Regional Impact of a Biomass-fueled Co-generation Boiler on Ambient PM2.5 and Ultrafine Particle (UFP) Concentration

Authors: Odelle Hadley[1], Lauren Whybrew[1], Cassandra Gaston[2], Honglian Gao[3], Dan Jaffe[3], and Joel Thornton[2]

1 – Olympic Region Clean Air Agency; 2 – University of Washington, Seattle; 3 – University of Washington, Bothell

Abstract

The Nippon Paper Company, located in Port Angeles, WA, applied for a permit in 2012 to replace a decades-old, mixed hog-fuel and diesel boiler with a 20-megawatt, biomass fueled, co-generation plant. Some residents were concerned that emissions from the new boiler would degrade air quality and cause health problems. This study evaluated the effect of Nippon's biomass co-generation facility on ambient PM2.5 and ultra-fine particles (UFP) and identified the primary sources of both pollutants in the region. Nippon stopped using the old boiler and began operating the biomass co-generation boiler in December 2014. Particulate and gas-phase measurements were made between January 2014 and June 2015, ensuring that both winter and summer months prior to and following operational changes were represented. Gaston et al (2016) showed higher particle concentrations in winter, relative to summer months, were from residential wood-fired heaters and stoves. This was confirmed by high concentrations of levoglucosan, a tracer of residential wood combustion, measured at the monitoring site. In summer, winds more frequently blow from the mill toward the monitoring site and background PM2.5 concentrations are low. These conditions made summer an ideal time to evaluate air quality impacts of the biomass co-generation plant. UFP concentrations decreased by about 500 particles per cm^{-3} in summer 2015, relative to summer 2014. No statistical difference in ambient PM2.5 concentration (5 +/-3 µg m^{-3}) was found between summer 2014 and summer 2015. A comparison of the particulate data collected during operating hours with non-operating hours in May and June 2015, revealed that although air monitors detected carbon monoxide emissions from the boiler, no measurable effect on ambient PM2.5 or UFP concentrations was found.

Introduction

As global concerns about climate change and air quality grow, the modern energy sector relies increasingly on renewable energy sources. Green power is a subgroup of renewable energy sources defined as having the "highest environmental benefit" and includes solar, wind, geothermal, biogas, biomass, and low-impact hydroelectric (https://www.epa.gov/greenpower/what-green-power). EPA created the Green Power Partnership (GPP) in 2001 to promote expanded use of renewable power. In addition to technical assistance, government funded financial incentives in the form of grants, tax credits, and tax deductions are available to both industry and the public for demonstrated commitment to using renewable, green-power.

Not all green power sources are universally accepted as environmentally friendly or sustainable. Biomass co-generation facilities are recognized by the United States and the European Union as a carbon neutral, green power energy source. Whether or not biomass fueled power plants are truly carbon neutral, largely depends on where and how fuel is harvested and how forests are managed after harvesting [*Mitchell et al.*, 2012]. Alternative fates of the fuel and whether the green power generation will replace other less eco-friendly power sources are also relevant [*Lee et al.*, 2010; *Springsteen et al.*, 2011]. Public fears regarding the use of biomass fueled co-generation facilities range from increased de-forestation and greenhouse house gas (GHG) emissions to degraded air quality. The study presented here focuses on the regional air quality impacts from PM2.5 and ultra-fine particles (UFP) emitted by a biomass co-generation plant. It does not address GHG emissions and long-term climate effects of biomass combustion as a power source.

In 2012, the Nippon Paper Company filed a permit to construct a biomass co-generation facility in Port Angeles, WA. Nippon proposed to replace a decades-old, mixed hog-fuel and diesel boiler (boiler #8) with a new, 20-megawatt, co-generation plant that would provide power to the mill and potentially sell power back to the grid. Debris from nearby logging activities and waste

products from the paper-making process would provide most of the fuel. Logging debris is typically burned in clear cuts where the logs are harvested. This releases vastly more particle pollution to the atmosphere compared with combustion in a boiler equipped with emission controls. Clear-cuts, however, are generally remotely located where smoke emissions do not impact population centers. Increasing biomass combustion at Nippon and its proximity to residential neighborhoods had many members of the public concerned about the impact on local air quality.

In July 2014, Nippon ceased operation of boiler #8. The primary particulate control was a wet scrubber with a removal efficiency rated at 50%. The new biomass co-generation boiler came on-line January 2015 and was equipped with the following pollution controls: an electrostatic precipitator rated over 96% efficient for PM2.5 and UFP removal [*Lind et al.*, 2003]; a condensing scrubber to remove acid gases and fine particulate, and a selective non-catalytic reduction system to remove NOx. Despite the improved pollution controls, some residents worried that greater fuel consumption would increase ultra-fine particle emissions, degrade regional air quality and exacerbate health problems in the area. Air quality is a concern raised by many communities around the nation in response to biomass co-generation facilities. Several emission comparison studies have been conducted regarding the fate of woody debris [*Lee et al.*, 2010; *Mitchell et al.*, 2012; *Springsteen et al.*, 2011], however few studies have been completed looking at a biomass co-generation boiler's impact on ambient air quality. This study evaluated the effect of Nippon's biomass co-generation facility on ambient atmospheric particulate concentrations 2 km from the mill site and attempted to identify the primary sources of ambient PM2.5 and ultra-fine particles in the region.

The Nippon mill was purchased by McKinley Paper Company in April 2017 and is not currently operating. The mill site is west of the port and downtown area of Port Angeles and less than a mile to the north of residential neighborhoods.

Measurements and Methods

Ambient air monitors were installed at the Port Angeles Fire Station (PAFS; 48.115°N, 123.436°W), chosen for accessibility, site security, power and internet availability, and its central location relative to local emission sources (Figure 1). PAFS is two miles southeast of Nippon and other industrial sources and less than a mile south of the Port of Port Angeles. Residential neighborhoods lay west, south, and southeast of the air monitors. Commercial districts were north and east of the monitors.

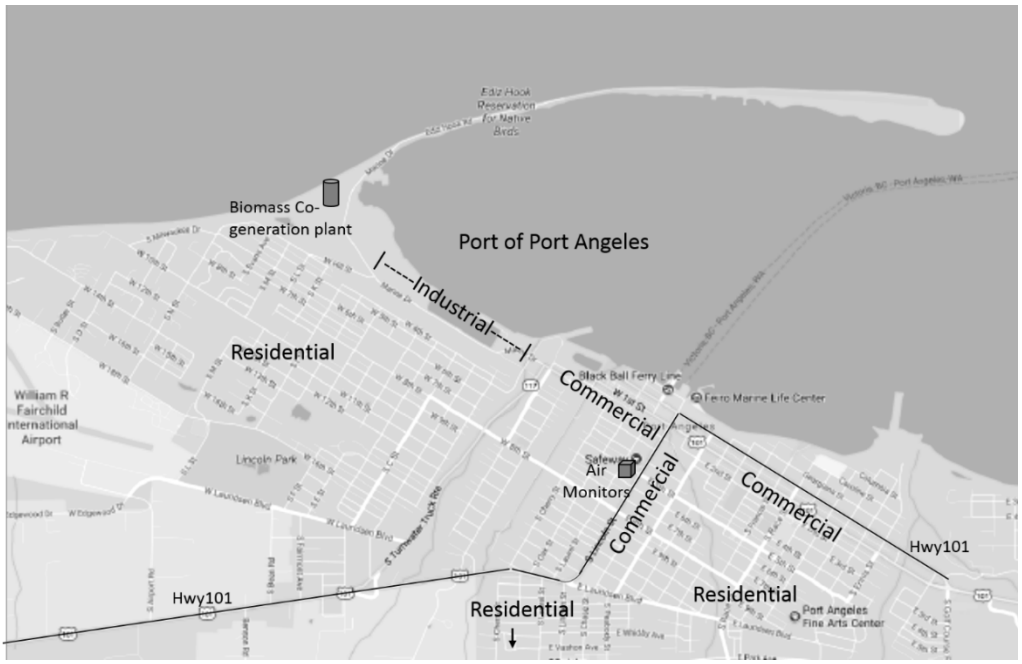


Figure 1) Map of Port Angeles: includes the location of the biomass co-generation plant, the air monitors and municipal zones

Particulate and gas-phase measurements were made between January 2014 and June 2015, ensuring that both winter and summer months prior to and following biomass co-generation facility operation were represented in the data. All instruments were placed in a secure room on the second floor of the fire station. Sample inlets ran from the instruments, through a vent to the roof and extended approximately 6 feet above the roofline. Meteorological instrumentation was

installed about 2.5 m from the sample inlets at a height of 3 meters above the roofline. Particle measurement sample lines were made of conductive silicone tubing or stainless steel 3/8" (OD) tubing. Gas phase samples were collected through 1/4" (OD) Teflon tubing. Data were collected at different temporal resolutions ranging from one to ten minutes. All data were converted to hourly averages and synchronized. Table 1 lists all measurements, instruments used, and dates of data collection.

Table 1) Measurements

Variable	Instrument	Dates of measurement
L50 (# particles w/ D< 50 nm)	TSI Scanning Mobility Particle Sizer Spectrometer 3938	01/01/2014 – 06/30/2015
L100 (# particles w/ D< 100 nm)	TSI Scanning Mobility Particle Sizer Spectrometer 3938	01/01/2014 – 06/30/2015
L300 (# particles w/ D< 300 nm)	TSI Scanning Mobility Particle Sizer Spectrometer 3938	01/01/2014 – 06/30/2015
Organic speciation	Chemical Ionization Mass Spectrometer	01/21/2014 – 03/6/2014
PM2.5 (µg/m3)	Optical Particle Counter (MetOne - Profiler 212)	01/01/2014 – 05/26/2014
PM2.5 (µg/m3)	Nephelometer, Radiance Research M903	07/17/2014 – 12/31/2014 04/06/2015 – current
Black carbon (ng/m3)	Aethalometer (Magee Scientific, Model AE22	01/01/2014 – 06/23/2015
SO_2	Thermo Environmental, Model 43C	04/20/2014 – 06/14/2015
CO_2	LiCOR Inc. Model LI-840A	01/01/2014 – 06/15/2015
CO	Thermo Environmental, Model 48C TL	01/01/2015 – 06/15/2015
Wind Speed/Direction	MetOne MSO Weather Sensor	01/01/2014 – 06/30/2015
Ambient temperature	MetOne MSO Weather Sensor	01/01/2014 – 06/30/2015

A scanning mobility particle sizer (SMPS) measured atmospheric particle number concentration at different sizes (10 to 600 nm). We did not use the more commonly used size distribution data

(dN/dlogdP) in this study. Instead the SMPS data were converted to a single UFP number concentration (# cm^{-3}) of all particles with diameters between 10 nm and 100 nm. Prior to field deployment, the SMPS was calibrated in the lab using various sizes of polystyrene spheres and then periodically checked throughout the experiment for quality assurance purposes.

The MetOne optical particle counter (OPC), which measured the number concentration of particles with diameters greater than 0.3 microns at 8 discrete size ranges, was calibrated and certified by MetOne prior to field operations. PM2.5 was estimated from the OPC particle number concentration using the following method. The OPC was collocated with a Tapered Element Oscillating Microbalance (TEOM), a federal equivalent method (FEM) for measuring PM2.5 (Equivalent Method: EQPM-0609-181), for several weeks prior to the study. The correlation coefficient (R^2) between the hourly TEOM PM2.5 and the OPC number concentration of all particles less than 2.5 microns was 0.92 and the relationship linear for PM2.5 concentrations less than 50 µg m^{-3}. We used a simple least squares linear regression to obtain an equation converting number counts to PM2.5 mass concentration. The uncertainty in PM2.5 data associated with this method is 2.5 µg m^{-3}.

The OPC was no longer available for this project after May 2014. PM2.5 measurements resumed in July 2014 using a Radiance Research M903 nephelometer. Light extinction measurements from the nephelometer were converted to PM2.5 using a correlation factor developed by the Olympic Region Clean Air Agency (ORCAA) in 1998. The correlation factor was determined by comparing light extinction to gravimetric PM2.5 filter measurements made in Port Angeles, WA (Federal Reference Method: RFPS-0498-117).

The aethalometer measured light absorbing carbon mass in ng m^{-3} [*Hansen*, 2003]. Aerosols collected on a filter and light attenuation through the filter was referenced to a blank. Light attenuation was measured at two wavelengths, 880 nm and 370 nm. Black carbon (BC) was inferred from light attenuation at 880 nm using a version of the Beer-Lambert law [*Hansen*,

2003; *Kirchstetter and Novakov*, 2007]. BC data were corrected to account for filter loading artifacts [*Preble et al.*, 2015].

A reference or zero value check was conducted on the carbon monoxide (CO) and carbon dioxide (CO_2) instruments every two hours and adjusted daily as needed. The SO_2 zero and span was checked prior to and following field deployment.

In addition to air quality data and meteorological measurements, ORCAA also collected CO and NOx stack emission data from Nippon's co-generation plant. These data were used to identify exact hours of boiler operations.

Results

Hourly data collected in 2014 showed both the average PM2.5 and ultrafine particle (UFP) concentrations were higher during the winter months (January and February) relative to summer (May and June) (Figure 2). PM2.5 hourly winter concentrations were on average 2.5 times greater in winter and maximum winter values were over 3 times higher than maximum summer values. The difference between summer and winter UFP concentrations was less extreme. Average winter UFP values were 1.5 times greater than summer values and, UFP concentrations exceeded 5000 particles per cm^3 in the winter about 4 times more often than in summer (Figure 2b). Extremely high concentrations of levoglucosan, a tracer for wood smoke, were highly correlated with the 2014 winter PM2.5 and UFP concentrations [*Gaston et al.*, 2016]. These data, along with wind direction and the diurnal profile of both pollutants, confirmed residential wood heating as the dominant source of UFP and PM2.5 in winter. PM2.5 was not collected in either January or February 2015 because of instrument malfunction and availability. A comparison of winter 2015 UFP concentrations to winter 2014 reveal similar diurnal variability and dependence on wind direction (Figure 3a). The average UFP concentration was 3744

particles per cm^{-3} for January and February 2014 and slightly lower at 3240 particles per cm^{-3} in 2015.

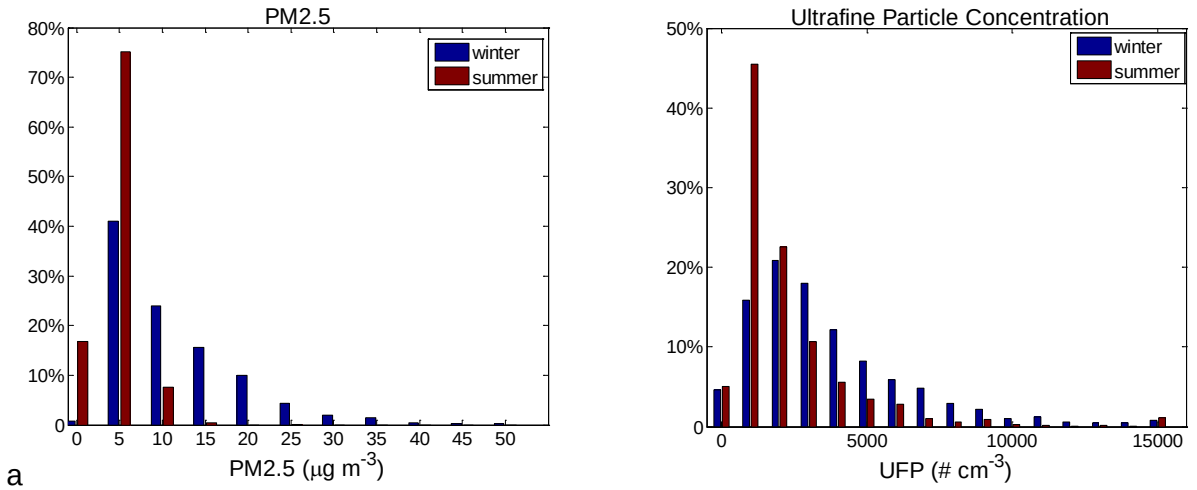


Figure 2) Relative frequency distribution for 2014 winter and summer hourly concentrations of a) PM2.5 and b) ultra-fine particles.

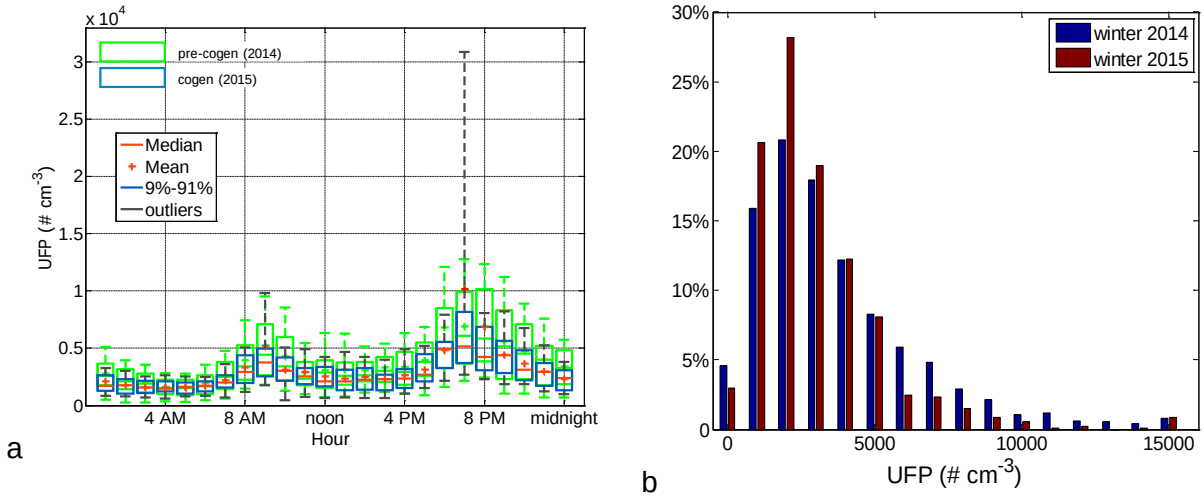


Figure 3 a) Average winter (Jan & Feb) diurnal concentration of UFP in 2014 and 2015 and b) winter frequency distribution of UFP concentration

A concentration frequency distribution analysis of winter UFP for both years shows UFP concentrations exceeded 5000 particles cm^{-3} twice as often in 2014 (Figure 3b; blue bars)

relative to 2015 (red bars). High concentrations of winter PM2.5 and UFP usually coincide with both low temperatures and windspeeds less than 1 MPH. Colder temperatures are generally associated with reduced boundary layer heights, winter stagnation events (no wind) and increased residential heating emissions [*Gaston et al.*, 2016]. While below-freezing temperatures were observed more frequently in January and February 2014 relative to 2015, average wind speeds were higher and the coldest temperatures in 2014 were not associated with the highest UFP or lowest wind speed. Based on the available data it is unclear to what degree meteorology affected the relative UFP concentrations.

Based on similarities in the meteorological and air quality data, we conclude the results from the 2014 winter analysis [*Gaston et al.*, 2016] are also applicable to winter 2015 measurements. The significant impact residential wood heat has on winter ambient air quality and prevailing southerly winds would have masked any air quality changes due to the biomass co-generation boiler in winter months

Figure 4 compares summer and winter wind direction and speed. In winter, winds were most often southerly and westerly. A higher frequency of low wind speed is indicative of stable air masses, which contributed to degraded air quality in winter [*Gaston et al.*, 2016]. Northerly and northwesterly winds were rarely observed in winter and monitoring instruments were rarely impacted by mill or port pollution. In contrast, summer winds frequently carried air from the direction of the mill and the port to the monitoring site. Favorable wind direction, combined with fewer pollution sources and cleaner background air, made May and June an ideal time frame to determine whether a measurable difference in PM or UFP concentrations due to changes in mill operation could be detected. This report therefore focuses on summer air quality data to determine if any changes in ambient UFP or PM2.5 could be detected after the biomass co-generation plant began operating.

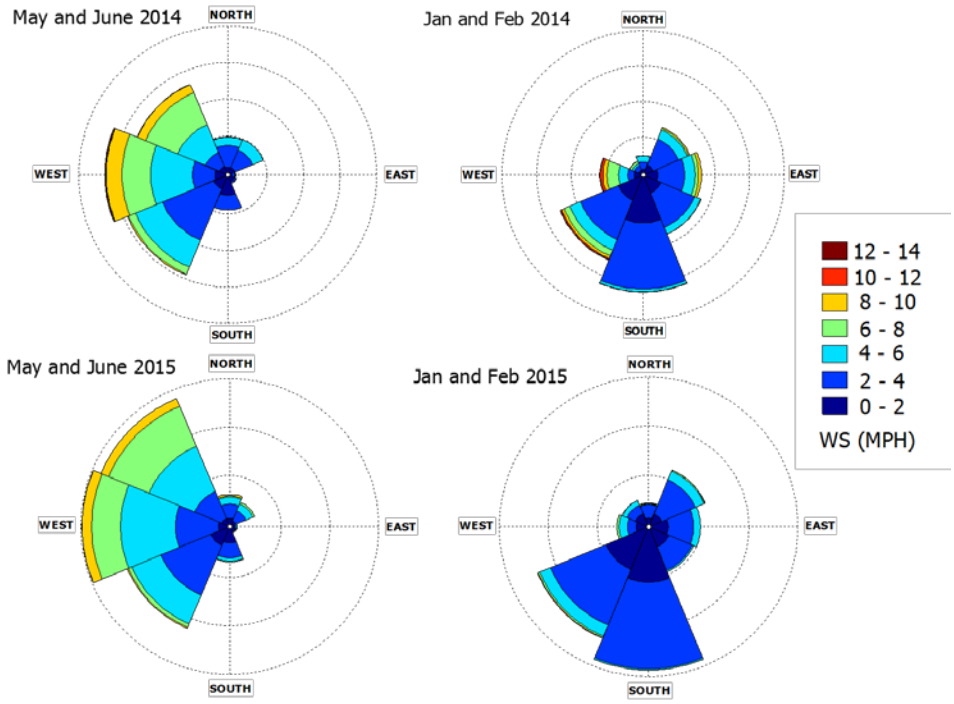
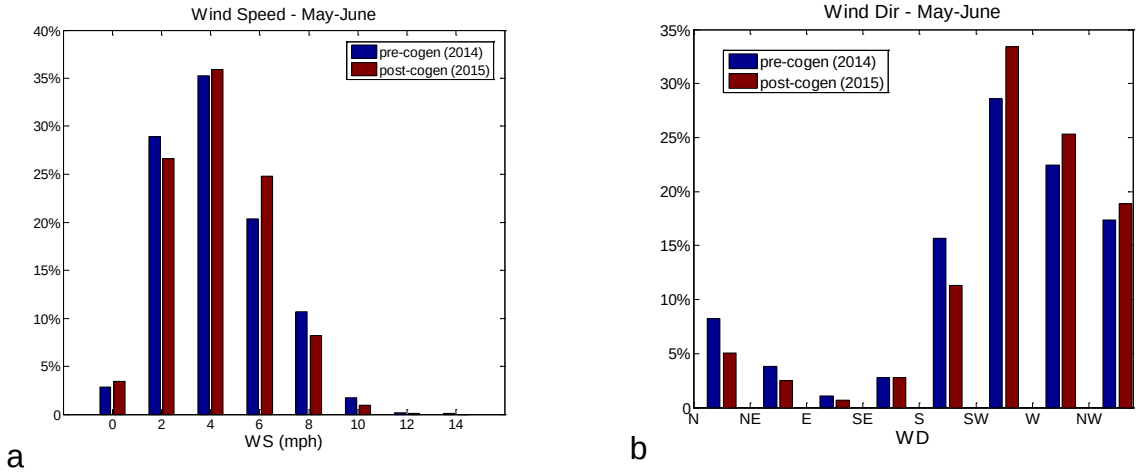


Figure 4 Port Angeles seasonal wind patterns for 2014 and 2015

2014 vs. 2015

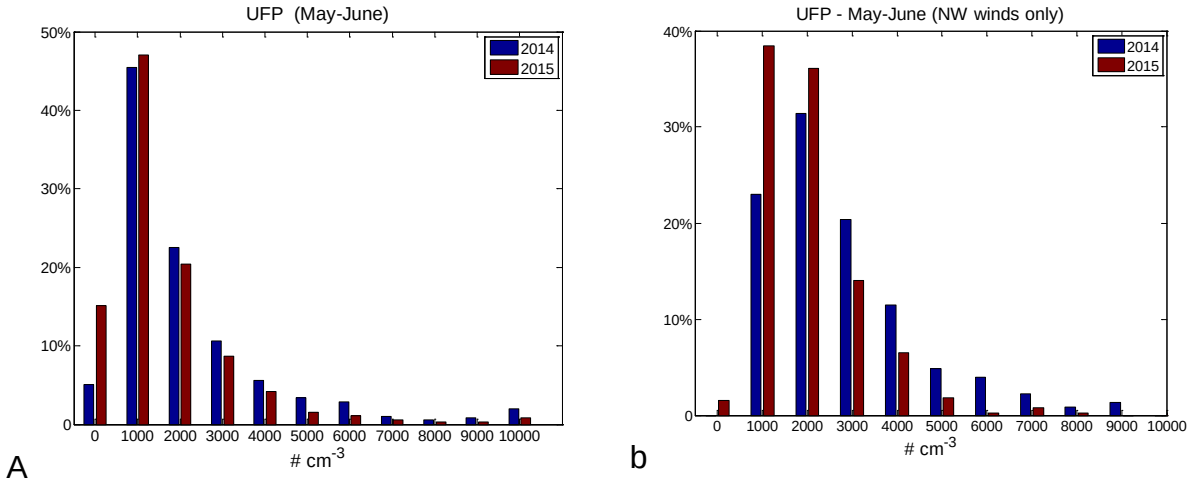
The biomass co-generation boiler came on-line November 2014. To specifically target the effect changes in mill operation had on ambient air quality, only UFP data from May and June 2014 and 2015 will be considered. Due to instrument malfunction and availability, PM2.5 was not collected in May or June 2014. Data from July and August 2014 and 2015 will be used for the interannual PM2.5 comparison instead. This ensures seasonal consistency between the two years and removes the overwhelming particulate signal from residential heating in the winter. Figure 5 demonstrates wind speed and direction were statistically similar during both years. May and June were chosen because all data collection, except PM2.5, ended June 30th, 2015.

The average UFP concentrations in May and June 2014, prior to co-generation boiler operation, were 31 (+/-15) % higher than those measured over the same time span in 2015, after operation began (Figure 6a). The difference between the two years is largely driven by less than 5% of the measurements when UFP spiked to higher than normal values. UFP concentrations exceeded 5000 particles cm^{-3} twice as often in 2014 relative to 2015.



a b

Figure 5 Frequency distribution of wind speed (a) and direction (b) for May and June in both 2014 and 2015.



A b

Figure 6) Frequency distribution of: (a) all UFP concentrations and (b) UFP concentrations when winds were northwesterly (from the direction of Nippon)

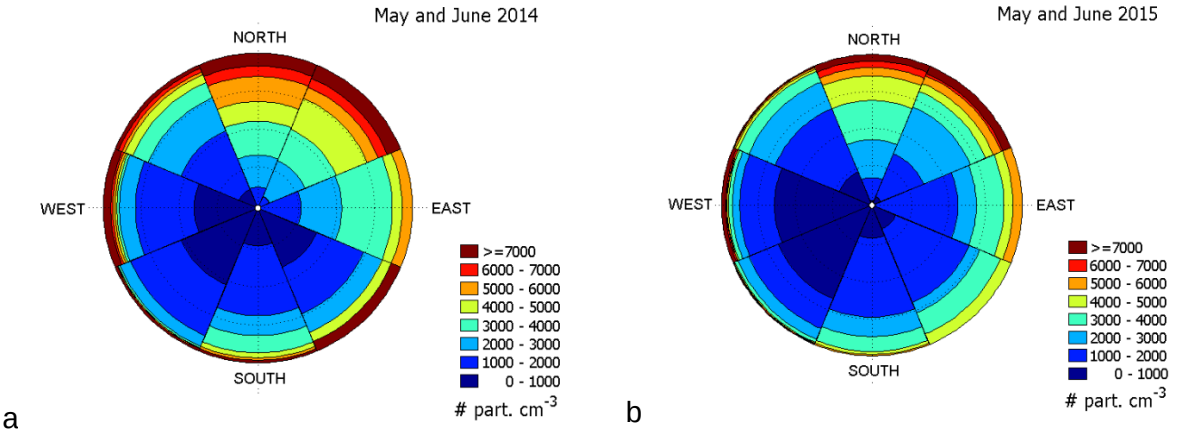


Figure 7) Distribution of summer UFP as a function of wind direction in a) 2014 and b) 2015.

Figure 6b considers UFP data collected only when winds were northwesterly (UFP_NW), i.e. from the direction of Nippon's co-generation boiler relative to the monitoring site. Average UFP_NW were 23 (+/-9) % lower in 2015 relative to 2014, however the differences are more strongly noted at lower concentrations. Figure 7 (a & b) shows the highest concentrations of summer UFP were associated with northerly and northeasterly winds during both years.

Table 2 Average pollutant concentration in May and June 2014 and 2015

Pollutant	May and June 2014	May and June 2015	% change
UFP (# cm^{-3})	2439 (+/-430)	1675 (+/- 201)	-31 (+/-15) %
PM2.5 (µg m^{-3}) (Jul & Aug)	5.8 (+/- 2.6)	5.6 (+/- 2.7)	--
BC (ng m^{-3})	247 (+/- 38)	213 (+/- 21)	-14 (+/- 15) %
SO_2 (ppb)	0.54 (+/- 0.06)	0.45 (+/-0.04)	-16 (+/- 12) %

Table 2 lists the average concentrations of UFP, PM2.5, BC, and SO_2 in May and June 2014 and 2015. As previously mentioned PM2.5 data is from July and August and there is no statistically significant change between the two years. Average ambient black carbon (BC)

mass and SO_2 gas concentrations decreased by 34 ng m^{-3} and 0.09 ppb respectively. Both difference values are below instrument detection levels and not considered significant. When data corresponding to northwesterly winds were considered no difference in SO_2_NW was detected between the two years. The average BC_NW concentration dropped from 294 to 236 ng m^{-3}, a difference of 57 ng m^{-3}, a value below the detection limit of the aethalometer. As with UFP, the highest concentrations of BC and SO_2 were associated with northerly and northeasterly winds and generally displayed the same patterns observed in the UFP data. If wind direction alone were the only determinant, these data would suggest port activity and traffic on Highway 101 were the dominant source of summertime pollutants in both years, however sources corresponding to wind direction are not the only variables to consider.

Wind direction in Port Angeles has a strong diurnal cycle during the summer. Warm land relative to the nearby water sets up an on-shore breeze during the day and then switches to off-shore after sunset (Figure 8). Traffic and other pollution-generating activities occur predominantly during the day and therefore elevated pollution levels appearing to correspond with northerly winds may actually be more influenced by daytime activity.

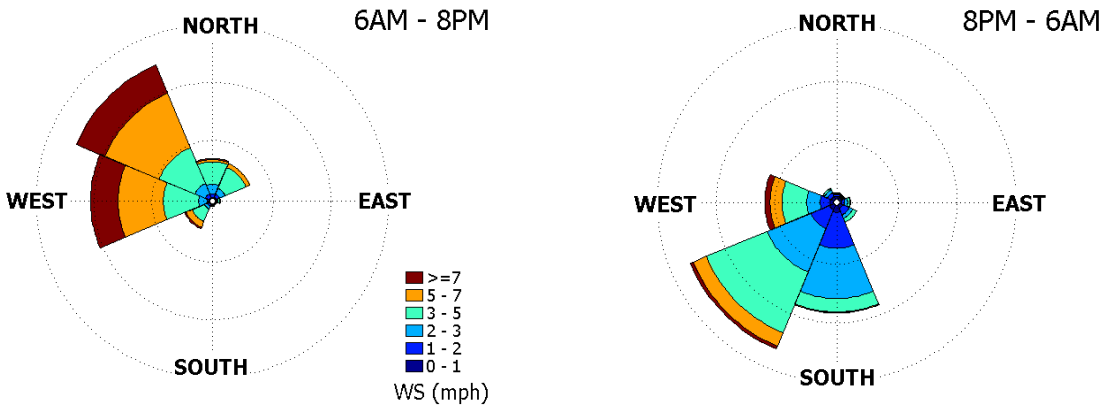


Figure 8) Diurnal on-shore and off-shore winds in May and June.

Based on the UFP measurements obtained in both years, replacing the oil fueled boiler #8 with the biomass co-generation boiler #11 did not measurably increase the ambient concentration of UFP in Port Angeles, WA and possibly caused UFP concentrations to decrease. A null hypothesis (there was no statistical change in average UFP concentrations between May and June 2014 and May and June 2015), paired t-test indicated the hypothesis could be rejected at the 5% confidence levels. When May and June data were considered independently, the null hypothesis for the May data could be rejected at the 5% confidence levels, however the null hypothesis for the June data could not. In other words, May UFP concentrations were statistically and significantly lower in 2015 relative to 2014, but the June data exhibited no statistical difference between the two years.

May and June 2015

The second half of this study attempts to determine the contribution of Nippon's co-generation boiler emissions to UFP and PM2.5 measured at the Port Angeles Fire Station in summer 2015. Data corresponding to northwesterly winds were isolated and divided into two groups: periods of time when the co-generation plant was operating and those hours when it was not. Nippon's co-generation boiler ran for 1061 hours in May and June 2015. Northwesterly winds coincided with operation for 280 of those hours. Northwesterly winds were recorded for 79 hours when the co-generation boiler was not in use.

Data from the co-generation boiler's continuous emission monitors (CEMS) were used to determine when the boiler was operating. CEMS recorded NOx and CO concentrations leaving the stack. The co-generation boiler was considered not operating when both the CO and NOx CEMS reported no values. If only one of the CEMS reported values indicative of operation, data were flagged as boiler operating.

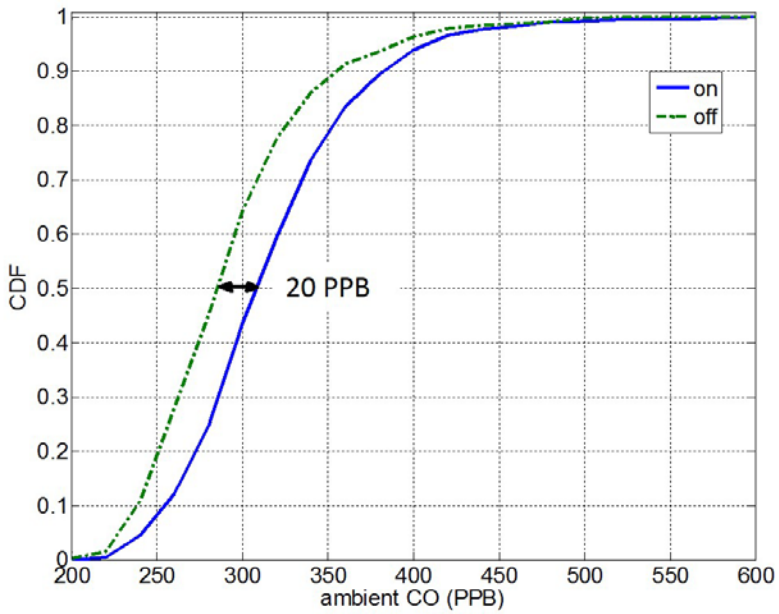


Figure 9) Cumulative distribution function (cdf) for ambient CO concentrations measured at the PAFS in May and June 2015. Data correspond to northwesterly winds.

Ambient CO concentrations and stack emissions of CO were continuously monitored throughout this study period. The CO_CEMS provided data on CO concentrations the co-generation boiler stack emission point. Figure 9 shows the cumulative distribution of ambient CO concentrations measured at the fire station for times when the co-generation boiler was operating and during downtime. Only data collected when winds were northwesterly were analyzed, focusing on the likeliest times the boiler plume would be detected at the monitoring site. The minimum CO concentration for both modes was 200 ppb, making this the baseline concentration of ambient CO in the region. The cumulative frequency distribution curves for operating and non-operating modes show a clear shift in ambient CO about 20 ppb higher during operation (Figure 9). There is little difference between the two operation modes for the highest concentrations of CO, indicating the co-generation boiler was not responsible for the highest observed CO levels. The same analysis for southwesterly winds showed no difference in concentration between the two operating modes. Data corresponding to westerly winds showed a slight shift to higher CO

concentrations (< 10 PPB) during boiler operation. As these results are based on hourly averages, it is likely the west wind data included a small fraction of northwest winds, which could have influenced the observations.

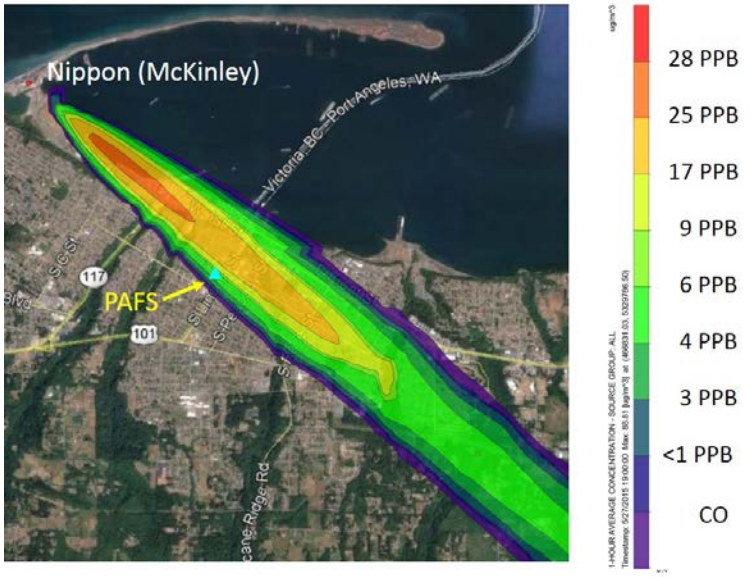


Figure 10) AERMOD result for CO concentrations in the biomass co-generation boiler plume. The plume height was set at 7.6 meters, the height of the air sampling probe. The stack height is 38.1 meters and therefore CO emissions in the immediate vicinity of the stack are above the atmospheric layer shown here.

The CO_CEMS data and AERMOD (a short range atmospheric dispersion model) [*EPA United States Environmental Protection Agency*, 2018] was used to determine if the observed increase in ambient CO was consistent with boiler CO emissions. AERMOD indicated that where the plume intercepted the PAFS measurement site, the CO contribution from the boiler to atmospheric CO was between 1 and 25 PPB. Figure 10 shows a one-hour snapshot when AERMOD predicted the monitoring site was intercepting the plume from Nippon and demonstrates the detected 20 PPB increase shown in Figure 9 is consistent with modeled CO concentrations at the monitoring site. Uncertainties in the model due to surface roughness are

not well quantified. These results provide evidence the air monitors did intercept and detect the emission plume from the co-generation boiler.

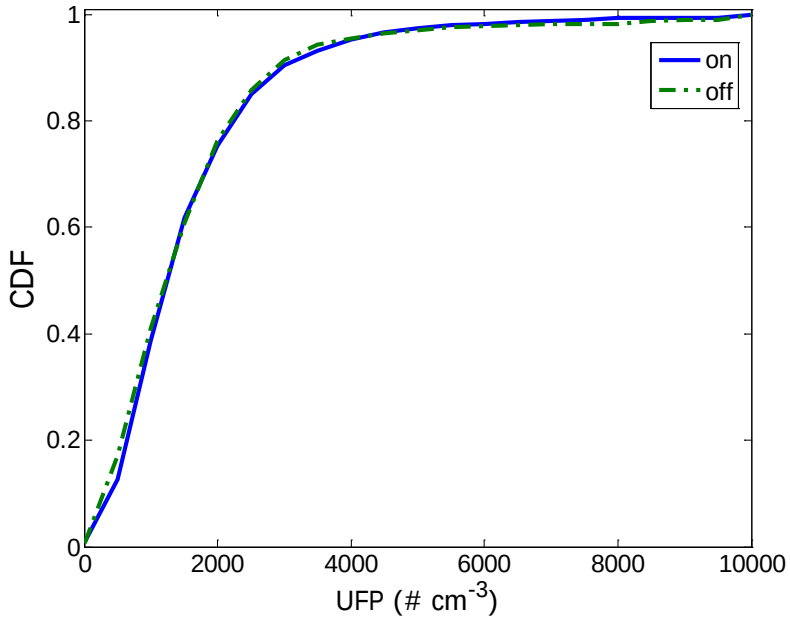


Figure 11) Cumulative distribution function (cdf) for ambient UFP concentrations measured at the PAFS in May and June 2015. Data correspond to northwesterly winds.

The same analysis applied to UFP measurements at PAFS indicates the biomass co-generation boiler did not contribute significantly to ambient UFP concentrations. There was no statistical difference in UFP concentration during hours of operation compared to periods when the boiler was shut down (Figure 11). As with the CO analysis, only data corresponding to northwesterly winds were examined.

Although there was no observed increase in PM2.5 due to co-generation boiler operation (Figure 12a), data indicate between 50 and 100 ng m^{-3} of black carbon (BC) data may have been from boiler emissions (Figure 12b). The aethalometer BC detection limit is between 50 and 100 ng m^{-3} and therefore assigning the perceived BC concentration difference to the boiler cannot be done with a high degree of confidence.

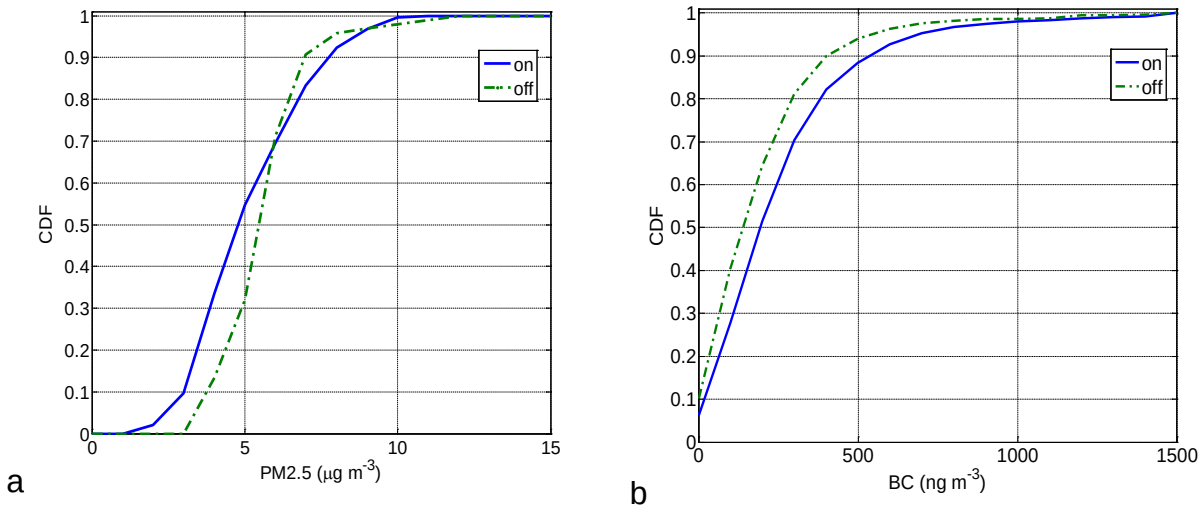


Figure 12) CDF plots for a) PM2.5 and b) BC during boiler operation (solid line) and shutdown (dash dot line). Data represent only measurements corresponding to northwesterly winds.

Discussion

When Nippon submitted a permit application to replace a 1950's era, oil-fueled boiler with a biomass co-generation boiler equipped with the best available control technology (BACT), the final determination predicted PM2.5, sulfur dioxide (SO_2), and sulfuric acid mist to decrease by 78 tons-per-year (TPY) (-68% relative to the old boiler), 133 TPY (-47%), and 8.3 TPY (-50%) respectively, while NOx emissions were forecast to increase by 6.3 TPY (+3%) (*ORCAA FD_10NOC763*). UFP emissions from neither the old nor the new boiler were considered in the final determination as UFP is not a regulated pollutant and no state or federal ambient air quality standards for UFP exist [*Baldauf et al.*, 2016]. This study attempted to measure differences in ambient concentrations of UFP and PM2.5 in response to operational changes at the Nippon mill. At 2.7 km from the source no significant difference in ambient concentration was detected for SO_2, BC, PM2.5 or UFP. NOx was not measured, and CO data were only available after the biomass boiler began operating.

Previous studies have consistently shown UFP concentrations can be very high near their source, but exponentially decay with distance [*Choi and Paulson*, 2016; *Zhu et al.*, 2002a; b]. UFP concentration downwind of a source begins to approach background levels within 300 to 1000 meters. This rapid decrease in UFP concentration is primarily due to coagulation and evaporation. CO and BC were also shown to decrease exponentially with distance, however not quite to the same degree as UFP [*Choi and Paulson*, 2016]. Both remained slightly elevated relative to upwind concentrations. Our data are consistent with published results. Both CO and BC were slightly enhanced during times when the boiler was operating, and favorable winds carried the mill's plume to our monitoring location. No change was detected in coincident UFP concentrations.

UFP, CO, and BC data heavily influenced by known point sources at the fire station were omitted for this analysis. This included 30 minutes of back-up generator testing on Sunday mornings and up to one hour of equipment testing on Saturdays. After these data were removed, spikes in UFP number concentration on the order of 5 to 10 thousand particles per cm^3 above background levels regularly occurred in May and June in 2014 and 2015 on Friday afternoons between 5PM and 7PM. These UFP spikes were tracked to the outdoor grill frequently used Friday evenings by the on-duty firehouse staff and were the highest UFP concentrations measured in the summer.

During the summer, nighttime (10PM to 5AM) UFP concentration were typically between 1 and 2 thousand particles per cm^3, while daytime (5AM to 10PM) values ranged from 2 to 4 thousand particles per cm^3.

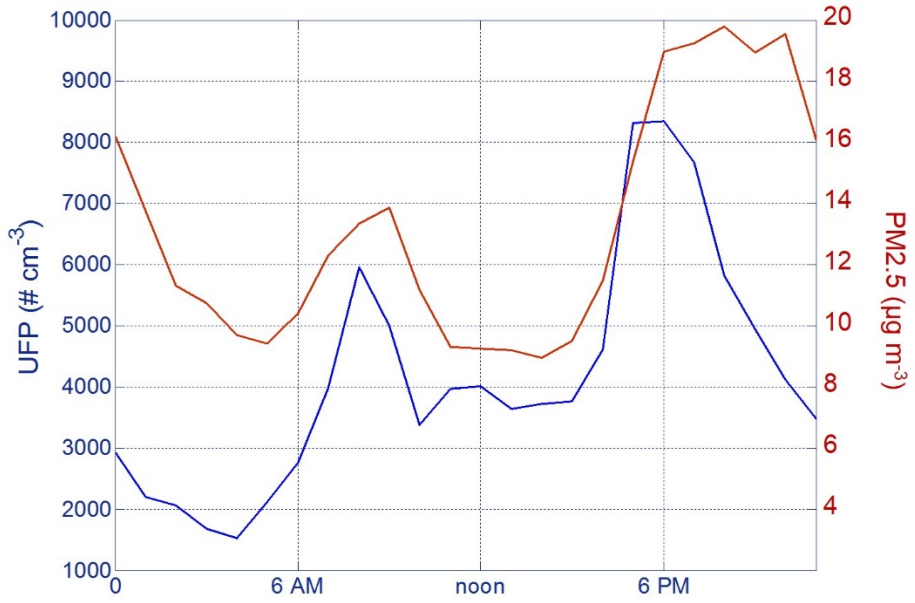


Figure 13) Average diurnal concentration of UFP and PM2.5 in January 2014.

Winter UFP, CO, BC, and PM2.5 daily profiles were all strongly tied to local residential heating practices and meteorology [*Gaston et al.*, 2016]. During cold stagnant periods, average evening UFP concentrations reached 8000 # cm^{-3} and PM2.5 levels climbed to 20 µg m^{-3}. UFP levels typically spiked between 5 and 6 PM before gradually dropping below 2000 # cm^{-3} in early morning hours. PM2.5 concentrations, however, remained elevated throughout the night (Figure 13). These data are consistent with our understanding of the short atmospheric lifetime of UFP. As woodstoves are ignited, smoke emissions cause a rapid increase in UFP and PM2.5. Particle coagulation rate is proportional to particle number concentration [*Finlayson-Pitts and Pitts*, 2000]. As UFP increases so does the rate of larger particle formation, acting as a sink for UFP and total particle number concentration. Larger particles more efficiently scavenge new UFP and cause UFP concentrations to drop further while PM2.5 remains elevated. In a warmer, summer environment, emitted UFP is also subject to evaporation, another mechanism for particle loss.

Average UFP and PM2.5 concentration in Port Angeles in 2014 and 2015 summer months was roughly 2000 (+/-500) # cm^{-3} and 5 µg m^{-3} respectively. Average daily winter values were about 3600 (+/-500) # cm^{-3} and 13 µg m^{-3}. Residential and rural measurements of UFP in Switzerland over 3 different seasons revealed a median concentration of 11,000 (+/- 7800) # cm^{-3} [*Meier et al.*, 2015]. roadside UFP measurements in several US cities ranged from averages of 25,000 to 65,000 # cm^{-3} [*Kumar et al.*, 2014].

Conclusions

In Port Angeles, WA the highest concentrations of UFP and PM2.5 were observed in winter due to an increase in residential wood heating and a higher frequency of air stagnation episodes. During winter, the highest PM2.5 and UFP concentrations were associated with southerly winds. Summer maximum concentrations of UFP and PM2.5 correlated with northerly and northeasterly winds.

In winter, UFP and PM2.5 reached maximum concentrations between evening and early morning hours reflecting both enhanced stagnation at night combined with peak hours for woodstove operation. In summer, both PM2.5 and UFP were highest during the day when there is more traffic, industrial activity, and outdoor burning activity. Although statistically significant, but small, atmospheric increases in BC and CO concentrations were detected when northwesterly winds carried Nippon's emission plume toward the monitoring site, the biomass co-generation facility did not have a measurable effect on ambient air concentrations of UFP or PM2.5 at a monitoring site almost 3 km away.

References

Baldauf, R. W., R. B. Devlin, P. Gehr, R. Giannelli, B. Hassett-Sipple, H. Jung, G. Martini, J. McDonald, J. D. Sacks, and K. Walker (2016), Ultrafine Particle Metrics and Research Considerations: Review of the 2015 UFP Workshop, *International Journal of Environmental Research and Public Health*, *13*(11), 1054, doi:10.3390/ijerph13111054.

Choi, W., and S. E. Paulson (2016), Closing the ultrafine particle number concentration budget at road-to-ambient scale: Implications for particle dynamics, *Aerosol Science and Technology*, *50*(5), 448-461, doi:10.1080/02786826.2016.1155104.

EPA United States Environmental Protection Agency, A. Q. M. G. (2018), User's Guide for teh AMS/EPA Regulatory Model (AERMOD), edited by A. Q. A. D. Office of Air Quality Planning and Standards, Research Triangle Park, North Carolina.

Finlayson-Pitts, B., and J. Pitts (2000), Chemistry of the Upper and Lower Environment: Theory, Experiments, and Applications, in *Chemistry of the Upper and Lower Environment: Theory, Experiments, and Applications*, edited, pp. 375-379, Academic Press.

Gaston, C., F. Lopez-Hilfiker, L. Whybrew, O. Hadley, F. McNair, H. Gao, D. Jaffe, and J. Thornton (2016), Online molecular characterization of fine particulate matter in Port Angeles, WA: Evidence for a major impact from residential wood smoke, *Atmospheric Environment*, *130*, 99-107, doi:10.1016/j.atmosenv.2016.05.013.

Hansen, A. D. A. (2003), *The Aethalometer*, Magee Scientific Company, Berkely CA.

Kirchstetter, T. W., and T. Novakov (2007), Controlled generation of black carbon particles from a diffusion flame and applications in evaluating black carbon measurement methods, *Atmospheric Environment*, *41*(9), 1874-1888.

Kumar, P., L. Morawska, W. Birmili, P. Paasonen, M. Hu, M. Kulmala, R. M. Harrison, L. Norford, and R. Britter (2014), Ultrafine particles in cities, *Environment International*, *66*, 1-10, doi:https://doi.org/10.1016/j.envint.2014.01.013.

Lee, C., P. Erickson, M. Lazarus, G. Smith, and M. Goodin (2010), Greenhouse Gas and Air Pollutant Emissions of Alternatives for Woody Biomass Residues*Rep.*, Stockholm Environment Institute.

Lind, T., J. Hokkinen, J. K. Jokiniemi, S. Saarikoski, and R. Hillamo (2003), Electrostatic Precipitator Collection Efficiency and Trace Element Emissions from Co-Combustion of Biomass and Recovered Fuel in Fluidized-Bed Combustion, *Environmental Science & Technology*, *37*(12), 2842-2846, doi:10.1021/es026314z.

Meier, R., et al. (2015), Ambient Ultrafine Particle Levels at Residential and Reference Sites in Urban and Rural Switzerland, *Environmental Science & Technology*, *49*(5), 2709-2715, doi:10.1021/es505246m.

Mitchell, S. R., M. E. Harmon, and K. E. B. O'Connell (2012), Carbon debt and carbon sequestration parity in forest bioenergy production, *GCB Bioenergy*, *4*(6), 818-827, doi:10.1111/j.1757-1707.2012.01173.x.

Preble, C., T. Dallman, N. Kreisberg, S. Hering, R. Harley, and T. Kirchstetter (2015), Effects of Particle Filters and Selective Catalytic Reduction on Heavy-Duty Diesel Drayage Truck Emissions at the Port of Oakland, *Environmental Science and Technology*, *49*(14), 8864-8871, doi:10.1021/acs.est.5b01117.

Springsteen, B., T. Christofk, S. Eubanks, T. Mason, C. Clavin, and B. Storey (2011), Emission Reductions from Woody Biomass Waste for Energy as an Alternative to Open

Burning, *Journal of the Air & Waste Management Association*, *61*(1), 63-68, doi:10.3155/1047-3289.61.1.63.

Zhu, Y., W. Hinds, S. Kim, S. Shen, and S. Constantinos (2002a), Concentration and Size Distribution of Ultrafine Particles near a Major Highway, *Journal of Air & Waste Management Association*, *52*, 1032-1042.

Zhu, Y., W. Hinds, S. Kim, S. Shen, and S. Constantinos (2002b), Study of ultrafine particles near a major highway with heavy duty diesel traffic, *Atmospheric Environment*, *36*(27), 4323-4335, doi:10.1016/S1352-2310(02)00354-0.