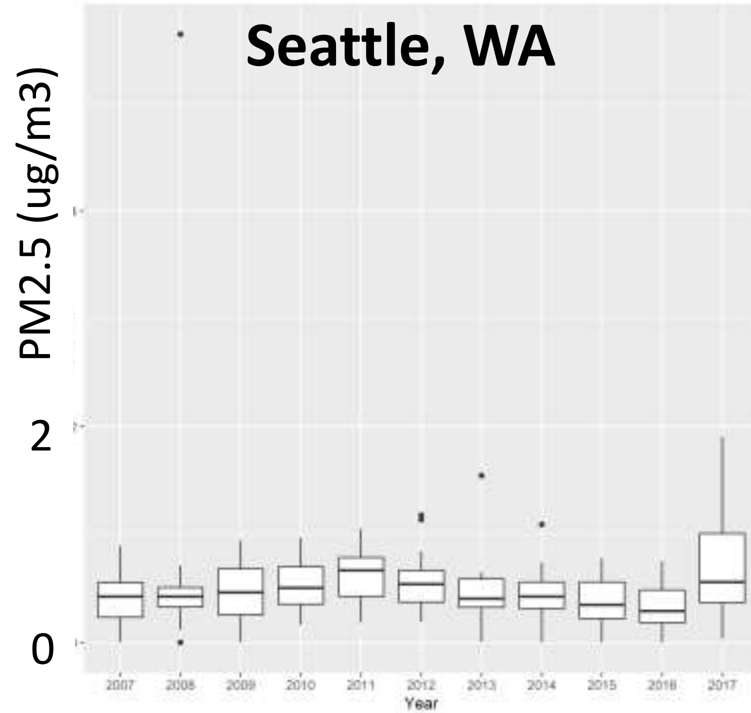
SO4 + EC2

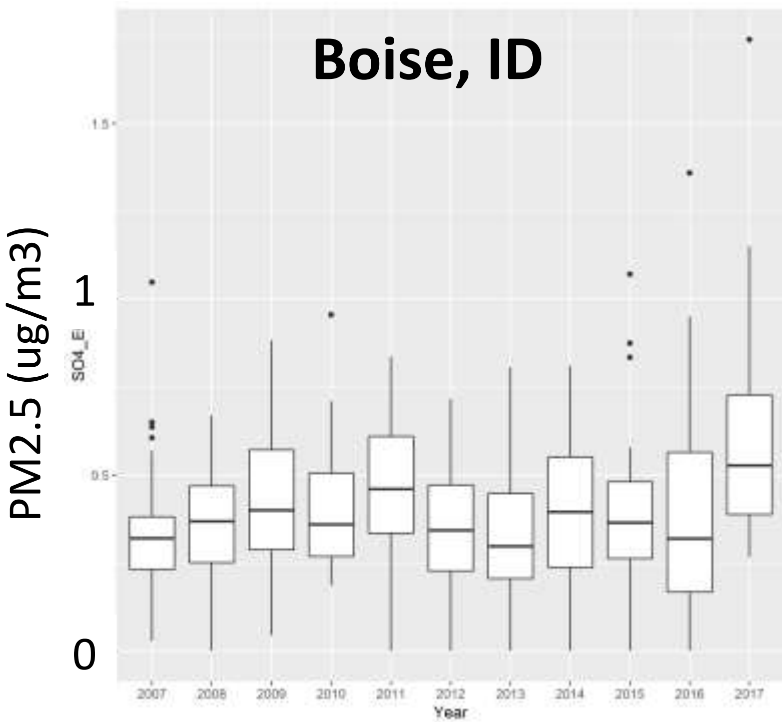


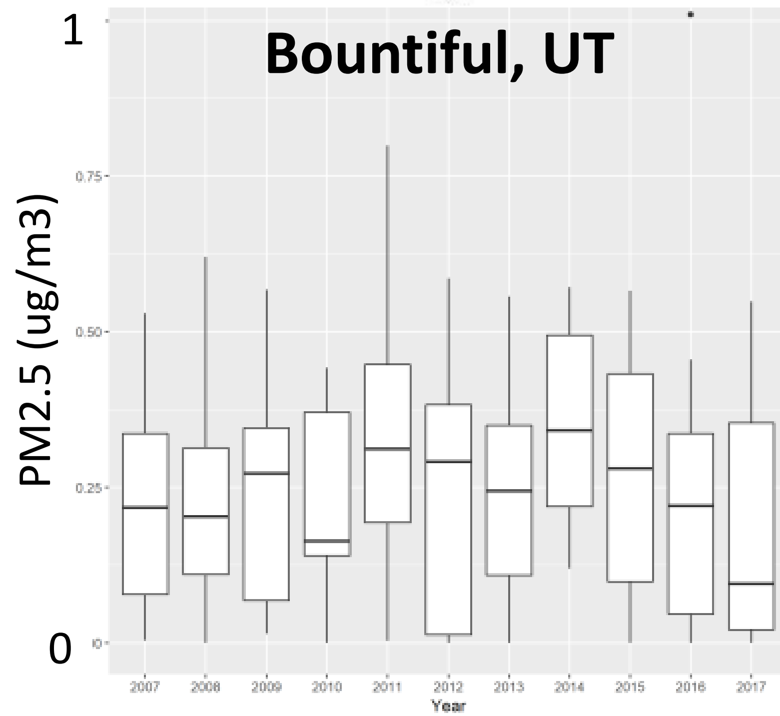




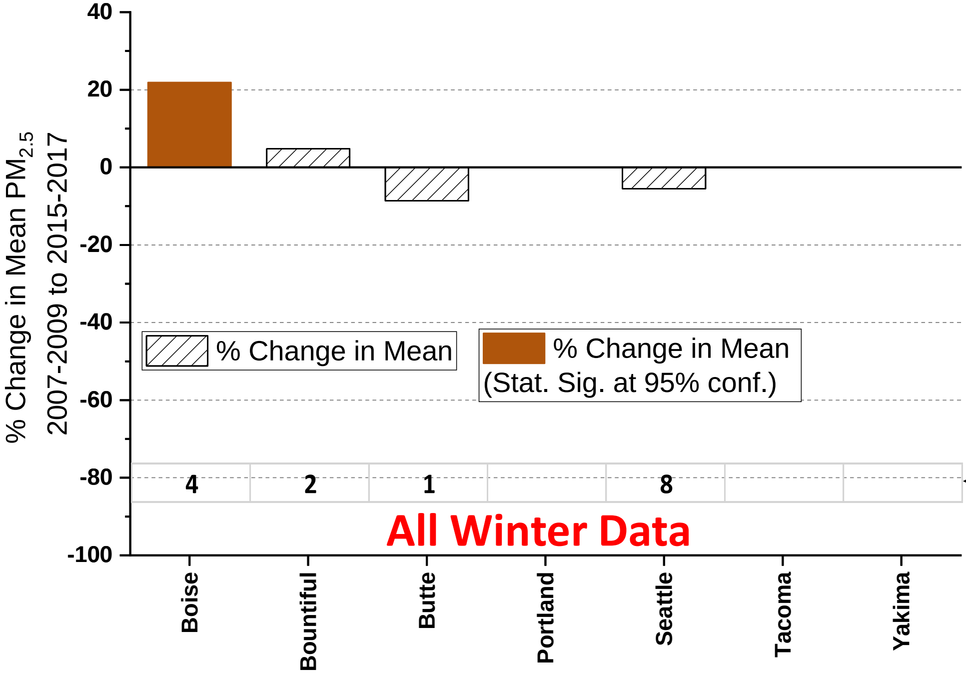
Boxplot examples, winter diesel engine PM2.5





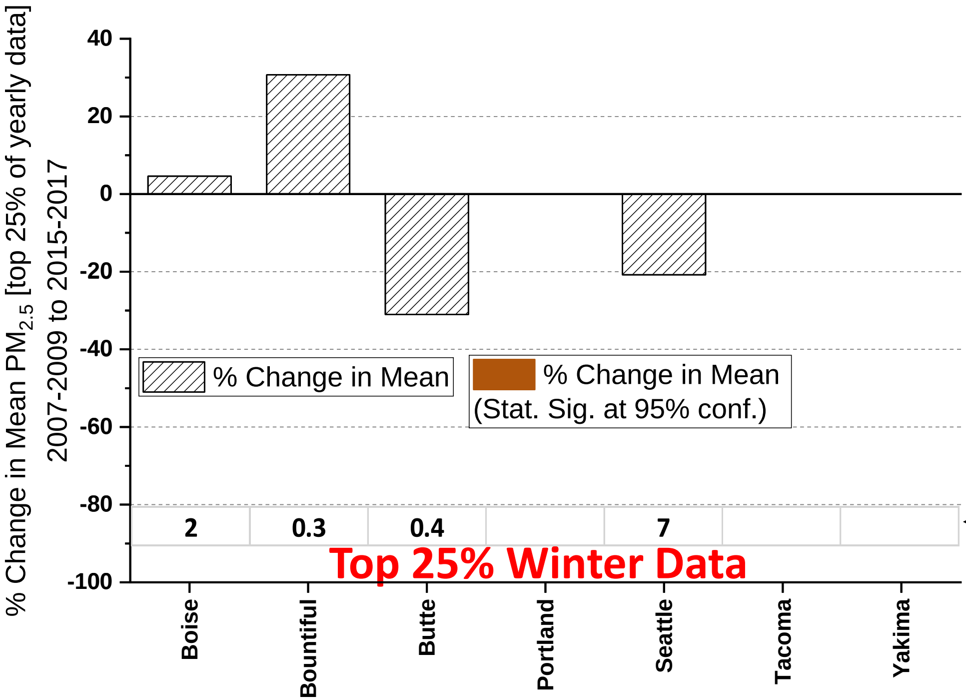


Change in 3-year average winter $PM_{2.5}$ from diesel engines [2007-2009 to 2015-2017]



Percent contribution of beginning 2007-2009 winter PM2.5 mass

Change in 3-year average winter $PM_{2.5}$ from diesel engines [2007-2009 to 2015-2017] for top 25% of yearly data



Percent contribution of beginning 2007-2009 winter PM2.5 mass (top 25% of yearly data)