PM2.5 Saturation Study

Clallam County, Washington

January 2013 through March 2014

Prepared by:

Odelle Hadley, PhD

Olympic Region Clean Air Agency

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1.0 EXECUTIVE SUMMARY

The Olympic Region Clean Air Agency (ORCAA) maintains at least one air quality monitor in each of its six regulated counties: Clallam, Grays Harbor, Jefferson, Mason, Pacific, and Thurston. A saturation study is one method used to determine the best location for placing the air monitoring stations. Prior to this study, the most recent Clallam County air quality saturation study was conducted during winter 1996/97. Data from the 1996/97 study identified Stevens Middle School as an ideal location for the Clallam County monitor. The following report describes the saturation study conducted in Clallam County between January 2013 and March 2014.

The goals of this study were:

1. Determine whether the Stevens Middle School nephelometer data was an accurate representation of regional ambient air quality in Clallam County
2. Evaluate neighborhood scale air quality variability
3. Identify the primary sources of PM2.5 in the region
4. Identify whether a new location would be more appropriate for the permanent monitor

Three monitoring sites were placed in Port Angeles and a fourth was established in Sequim. All four air monitoring sites in Clallam County were significantly correlated with each other. Stevens Middle School (SMS), the current long term monitoring site, generally reflected area wide air quality, however data collected during the saturation study revealed the Port Angeles Fire Station (PAFS) monitor correlated most highly with the other three sites and typically measured higher levels of particulate matter. The PAFS site best represented the region and statistically registered the highest PM2.5 concentrations.

Analysis of particle absorption properties, seasonal and daily PM2.5 variability, and meteorology in conjunction with PM2.5 helped identify the primary area sources. Residential wood heating is the dominant source of ambient PM2.5 during cold winter months. Concentrations are highest in the mornings and evenings when ambient air pollution levels are considered “moderate” under the Washington Air Quality Advisory (WAQA). Morning and evening maxima coincide with the time of day when fire places and wood stoves are lit: between 6:00 AM and 7:00 AM when many people wake up and between 4:30 PM and 6:00 PM when people return to their homes. Winter midday air quality is similar to summer air quality and is generally “good” in the WAQA index. Sequim has a similar winter daily pattern, but evening levels are not as high as in Port Angeles and on average, air quality is always good.

The average summertime PM2.5 concentrations were “good” at all four monitoring sites and data showed no significant daily variability. Summertime air pollution sources are a mix of diesel exhaust from trucks and ships in port, local industry, wind-blown dust, and salt from sea spray. Sequim’s summer PM2.5 concentrations were elevated compared to Port Angeles, however were still considered “good” over 95% of the time. There were two episodes extending over several days in the summer when Sequim’s daily PM2.5 concentrations reached “moderate” levels. It is unclear whether these concentrations were typical for this time of year, or due to a massive highway expansion project occurring a few miles upwind of Sequim.

Following this study, the Clallam County air monitor was moved from SMS to PAFS.

2.0 ACKNOWLEDGEMENTS

ORCAA would like to acknowledge Port Angeles Fire Department, the North Olympic Library System, Sequim Fire Department, and Stevens Middle School for hosting air quality monitoring equipment and providing power and internet access. In particular we thank Chief Ken Dubuc and Keith Bogues from the Port Angeles Fire Department for going above and beyond in allowing us to install additional circuits and providing assistance with rooftop installation of equipment. We also thank Assistant Chief Tony Hudson of the Sequim Fire Department for using the fire ladder to install the inlet tube on the tower, where it would otherwise have been impossible for us to reach, and again to remove it. We thank Brian Phillips at North Olympic Library System for helping install monitoring equipment and remote communications. Finally we thank Matt Harper and Puget Sound Clean Air Agency for allowing us to evaluate and validate our saturation study instruments using their air monitoring site and federal equivalent method (FEM) monitors in Kent.

Table of Contents

3.0 LIST OF ACRONYMS

CPO – Cheeka Peak Atmospheric Observatory

ECY – Department of Ecology, Washington State

EPA – Environmental Protection Agency

FRM – Federal Reference Method

FEM – Federal Equivalent Method

NAAQS – National Ambient Air Quality Standard

NOC – Notice of Construction

NOLS – North Olympic Library

OPC – Optical Particle Counters

ORCAA – Olympic Region Clean Air Agency

PAFS – Port Angeles Fire Station

PM – Particulate matter

PM2.5 – Mass concentration of all atmospheric particles with diameters less than 2.5 microns

PM10 – Mass concentration of all atmospheric particles with diameters less than 10 microns

SFS – Sequim Fire Station

SMS – Stevens Middle School

TEOM – Tapered Element Oscillating Microbalance

USG – Unhealthy for Sensitive Groups

WAQA – Washington Air Quality Advisory

UNITS

nm – nanometer (meter^{-9})

m – meter

MPH – miles per hour

µg – microgram (gram^{-6})

4.0 INTRODUCTION

The Olympic Region Clean Air Agency (ORCAA) maintains at least one air quality monitor in each of its six regulated counties: Clallam, Grays Harbor, Jefferson, Mason, Pacific, and Thurston. Clallam County has one additional monitoring site at the Cheeka Peak Observatory (CPO) located in the Olympic National Forest, 400 meters above Neah Bay, WA. This site is part of the National Core (NCore) monitoring network and is a rural, or background, site. The other six county air monitors are placed in populated neighborhoods where ambient air quality is generally the poorest for that region. A saturation study is one method used to determine the best location for placing the air monitoring stations.

During a saturation study several air quality monitors are placed in various locations in a community or communities. The study may last a couple of months, a year, or multiple years. Supplemental measurements, such as meteorology, may be used to provide additional information about air pollution sources and air quality response to weather conditions. Saturation studies are used to:

- determine the best location for each county’s air monitor
- assess regional air quality variability
- evaluate daily and seasonal air quality variability
- identify primary sources of air pollution in each region
- determine how changes in industry and/or population have affected air quality

The following report describes the saturation study conducted in Clallam County between January 2013 and March 2014. A brief discussion of the regional air quality, industry, population, and climate is followed by a detailed description of the study design, the monitoring network, data analysis, and final results.

5.0 BACKGROUND

Prior to the saturation study reported here, the most recent Clallam County air quality saturation study was conducted during the 1996/97 winter [*Moody and Werner*, 1998]. The study ran from December 1, 1996 through April 17, 1997. Since PM2.5 (mass of atmospheric particles with diameters less than 2.5 microns per cubic meter of air) National Ambient Air Quality Standards (NAAQS) were not implemented until 1997, the previous saturation study measured ambient PM10 (mass of atmospheric particles with diameters less than 10 microns per cubic meter of air). On July 18, 1997, shortly after the study ended, EPA introduced the national PM2.5 standard. The PM2.5 standards were more stringent than previous PM10 standards and initiated a switch from PM10 measurements to PM2.5 in Washington State’s air quality monitoring network plan. This current saturation study evaluated ambient PM2.5 concentration, rather than PM10.

In the seventeen years since the PM10 saturation study was completed, Clallam County population experienced a 14% increase from 63,000 to 72,000 and there have been significant changes in number and placement of industrial emissions. Rayonier Pulp Mill closed in May 1997, just after completion of the PM10 saturation study. K-Ply, a plywood mill operating near the port, shut down in 2007 after operating over 70 years in Port Angeles. It re-opened again in March 2010 as Peninsula Plywood (PenPly), but closed down less than 2 years later in December 2011. Both Rayonier and K-Ply/PenPly were Title V air pollution sources and their closures represent significant PM10 and PM2.5 emission decreases in the region. Between 2003 and 2006, two new sawmills, Interfor and Port Angeles Hardwood, opened west of Port Angeles. Both mills operate boilers whose emissions are generally upwind of the city. Population changes affect residential burning emissions, which comprise a significant fraction of winter PM2.5

In 2011, the Nippon paper mill filed a Notice of Construction (NOC) to build a 20 megawatt (MW) biomass cogeneration boiler. The new boiler's air operating permit stipulated better pollution control technology relative to the old boiler, but also allowed roughly 3 times as much fuel combustion. New controls, combined with higher capacity, mean lower emissions for some pollutants (PM2.5, CO, and SO_2) and increases for others (NOx and VOC)[*CH2MHill*, 2010]. In light of these operational changes, citizens became concerned that the current air monitor location did not sufficiently represent regional ambient air quality. To address citizen concern, potential changes in distribution and amount of ambient air pollution, and the lack of PM2.5 regional air quality studies throughout their jurisdiction, ORCAA proposed a one year saturation study be conducted in each county under its authority beginning with Clallam County. These results are placed in context with historical trends in local air quality, population and industry, and regional climate.

5.1 Air Quality

ORCAA has monitored several air quality parameters in various locations around Clallam County since 1971. Initially the primary measure of air quality was total suspended particulate matter (TSP), for which Port Angeles was classified as non-attainment between 1978 and 1982 *(http://www.epa.gov/oaqps001/greenbk/phisttsp_wa.html)*. In 1987, EPA instigated a new national ambient air quality standard (NAAQS) for PM10. On July 18, 1997, annual and 24-hour PM2.5 NAAQS were established. The 24-hour PM2.5 standard was updated in 2006 and the annual PM2.5 standard was updated in 2012 (Table 1).

ORCAA has used a nephelometer to monitor ambient particulate matter at Stevens Middle School (14th & D), Port Angeles since December 1996. The nephelometer provides data on the amount of light scattered by ambient air particle concentrations.

In October 1999, ORCAA collocated a Rupprecht & Patashnick Co Partisol – FRM (Federal reference method) PM2.5 Air Sampler with the Nephelometer at Stevens Middle School

(SMS). The FRM sampler directly measured 24-hour average PM2.5 concentrations once every 6 days and operated through December 2001. This instrument sampled air through a filter for 24 hours on each sample day. The filters were collected weekly and sent to Manchester environmental lab for gravimetric analysis. Data were available to ORCAA and the public after the filters were validated and weighed. This could take up to three weeks after the day a sample was collected.

Table 1: Historical PM10 and PM2.5 NAAQS.

Date	Pollutant	Averaging Time	Standard
1987	PM10	24-hour	**150 μg m^{-3}**
		annual	**50 μg m^{-3}**
1997	PM2.5	24-hour	**65 μg m^{-3}**
		annual	**15 μg m^{-3}**
2006	PM2.5	24-hour	**35 μg m^{-3}**
		Annual	15 μg m^{-3}
	PM10	24-hour	150 μg m^{-3}
		Annual	Revoked
2012	PM2.5	24-hour	35 μg m^{-3}
		annual	**12 μg m^{-3}**

Changes from a previous standard are bold.

Correlations between the FRM PM2.5 concentrations and the nephelometer's light scatter data provided a factor to convert the hourly nephelometer data into equivalent PM2.5 (Figure 1). For the range of PM2.5 values observed at this site, a linear regression is sufficient [*Charlson et al.*, 1968; *Chow et al.*, 2006]. Although the FRM measurements were discontinued after 2001, the nephelometer continues to produce hourly PM2.5 data that are made available to ORCAA staff and the public in near real time.

Figure 2 depicts monthly averaged ambient PM2.5 measured at Stevens Middle School, Port Angeles, WA between January 1999 and December 2013. Also shown are the maximum and minimum daily average PM2.5 for each month. PM2.5 concentrations follow a strong annual cycle wherein concentrations are highest in the winter and lowest in the summer. Prior to 2007, average monthly concentrations in the winter exceeded 15 μg m^{-3}, 3 times the average summer monthly concentrations. Wintertime air quality was considered moderate most of the time and occasionally crept into the unhealthy for sensitive groups. While a winter maximum was still observed after 2007, average monthly concentration did not exceed 15 μg m^{-3}, with the exception of 2012/2013. Daily maximum values likewise decreased after 2007. Summertime monthly averaged PM2.5 concentrations have consistently remained at or below 5 μg m^{-3}. Summertime PM2.5 values have not changed significantly since 1999.

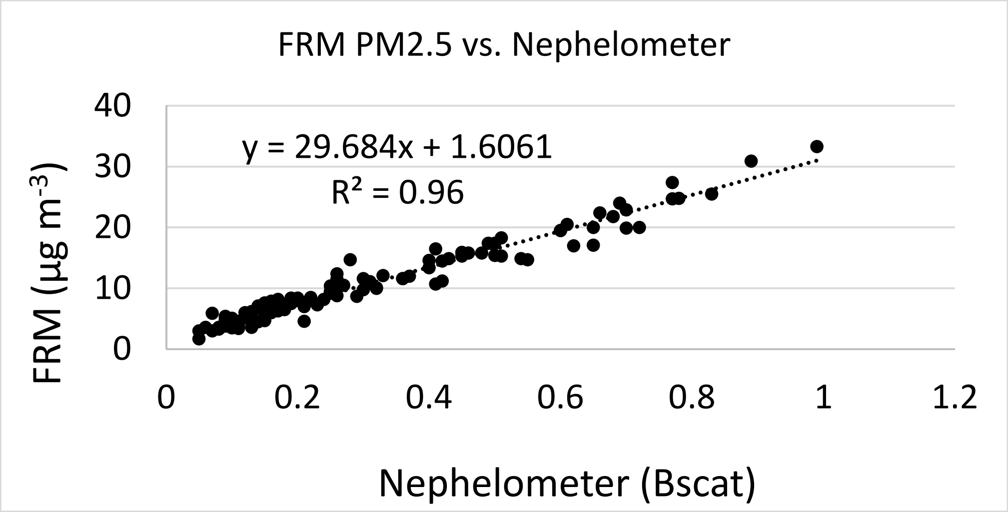

Figure 1) Correlation between Federal Reference Method and nephelometer light scatter data used to infer PM2.5 concentrations from nephelometer data.

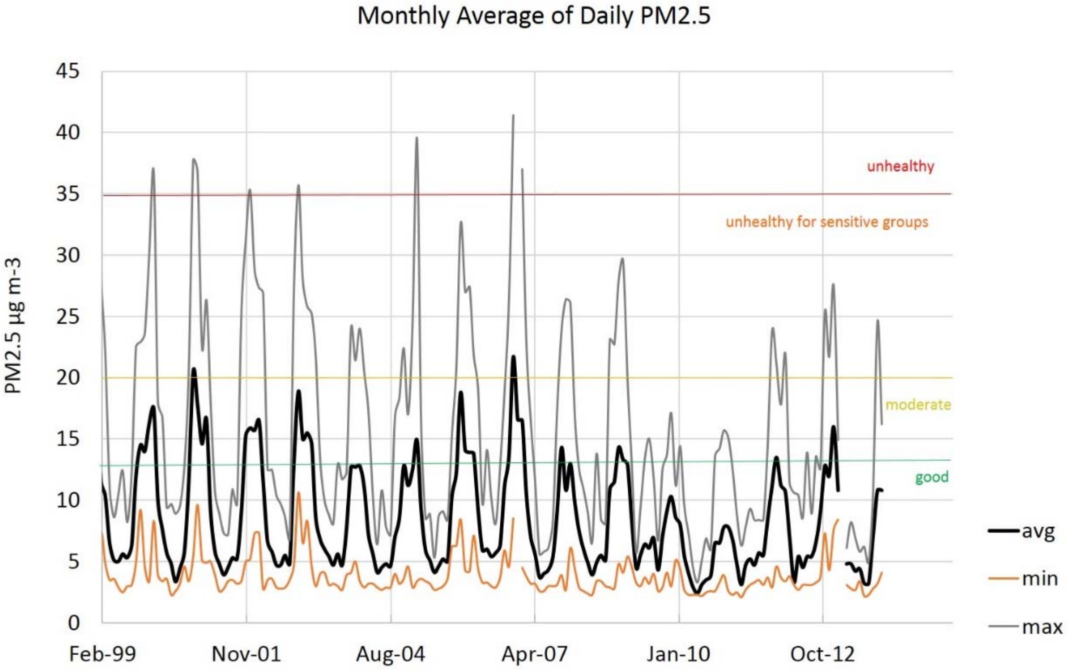

Figure 2) Historical air quality record for Port Angeles, WA. Maximum and minimum values correspond to the maximum and minimum 24-hour average in each month.

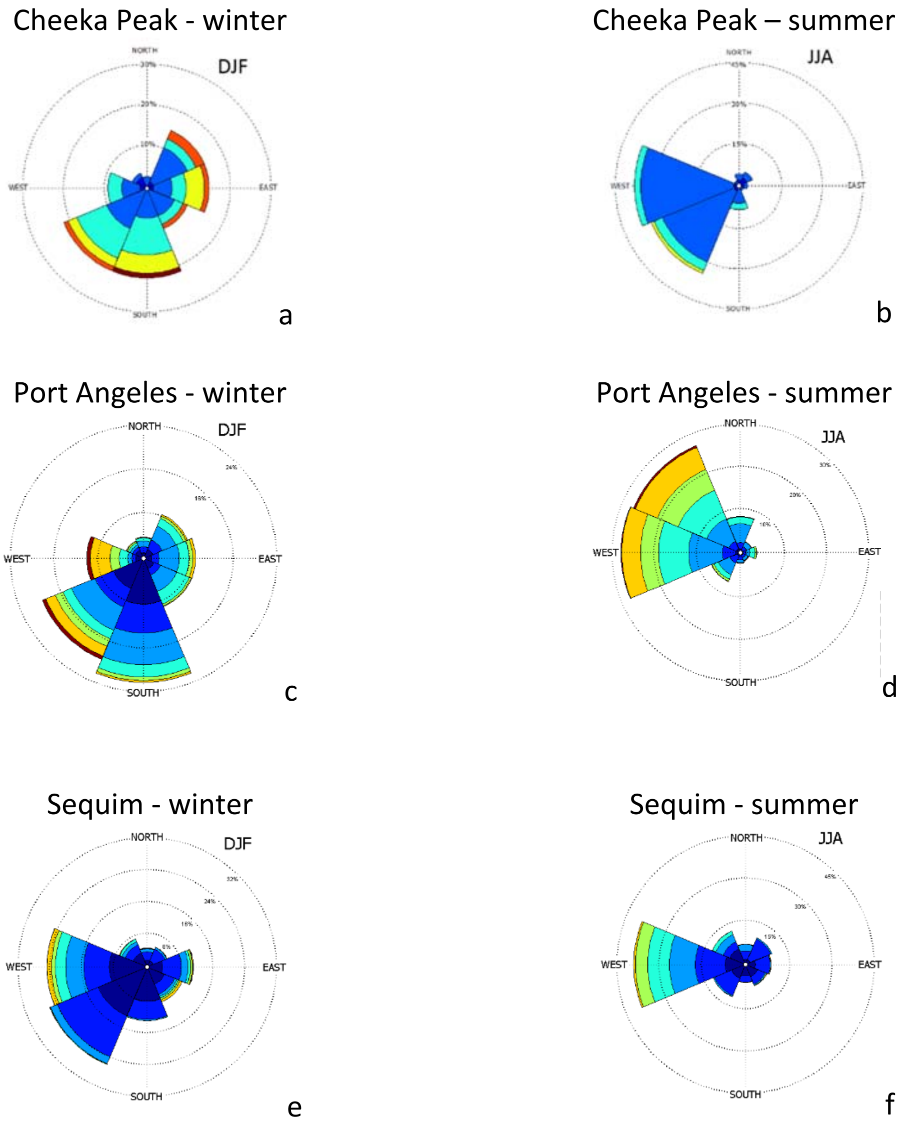

Figure 3) Wind roses for Cheeka Peak, Port Angeles, and Sequim. Colors correspond to wind speeds: 0-10 MPH (dark blue - cyan), 10-15 MPH (yellow), and 15-20 MPH (dark red).

5.2 Climate

Clallam County stretches along the northern coastline of the Olympic Peninsula, with the vast majority of its population sandwiched between the Olympic Mountains and the Pacific Ocean or the Strait of Juan de Fuca. Western Clallam County has a very wet coastal climate, strongly impacted by direct winds and storms from the Pacific Ocean. The eastern county is much dryer, residing in the rain shadow of the Olympic Mountains. Forks Washington, sitting about 10 miles inland on the west coast, receives on average 119” of precipitation per year. Port Angeles and Sequim, which are respectively 50 miles and 70 miles east of Forks,

receive only 26" and 16" of annual precipitation. Summer high and winter low temperatures (72°F and 32°F) are similar in all three communities.
Wind patterns are seasonally dependent and complicated by mountain ranges, land and sea breezes, and diurnal cycles. On the northwestern point of the Olympic Peninsula (Cheeka Peak), winter winds are bimodal, typically either northeasterly or southwesterly (Figure 3a). Summer winds are lighter and predominantly westerly and southwesterly (Figure 3b). In contrast, Port Angeles winter winds are typically south and southwesterly and summer winds are most often north and northwesterly (Figure 3c and 3d). While the strongest winds tend to occur in the winter, statistically summer winds are generally faster. Just 20 miles east of Port Angeles, Sequim's winds are usually westerly, regardless of season (Figure 3e and 3f) and wind speeds are about the same in both summer and winter. A meteorology station was not installed at the Sequim monitoring site and these data were obtained from Washington State University AgWeatherNet (http://weather.wsu.edu/awn.php).

5.2 Population and Industry

Port Angeles is not only the most densely populated region of Clallam County, but also has the highest number of industrial air pollution sources. Since 1996, the number of county residents increased by about 9000, while Port Angeles population has remained almost the same *(US Census Bureau)*. Port Angeles residents currently account for 26% of the county's population compared to 30% in 1996. Sequim, the second largest community in Clallam County has grown by 40%, from 4752 to 6624. Forks, the third incorporated town in Clallam County has grown by about 10%, or 300 people, contributing little to total county population growth. Most of the population growth has been in the over 65 age group in small communities outside Sequim and Port Angeles city limits (*Sequim Gazette*, *August 15, 2014*). Based on these numbers, residential PM2.5 emissions likely have not changed significantly in Port Angeles, however may have increased in and around Sequim. Residential emissions from the Port Angeles Urban Growth Area (UGA) may also have increased.

Major area industrial pollution sources have changed since the original saturation study. In February 1997, Port Angeles's largest mill, Rayonier, closed. This reduced emitted PM10 in the area by about 25 tons per year. The closure of K-Ply/PenPly, a mill that operated from 1941 to 2012, reduced area emissions by roughly 40 tons per year. Changes in some industrial procedures have also reduced emissions. Stack tests at Nippon in 2003 indicated facility PM2.5 emissions were around 100 tons per year. More recent stack tests conducted in 2010 showed that emission factors had changed and PM2.5 emissions were closer to 35 tons per year. This is a significant change in area PM2.5 emissions between 2003 and 2010.

Although mill closures and changes in operations marked a significant decrease in area emissions, several new area sources began operating during the last two decades and increased emissions. These new sources include Interfor Inc. (1998, formerly Crown Pacific), Evergreen Fibre (2001), and Port Angeles Hardwood (2006). Together they have increased area PM10 emissions by approximately 40 tons/year. Since the last saturation study, overall industrial PM10 emissions in the region have decreased by about 90 tons/year.

Rayonier, K-Ply/PenPly, and Nippon all represent emission reductions north of town near the port. Nippon is located northwest of downtown Port Angeles, K-Ply/PenPly lay just east of Nippon, and Rayonier sat northeast of town, on the water. In contrast, all previously mentioned new sources are located southwest of town near Highway 101. Based on seasonal dominant wind directions, mill closures near the port represent potential reductions in summer ambient particles measured by the monitor at Stevens Middle School (SMS), while new sources to the west may increase ambient particulate measured at SMS all seasons.

As the original saturation study focused on PM10 concentrations, only changes in industrial PM10 emissions were described. Emissions inventories for these sources indicate PM2.5 generally comprised between 60 and 90% of the PM10.

6.0 STUDY DESIGN

The goals of this study were:

a) Determine whether the Stevens Middle School (SMS) nephelometer data was an accurate representation of regional ambient air quality in Clallam County
b) Evaluate neighborhood scale air quality variability
c) Identify the primary sources of PM2.5 in the region
d) Identify whether a new location would be more appropriate for the permanent monitor

In addition to the permanent air monitor at SMS, four air quality monitors and two meteorological stations were placed in Clallam County. In the following sections, instrumentation, monitoring sites, and data analysis strategies are described.

6.1 Instrumentation

ORCAA purchased four additional air monitors to conduct the saturation study. The monitors were optical particle counters (OPC) manufactured by MetOne (Profiler 212). The OPC operates similarly to the nephelometers most commonly used in Washington State’s air quality network. Both instruments use light scattering off particles to determine PM concentrations. The nephelometer produces a measure of light extinction due to scattering [*M Z Hansen and Evans*, 1980]. The OPC converts the light scattered from individual particles to a specific size bin and produces particle number concentrations at 8 different diameters between 0.3 and greater than 10 microns [*Iwasaki et al.*, 2007].

Prior to the study, an OPC was collocated with both a nephelometer and a tapered element oscillating microbalance (TEOM), a federal equivalent method (FEM) for monitoring PM2.5. The instrument comparison was performed in February to ensure the highest range of PM2.5 values from both traffic and residential burning. Hourly averaged data from the OPC was compared to both the nephelometer and TEOM data to ensure it was a good proxy for measuring PM2.5 (Figure 4).

Total particle volume was estimated from the OPC data assuming all measured particles were spheres. Light scatter, or nephelometer data, correlates most highly with total particle number concentration. Particle mass concentration (PM) corresponds directly to volume concentration (V) using the following equation PM = ρV, where ρ is the particle density. The scattering efficiency of a particle strongly depends on size and the relationship between particle mass concentration and light scatter will differ depending on the aerosol density and size distribution [*Chow et al.*, 2006]. This may be observed in the bottom two panels of Figure 4, where the OPC volume concentration correlates more highly with TEOM mass measurements than the OPC counts. Both OPC particle counts and volume concentration correlate most highly with the nephelometer, because both instruments rely on aerosol light scattering properties.

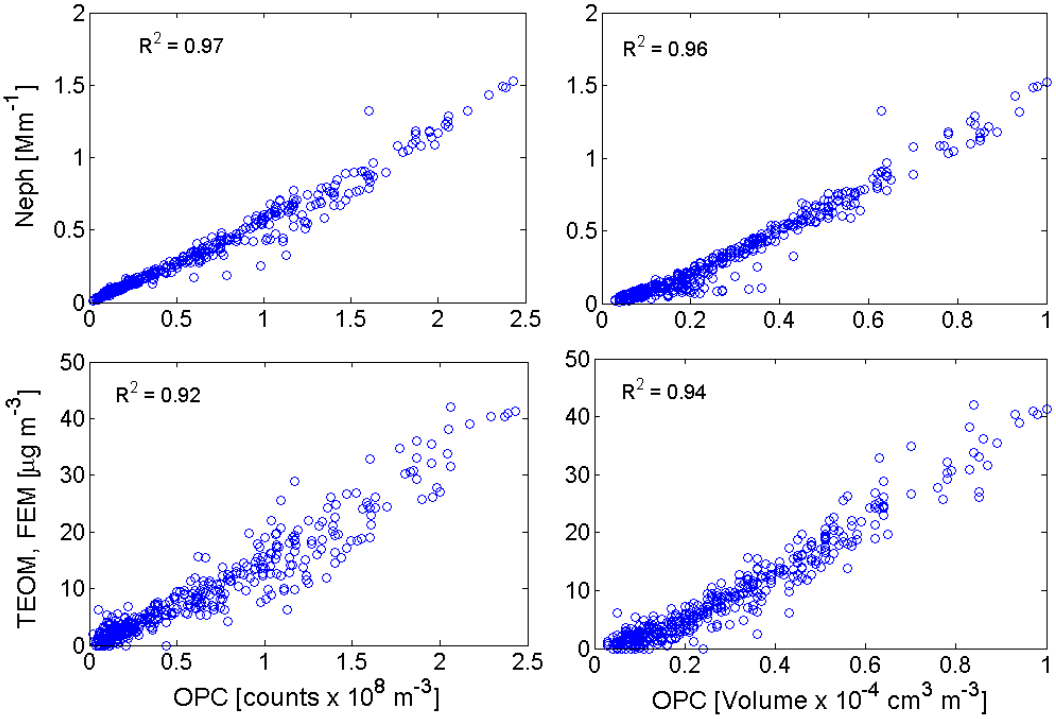

Figure 4) Linear regression comparison between the OPC, the nephelometer and the TEOM.

In addition to the OPC, ORCAA installed two meteorology stations and an aethalometer, which measures black carbon mass concentration. The meteorology stations provide high resolution measurements of temperature, pressure, relative humidity, and wind direction and speed. The aethalometer and one meteorology station were installed at Port Angeles Fire Station (PAFS). The other meteorology station was installed at North Olympic Library System (NOLs).

The aethalometer measures the amount of light absorbed by black or brown particles. These light absorbing particles are emitted during combustion and their color provides information on the fuel type burned. Diesel and oil based fuels produce very black particles that absorb light strongly across the visible spectrum. Biomass fuels tend to produce particulate that absorbs ultraviolet and visible light of shorter wavelengths (blue to green) much more strongly than at longer wavelengths (yellow to red). These particles appear brown in the atmosphere.

The OPCs, data loggers, modems, and an uninterrupted power supply (UPS) were integrated in portable boxes (Figure 5). Meteorological and OPC data were both stored on the computer and transmitted daily to a cloud server owned by ORCAA. This allowed ORCAA staff to remotely check on instrument performance and ensured better data recovery, as well as providing back-up storage for the data.

Figure 5) Housing for OPC, data logging system, modem, and UPS.

6.2 Monitor Locations

The Environmental Protection Agency (EPA) and the Washington State Department of Ecology (ECY) have strict guidelines on where and how neighborhood scale PM air monitoring sites are placed [*Thompson*, 2008]. ORCAA followed these guidelines in choosing and placing the saturations study monitors. The main requirements are as follows:

1) The sample inlet must be between 2 and 15 meters above the ground and at least 1 meter from a supporting structure.
2) Sample inlet should be placed at least 100 meters from a wood burning device and a quarter mile from any fugitive dust source.
3) Distance to nearest road should be at least 10 meters for every 1000 vehicles driven per day. This is based on average daily traffic counts.
4) An open horizontal arc of at least 270° must surround the sample inlet.
5) The sample inlet must be at least 10 meters from the tree drip line.

In addition to state and federal monitoring station placement requirements, practical logistics also needed to be taken into account. Sites were required to have access to power and internet connection. The instruments and data logger needed an area, easily accessible by ORCAA staff, but secure from tampering and vandalism. The chosen monitoring locations should represent ambient air quality in communities with the highest population and therefore exposure. Lastly, one of the sites should duplicate the current permanent monitoring station to ensure data consistency and continuity. One of the new OPC monitors was collocated with the nephelometer at Stevens Middle School (SMS), providing a continuous comparison between the two instruments. The other three were placed at: North Olympic Library (NOLS), 2210 S Peabody St, Port Angeles; Port Angeles Fire Station (PAFS), 102 E. 5th St, Port Angeles; and Sequim Fire Station (SFS), 323 N 5th Ave, Sequim (Figure 6).

Figure 6) Clallam County Saturation Study map

SMS is located in a residential neighborhood, southwest of downtown Port Angeles and has been the location of an air quality monitor in Clallam County since 1998. Previous saturation study results demonstrated that this site represented regional ambient PM10. SMS is 1.3 miles south of Nippon Paper Mill and 1.5 miles northeast of Port Angeles Hardwood and the Eclipse industrial site. NOLS is in a mixed residential and commercial area, almost 2 miles south of the port and downtown and 2 miles east of Eclipse industrial park. PAFS is 5 blocks southwest of the port and about 60 meters west of S. Lincoln Street (also Highway 101 and the main road through town). PAFS is over 1.5 miles southeast of Nippon Paper Mill and just over 2 miles northeast of the Eclipse industrial park. SFS is in the heart of Sequim, a few blocks east of downtown and at the interface between commercial and residential neighborhoods. It is about 182 meters north of W. Washington Street, the main thoroughfare through Sequim. All four locations are centrally located with respect to regional air pollution sources.

6.3 Data

Raw PM and meteorological data are collected at a one minute temporal resolution. Aethalometer data is produced at five minute intervals. All data are validated and processed to hourly and daily averages. Hourly data provide information on daily trends and sources of air pollution. Similarly, daily averages provide information on weekly, monthly and seasonal air pollution patterns. Meteorological data, including wind direction, wind speed, and temperature, analyzed in conjunction with the OPC data offers further evidence of potential sources and is used to isolate meteorological effects on air pollution from changes in source emissions. For example, persistent temperature inversions lead to poor atmospheric ventilation (Wind Speed = 0) and this allows PM2.5 concentrations to increase, even if source emissions remain the same. People who primarily heat their homes with wood burning devices generally light them when they arrive home from work between 4 PM and 5 PM. This can greatly increase evening and night PM2.5 concentrations relative to daytime hours. Wind direction carries pollution from different sources either to or away from the air pollution monitors. PM2.5 spikes corresponding to specific wind direction indicate likely source or sources.

7.0 RESULTS

7.1 Inter-Site comparison

PM2.5 data from the three Port Angeles sites were significantly correlated (Table 3 & 4). Data collected at the Sequim Fire Station (SFS) was significantly correlated with both the Port Angeles Fire Station (PAFS) and the North Olympic Library (NOLs) sites, but much less so with Stevens Middle School (SMS). Correlation decreases as the temporal resolution is increased; i.e. the correlation between the daily values is higher than the hourly values. This is most likely due to variations in daily source emissions close to each station. For example, some residents near SMS may start their stoves earlier in the day than residences near NOLs. This would create very different hourly PM2.5 profiles at the two stations, but still have similar concentrations when averaged daily. Correlation coefficients of site-to-site comparison is a strong indicator as to whether or not a site is regionally representative. Since NAAQS is based on daily averages, the results in Table 4 are of greater importance when choosing the best monitoring location.

NOLs had the highest average daily correlation with the other three sites at $R^2 = 0.66$. PAFS was just slightly lower at $R^2 = 0.63$. SMS and SFS were considerably lower at $R^2 = 0.48$ and $R^2 = 0.34$ respectively. Including only those sites in the city of Port Angeles, PAFS had the highest average correlation with the other two sites.

Table 2: Correlation coefficients (R^2) for hourly data between saturation study sites

	SMS	**PAFS**	**NOLS**	**SFS**
SMS	1.0	0.55	0.41	0.12
PAFS	0.55	1.0	0.60	0.26
NOLs	0.41	0.60	1.0	0.43
SFS	0.12	0.26	0.43	1.0

Table 3: Correlation coefficients (R^2) for daily data between saturation study sites

	SMS	**PAFS**	**NOLS**	**SFS**
SMS	1.0	0.71	0.61	0.12
PAFS	0.71	1.0	0.82	0.35
NOLs	0.61	0.82	1.0	0.54
SFS	0.12	0.35	0.54	1.0

The magnitude and frequency of low and high pollution days is similar at all 4 locations (Figure 7). Air quality classifications are based on Washington Air Quality Advisory (WAQA) values and are termed good, moderate, and unhealthy for sensitive groups. The average daily PM2.5 was less than 7 $\mu g\ m^{-3}$ over 60% of the time and between 7 and 13.5 $\mu g\ m^{-3}$ over 30% of the time at SMS and SFS. At PAFS and NOLs, the daily PM2.5 was less than 7 $\mu g\ m^{-3}$ approximately 50% of the time and between 7 and 13.5 $\mu g\ m^{-3}$ about 42% of the time. PM2.5 concentrations were classified as “good” over 90% of the days at all 4 sites. “Moderate” air quality was observed less than 10% of the days at all locations. PAFS exhibited the highest percentage of days (9%) at moderate PM2.5 levels and was the only site to register one day at USG (unhealthy for sensitive groups).

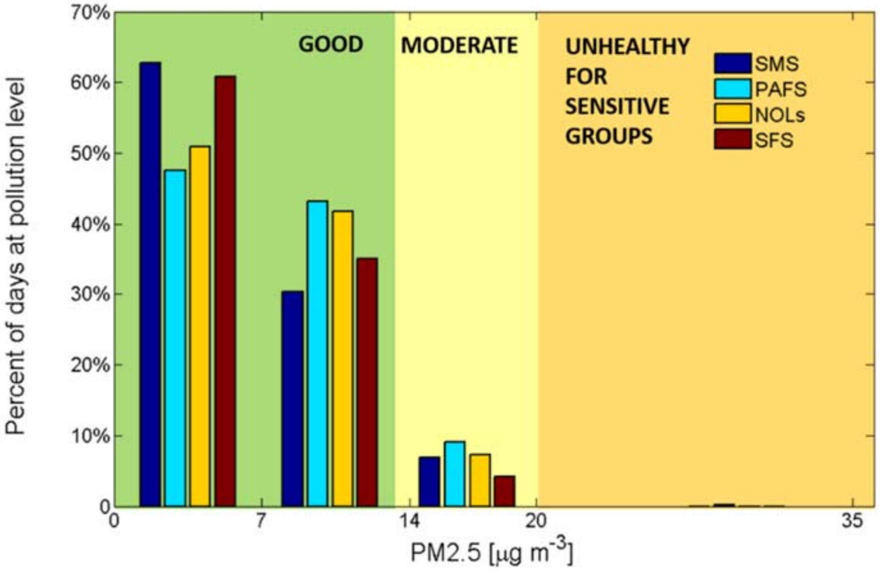

Figure 7) Probability distribution of days at different PM2.5 levels for all 4 monitoring sites

7.2 Seasonal Variability

The seasonal variability found in the historical data (Figure 2) remained consistent in Port Angeles during the saturation study. Daily winter PM2.5 concentrations (10 +/- 7 μg m^{-3}) were twice that observed during summer (5 +/-2.5 μg m^{-3}). In Sequim, the summer and winter PM2.5 averages concentrations were almost identical at 7.5 +/- 2 μg m^{-3} and 7.3 +/- 4 μg m^{-3} respectively.

Compared with the Port Angeles sites, Sequim's summer PM2.5 concentrations were split nearly equally between the lower (0-7 μg m^{-3}) and higher range (7 – 13.5 μg m^{-3}) of what is considered "good" air quality (Figure 8). Port Angeles PM2.5 summer concentrations were below 7 μg m^{-3} more frequently: 92% at SMS and around 70% at PAFS and NOLs. In Port Angeles daily summer PM2.5 concentrations never exceeded "good" levels. Although over 95% of the summer daily PM2.5 levels were categorized as "good" in Sequim, 3 days registered "moderate" air quality.

During the winter, Port Angeles monitoring sites were more heavily skewed toward higher PM2.5 concentrations when compared with the summer data (Figure 8). PAFS had the highest percentage of days registering as moderate and one day in the "unhealthy for sensitive groups (USG)" category. SMS and NOLs recorded 80% of the days as "good" air quality and approximately 20% as "moderate". At PAFS, 73% of the days were considered "good", 26% moderate, and 1% as "USG". Although Sequim's summer and winter "good" air quality days were about the same, Sequim's winter air quality was skewed toward the lower end of the "good" range, with 58% of the winter days below 7 µg m^{-3}, 38% between 7 and 13.5 µg m^{-3}, and 4% between 13.5 and 20 µg m^{-3}, or “moderate”.

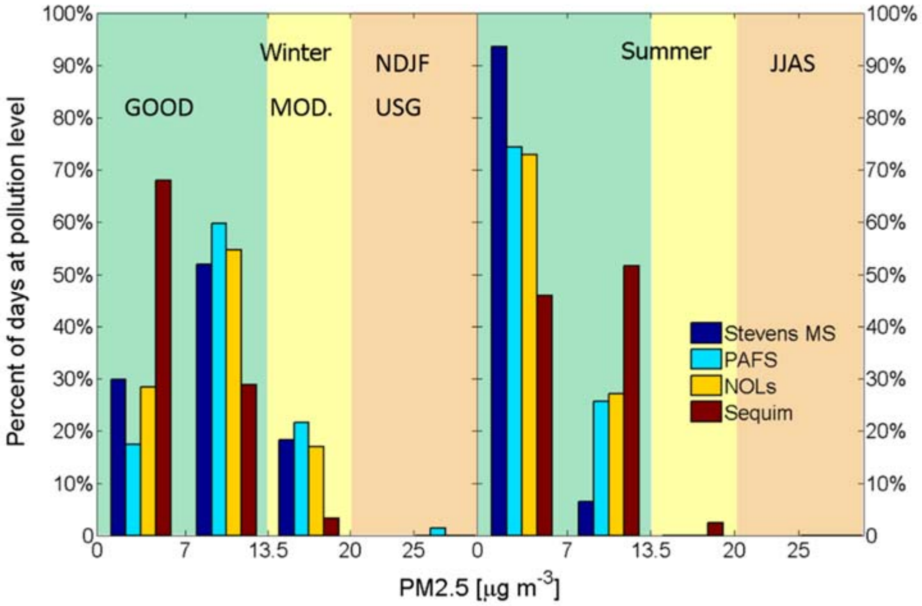

Figure 8) Seasonal probability distribution of days at different PM2.5 levels. Winter months include November, December, January, and February. Summer months are June, July, August, and September. Air quality classifications are good, moderate, and unhealthy for sensitive groups.

Port Angeles air quality was worse in the winter with greater variability than in summer. While Sequim air quality was statistically similar in both seasons, a higher percentage of winter days had better air quality. At the same time, a slightly higher number of Sequim’s winter days also registered as “moderate”. Possible reasons for differences between Sequim and Port Angeles seasonal air quality trends are discussed in section 7.4.

7.3 Daily Variability

Winter and summer hourly data were averaged by hour of day and are shown in Figure 9. The solid line symbolizes winter data and the dashed line is for summer data. A persistent winter, daily pattern was evident at all monitoring sites. In winter, PM2.5 peaked twice during the day: a smaller spike between 8AM and 10AM, followed by a much larger increase beginning around 4PM and lasting through the night. Sequim winter data also followed this pattern, but exhibited much lower morning and evening concentrations relative to Port Angeles. The summer data did not display significant daily variability at any of the sites. Port Angeles summer PM2.5 concentrations were less than the corresponding winter data at all hours of the day. Relative to winter, Sequim's summer PM2.5 concentrations were typically higher in the morning and afternoon, but fell below the evening (5 PM– 10 PM) winter levels.

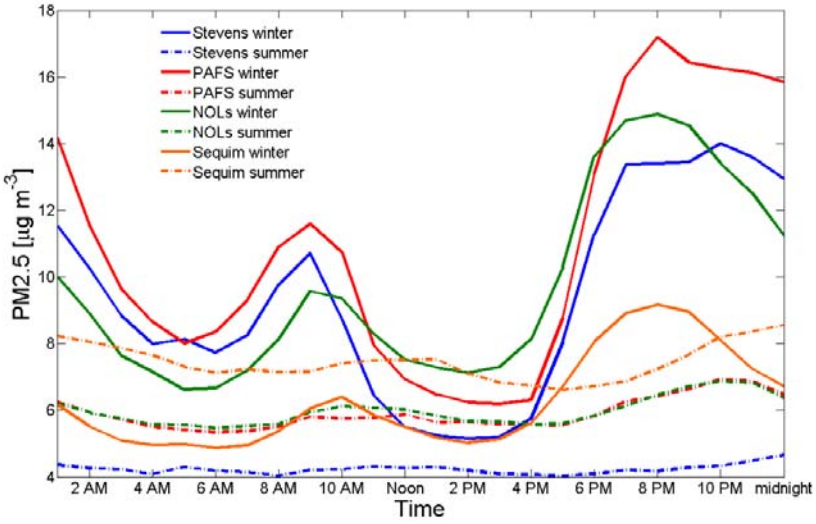

Figure 9) Average hourly PM2.5 for winter and summer days at the 4 saturation study sites

These data demonstrate Port Angeles's high winter daily PM2.5 values were largely driven by the nighttime, and to a lesser extent, morning peaks in concentration. The winter midday lows in PM2.5 concentrations were only slightly higher (< 2 µg m^{-3}) than the summer midday values. Sequim's summer PM2.5 concentration was 33% higher than that observed at PAFS and NOLs and nearly 100% greater than at SMS.

The higher baseline in Sequim was driven largely by two separate periods where air quality in Sequim was considerably higher than Port Angeles. The first happened in July 19, 20, and 21 during Sequim's annual Lavender Festival. Hourly PM2.5 concentrations were usually between 15 and 20 µg m^{-3} during those three days. Port Angeles also saw a spike in PM2.5 that weekend, but levels did not exceed 15 µg m^{-3} and were rarely above 10 µg m^{-3}. Most likely, enhanced vehicle traffic and emissions from street food trucks at the fair caused the higher PM levels in Sequim. The second occurred between September 19 and 26, Sequim's baseline PM2.5 concentrations were steady at 10 µg m^{-3}, while PM2.5 in Port Angeles was approximately half that concentration. Based on time of year, late summer and early fall, elevated PM levels in Sequim were likely a result of outdoor burning downwind of the Port Angeles monitors, but upwind of Sequim. Air quality in both locations was categorized as "good" during this period. The rest of the summer, Sequim experienced air quality that was similar in magnitude and variability to PAFS and NOLS, both of which were typically higher than Stevens Middle School.

7.4 Sources

The daily and seasonally resolved PM2.5 data provide insight to the largest regional sources. The higher winter concentrations, combined with the seasonally resolved daily pattern, suggest a significant wintertime PM2.5 emission source that is most prevalent for a few hours in the winter morning and again in the evening. This source is insignificant in the summer months. The daily resolved, aethalometer black carbon data indicate wood combustion as the primary winter PM2.5 source and the daily PM2.5 and black carbon pattern further isolate this combustion to residential heating.

The aethalometer measures the mass of light absorbing particles at two wavelengths, 880 nm and 370 nm [*A D A Hansen*, 2003]. Aside from dust, light absorbing particles are generally the product of combustion [*Andreae*, 1983; *Bergstrom et al.*, 2007; *Bond and Bergstrom*, 2006; *Bond et al.*, 2004; *Streets et al.*, 2003]. High efficiency combustion and fossil fuels produce very black soot that has a light absorption spectrum closely following the relationship: abs = λ^{-1}, where λ is the wavelength of light [*T. W. Kirchstetter and Novakov*, 2007]. Wood smoke and other biomass fuels burn less efficiently and produce more organics. These particles absorb ultraviolet and shorter wavelengths (370 nm) more efficiently than longer wavelength light (i.e. 800 nm) [*Hadley et al.*, 2008; *T. W. Kirchstetter et al.*, 2004; *T.W. Kirchstetter and Thatcher*, 2012]. A comparison of particle absorption at both wavelengths can be used to differentiate between woodstove (fireplace) and fossil fuel (diesel) emissions.

The aethalometer assumes the absorption (abs_{ff}) spectra of all particles follows the relationship:

$$abs_{ff} = k_{ff} * \lambda^{-1} * BC_{ff} \qquad (1)$$

where k is a constant and BC is the mass of black carbon. The subscript ff denotes "fossil fuel". While this is generally true for combustion particles derived from diesel and other fossil fuels [*Bergstrom et al.*, 2007; *A D A Hansen*, 2003], wood smoke particles contain a significant fraction of organics, which absorb strongly at short wavelengths and very little at longer wavelengths. The wood smoke absorption (abs_{ws}) spectra follow the relationship [*T.W. Kirchstetter and Thatcher*, 2012] :

$$abs_{ws}=k_{ws}\, \lambda^{-1.89} * BC_{ws} \qquad (2)$$

When a significant amount of wood smoke is present, the aethalometer BC data from the 370 nm wavelength channel are enhanced relative to the data at 880 nm. Equations

1 & 2 were used to calculate the relative fraction of absorbing particles from wood smoke and from fossil fuel combustion. Since the enhancement is only evident in the 370 nm channel, abs_{ff} and abs_{ws} are equal when λ = 880 nm. To calculate the relative absorption enhancement in the 370 nm channel, kws must first be solved; k_{ff}, BC_{ff}, and BCws are set equal to 1, λ = 880 nm.

$$k_{ws} = 880^{0.89} = 417.4 \qquad (3)$$

$$abs_{ws}/abs_{ff} = k_{ws}*(370^{-1.89+1}) = 2.16 \qquad (4)$$

From the Beer-Lambert law, absorption is linearly related to concentration and thus a factor of 2 enhancement in absorption translates to double the BC. Thus, when the BC concentration from the 370 nm channel was more than twice that of the 880 nm channel, the light absorbing material was determined to be entirely from wood smoke. When the two channels were equal, the particles were assumed to be from fossil fuel combustion. A mix of diesel and wood smoke was calculated when the absorption enhancement was greater than 1 and less than 2. There are significant and undetermined uncertainties associated with these calculations and these results are a best estimate. The uncertainties arise from varying degrees of woodstove and fireplace combustion efficiencies that alter the emitted particles' absorption characteristics. For example a small amount of wood smoke that absorbs more strongly at shorter wavelengths mixed with diesel soot would appear to have a higher wood smoke fraction.

The daily resolved pattern of black carbon in December, January, and February (Figure 10a) is the same as observed in winter PM2.5 (Figure 9). As winter transitions to summer, the morning and evening spikes in black carbon diminish and by June look like the summer PM2.5 daily pattern. Figure 10b illustrates how morning and evening black carbon concentrations can be traced almost entirely to woodstoves in the winter, while the mid-afternoon concentrations look more like diesel emissions, similar to summertime values.

Past studies have published chemically resolved PM2.5 emission factors for woodstoves and fireplaces, as well as for diesel fueled trucks, cars and ships. Black or "elemental" carbon accounts for 5 to 30% of the PM2.5 emissions from woodstoves and fireplaces [*Fine et al.*, 2002; *Rau J. A.*, 1989]. Fire temperature and fuel differences account for the large range in values. Fireplaces produce more PM2.5 relative to BC than woodstoves. In this study a median BC contribution to wood smoke of 12% was estimated. BC makes up about 50 to 60% of the mass of emissions from diesel fueled trucks [*Wang X. et al.*, 2011], but only about 5% from ships [*Petzold A. et al.*, 2010]. Assuming that trucks and ships are the largest sources of diesel emissions in Port Angeles, the BC percentage was set in the middle. 25% of ship and truck combined particulate emissions was estimated to be black carbon. Dividing the wood smoke BC mass and the diesel BC mass by their respective contributions to each source's average particulate emissions provided an approximate contribution to total measured PM2.5 (Figure 11). After subtracting the wood smoke and diesel particulate contribution from measured particulate, the remaining PM2.5 accounts for all other sources and background particulate. Other sources include dust, sea salt, and condensed organic vapors.

In winter, wood smoke accounts for 50% to 70% of the morning PM2.5 and 85% to 95% of the evening and night time PM2.5. Wood smoke only makes up about 20% of the particulate between noon and 4 PM. PM from diesel combustion contributes between 20% and 30% (about 2 µg m^{-3}) between 8 AM and 7 PM and is negligible at night. All other sources contribute roughly 30% +/- 17% throughout the day. The average winter concentration of "other" PM is 2.6 +/- 1.2 µg m^{-3} (Figure 11a)

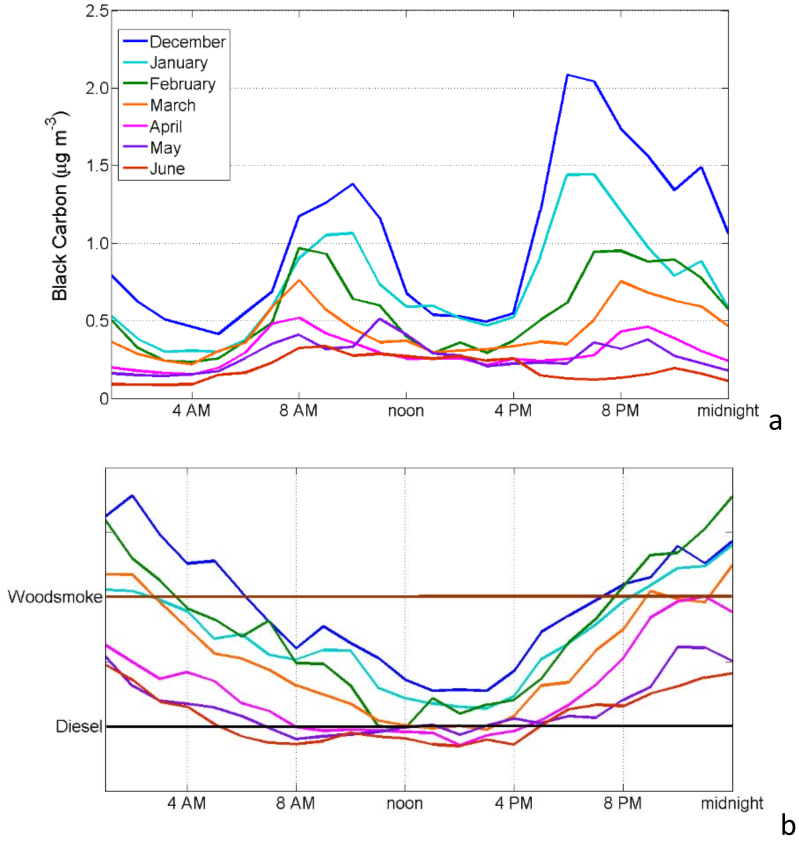

Figure 10) Diurnal pattern of: (a) black carbon concentrations and (b) diurnal pattern of diesel vs. woodstove derived black carbon

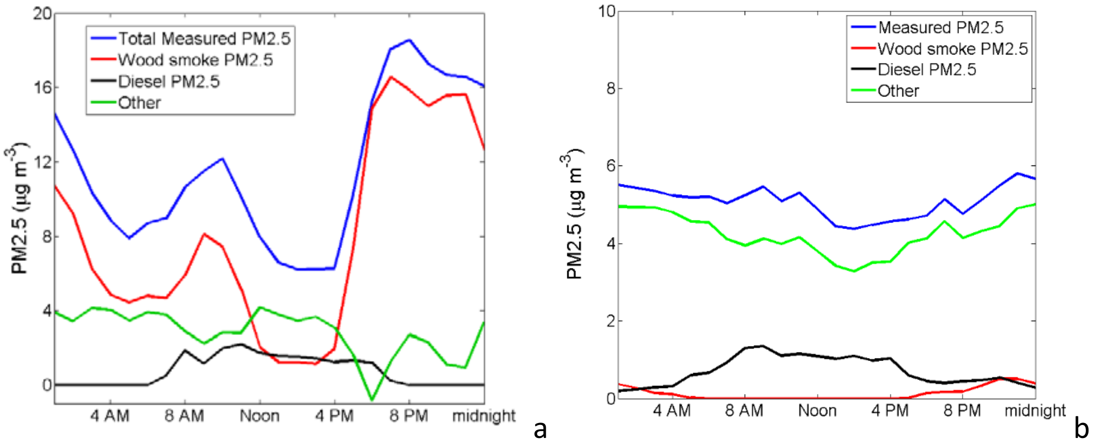

Figure 11) Contribution of wood smoke and diesel to total PM2.5 concentration in the a) winter and b) summer.

In the summer months, wood smoke is almost non-existent (Figure 11b). Summer diesel particulate follows a similar daily cycle to that observed in winter. The summer diesel contribution is about 30% between 7 AM and 5 PM. Other summer PM sources make up 70% to 90% of the summer particulate concentration. The average summer concentration from other sources is 4 +/-0.5 μg m^{-3}, about 50% higher than in winter. Increased dust generation in hot, dry summer months may well account for the increase.

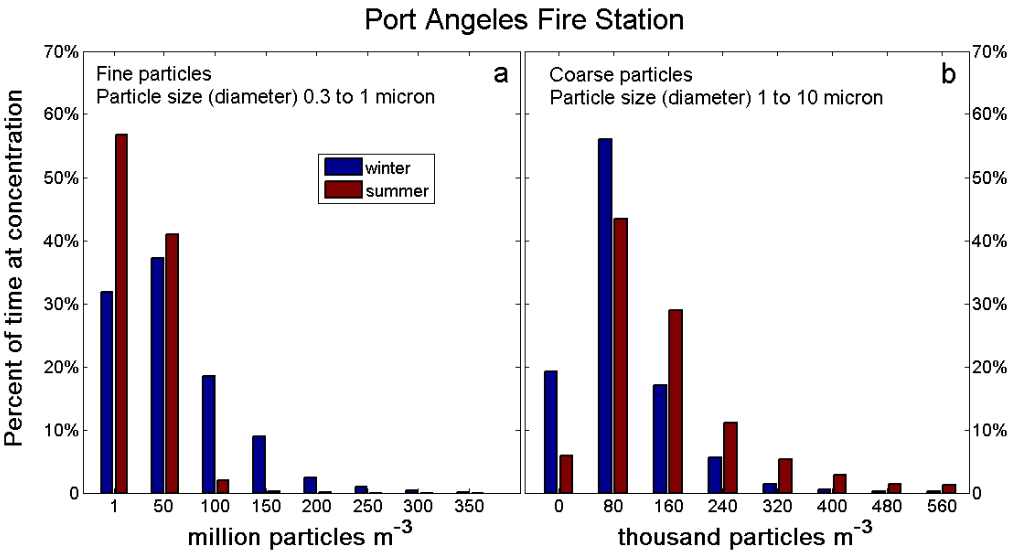

Figure 12) Seasonal concentration frequency distribution for a) fine particles and b) coarse particles at PAFS

The optical particle counters (OPC) provided particle size number concentration between 0.3 and 10 μm. Where combustion particles are typically smaller than 1 μm

[*Rau J. A.*, 1989], dust particle diameters generally exceed 1 µm [*Mahowald et al.*, 2014]. Particle number concentration frequency distribution for fine (Figure 12a) and coarse particles (Figure 12b) are shown below for both summer and winter months. In the winter, fine particle concentrations exceeded 100 million per meter cubed about 30% of the time compared with summer concentrations that reached this threshold less than 5% of the time. This is consistent with increased woodstove use in the winter. Despite coarse particle number concentration being several orders of magnitude smaller than that of fine particles in either season, their contribution to PM (mass concentration) can be significant as the mass of each particle increases as the cube of the radius. In summer months, coarse particle number concentration exceeded 160 thousand particles per cubic meter 50% of the time compared to only 25% in winter. The relatively higher coarse particle concentration in summer is typical of increased dust generation.

In Sequim, the seasonal difference in particle size distribution was even more pronounced. As expected, the frequency distribution of fine particle number concentration (Figure 13a) looks very similar to Sequim's PM2.5 concentration distribution. Unlike Port Angeles, coarse particles are only observed in Sequim during the summer. The number of coarse particles observed in Sequim during the summer are only a small fraction of what is seen in Port Angeles (Figure 12 b). Enhanced dust generation from the highway expansion project west of town does not explain Sequim's elevated summer PM2.5 concentrations relative to Port Angeles.

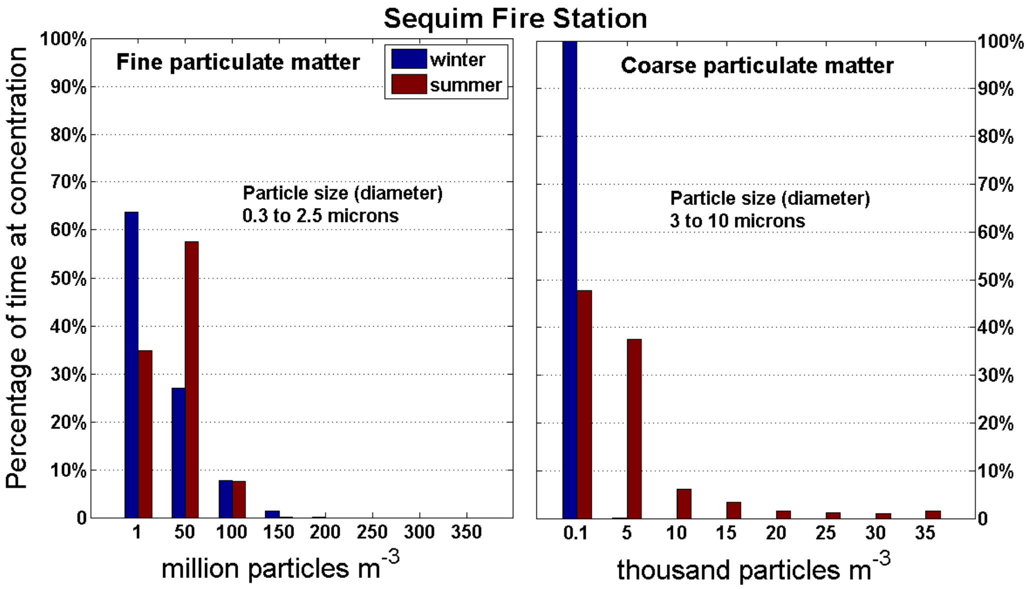

Figure 13) Seasonal concentration frequency distribution for fine and coarse particles

As previously stated, the Lavender Festival and outdoor and land clearing burns between Port Angeles and Sequim in late summer are the most likely reasons for the elevated PM2.5 observed in Sequim's summer months.

7.5 Meteorology

Seasonal winds also provide information on regional PM2.5 sources. Both wind direction and wind speed strongly affect air pollution concentrations. Strong winds indicate good air ventilation and prevent pollution from building high concentrations. Winter winds in Port Angeles are most often southerly with wind speed below 10 MPH (Figure 3). Higher wind speeds are associated with westerly and south westerly winds. Summer winds are almost always westerly or north westerly and more frequently are between 10 and 15 MPH (Figure 3). Figure 14 depicts the PM2.5 concentration distribution associated with wind direction in both winter (a) and summer (b). *Note that the PM2.5 color scale is different for each season.

In winter, the highest concentration of PM2.5 is associated with southerly winds. Port Angeles residential neighborhoods are almost all south of the air monitors, while most of the industrial emissions are north along the waterfront. Increased residential heating in winter, combined with low wind, more frequently cause PM2.5 concentrations to increase to moderate and sometimes unhealthy levels. Summertime PM2.5 levels do not have a strong dependence on wind direction, although a slightly higher concentration may be associated with easterly and north easterly winds. Higher wind speed and greatly reduced emissions in the summer create "good" air quality 99% of the time.

A meteorology station was also collocated with the NOLs monitoring site and showed consistent results with those obtained from the PAFS data. All meteorology parameters were highly correlated and the relationship between PM2.5 and wind direction was the same.

Sequim's seasonal wind pattern is different from Port Angeles, with most of the winds blowing from the west in either season, although in winter they are more southwesterly. Summer winds are almost always due west. Seasonal differences in Sequim's wind speed are not pronounced.

The winter dependence of PM2.5 on wind direction was not as distinct in Sequim, although higher PM2.5 concentrations coincided slightly more with northerly wind (Figure 15a and 15b). Sequim's summer PM2.5 concentrations appeared independent of wind direction. Wind measured in Sequim nearly always blows from the west, so the PM2.5 data for other directions may not be statistically representative.

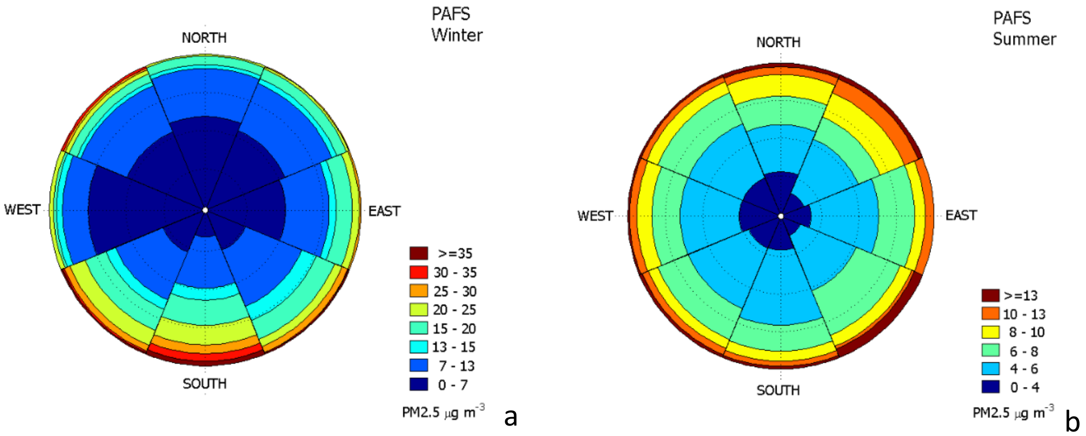

Figure 14) PAFS hourly PM2.5 concentrations normalized by wind direction for: a) winter and b) summer

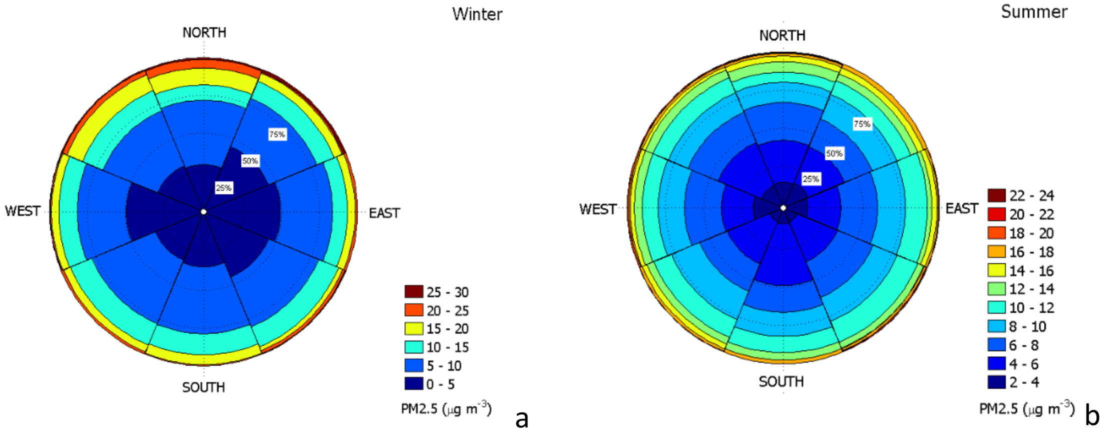

Figure 15) Sequim hourly PM2.5 concentrations normalized by wind direction for: a) winter and b) summer

8.0 CONCLUSIONS

Winter air quality in Port Angeles was most heavily influenced by residential heating using woodstoves and fireplaces. The PM2.5 and black carbon from wood smoke both followed a similar daily pattern. A small mid-morning spike between 7 AM and noon aligned with morning start-up of wood heaters. A much larger increase was observed in the evening beginning around 4 PM and lasting until after midnight. Emissions from heaters in the evening combined with increased nighttime stability in the winter created conditions conducive to build-up of pollutants. The highest levels of PM2.5 are associated with southerly winds. The area to the south of the air monitors was primarily residential.

Sequim is located 20 miles east of Port Angeles and experienced somewhat different air quality. The highest number of “moderate” air quality days in Sequim were observed in winter, but Sequim also had more “moderate” air days in the summer than did Port Angeles. Although over 96% of Sequim’s summer PM2.5 concentrations were ““good” air quality days, they were shifted more frequently toward the higher side of the “good” classification relative to Port Angeles. Annually, Sequim statistically had cleaner air than any of the three stations in Port Angeles (Figure 7).

9.0 RECOMMENDATIONS

Based on the data presented, ORCAA has determined that the Port Angeles Fire Station (PAFS) is the most ideal location for measuring ambient air quality that is representative of the region. Clallam County experiences impaired air quality almost exclusively in winter and PAFS experienced the highest PM2.5 concentrations of all the sites. Although PAFS PM2.5 was statistically higher than at the other sites, all of the site data were significantly correlated and PAFS data was representative of air quality in the region.

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