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RESEARCH ARTICLE

Pilot study investigating ambient air toxics emissions near a Canadian kraft pulp and paper facility in Pictou County, Nova Scotia

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Abstract Air toxics are airborne pollutants known or suspected to cause cancer or other serious health effects, including certain volatile organic compounds (VOCs), prioritized by the US Environmental Protection Agency (EPA). While several EPA-designated air toxics are monitored at a subset of Canadian National Air Pollution Surveillance (NAPS) sites, Canada has no specific "air toxics" control priorities. Although pulp and paper (P&P) mills are major industrial emitters of air pollutants, few studies quantified the spectrum of air quality exposures. Moreover, most NAPS monitoring sites are in urban centers; in contrast, rural NAPS sites are sparse with few exposure risk records. The objective of this pilot study was to investigate prioritized air toxic ambient VOC concentrations using NAPS hourly emissions data from a rural Pictou, Nova Scotia Kraft P&P town to document concentration levels, and to determine whether these concentrations correlated with wind direction at the NAPS site (located southwest of the mill). Publicly accessible Environment and Climate Change Canada data (VOC concentrations [Granton NAPS ID: 31201] and local meteorological conditions [Caribou Point]) were examined using temporal (2006–2013) and spatial analytic methods. Results revealed several VOCs (1,3-butadiene, benzene, and carbon tetrachloride) routinely exceeded EPA air toxics-associated cancer risk thresholds. 1,3-Butadiene and tetrachloroethylene were significantly higher ($p < 0.05$) when prevailing wind direction blew from the northeast and the mill towards the NAPS site. Conversely, when prevailing winds originated from the southwest towards the mill, higher median VOC air toxics concentrations at the NAPS site, except carbon tetrachloride, were not observed. Despite study limitations, this is one of few investigations documenting elevated concentrations of certain VOCs air toxics to be associated with P&P emissions in a community. Findings support the need for more research on the extent to which air toxics emissions exist in P&P towns and contribute to poor health in nearby communities.

Keywords Air toxics · Air quality · Volatile organic compounds (VOCs) · Community health · Pulp and paper · Cancer risk

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Introduction

Poor ambient air quality is an increasing global concern with recent revelations that 92% of the world's population is exposed to air pollution levels above the World Health Organization (WHO) air quality guidelines (WHO 2006, 2016; Kelly and Fussell 2015). Ambient air pollution is widely recognized and increasingly associated with a wide range of acute and chronic adverse health effects, including cancer, cardiovascular, respiratory, and mortality outcomes (IOM 2011; Villeneuve et al. 2013; ECCC 2015a). The pathological mechanisms by which these toxic exposures exert their effects are not well understood. WHO highlights the need for research in order to better inform exposure-response relationships (WHO 2016).

Most air pollution surveillance activities are limited to measurement of respirable fine particulate matter ≤2.5 μm ($PM_{2.5}$), without regard to their specific chemical composition and criterion air contaminant (CAC) gases (i.e., nitrogen oxides [NO_x] and ground-level ozone [O_3]). There is growing concern about the toxicity of volatile organic compounds (VOCs) (Cicolella 2008) and the consequences of long-term, low-dose exposure to these agents. VOCs are varied and widespread air pollutants (e.g., hydrocarbons, aromatics, and some chlorinated compounds) that are increasingly recognized as important precursors to $PM_{2.5}$ and ground-level O_3 formation through photochemical reactions (Ryerson et al. 2001). Atmospheric deposition of VOCs may contaminate other environmental media (e.g., soils, sediments, and biota) (ATSDR 2014a; MacAskill et al. 2016). Many VOCs are included in the US Environmental Protection Agency (EPA) "air toxics" list. "Air toxics" are defined as "those pollutants that cause or may cause cancer or other serious health effects […] or adverse environmental and ecological effects" (EPA 2015a).

According to the Canadian Environmental Protection Act 1999 (CEPA), VOC releases are acknowledged as a health concern, but, due to their highly volatile properties, are challenging to monitor and manage (CCME 2011). Although no specific Canadian legislative or regulatory tools address ambient VOC levels, emissions are indirectly controlled through regulatory mitigation of $PM_{2.5}$ and ground-level O_3 under the Canadian Ambient Air Quality Standards (CAAQS). Under CEPA, it is mandatory for owners or facility operators, who meet reporting requirements, to self-report pollutant releases to air, water, and land to Environment and Climate Change Canada (ECCC)'s National Pollutant Release Inventory (NPRI) (ECCC 2014). While this provides a disincentive to those industries releasing these agents, there is less regulatory control or routine monitoring of these agents in Canada which, in turn, limits scientific understanding of sources, exposures, and the effectiveness of current control measures across the country.

Ambient air monitoring in the US is conducted in accordance with the *Clean Air Act* (CAA) (Clean Air Act 1970). CAA amendments identify 187 air toxics, which form the basis for EPA's approach to regulating emissions (EPA 2015a). Of these, EPA identified 30 air toxics that pose the greatest potential health threat in urban areas (EPA 2015b). Although many CEPA-toxic or equivalent agents are monitored by the National Air Pollution Surveillance (NAPS) network, it includes a selection (not all) of EPA's list of prioritized air toxics, and the main criteria for air toxics monitoring in Canada has been their potential contribution to ambient PM and ground-level O_3 (Galarneau et al. 2016). Consequently, there are gaps in understanding of air toxics concentrations across the NAPS network. Using the risk-based principles outlined in CAA, EPA developed the National Air Toxics Assessment (NATA), a comprehensive evaluation tool that prioritizes efforts to regulate emissions of air toxics (EPA 2015c). Such a rigorous initiative has yet to be implemented in Canada, where no federal guidelines exist for ambient air toxics.

Despite economic benefits of the P&P industry, it generates large quantities of atmospheric and effluent emissions, resulting in environmental degradation (Hewitt et al. 2006; Hoffman et al. 2015; Hoffman et al. 2017). P&P mill emissions vary depending on the pulping method, wood species, and by the age and technology used (Soskolne and Sieswerda 2010). P&P mills are industrial emitters of air toxics, although few investigations (e.g., the Nez Perce National Air Toxics Program, funded by EPA [STI 2009]) have characterized ambient concentrations in nearby communities.

Potential adverse health effects associated with exposure to air pollutants in the vicinity and downwind from P&P facilities include respiratory disease, neurophysical symptoms, and higher risks of contracting lung cancer (Soto et al. 1991; Toren et al. 1996; Mirabelli and Wing 2006). Yet, few investigations reported adverse health effects from chronic community-level ambient exposures to P&P mills emissions in Canada (Mirabelli and Wing 2006; Soskolne and Sieswerda 2010). While there have been a number of occupational epidemiological studies of P&P workers, these investigations have not been extended to examine community exposures, due to research design challenges including ecological fallacy (i.e., inferences made about individuals deduced from the population) in community studies (Soskolne and Sieswerda 2010). Additionally, most of these studies focused on respiratory disease outcomes; there is a dearth of epidemiological studies of cardiovascular effects or cancer effects in these communities.

Decades-long concerns for perceived higher incidence and mortality rates for all-cause cancer, cardiovascular disease, chronic respiratory disease, and diabetes (Reid 1989; PCHA 2008; Statistics Canada 2013) in PC, Nova Scotia, have generated considerable community antipathy among residents towards a local P&P mill (Hoffman et al. 2015). This bleached kraft P&P mill ("the mill") is located approximately 3 km south of the town of Pictou (population 3500) and produces approximately 280,000 t of bleached kraft pulp annually from softwood and hardwood chips (NP 2016b) (Fig. 1) and has been in production since 1967 (Ogden 1972). Public backlash gained momentum during 2014 due to the failure of the recovery boiler electrostatic precipitator (i.e., particulate filtration device).

Environmental reporting by the mill, when compared against provincial and federal regulatory compliance standards, contrasted to local perceptions of impacts. Most environmental monitoring reports indicated some

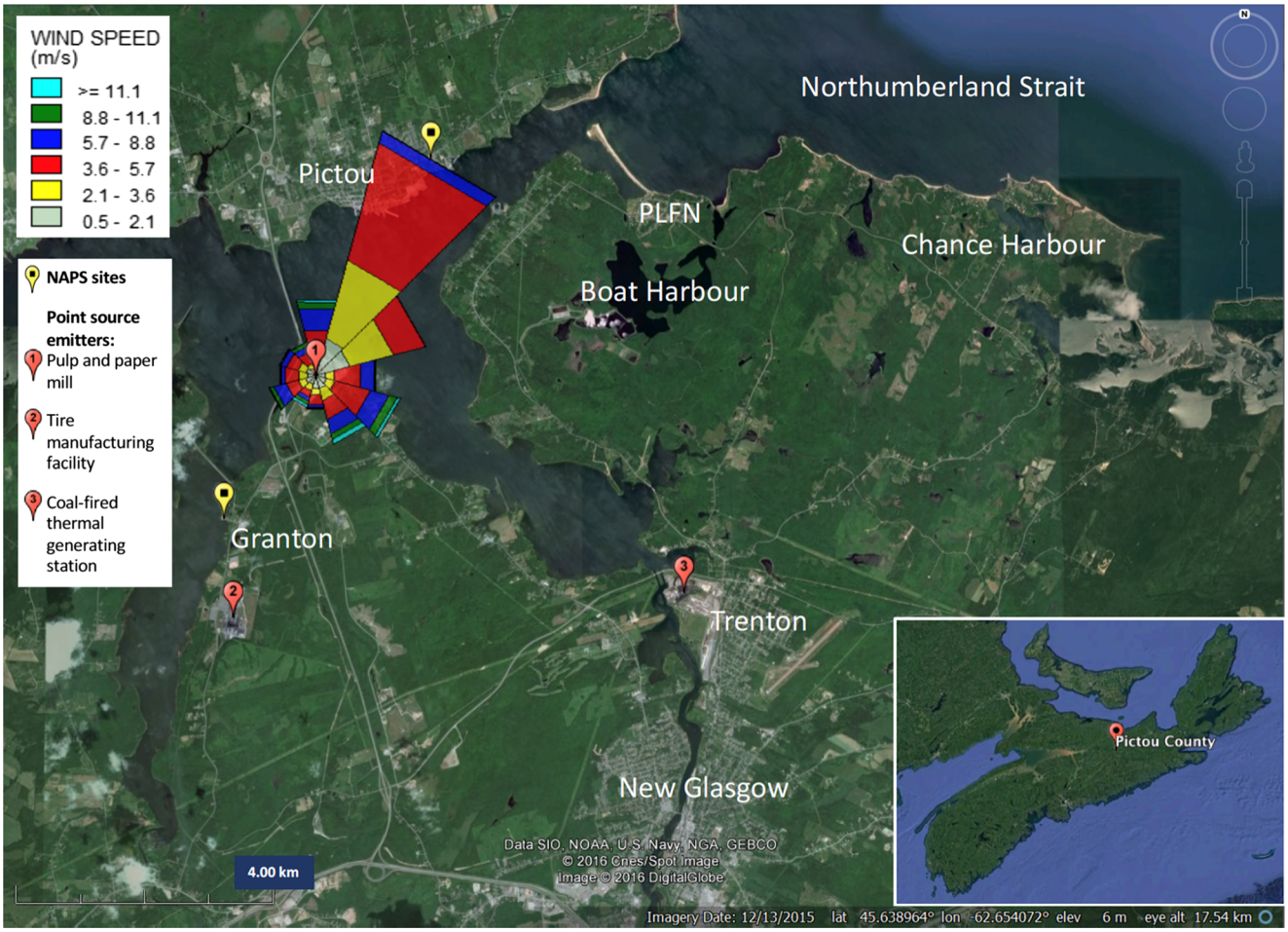

Fig. 1 Summer (2006–2013) wind rose simulation using WRPlot View™ (blowing to direction) with the mill as the focal point (*1*), relative to communities (e.g., Pictou and Pictou Landing First Nation [PLFN]), NAPS discrete receptor sites, and other local point source emitters (e.g., tire manufacturing facility (*2*), coal-fired thermal electrical generating station (*3*)). The length of each radial spoke represents the relative frequency of wind direction (©Google Earth)

level of compliance in atmospheric emissions, but when compliance targets were exceeded, there were inconsistent regulatory enforcement (Hoffman et al. 2015). The mill is required to report emissions to NPRI: conduct third-party stack testing, continuous emission monitoring of total reduced sulfur (TRS), and ambient air monitoring for pollutants found in the Nova Scotia Air Quality Regulations pursuant to the *Environment Act* (NSE 2015). The mill's air emission monitoring data are reviewed by provincial and federal regulators to ensure compliance with applicable environmental permits and air quality objectives (ECCC 2014). Hoffman et al. (2015) provided detailed information on new and existing environmental policies that impose pollution abatement in the P&P industry in Canada, particularly the PC mill (e.g., 2015 Industrial Approval).

These are critical research gaps both in relation to community exposures to VOCs in P&P communities and in regard to adverse health effects resulting from chronic exposure to P&P emissions which are a concern given the potential adverse health outcomes that VOCs and other P&P air emissions pose. This further justifies the need for more research to characterize air quality in this particular subset of industrial communities, which have often been neglected because of their remote locations.

An intensive study of specific ambient air toxic emissions in PC has not been undertaken. The aim of this pilot study was to assess levels of PC community exposures to VOC air toxics emissions from 2006 to 2013, and to evaluate these data in relation to potential risks suggested by EPA air toxic guidelines. The main objective of this study was to determine whether wind direction correlated with prioritized air toxic ambient VOC concentrations at a nearby NAPS site (Granton). As the Granton NAPS site is positioned southwest of the mill, it was hypothesized that prevailing winds (PW) from northerly and northeasterly directions would

positively correlate with an increase in ambient VOC concentrations, as capturing potential VOCs from the mill's plume would be optimized (Fig. 1).

Materials and methods

Spatial and temporal sampling

Historical meteorological and NAPS data from the Granton NAPS site discrete receptor (ID: 31201) were collected from publically assessable ECCC databases (http://climate.weather.gc.ca/climateData/; http://maps-cartes.ec.gc.ca/rnspa-naps/). Hourly surface wind observations (i.e., speed and direction to the nearest 10°) were obtained from the closest EC meteorological station, Caribou Point (45.767° N; 62.683° W), located ~10 km north of the mill (45.652° N; 62.718° W). Temporal data for ambient VOCs monitored at the Granton NAPS station were limited to 2006 to 2013.

Nova Scotia Environment (NSE) operates both NAPS monitoring stations in PC: (i) downtown Pictou (ID: 30901) located 3.5 km northeast of the mill and (ii) Granton (ID: 31201) located 2.5 km southwest of the mill (Fig. 1). The Pictou NAPS site routinely monitors NO, NO_2, NO_x, O_3, $PM_{2.5}$, TRS (not VOCs), and wind characteristics, whereas the Granton site monitors 36 VOC species. Multi-component VOC monitoring at NAPS sites are conducted using canister sampling and gas chromatography/mass spectrometry (GC/MS) (CCME 2011). Sampling of 24 h (midnight to midnight) cumulative ambient air samples ($\mu g/m^3$) are taken on a 1-in-6-day schedule by pumping ambient air into pressurized stainless steel SUMMA® canisters and analyzed by an EC accredited Laboratory (CCME 2011; Galarneau et al. 2016).

Statistical analyses

Variation of meteorological conditions and VOC concentrations were assessed by conducting a spatiotemporal analysis to characterize ambient air toxics emissions in PC from 2006 to 2013. Various analytical methods can be applied to concentration data to estimate source apportionments of air pollutants to provide additional insights into the source/receptor relationships to guide development of more effective air quality management strategies (Hopke 2016). However, given the limitations of having complete VOC data from only one NAPS monitoring station in the region, a full chemical mass balance analysis to identify and apportion sources of atmospheric contaminants were not conducted in this study.

Wind rose plots were generated with WRPlot View™ (©Lakes Environmental Software) to simulate seasonal and spatial variation of wind direction (°) frequency and wind speed (m/s) with the mill as the focal point. Although simplified, wind rose models have proven utility for estimating spatial gradients for fate and transport of pollutants from emission sources (Gibson et al. 2013). Summer, when local people spend more time outdoors (and more vulnerable to outdoor pollution exposure), was a focus of this study (Figs. 1 and 2).

This pilot study was conducted to determine whether ambient concentrations of VOCs exceeded their EPA-associated cancer and/or noncancer risk thresholds, to help identify potential human health concerns in PC. VOCs selected for analysis were based on EPA's list of 30 urban air toxics (EPA 2015b) and *National Air Toxics Trends Station Work Plan Template* (EPA 2015d). Health Canada and the province of Nova Scotia currently do not have specific guidelines for air toxics exposures. Therefore, EPA thresholds were considered a more acceptable standard for carcinogenic exposures in this study. Cancer risk threshold refers to the probability of contracting cancer if exposed to a substance every day over the course of a lifetime (assumed to be 70 years for the purposes of NATA risk characterization). Lower threshold values correspond with higher toxicity. Noncancer risk threshold is associated with effects other than cancer, based on reference concentrations via the "hazard quotient" ratio (HQ; exposure divided by appropriate chronic or acute value) (EPA 2015c). The HQ should not be interpreted as a probability of adverse effects. Noncancer risk thresholds are typically higher compared to cancer risk thresholds, as lower concentrations can elicit a carcinogenic response, whereas other diseases are not triggered until higher exposure thresholds are reached. US and Canadian method detection limits (MDL) are provided (Health Canada 2010; EPA 2015d) (Table 1).

The mill is located approximately 40° northeast of the Granton NAPS site. The selected PW range expected to result in increased VOC concentrations at the NAPS site (±40° either side of the mill [80° total]). A narrower range may be more accurate; however, due to the sample size of VOC samples, the selected range captured more data. All other wind directions (AOWD) represent ranges outside PW (i.e., >80°, <360°). AOWD represent sampling days when no time PW blew from the selected range (i.e., 0 h). VOC concentrations for AOWD were compared to when PW were present for at least 1 h.

Hourly meteorological data were compiled to correspond with ambient VOC sampling. Hourly wind direction within defined PW range (i.e., 360°–80°) was assigned a value of 1; AOWD were assigned a value of 0. Daily totals represented the proportion of time with PW (i.e., 1–24 h) compared to AOWD. Daily

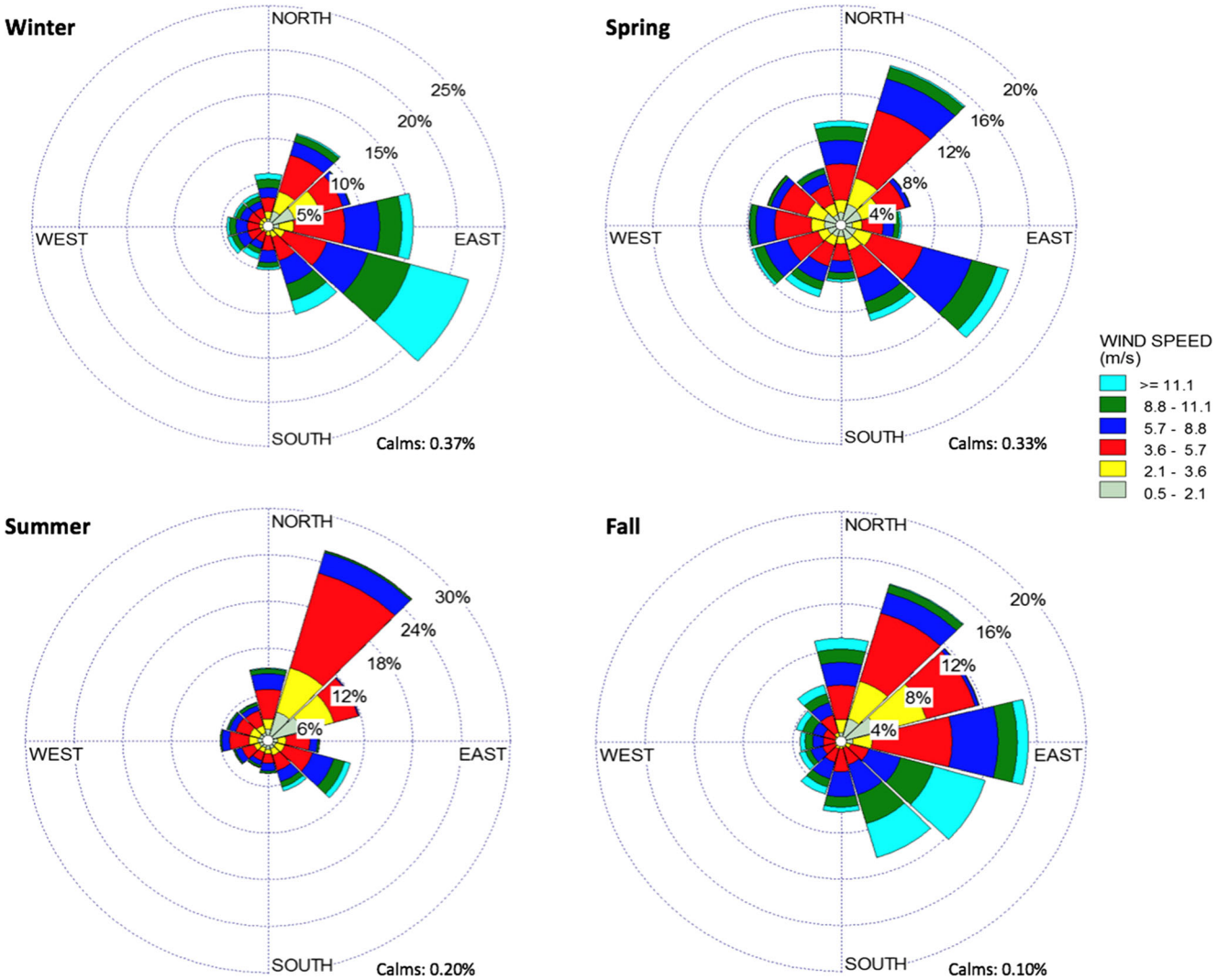

Fig. 2 Seasonal (2006–2013) wind rose simulations using WRPlot View™ (blowing to direction). Percentages represent frequency of wind direction

totals corresponding to VOC sampling were paired. Increasing proportions of PW (AOWD [0 h], ≥1 h, ≥4 h, ≥8 h, ≥12 h) categorized VOC concentrations, which were predicted to correlate with higher VOC concentrations. To test the effect of wind direction and season on ambient VOC concentrations, multivariate analysis of variance (MANOVA) and univariate analysis of variance (ANOVA) were applied in ©R. Due to right-skewed distributions for all VOCs, except for carbon tetrachloride, statistical procedures were performed on both raw and log-transformation of VOC concentrations (Supplementary material Table S1).

Box plots and histograms were used to compare VOC concentrations with PW ≥ 1 h to AOWD on an annual and seasonal basis in relation to their respective cancer and noncancer risk thresholds. See Supplementary material for histograms, and additional box-and-whisker plots illustrating VOC concentrations with increasing time categories with PW (AOWD [0 h], ≥1 h, ≥4 h, ≥8 h, ≥12 h) (Figs. S2 and S4). Box plots display the distribution of data based on a five-number summary: minimum, first quartile, median, third quartile, and maximum. The central rectangle ("box") spans the first to the third quartile (i.e., interquartile range [IQR]). The horizontal line segment within the box represents the median, and "whiskers" above and below the box represent the minimum and maximum. Radar plots consist of a sequence of angular spokes, whose length extending from the center along a separate axis is proportional to the magnitude of the variable relative to the magnitude of the variable across all data points. Lines connect the data values for each spoke. Radar plots were used to display seasonal variation of median VOC concentrations for PW ≥ 1 h and AOWD. One-tailed *t* tests, assuming unequal variance, were performed to determine whether seasonal variation associated with increasing proportions of time

Table 1 List of priority air toxics (i.e., VOCs), associated cancer/noncancer risk thresholds (μg/m^3), and method detection limits (MDL) (μg/m^3) (Health Canada 2010; EPA 2015d)

VOC	Cancer risk[a] (μg/m^3)	Noncancer risk at HQ = 0.1[b] (μg/m^3)	MDL[c] (NATS) (μg/m^3)	MDL (Health Canada) (μg/m^3)
Chloroform	–	9.8	0.50	0.089
1,3-Butadiene	0.0300	0.2	0.10	0.055
Vinyl chloride	0.1100	10.0	0.11	0.046
Benzene	0.1300	3.0	0.13	0.038
Carbon tetrachloride	0.1700	19.0	0.17	0.123
Trichloroethylene	0.2083	0.2	0.20	0.190
Tetrachloroethylene	3.8462	4.0	0.17	0.120

[a] Cancer risk threshold: the probability of contracting cancer over the course of a lifetime (assumed to be 70 years for the purposes of NATA risk characterization). Lower threshold values correspond with higher toxicity (EPA 2015c)

[b] Noncancer risk threshold: the risk associated with effects other than cancer, based on the reference concentration via a ratio known as the "hazard quotient" (HQ; the exposure divided by the appropriate chronic or acute value)

[c] MDL: the lowest concentration that can be detected with confidence. NATA and Health Canada's MDLs are listed for comparison (Health Canada 2010; EPA 2015d)

with PW (i.e., ≥1 h, ≥4 h, ≥8 h, ≥12 h) resulted in a significant increase ($p < 0.05$) in VOC concentrations compared to AOWD (see Supplementary material, Table S3).

Quality control

Standard procedures of the Meteorological Service of Canada have been developed in accordance with internationally recommended procedures established by the World Meteorological Organization (ECCC 2013a). As part of the quality assurance and quality control (QA/QC) program, observational meteorological data are subjected to a computer analysis or review to reveal possible errors. EC and the operating agency are jointly responsible for the NAPS network QA/QC program. Elements of the program include site selection, sampling system requirements, instrument calibration and reference standard requirements, and inter-laboratory testing and performance audits. With few exceptions, analyzers are accorded with EPA designation as either a reference or equivalent method for ambient air monitoring (ECCC 2004). ECCC's air quality laboratories use International Organization for Standardization requirements (i.e., ISO 9001:2008 or ISO/IEC 17025:2005) (ECCC 2013b).

Results and discussion

Meteorological observations

Wind rose simulations illustrate seasonal variability with respect to wind direction, with the mill as the focal point (Fig. 2). During summer, wind blew NNE (25.47%) and ENE (11.80%) directions (aggregate range 15°–75°) towards Pictou and Pictou Landing First Nation (PLFN) (Figs. 1 and 2). Wind blew less frequently towards S-NNW (aggregate range 165°–345°) towards the Graton NAPS site. During winter, wind typically prevailed from the north; the highest frequency (21.99%) blowing ESE, followed by E (15.37%) directions (aggregate range 75°–135°) (Fig. 1). Spring and fall have meteorological characteristics that are similar to summer and winter and were considered transitional periods. Pictou, PLFN, Chance Harbour, Trenton and New Glasgow are communities close to the mill that are downwind of annual PW (range 15°–165°). The Granton NAPS site correlates poorly with seasonal or annual wind directions (Fig. 2).

VOC concentrations

Carbon tetrachloride had the highest median concentration (0.5452 μg/m^3) and vinyl chloride had the lowest (0.0014 μg/m^3) (Fig. 3). Carbon tetrachloride concentrations exceeded its EPA cancer risk threshold (0.1700 μg/m^3) for all samples, with maximum and minimum concentrations of 0.7047 and 0.2892 μg/m^3, respectively. Benzene concentrations exceeded its cancer risk threshold (0.1300 μg/m^3) for most samples, with maximum and minimum concentrations of 1.889 and 0.0266 μg/m^3, respectively. Concentrations of 1,3-butadiene concentrations occasionally exceeded its cancer risk threshold (0.0300 μg/m^3), with maximum and minimum concentrations of 0.1062 and 0 μg/m^3, respectively (Fig. 3). Consequently, 1,3-butadiene, benzene, and carbon tetrachloride were air toxics of primary concern in terms of local

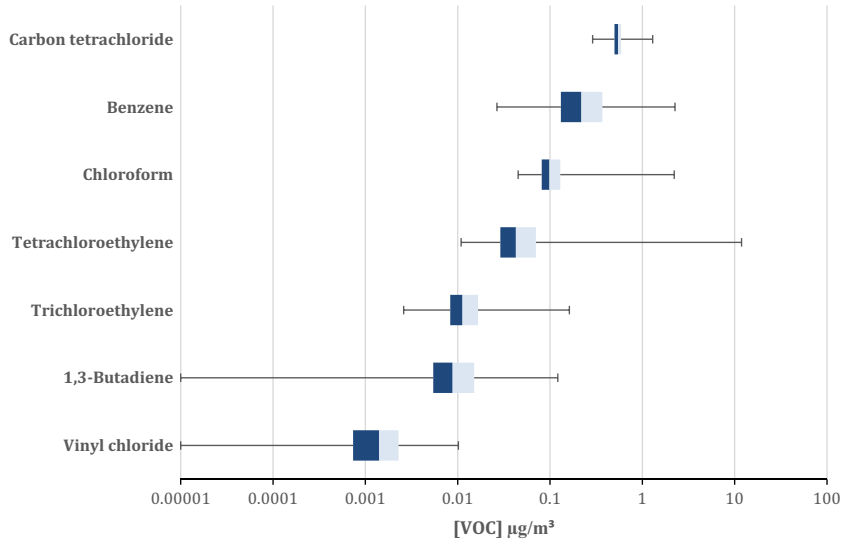

Fig. 3 Relative VOC concentrations (µg/m^3) (2006–2013). Should not be interpreted as orders of magnitude of toxicity. Minimum concentration for 1,3-butadiene and vinyl chloride is 0 µg/m^3 or undetectable

population risk. Other VOCs are presented in Supplementary material (Fig. S5).

ANOVA and MANOVA results revealed that 1,3-butadiene was significantly higher with the presence of PW ≥ 1 h (p = 0.001and p = 0.01 for raw and log-transformed data, respectively). Tetrachloroethylene was also statistically higher with the presence of PW ≥ 1 h (p < 0.01) for log-transformed data. Benzene approached significance with the presence of PW ≥ 1 h (p = 0.07) for log-transformed data. Although not statistically significant, median concentrations of other VOCs, except carbon tetrachloride, were equal or marginally higher with presence of PW ≥ 1 h compared to AOWD. Season had a consistent significant effect on VOC concentrations, except chloroform and tetrachloroethylene (Supplementary material Fig. S2 and Table S1).

Box plots combined with radar graphs illustrate seasonal variation of VOC concentrations of primary concern (i.e., 1,3-butadiene, benzene, carbon tetrachloride) under PW ≥ 1 h and AOWD conditions (Figs. 4, 5, and 6). Median VOC concentrations associated with PW ≥ 1 h and AOWD display parallel

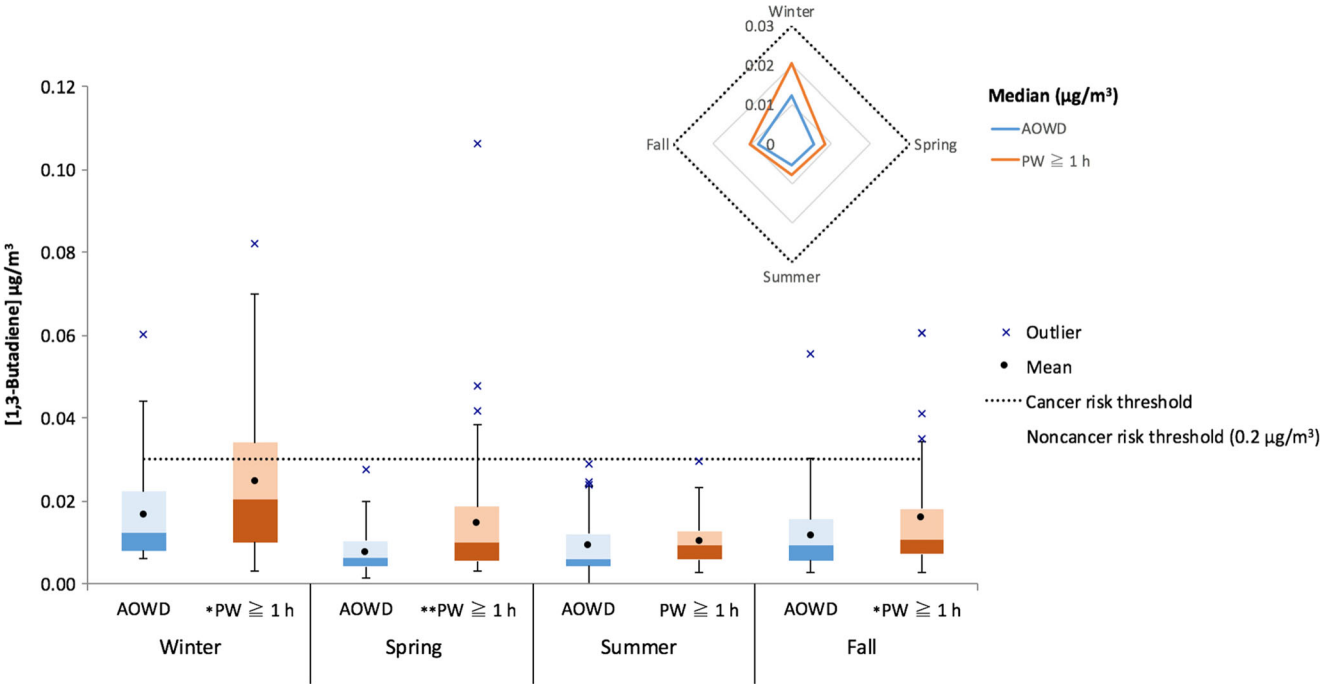

Fig. 4 Seasonal variation (2006–2013) of [1,3-butadiene] (µg/m^3) comparing AOWD to PW for at least 1 h on sampling days (i.e., 360° − 80°), relative to associated cancer and noncancer risk thresholds. Minimum concentration is 0 µg/m^3 or undetectable. Significant differences indicated as *<0.05; **<0.01

seasonal trends. Concentrations of 1,3-butadiene (Fig. 4) and benzene (Fig. 5) exhibit seasonal variation. Both have evidently higher concentrations during winter, with summer having overall lowest concentrations. In addition to exceeding cancer risk thresholds, *t* test results revealed that 1,3-butadiene and benzene concentrations were significantly higher with the presence of PW ≥ 1 h compared to AOWD during at least two seasons, including spring and fall. Conversely, median carbon tetrachloride concentrations showed little variation (Fig. 6). Regardless of season or wind direction, all carbon tetrachloride samples exceeded its associated cancer risk.

This pilot study presents findings of a secondary analysis of 8 years of air toxic VOC exposure data associated with ambient air quality in a Canadian P&P town. Concentrations of three ambient outdoor air toxics routinely exceeded EPA air toxics-associated cancer risk thresholds and are consequently of primary health concern in relation to population health risk in PC: 1,3-butadiene, benzene, and carbon tetrachloride. Exceedance in cancer risk thresholds for these air toxics is consistent in the literature (e.g., Morello-Frosch et al. 2000). The extent to which threshold exceedances of 1,3-butadiene adversely affect human health is poorly understood, with little toxicity information available to compare with cancer risk estimates (Morello-Frosch et al. 2000). With respect to benzene exposure, most monitoring data are associated with occupational studies (ATSDR 2007a), where long-term exposure can cause leukemia (ATSDR 2007b). High exposure to carbon tetrachloride can cause liver, kidney, and central nervous system damage (ATSDR 2005). Combinations of air toxics may have additive or synergistic adverse health effects (Morello-Frosch et al. 2000). Therefore, exposure to mixed VOCs might pose health risks to facility employees and neighboring residents (An et al. 2014; He et al. 2015).

Emission sources within the defined PW range, N to ENE of the Granton NAPS site, may be a causal factor for the increase in VOC concentrations, except carbon tetrachloride. The largest point source emitter within this range is likely the mill; however, the origin(s) of VOCs are inconclusive. According to the mill's most recent substance report submitted to NPRI in 2012, 143.18 t of VOCs were atmospherically emitted on-site (ECCC 2012). An estimated 3.195 t of benzene were released to the air from a stack higher than 50 m and 0.022 t were released within 50 m of the ground. Additionally, benzo(a)anthracene and of benzo(a)phenanthrene were emitted to the air (9.7 and 6.7 kg, respectively) and deposited on-site (0.753 and 0.142 kg, respectively) (ECCC 2012). Although trichloroethylene, tetrachloroethylene, and carbon tetrachloride were not reported to have been released, they may become airborne through evaporation from P&P wastewater (Soskolne and Sieswerda 2010). Boat Harbour (the mill's effluent treatment facility) may therefore contribute to ambient concentrations of VOCs. Collectively, these emissions may have contributed to the ambient atmospheric levels of VOCs measured at the Granton NAPS site. While

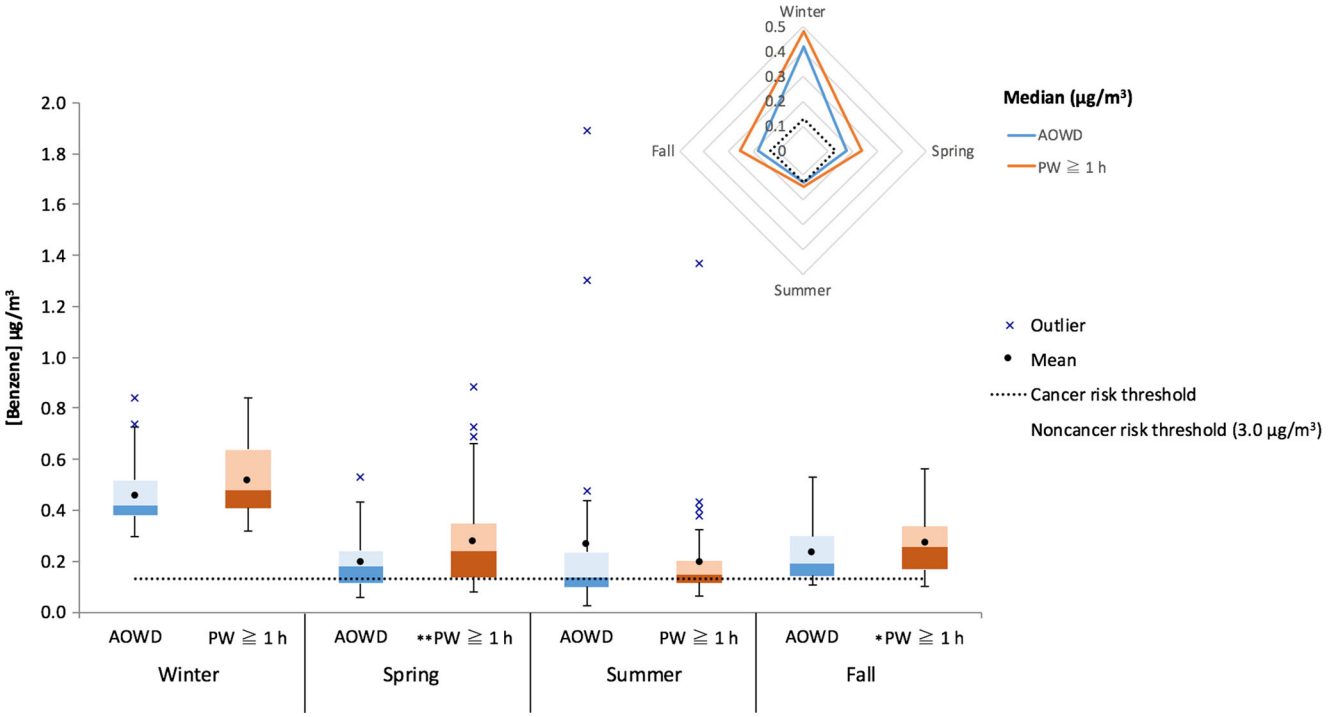

Fig. 5 Seasonal variation (2006–2013) of [benzene] (µg/m^3) comparing AOWD to PW for at least 1 h on sampling days (i.e., 360° − 80°), relative to associated cancer and noncancer risk thresholds. Significant differences indicated as *<0.05; **<0.01

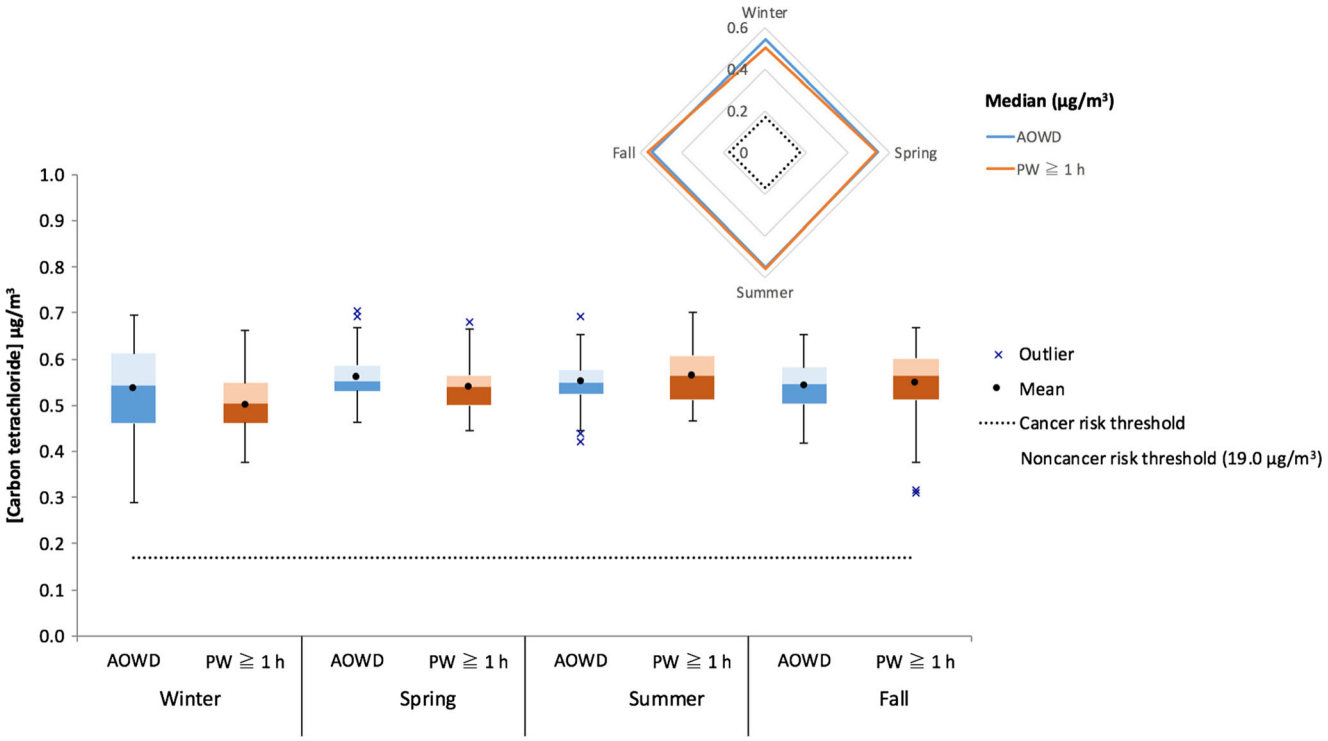

Fig. 6 Seasonal variation (2006–2013) of [carbon tetrachloride] (µg/m^3) comparing AOWD to PW for at least 1 h on sampling days (i.e., 360° − 80°), relative to associated cancer and noncancer risk thresholds

NPRI provides detailed information on pollutant releases, data are self-reported by facilities, with no audits to ensure accuracy. Consequently, data quality may be compromised.

The major chlorinated hydrocarbon emitted into the air from bleached kraft pulp mills of concern is chloroform, which is produced by heating a mixture of chlorine and either chloromethane or methane (EPA 1985). Although chloroform is a recognized by-product of the chlorination process in the P&P industry, it has been suggested that up to 90% of total emission sources may be natural in origin and is widely dispersed in marine environments (McCulloch 2003). As PC is located along the coast of the Northumberland Strait, marine environments may have contributed to the observed ambient chloroform concentrations (see Supplementary material).

Results implicate the mill as a source of air toxics (particularly 1,3-butatdiene and tetrachloroethylene); however, other local sources likely contribute to air toxics emissions. Area and mobile sources have been reported to largely contribute to concentrations of 1,3-butadiene (ATSDR 2014b) and benzene (ATSDR 2007a). Because the Granton NAPS site is located near a highway and access roads, vehicle emissions may have contributed to the observed concentrations of these compounds. A coal-fired thermal generating station and a tire manufacturing facility (located 7 km E and 1.5 km S from the Granton NAPS site, respectively) may be other local point source emitters of VOCs (e.g., 1,3-butadiene is used to make synthetic rubber [ATSDR 2014b]) (Fig. 1). According to the latest NPRI substance reports: the tire manufacturing facility released 220 t of atmospheric VOCs, whereas no VOC releases were reported by the thermal generating station (ECCC 2015b), despite that coal combustion is a significant contributor (Chagger et al. 1999). Direct links between 1,3-butdiene and vinyl chloride with P&P industries were not found in the literature.

Major monitored pollutants at the mill include NO_x, sulfur dioxide (SO_2), and total PM ([TPM] upper size limit of 100 µm diameter) (NP 2016a). A 2013 study concluded that $PM_{2.5}$ concentrations were highest (0.88 µg/m^3) downwind from the mill from using an AERMOD atmospheric dispersion model (Gibson et al. 2013), though this investigation used Halifax wind speed and direction meteorological data (130 km to the south). Hoffman et al. (2015) reported an analysis of 2013 data showing that when Pictou is downwind of the mill, average 1 h ambient $PM_{2.5}$ concentrations result in a twofold increase (12.96 µg/m^3), compared to all other wind directions (5.73 µg/m^3), suggesting the mill is likely the primary contributor of ambient $PM_{2.5}$ in the community. Additionally, TRS, TPM, $PM_{2.5}$, and coarse particulate matter ≤10 µm (PM_{10}) emission exceedances at the mill during 2012 were two to three

orders of magnitude higher than five similar Canadian kraft P&P mills; however, VOC emissions were comparable (Hoffman et al. 2015).

A comparable ambient air toxics monitoring study of a P&P community was conducted in the metropolitan Lewiston, Idaho area and the Nez Perce Reservation (STI 2009). Findings revealed that concentrations of formaldehyde and acetaldehyde were much higher than expected relative to Lewiston's size. Chloroform, tetrachloroethylene, and trichloroethylene were highest at monitoring sites nearest the mill, which presumably contributed at least 50% to pollutant concentrations (STI 2009). However, due to insufficient information on local concentrations of anthropogenic and biogenic VOCs, it was inconclusive whether the mill was a causal factor.

Recent (2009–2013) measurements of ambient CEPA-toxic or equivalent agents monitored at NAPS sites revealed that 11 air toxics, including benzene, chloroform, trichloroethylene, and tetrachloroethylene exceeded ambient air quality guidelines set by respective Canadian jurisdictions (Galarneau et al. 2016). An additional 16 air toxics approached guidelines. Although these guidelines are not necessarily enforceable, CEPA outlines provisions for toxic compounds and are thus subject to risk management actions. Air toxics' contribution to poor health on a regional and national scale has not been thoroughly investigated; therefore, calls into question the effectiveness of current toxic substance management in Canada.

Nova Scotia is known as the "tail pipe of North America," due to being within the trajectory of long-range transport of emissions from transboundary sources along the Eastern Seaboard, plus central and eastern Canada (NSE 2014). Background levels of air pollution that originate from resuspension and natural sources has been found to be major contributors to concentrations of carbon tetrachloride and benzene (Morello-Frosch et al. 2000). Background levels, in combination to carbon tetrachloride's capacity to persist in the atmosphere for a least a year, may explain why observed concentrations are consistently above its associated cancer risk threshold at the Granton NAPS site, regardless of wind direction.

Atmosphere circulation plays a complex role in dispersion, transformation, and removal of pollutants. The dispersion of pollutants from source emitters (e.g., smokestacks) is affected by crosswind mixing in both horizontal and vertical directions. Meteorological variables, including wind speed, wind direction, temperature, humidity, precipitation (process of removal), and atmospheric pressure are the main drivers of variation in pollutant concentrations and dispersion (Bates and Caton 2002). Furthermore, gravitational settling is important for pollutants with larger molecular weights (Oliver 2008); heavier particles settle or deposit closer to emission sources (Walker et al. 2003a, 2003b). Gravitational settling may also explain the high concentrations of carbon tetrachloride.

Topography and coastal conditions can affect wind characteristics (e.g., direction, speed) and the behavior of pollutant transport. A sea breeze that is trapped under descending warmer air from land can exaggerate conditions at coastal zones, a phenomenon known as coastal inversion (Bates and Caton 2002). In addition, turbulent winds along the coast may influence wind characteristics at the Caribou Point meteorological station, and the fate and transport of pollutants. Such coastal conditions, in combination with transboundary air pollution, may be occurrences that coastal areas experience in Nova Scotia, including PC.

Seasonal variability

Seasonal variability exists for both meteorological conditions and VOC concentrations. Variations in meteorological conditions, the nature and intensity of emissions from nearby sources, and photochemical activity are factors that could have led to the observed seasonal variability of outdoor VOC levels (Al-Khulaifi et al. 2014). Of the three VOCs considered particular concern in this study, 1,3-butdaiene and benzene exhibited the highest concentrations during winter. Photochemical reactions involved with ground-level O_3 formation are catalyzed by ultraviolet radiation and temperature. Therefore, peak ground-level O_3 levels typically occur during warm days with sufficient sunlight exposure; thus, people are more vulnerable to exposure during summer. The opposite is true during winter, when available light is diminished, and temperatures are colder (ATSDR 2014b).

Demographic behavior and technological improvements that aim to mitigate emissions (e.g., smokestack precipitator installation in 2015) also need to be considered when evaluating pollutant concentrations. For instance, households in the Atlantic provinces are heated primarily with oil, electricity, and wood or wood pellets (Statistics Canada 2011); therefore, as residential heating increases during winter, biogenic VOCs (e.g., benzene [ATSDR 2007a], 1,3-butadiene [ATSDR 2014b]) from wood burning may have contributed to higher concentrations of these compounds observed in this study. As the mill operates on a 24/7 schedule (ECCC 2012), atmospheric VOC emissions were assumed consistent throughout the year.

Implications

Location of ambient air quality monitoring stations has a direct impact on the observed concentrations of pollutants

(Craig et al. 2008). Based on the time series and spatial analyses, wind direction appears to play a key role in the Granton NAPS site's ability to monitor ambient VOCs from the mill. PW ≥ 1 h from the selected range (360°–80°) typically resulted in equal or higher VOC concentrations for all compounds, except carbon tetrachloride, compared to AOWD (Figs. 4, 5, and 6; Supplementary material), suggesting that the mill is likely a causal factor. Furthermore, as there is a higher frequency of northerly winds blowing towards the south during winter (Fig. 2), the Granton NAPS site is more likely to capture ambient pollutants from the mill's atmospheric emissions. Southwest PW blowing towards Pictou dominate during the summer months when people are more vulnerable to ambient air pollution exposure. Due to Pictou's geography, air toxics from the Eastern Seaboard in combination with local emission sources, including the mill, converge there; hence, higher concentrations of VOCs are expected in Pictou during summer. Subsequently, southwest PW are expected to result in lower VOC concentrations at the Granton NAPS site, as capturing the mill's atmospheric emissions would not be optimized. Therefore, VOC concentrations at the Granton NAPS site during winter would likely be representative of ambient VOC concentrations in Pictou during summer. Moreover, Pictou's considerably larger population base compared to the rural area of Granton further confirms that the NAPS site is not strategically positioned to accurately represent ambient levels of air toxics where there is higher residential exposure.

Study limitations

This study only evaluated exposure to ambient VOC air pollutants. Human exposure to air pollution is a combination of both outdoor and indoor environments and varies according to daily activity patterns and the conditions of specific settings. Secondary data analysis was used in this study; therefore, the ecological nature of these findings limit the explicit attribution of ambient air toxic exposures to the risk potential for cancer for community residents. Personal exposure monitoring, more detailed spatial analysis of ambient conditions, and source apportionment studies would be required to establish more explicitly the health risk associated with these exposures. The analysis was limited by the inability to examine the interaction of local meteorological conditions. Meteorological data were retrieved from Caribou Point, located approximately 10 km from the mill; consequently, coastal conditions may cause differences in meteorological measurements between sites.

Future research and monitoring

A *field component* consisting of real-time measurements of ambient air toxics would improve the rigor and validly of the present study. Although labor intensive, air toxics samples can be analyzed with a high degree of accuracy (Craig et al. 2008). Because monitoring stations are typically fixed, government-approved *atmospheric dispersion modeling* that considers landscape dynamics and seasonal meteorological variability (e.g., ©CALPUFF, ©AERMOD) would more accurately estimate spatial patterns of air toxics dispersion, and human exposure at the population or individual level (EPA 2013). This would require numerous stations within the community so would likely only be feasible for a specific research investigation. Further, installation of a new precipitator in 2015 has likely changed in ambient conditions. A follow-up assessment would provide a comparison to these findings to determine if VOC levels have improved. Additional research includes applying a *Conditional Probability Function* to calculate the probability that an air pollution source is located within a particular wind direction sector to help determine direction of a source from a NAPS discrete receptor site, and conducting an analysis of the effect of mixing height on measured VOC concentrations to further investigate seasonal patterns.

Investigation of health outcomes might involve longitudinal epidemiological research of human exposures to air toxics emissions in the ambient Pictou environment with appropriate consideration for latency of health outcomes, while controlling for indoor and occupational sources and other contextual factors. Several recent Canadian nationwide cohort studies that may provide a foundation for such investigations have been described (e.g., Crouse et al. 2012).

A *comprehensive risk assessment* investigates uncertainties that have implications for risk estimates in the present study, including those surrounding toxicity information (Morello-Frosch et al. 2000). More research is required to determine what cancer and noncancer risks are from ambient air toxics exposure. Further, it is important to consider synergistic effects of a full suite of ambient pollutants, and physical and chemical processes involved in fate and transport of these compounds. Comprehensive emission inventories are necessary to thoroughly address (i.e., characterize, model, and manage) air quality issues (CEC 2009). Collectively, these research efforts aim to better inform air quality management, composed of federal (e.g., ECCC, Health Canada) and provincial (e.g., NSE, Nova Scotia Department of Health and Wellness) government and public health agencies, how best to proceed to ensure the health of residents in industrial communities is prioritized. Implications of the current findings warrant further investigation.

Given the contribution emissions from local sources have to regional, national, and global airsheds, local mitigation initiatives should be an integral part of air quality strategies. There is no common approach to assess health effects of a mixture of pollutants, as they tend to be site specific; hence, an assortment of effective measures may be required. Case studies that provide evidence of effective of air quality

management interventions and guidance documents for risk managers may help inform air quality management for stakeholders (Craig et al. 2008).

To address potential adverse health effects associated with degraded air quality, Health Canada, the Public Health Agency of Canada (PHAC) and provincial partners might work collaboratively with local stakeholders to mitigate health risks and improve efficient industrial technology, while balancing economic, political, and social factors in development and implementation of air quality management. Mitigating industrial emissions has beneficial outcomes for wellbeing (Clougherty 2010); environmental stewardship and governance fosters a more proactive and cleaner environment, while building trusting relationships between industrial stakeholders (Pascal et al. 2013). "A comprehensive enforcement program with mandatory reporting of emissions, […] and meaningful penalties for noncompliance ensures that emission standards are being met" (Craig et al. 2008), and facility operators are held accountable. Data collected internally by the mill is not readily available. To improve transparency, siting rationale for air quality monitoring stations and accompanying data should be provided as part of a commitment to corporate responsibility of the mill (Hoffman et al. 2015).

To improve air quality conditions, stakeholders could increase the capacity for surveillance, assessment, and response to air quality. Furthermore, evaluation of a wide-suite of air toxics, including NATA compounds not measured by the NAPS network (particularly prioritized air toxics) would contribute to ensuring that air quality in Canada is adequately studied. Therefore, ECCC and NSE should consider implementation of a long-term monitoring program for priority air toxics that is comparable to the NATA network monitored by EPA to characterize air toxics exposure on local, regional, and national scales. Data will be useful to help mitigate emissions and achieve acceptable air quality standards that do not exceed cancer or noncancer risk thresholds.

ECCC should also consider the feasibility of installing and maintaining additional strategically placed NAPS sites to improve air pollution evaluation in both rural and urban areas, as well as in microenvironments (e.g., near point source emitters, high-traffic areas) (Craig et al. 2008). More effective communication of the results is required to increase transparency among stakeholders, including the public (Hoffman et al. 2015). Based on the population's risk of exposure, it is strongly recommended that ambient air toxics monitoring to be incorporated at the established NAPS station in Pictou to optimize capturing of said air toxics, and to best correlate pertinent results. Additionally, atmospheric dispersion modeling should use local meteorological data; therefore, meteorological data should also be collected concurrently at NAPS sites to help identify source emitters.

Measurement of individual VOC compounds is necessary to provide insight into their contribution to $PM_{2.5}$ and ground-level O_3 formation. Data would be useful to help target large source emitters and aid regulatory enforcement. Establishment of stringent and/or adapted air quality standards that encompass more air toxics (e.g., VOCs) fosters strong public support and political engagement to address air quality issues. Moreover, health impacts associated with background air pollution should be estimated. Air quality management programs are human resource intensive; therefore, they must have clear and feasible short- and long-term objectives. These initiatives gain predictive insights on atmospheric chemistry, and engage and support relevant sectors in the development and implementation of policies to reduce health risks associated with air pollution exposure (Craig et al. 2008).

Conclusions

Findings reveal that 1,3-butadiene, benzene, and carbon tetrachloride exceeded their respective cancer risk thresholds and are of primary health concern in terms of population risk. Results highlight associations with wind direction and the Granton NAPS site's ambient VOC concentrations in relation to location of the pulp mill. Compared to AOWD, PW from the selected range (360°–80°) typically resulted in higher VOC concentrations for all compounds, except carbon tetrachloride, suggesting that the mill is likely a contributor to increased concentrations. In addition, there are clear seasonal variations of meteorological conditions and VOC concentrations. Southwest PW blowing towards Pictou dominate during summer months, when people spend more time outdoors, and consequently are exposed to higher concentrations. Due to Pictou's geography, air toxics from transboundary and local sources may converge in summer, resulting in higher VOC concentrations. Findings suggest the Granton NAPS site is not positioned to accurately represent ambient levels of toxicity in PC. Therefore, ECCC and NSE should consider incorporating ambient air toxics (e.g., VOCs) monitoring at the established Pictou NAPS site where there is higher residential exposure.

Future research will provide air quality management with a comprehensive characterization of air toxics to support informed public health decisions. Moreover, this pilot study may serve as a precursor to gaining awareness, so that government agencies adopt more stringent air quality regulations and monitoring programs to ensure health of citizens is safeguarded and prioritized.

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