CHARACTERIZATION OF AIR EMISSIONS FROM PULP AND PAPER MILL FACILITIES IN ATLANTIC CANADA

by

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ABSTRACT

A kraft pulp mill that operated in Pictou (KPMP), Nova Scotia for more than 50 years is considered a major source of environmental pollution, although its air emissions have not been widely studied. To address this gap, this research compares air releases from the KPMP with other industrial facilities releases and with regulations. A comparison between nine different pulp and/or paper mills in Atlantic Canada between 2002-2019 suggested that pulp mills emit higher pollutant loads than those producing paper and that the highest release was fine particulate matter ($PM_{2.5}$) from the KPMP. The KPMP releases were also 10 and 1000 times the releases of a tire manufacturing facility and a coal-fired power plant nearby, and results suggest that the KPMP was likely the primary source for high $PM_{2.5}$ concentrations in Pictou. Air quality $PM_{2.5}$ management levels in Pictou became green following the installation of a precipitator by the KPMP in 2015.

LIST OF ABBREVIATIONS USED

APEI	Air Pollutant Emission Inventory
ARL	Annual Recommended Limits
AT	Atholville
BH	Boat Harbour
CAAQS	Canadian Ambient Air Quality Standards
CB	Corner Brook
CO	Carbon Monoxide
CFPGS	Coal-fired power generating station
CPMAEPPF	Code of Practice for the Management of Air Emissions from Pulp and Paper Facilities
DRT	Differences from the Reporting Threshold
ECCC	Environment and Climate Change Canada
ED	Twin Rivers Paper
HA	Port Hawkesbury
HYSPLIT	Hybrid Single-Particle Lagrangian Integrated Trajectory
IA	Irving Paper Limited
IP	Irving P&P Limited
KPM	Kraft Pulp Mill
Mt	Mega Tonnes
NA	Nackawic
NAICS	North America Industry Classification System
NAPS	National Air Pollution Surveillance
NB	New Brunswick
NL	Newfoundland and Labrador
NO_x	Nitrogen Oxides
NO_2	Nitrogen Dioxide
NP	Northern Pulp
NPRI	National Pollutant Release Inventory

NS	Nova Scotia
O3	Ozone
P&P	Pulp and Paper
PC	Pictou County
PM	Particulate matter
$PM_{2.5}$	Particulate matter with a diameter below 2.5µm
PM_{10}	Particulate matter with a diameter below 10µm
PLFN	Pictou Landing First Nation
SO_2	Sulphur Dioxide
SW	Southwest
SSW	South-southwest
TMF	Tire Manufacturing Facility
TPM	Total Particulate Matter
TPY	Tonnes per year
TRS	Total Reduced Sulfur
UT	Lake Utopia
VOCs	Volatile Organic Compounds
WD	Wind direction
WS	Wind speed

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CHAPTER 1 INTRODUCTION

1.1 Air pollution from industrial sources

Environmental issues affect nature, communities, human and animal health, and natural resources. Air pollution is one of the greatest threats to human health, as it is considered to cause have caused 6.7 million premature death in 2019 globally (Health Effects Institute 2020). Air pollutants, such as particulate matter (PM), are among the biggest environmental risks to human health as they have the potential to increase mortality from stroke, heart disease, or pulmonary disease, among others (World Health Organization 2019). Continuous monitoring of air pollutants enables governments worldwide to evaluate the compliance of air quality guidelines and regulations, and to act in case high emissions are detected. However, the readily available evidence of environmental degradation does not warrant that air emissions will be controlled or reduced to prevent more damage (Westwood et al. 2019). There are many other factors that influence the process of reducing air emissions, such as commitment from different stakeholders, industry pressure on the government, and social and economic aspects (Jacob et al. 2018; Johnston Edwards and Walker 2020). Thus, it is important to gather scientific evidence that supports environment protection to be considered in the decision-making process to reduce air emissions when there are multiple stakeholders involved.

Outdoor air pollution affects all regions of the world and is mainly caused by, although not limited to, fossil fuel combustion, industrial atmospheric emissions, forest fires and natural dust (Ghosal, Stephan, and Weiss 2019; Kelly and Fussell 2015; Stewart et al. 2019; Wang et al. 2021, 2022). Emissions from the pulp and paper (P&P) industry are extensive, comprising both air pollutants and greenhouse gases, resulting from considerable energy consumption as part of the production process (Cheremisinoff and Rosenfeld 2010). Pollution from P&P industries occur primarily because of the process of converting wood to pulp and paper, bleaching, and the application of chemicals in the papermaking process that result primarily in air and wastewater effluent emissions (Torén, Persson, and Wingren 1996) . Wastewater created from pulp and paper operations mainly contains suspended

solids, dissolved inorganic substances and blanch agents generated mostly by washing, bleaching, and from the chemical recovery system (Mandeep, Kumar Gupta, and Shukla 2020). Although water discharges can be treated and reused, landfill and food chains can end up being affected if adequate pollution prevention procedures are not implemented (Kamali and Khodaparast 2015; Mandeep et al. 2019, 2020). While health and environmental impact from water pollution from P&P mills has been well documented, few studies have focused on air pollution emissions and their potential to cause human health impacts (Dionne and Walker 2021).

1.1.1 Air emissions from a Kraft Pulp Mill in Nova Scotia

Historically, the P&P industry has been challenged to comply with emission regulations worldwide (Kamali and Khodaparast, 2015). In Pictou County (PC), Nova Scotia (NS), Canada, a kraft pulp mill that operated for more than 50 years is considered a major source of environmental pollution (Hoffman et al., 2015). Boat Harbour (BH), a tidal estuary that received the wastewater effluent, presented dioxins, furans and metals above the Canadian Sediment Quality Guidelines and Canadian Water Quality Guidelines (Group Pictou Landing Native Women's (c/o Sheila Francis Past President) et al., 2016; Hoffman et al., 2017). In 2012, Total Reduced Sulfurs (TRS) and PM emissions were between five to 20 times higher than other kraft pulp and paper mills in Canada (Hoffman et al. 2015).

The pulp mill in Pictou, currently named Northern Pulp Nova Scotia Corporation (NP) and currently owned by Paper Excellence, began its operation in 1967. Since the beginning, the local population expressed concern about air and water pollution, the decrease in the number of fish, an increase in odours, the loss of landscape and the destruction of a sacred site for the Mi'kmaq community (Baxter, 2017; Castleden et al., 2017; CBC News, 2014a; Group Pictou Landing Native Women's (c/o Sheila Francis Past President) et al., 2016; Lewis et al., 2020b; Reid, 1989; The Canadian Press, 2014). Also, the promise of keeping Boat Harbour (BH) lagoon clean of discharge was not kept (Castleden et al., 2017). Since its more than fifty years of operation the mill was nearly close several times due to non-compliance with environmental regulations, but the provincial government of NS granted extensions (Baxter, 2021; Hoffman et al., 2015; University of King College Journalism Students, 2009). However, in 2014 an effluent spill triggered the *Boat Harbour Act* (2015),

which stated that the NP should stop discharging effluents into BH and remediate the estuary (Hoffman et al., 2015; Quanz et al., 2021). NP's failure to present an adequate alternative lagoon of discharge resulted in the closure of the mill on 31 January 2020 (Davidson et al., 2021). Although the mill is still closed, its authorities are currently suing the provincial government of NS for almost half a billion dollars, alleging that they were forced to close losing millions as a result (Beswick, 2021; Gorman, 2022).

Previous studies documented that the most concerning air emissions from NP were PM and volatile organic compounds (VOCs) (Hoffman et al., 2017a, 2015). A field campaign with dispersion modeling showed that PM was affecting Pictou Landing First Nation (PLFN), although in small quantities, and that other air pollutants were not measured in quantities to be considered a threat for human health (Group Pictou Landing Native Women's (c/o Sheila Francis Past President) et al., 2016). In 2014, a high emission of PM was recorded near Pictou and NP was identified as the source (Hoffman et al., 2015). Aiming to improve air emissions, in 2015 the pulp mill installed a recovery boiler electrostatic precipitator to reduce PM emissions from the combustion fumes (Hoffman et al., 2017a). Although a follow-up study was recommended, there is no research focused on analyzing the effectiveness of the precipitator even though the data is publicly available.

1.2 Research overview

The pulp mill in Pictou represents a controversial facility that has been historically identified as a major local source of environmental pollution and is potentially responsible in part for several environmental and human health impacts in nearby communities. While the mill has been criticized for different issues, this thesis focuses only on air pollution emissions and compares pollutant emissions from NP with other industrial facilities in Atlantic Canada.

The study is divided into two stages. The first stage consists of a comparison of annual self-reported industrial emissions from the National Pollutant Release Inventory (NPRI) of seven different air pollutants among nine existing pulp, paper, and P&P mills in NS, New Brunswick, and Newfoundland and Labrador. As the NPRI does not present an upper cap

for annual emissions, the reported releases were compared to the reporting threshold (when annual emissions exceed this lower reporting threshold, the facilities must report the total releases). This part provides long-term insights into how the mill is performing compared to similar facilities to understand whether the continuous complaints about NP's pollution were appropriate. The second stage compares hourly, daily, and annual concentrations of PM with a diameter of ≤2.5 μm ($PM_{2.5}$) recorded on the Pictou station as part of the National Air Pollution Surveillance (NAPS) program. This analysis seeks to understand the contribution of three industrial facilities in PC (NP, a coal-fired generation station, and a tire manufacturer) to atmospheric concentrations of air pollution. The data used on both stages, NPRI and NAPS, consist of government publicly available data from Environmental and Climate Change Canada (ECCC).

The aims of this project are i) to research the effectiveness of environmental mitigation and management strategies to improve air quality by NP; and ii) to generate an updated comparison of air quality among industries in Atlantic Canada, providing a general overview of how the facilities comply with regulations.

The specific purpose of this research is to answer the following questions:

1. Did the precipitator installed in NP in 2015 effectively remove the intended atmospheric pollutants?
1. How was air emission compliance regulation in NP in comparison to similar pulp and/or paper mills in the region?
1. How much did NP contribute to the PM monitored in Pictou air quality station?

1.3 Thesis organization

The thesis is organized as follows. Chapter 1 briefly introduces the research topic and summarizes the main aspects of the manuscript. Chapter 2 presents the literature review and background information. Chapter 3 and Chapter 4 are presented as stand-alone journal articles with specific journal structures (abstract, introduction, data, methodology, results, discussion, and conclusions). This layout may result in some repetition between chapters. Chapter 3 compares annual air emissions from pulp and paper mills in Atlantic Canada and

has been accepted in the journal Pollutants on March 29, 2022. Chapter 4 focuses on fine particulate matter emission in Pictou and identifies potential sources of emissions. This paper is currently under preparation to be submitted to Atmospheric Pollution Research. Finally, Chapter 5 summarizes the key findings, the main recommendations, and the conclusions of the research.

CHAPTER 2 AIR POLLUTION FROM PULP AND PAPER MILLS

2.1 Air pollution

Air pollutants are emitted to the atmosphere by natural sources (such as volcanic eruption and biological decomposition) and by anthropogenic emissions (such as industrial activities and the transport sector) (Muralikrishna and Manickam 2017; Nadadur et al. 2007; Stanek et al. 2011; Strum and Scheffe 2016). The industrial sector is one of the principal sources of air pollutants emissions mainly through the combustion of fossil fuels and biofuels during energy-intensive production processes (Ghosal et al. 2019; Strum and Scheffe 2016) as shown in Figure 1 (International Energy Agency 2016). Even when air emissions are released from point sources, like industrial facilities, the interaction of air pollutants with sunlight, with other pollutants, and with water vapour plus the effect of the local meteorology through atmospheric circulation can modify the nature of the pollutant (i.e., its toxicity) and its region of influence (European Environment Agency 2016; Nadadur et al. 2007; Yang et al. 2015).

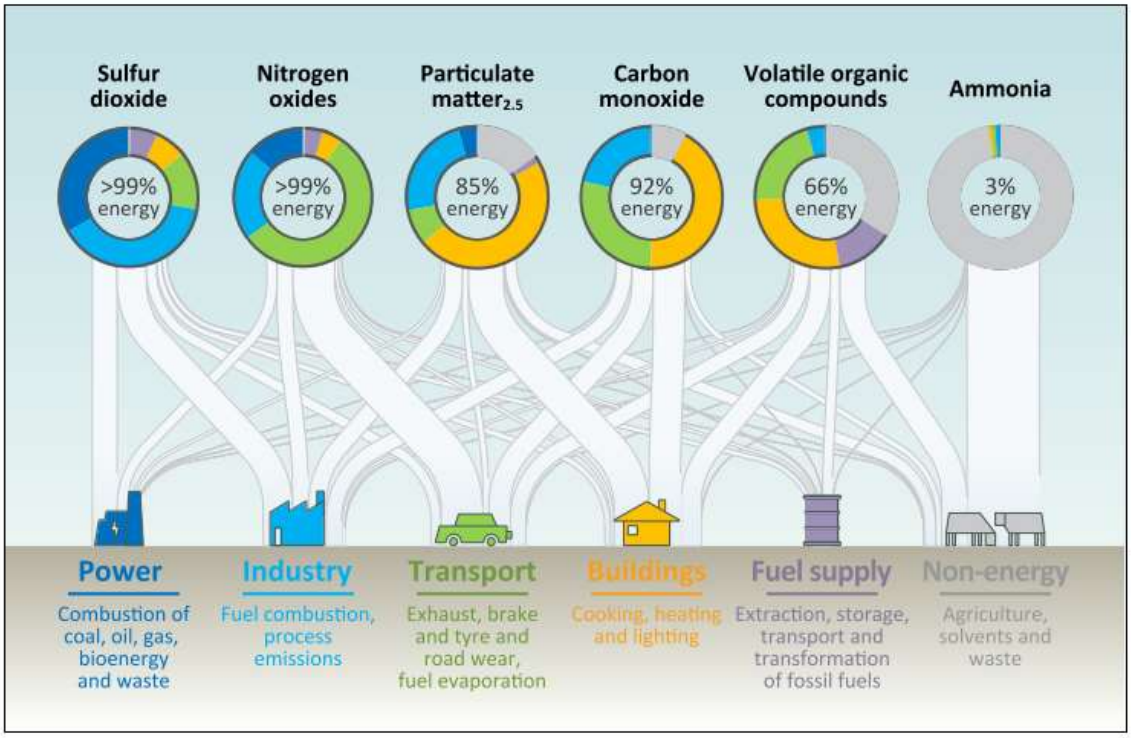

Figure 1. Common sources of air pollutants (from International Energy Agency, 2016).

An important consideration when analyzing air pollution concentrations is meteorology (Neiburger, 1969). Figure 2 illustrates how meteorological factors and topography influence the rate of dilution and dispersion of contaminants; in places where dispersion is more unfavourable, the emission rates should be low (Yang et al., 2015). Prevailing winds and atmospheric circulation should be analyzed before the siting of any new industry to ensure unnecessary exposure to air pollution emissions (Neiburger, 1969). In addition, there are several ways in which local meteorological events can produce severe air pollution events. For example, temperature inversions in winter could increase the probability of severe PM pollution by a factor of more than two (Hou & Wu, 2016). Every compound may have its unique (daily or annually) cycle and the prevailing winds may change throughout the year (Hoffman, Guernsey, et al., 2017; Neiburger, 1969; Stewart et al., 2019). According to Neiburger (1969), a detailed study of the local meteorology of the region can help to locate i) industrial developments in places where pollutant dispersion is favourable and ii) monitoring stations to ensure they give representative values of emissions and therefore regulation is fairly applied to all industries.

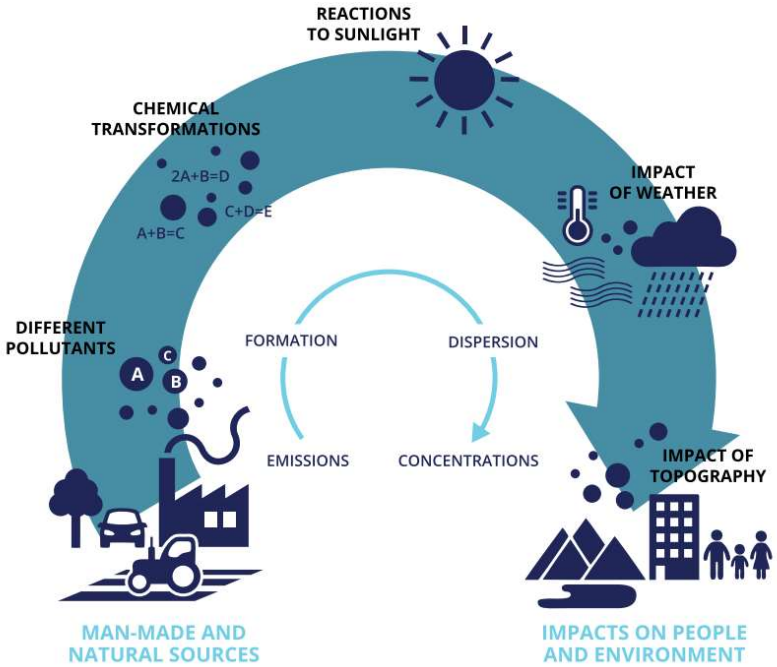

Figure 2. Lifecycle of air emissions (from European Environment Agency, 2016).

Air pollution is responsible for around 6.7 million deaths per year globally (Health Effects Institute 2020; World Health Organization 2019), has the potential to affect human health with cancer or lung diseases (Soskolne and Sieswerda 2010; Torén et al. 1996), and can lead to depression or psychological problems (Kelly and Fussell 2015). Air pollutants come from diverse sources, and the health impacts depend on the nature of the air pollutant. The following subsections briefly describe each of the air pollutants that are analyzed in this thesis and include PM, carbon monoxide (CO), nitrogen oxides (NO_x), sulphur dioxide (SO_2), and VOCs.

2.1.1 Particulate matter

The mixture of solid and liquid particles of different sizes, chemical compositions, shapes, and origins, when in the air, is known as PM (Pope & Dockery, 2006; World Health Organization, 2019). The effect of PM on human, animal and environmental health will depend on the chemical composition and the particle size (Nadadur et al., 2007). Total particulate matter (TPM) includes solid, liquid and suspended particles of all sizes. Size classifications include PM10 (particles with a diameter below 10µm) and PM2.5 (particles with a diameter below 2.5µm), as illustrated in Figure 3 (Environmental Protection Agency, 2022b; Pope & Dockery, 2006).

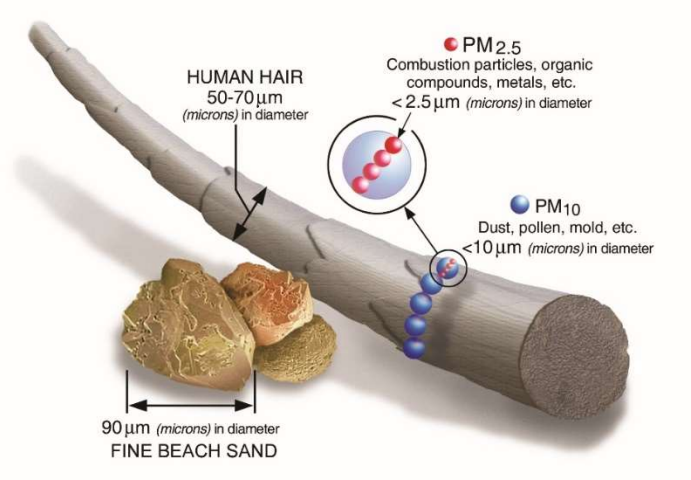

Figure 3. Illustration of particulate matter distribution size (from Environmental Protection Agency, 2022a)

The origin of PM includes several sources such as fossil fuel combustion, automobiles, unpaved roads, fields, and construction sites; or it could be the result of a complex chemical reaction from SO_2 and NO_x (Kelly and Fussell 2015; Nadadur et al. 2007; Stanek et al. 2011).

The smaller the particle, the higher potential of causing health problems, especially when the diameter is below 10 μm as it can penetrate the lungs, the brain and the heart, and get into the bloodstream (Environmental Protection Agency 2022a; Pope and Dockery 2006). Exposure to fine particles can result in heart attacks, aggravated asthma, and irregular heartbeat (Kelly and Fussell 2015). People with a pre-existent condition (like a heart or lung disease) could suffer premature death (Pope and Dockery 2006; Stanek et al. 2011). PM can reduce visibility, creating a haze, mainly in the presence of $PM_{2.5}$ (negatively affecting transport and tourism) (Stanek et al. 2011; Yang et al. 2015). It can also damage construction materials (including cultural monuments and statues), and deplete or modify the nutrients in soil and water (Environmental Protection Agency 2022a).

2.1.2 Carbon monoxide

CO, a poisonous gas, can be produced by incomplete combustion and through the oxidation of atmospheric hydrocarbons (Manisalidis et al. 2020; Zhang et al. 2019). Odourless, colourless, and tasteless, CO is produced when burning fossil fuels, propane, and wood (or wood pellets); CO could be released by unmaintained or malfunctioning equipment like furnaces, vehicle exhausts, charcoal grills, blocked chimney flues, and gas-powered cooking appliances and dryers (Government of Canada 2022). Atmospheric CO is a precursor of ground-level ozone (O_3) and one of the main sinks for hydroxyl radicals (Chen et al. 2017; Zhang et al. 2019).

Low CO exposure levels can cause flu-like symptoms like muscle weakness, headaches, and tiredness (Government of Canada 2022). The personal exposure limit for workers established by the Occupational Safety and Health Administration is 50 ppm (Occupational Safety and Health Administration 2012). After exposure to high levels of CO (which depend on the person's susceptibility and can vary between 150 to 200 ppm), serious poisoning may occur, as CO competes with the oxygen in the bloodstream and CO's

affinity to hemoglobin is greater than oxygen's affinity; this causes poor vision, dizziness, nausea and vomiting before the loss of consciousness (Consumer Product Safety Commission n.d.; Manisalidis et al. 2020).

2.1.3 Nitrogen oxides

Tailpipe emissions, power plant emissions and road traffic are the main anthropogenic sources of NO_x (Chaloulakou, Mavroidis, and Gavriil 2008; Nadadur et al. 2007) and natural sources of NO_x include lightning and microbial processes (Zhang et al. 2019). After fuel combustion, nitrogen liberated in the atmosphere can form NO_x ($NO_x = NO + NO_2$) from interactions with the oxygen in the air (Chaloulakou et al. 2008; Zhang et al. 2019). When nitrogen dioxides (NO_2) interact with water vapour, they can form acids, which after reacting with other gases can result in nitrates and ammonia (Environment and Climate Change Canada 2013a). NO_x can also be a precursor of $PM_{2.5}$, O_3 and VOCs (Stanek et al. 2011).

Apart from damaging vegetation (i.e., by lesioning the tissue), NO_x can cause adverse impacts on the respiratory system of humans and animals. It may contribute to the acidification of ecosystems (Environment and Climate Change Canada 2013a) and to eutrophication of lakes (Environment and Climate Change Canada 2020a). People with respiratory diseases (like asthma or chronic obstructive pulmonary disease) are very sensitive to NO_2 and long-term exposure can aggravate the susceptibility (Chaloulakou et al. 2008). Long-term exposure to NO_2 has also been linked to reduced lung function (World Health Organization 2021).

2.1.4 Sulphur dioxide

Among the family of sulphur oxide gases, SO_2 is the most common and harmful gas (Environment and Climate Change Canada 2013b). Power plants are the most significant source of SO_2 emissions, along with other industrial facilities (Nadadur et al. 2007). The combination of SO_2 from coal combustion with natural fog can cause a highly toxic smoke capable of worsening existing heart or respiratory disease or even death (Kelly and Fussell 2015; Stanek et al. 2011). After SO_2 is dissolved in water vapour, it can form acids that in combination with other particles can threaten ecological and human health as well as be

the precursor of acid rain and sulphur acid (Environment and Climate Change Canada 2020a). After a mixture and reaction with other aerosols such as NO_x and VOCs, SO_2 can be the precursor of $PM_{2.5}$, which has a great potential to damage human health for its capacity of penetrating respiratory and heart tissues in the human body (Buteau et al. 2018; Dabek-Zlotorzynska et al. 2011).

Continuous exposure to SO_2 can increase the risk of asthma among children (Buteau et al. 2018; Delfino et al. 2003) and even cause emergency room visits and hospitalizations for aggravation of respiratory disease or infections of the respiratory tract (CCME 2019; World Health Organization 2021). Indirect effects from acid rain deposited into soil and water can interfere with photosynthesis negatively affecting plants growth (being a cause for deforestation) and damaging outdoor sculptures, statues and buildings (Manisalidis et al. 2020; World Health Organization 2021).

2.1.5 Volatile organic compounds

VOCs are primarily released into the atmosphere from chemicals, solvents, fuels, natural sources or from atmospheric reactions between O_3 and other pollutants and consist of around 70% of the air pollutants classified as hazardous (International Energy Agency 2016; Suh et al. 2018). Examples of VOCs include benzene, formaldehyde, toluene, isoprene, halocarbons, and propene (Chen et al. 2017; Delfino et al. 2003; Xiong et al. 2020). Under normal atmospheric conditions, these compounds can easily turn into vapour and after sun exposure can react with other pollutants to produce O_3 and $PM_{2.5}$ (Chen et al. 2017). The principal anthropogenic source is fossil fuel combustion from the transport sector and to a lesser extent oil and gas extraction, paints and solvents, and firewood burning (CCME 2019; Delfino et al. 2003).

Different VOCs have differential impacts on health (Suh et al. 2018). The effects can vary from mild (like eye, nose or throat irritation; headaches, nausea and dizziness) to more severe (worsening asthma symptoms or damage to the liver, kidneys or the central nervous system) (CCME 2019). Some VOCs (like formaldehyde and benzene) are classified as likely to be carcinogenic over a long time exposure (Suh et al. 2018; Xiong et al. 2020) and could be associated with adverse respiratory diseases (Delfino et al. 2003).

2.2 The pulp and paper industry

2.2.1 The process of making pulp and paper

Pulp and paper are refined from raw materials (generally wood, recycled paper, and agricultural waste) that contain cellulose, and may exist separately (i.e., only pulp making or only paper making) or integrated (Bernard et al., 2020). Raw material preparation includes wood debarking, chip making, and deinking (for recycled paper). A simplification of the pulp-making process (Figure 4) illustrates the main differences between producing pulp from recycled paper and wood. While the recycled paper needs to be deinked (i.e., to remove printing ink from paper fibers of recycled paper), the chips need to be washed before being processed on the bleaching tower. The paper-making process can be divided into four main steps described following (pulp manufacturing, pulp bleaching, paper manufacturing and fibre recycling) (Cheremisinoff and Rosenfeld, 2010; Mandeep et al., 2019; Soskolne and Sieswerda, 2010).

a. Pulp manufacturing. The process could be mechanical (including disk abrasion and billeting), thermomechanical (heat or steam is applied to raw materials plus the mechanical process), chemimechanical (mechanic abrasion plus chemicals) and chemical (the two majors processes include sulphite and kraft or sulphate).

b. Pulp bleaching. Washing (with water), bleaching, rolling, and drying of the resulting pulp.

c. Paper manufacturing. Pulp is deposited in a moving forming device where water is removed to form paper and cardboard. The finishing step could include chemical additives depending on the final product (like pigments to add colour).

d. Fiber recycling.

The P&P industry is one of the most energy-intensive and water-consuming industries (Bernard et al., 2020). However, most of the negative environmental impacts from pulp mills are due to the pulping and bleaching processes where lignin (an unwanted constituent) is removed from the wanted fibers (Cheremisinoff & Rosenfeld, 2010; Soskolne &

Sieswerda, 2010). The remaining solution (known as black liquor) is generally burned to recover chemicals and energy (Bernard et al., 2020). Pulp bleaching used to be done with chlorine, a major source of chlorinated dioxins and furans (Hocking, 1991) which has now been substituted with chlorine dioxide and/or hydrogen peroxide (Soskolne & Sieswerda, 2010).

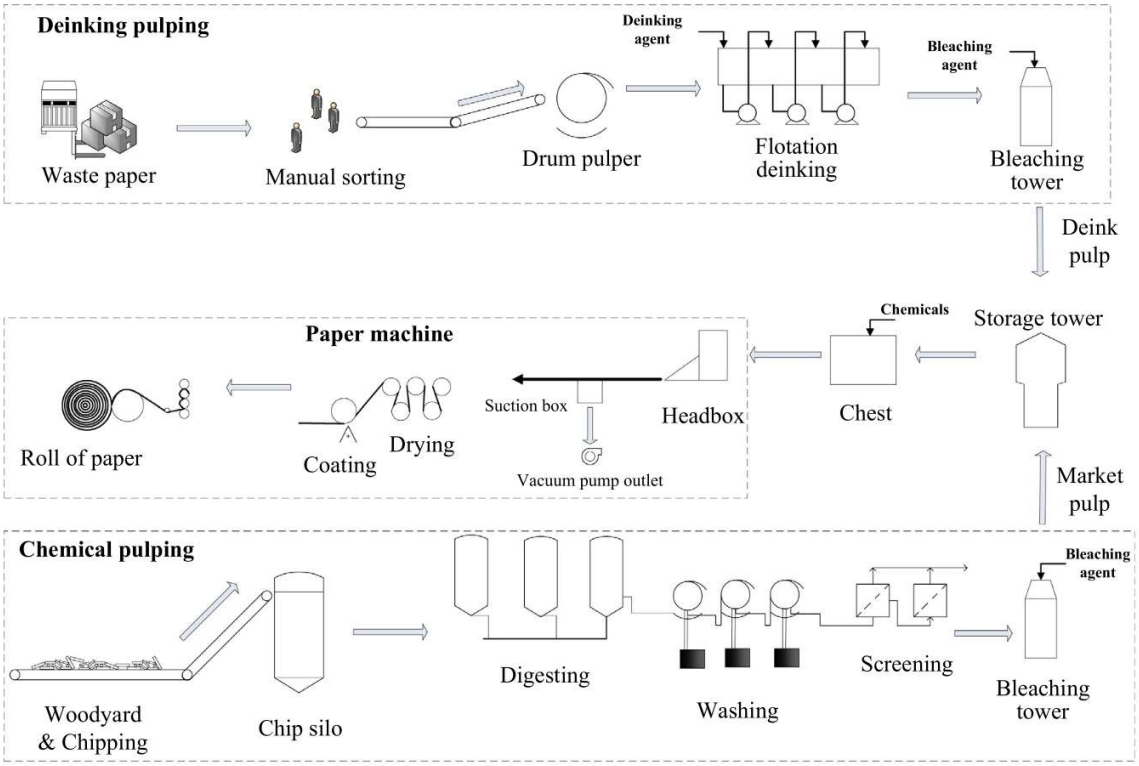

Figure 4. The process of pulp and paper making (from Tong et al., 2018).

2.2.2 Air pollution from pulp and paper mills

The environmental impacts of a P&P mill are not always proportional to the mill's production and waste generated; the environment's capacity to assimilate the releases will also shape habitat degradation (Colodey and Wells 1992). The local features where industry is located (including meteorological patterns, topography, population distribution and ocean currents) will influence how the pollution is dispersed and how local population health is affected (Neiburger 1969). For example, thermal inversions (a decrease in the planetary boundary layer), or transboundary transport of pollutants can favour the

occurrence of high pollution concentrations in specific locations (Mitchell, Wiacek, and Ashpole 2021; Vedal et al. 1998; Yang et al. 2015).

In P&P mills, air pollution is mainly caused by the intensive energy processes of the sulphide pulping process (in pulp mills). Energy consumption results in the production of various volatile sulphur compounds, SO_2, NO_x, CO, PM (of all sizes) and VOCs (Cheremisinoff and Rosenfeld 2010; Hoffman et al. 2015; Mandeep et al. 2019, 2020). Kraft pulp mills emit high amounts of malodourous emissions (total reduced sulphurs (TRS)) and sulphur oxides; these emissions are generally lower in mills with different pulping processes such as mechanical methods (Cheremisinoff and Rosenfeld 2010).

Air pollution impacts on human health from P&P facilities affect mill workers (Dionne and Walker 2021; Mandeep et al. 2019; Soskolne and Sieswerda 2010; Torén et al. 1996) and local populations exposed to the airborne pollutants (Hoffman et al. 2015; Lewis, Castleden, et al. 2020; Lewis, Francis, et al. 2020; Vedal et al. 1998). Although it is well known that carcinogenic compounds are generated in a P&P facility, how those compounds affect workers' health can be complex to determine as it depends on many factors (i.e., the socioeconomic status of the worker) (Coble, Lees, and Matanoski 2001; Soskolne and Sieswerda 2010; Torén et al. 1996). Torén et al. (1996) mention that lung cancer, malignant mesothelioma, malignant lymphomas, stomach cancer and leukemias should be considered occupational exposures for P&P workers. Children in a pulp mill community experienced peak expiratory flow reductions and increased sore throat and cough during high PM_{10} concentrations events (Vedal et al. 1998). Those effects were more visible in children with pre-existent asthma conditions.

2.2.3 The pulp and paper industry in Canada

In Canada, the P&P industry began in the early 1800s in Ontario, Quebec and NS, although the industry at that time was relatively small and localized as imports from the United States supplied most of the demand for paper (Kuhlberg 2006). A major expansion of the P&P industry in Canada occurred in the 20^{th} century and it lasted until the early 1990s (Bernard, Hussain, and Sinha 2020). The growth was driven by the diversification (i.e.,

book products, wallpaper, wrapping paper) and specialization of the industry, accompanied by an exponential increase in the newsprint demand (Kuhlberg 2006).

Since the beginning of the 21st century, the P&P industry in Canada has declined as pulp production exploded in industrializing countries in the last two decades, particularly in some countries in South America (Kuhlberg 2006). Between 2005 and 2013, the total number of operating mills in Canada declined by 47%; there are currently 102 pulp, paper, board, and/or tissue mills currently in operation or temporarily closed (Bernard et al. 2020; Pulp and Paper Canada 2021). Canada is still among the five leading exporting countries of wood pulp, paper and paperboard (FAO 2019). In 2019 Canada produced 16,815 MT of total wood pulp; half of this production, 8,813 MT, was from kraft-bleached pulp (Table 1). Exports of paper, paperboard, recovered paper, graphic papers and newsprint summed 15,367 MT (FAO 2019).

Table 1. Canadian pulp and paper statistics for 2019 (FAO 2019).

MT	Production	Imports	Exports	Consumption
Total wood pulp	16,815	398	9,676	7,537
Total chemical wood pulp	9,021	390	7,044	63
Chemical wood pulp, sulphate (kraft), unbleached	672	65	264	473
Chemical wood pulp, sulphate (kraft), bleached	8,141	313	6,510	1,944
Chemical wood pulp, sulphite	208	13	270	No Data
Mechanical and semi-mechanical wood-pulp	7,174	7	2,162	5,019
Paper and paperboard	9,473	2,434	6,698	5,209
Recovered paper	2,700	1,383	1,686	2,397
Graphic papers	5,274	594	4,569	1,299
Newsprint	2,570	27	2,414	183

2.3 The pulp mill in Pictou, Nova Scotia

2.3.1 Historical background

In Abercrombie Point in PC, a pulp Kraft mill operated from 1967 to 2020. Several companies owned the mill for more than fifty years of operation, and since 2011 it is owned by Paper Excellence who named the mill NP (Hoffman et al. 2015). The mill is located near Pictou Town and next to an Indigenous community, PLFN (Figure 5). The lagoon for the mill's effluents discharge, BH, used to be a sacred and cultural site for PLFN known by the community as A'se'k (translated as 'the other room' from the Mi'kmaw language) (Castleden et al. 2017). This former tidal estuary, a traditional place where PLFN members used to collect fruit, fish, hunt and perform recreational and religious activities, "became a place of disease and death" (Lewis, Francis, et al. 2020) after the mill became operative.

Figure 5. Location of Northern Pulp, Pictou Town, Pictou Landing First Nation, and Boat Harbour.

The following is a summary of the main events in the pulp mill since its establishment:

- 1964 to 1974
    - The building of the pulp mill started in 1964 and it became operative in 1967 (Hoffman et al. 2015).
    - A negotiation was signed with PLFN to discharge the mill's effluents on BH; the Indigenous community received a compensation of $60,000 and was assured that the estuary would remain clean (Paul 2006).
    - PLFN chief and council state reported concern about odours and pollution on BH; significant negative impacts were reported on the marine benthic community near the water discharge after two years of operation (Hoffman et al. 2015; Peer 1972)
- 1975 to 1984
    - PLFN was refused compensation from the provincial government for the damages that the effluent discharges created on the PLFN territory (Lewis, Francis, et al. 2020).
- 1985 to 1994
    - PLFN began action against the federal government and negotiated the Boat Harbour Settlement Agreement (1993).
    - The provincial government promised to abate the negative effects of the effluent discharge by 1995 (Hoffman et al. 2015).
    - Two studies reported contamination in Pictou Harbour (like dissolved metals) stating that it was not clear whether the pulp mill was the source and that the concentrations were not harmful to human health (Dalziel et al. 1993; Painter and Stewart. 1992).
    - The hospital in Pictou noticed a highly statistically significant incidence of respiratory diseases, and pharmacists in PC reported a decrease in respiratory aerosols and inhalers when the mill was not operating; further human health studies in PC were recommended (Reid 1989).

- 1995 to 2004
    - Kimberly-Clark Corporation acquired the mill in 1995 promising to close BH by 2005 and to pay the lagoon's environmental liability cost (Trombetta and Moritiwon 2020).
    - Neenah Paper became the new owner of the mill in 2004 (Group Pictou Landing Native Women's (c/o Sheila Francis Past President) et al. 2016).
- 2005 to 2014
    - After no alternative effluent treatment site is identified, the provincial government announced the closure of BH by the end of 2005 (Hoffman et al. 2015).
    - A group of Mi'kmaw women from the PLFN community initiated a community research campaign with a Two-Eyed Seeing approach to assess how the pulp mill in PC impacted the soil, air, water, and sediments, and to evaluate the community environmental and human health concerns (Group Pictou Landing Native Women's (c/o Sheila Francis Past President) et al. 2016).
    - Peaceful protests from residents after an effluent leak in 2014 led to the creation of the Boat Harbour Act; the mill would stop using BH as a discharge lagoon and BH would initiate its remediation by June 30, 2015 (Hoffman et al. 2015).
    - A failure in the stack precipitator and the release of fumes triggered a ministerial order stating the installation of a new recovery boiler precipitator by May 2015, which was completely installed by October 2015 (Hoffman, Guernsey, et al. 2017).
- 2015 to 2022
    - In 2015 The Boat Harbour Act was delayed for five more years (until 2020), and the mill continued operating. The pulp mill closed on January 31, 2020, as a result of its inability, after the five years extension, to present a proper environmental assessment plan and to relocate the effluent treatment plant (Quanz et al. 2021).

- After accessing health government data, many studies showed that human health impacts on PLFN (solicited several times by the community without a response from the federal nor the provincial government) required further investigation (Lewis, Castleden, et al. 2020; Lewis, Francis, et al. 2020).
- Community health and well-being were considered to have been negatively impacted since the mill's establishment (Castleden et al. 2017; Group Pictou Landing Native Women's (c/o Sheila Francis Past President) et al. 2016).
- The location of the mill was considered a deliberate decision and it was considered a case of environmental racism (Waldron 2018).
- VOCs and PM were considered the main air pollutants emitted from the pulp mill (Hoffman et al. 2015; Hoffman, Guernsey, et al. 2017).
- Dioxins and furans present in sediments were considered the main pollutants of concern in water sediments (Group Pictou Landing Native Women's (c/o Sheila Francis Past President) et al. 2016; Hoffman et al. 2019; Hoffman, Lyons, et al. 2017).
- After declaring insolvent in June 2020, NP sues the provincial government for $450 millions alleging that the closure resulted in losses of hundreds of million of dollars for the mill (Baxter 2022; Beswick 2021).

2.3.2 Environmental and social impacts attributed to the mill

Since the establishment of the mill, BH and its surroundings changed in many respects. The relationship between PLFN and the government regarding the pulp mill has been tense since the PLFN community felt that their concerns (about human health, environmental impacts, and cultural legacy) were not addressed (Castleden et al. 2017; Lewis, Castleden, et al. 2020; Lewis, Francis, et al. 2020). PLFN feel they suffered racism for more than 50 years (*Pictou Landing First Nation, 2018*) as the promise of keeping BH clean was 'an empty one' as soon as the mill became operational (Castleden et al. 2017). It has been considered a case of environmental racism (Baxter 2021; Burguesson and Ruffinengo 2020; Castleden et al. 2017; Paul 2006; Wood 2020) and it has also been mentioned that the location of the mill was deliberately chosen to be near an indigenous community (Castleden et al. 2017). The provincial government spent millions of dollars to allow the

mill to continue its operation and to pay the local community for the damage caused by the mill (Baxter 2017; University of King College Journalism Students 2009), but they also extended the BH relocation and restoration deadline several times over at least three decades (Baxter 2017; Hoffman et al. 2015; The Mill 2020; University of King College Journalism Students 2009).

The local population has long been worried about health issues and unpleasant odours (Henderson 2014; Hoffman et al. 2015; Lewis, Castleden, et al. 2020; Reid 1989). In the early stages of the mill's operation, a study from a local doctor showed that those effects were hazardous (Reid 1989). Reid compared the individual proportions of respiratory diseases in three years periods (from 1986 to 1989) in ten hospitals in Nova Scotia (all with 51-100 beds) and found out that Pictou was among the four sites where respiratory cases were statistically significant (the other three hospitals were located in active coal mining towns). Other observations made by Reid (1989) include the higher cancer incidence and higher respiratory illnesses in comparison with other counties in the province, a drop in the sales of respiratory inhalators when the mill was down of operation (for maintenance, prolonged strikes), eye irritation, nausea and other effects caused by the odour and air emission from the mill.

A six-year project bringing Indigenous and Western knowledge systems together in 2016 resulted in the report "Our Ancestors Are in Our Land, Water, and Air": A Two-Eyed Seeing Approach to Researching Environmental Health Concerns with Pictou Landing First Nation (Group Pictou Landing Native Women's (c/o Sheila Francis Past President) et al. 2016). In this process, the group combined oral histories, a community health survey, and environmental monitoring techniques to analyze the impact of the mill on PC and its surroundings. The water samples collected within BH and directly leaving BH were found to exceed the Canadian Water Quality Guidelines for the Protection of Aquatic Life. Total suspended solids, some metals (like aluminium, lead, zinc) and nitrogen were above guidelines for aquatic life, although pH levels were within the accepted range. Biological oxygen demand, phosphorous and conductivity levels indicate that BH is likely impacted by industrial discharge. The study showed that some metals in the soil exceeded federal guidelines, such as arsenic and copper, though they conclude that the soil is generally safe.

The report highlights the alarming low biodiversity observed in the ecotoxicology and sediments samples and the high density of species known for being tolerant to pollution. They recommended avoiding contact with sediments from BH as dioxins and furans were the primary organic pollutants of concern within the sediments; and warned about the consumption of beaver and muskrat tissues as it could result in exceeding the recommended consumption limits of dioxins and furans. None of the air pollutants sampled were above the existing safe levels, including the United States National Ambient Air Quality Standards, the Canadian Ambient Air Quality Standards (CAAQS), and occupational standards.

In the last decade, many studies have linked air, water and soil pollution findings with the pulp mill (Davidson et al. 2021; Hoffman et al. 2015, 2019; Hoffman, Guernsey, et al. 2017; Hoffman, Lyons, et al. 2017; Quanz et al. 2021; Romo, Chaudhary, and Walker 2019). However, only a few of them focus on air emissions. Hoffman et al. (2017) documented elevated concentrations of certain volatile organic compounds (VOCs) between 2006 and 2013 exceeded cancer risk thresholds (of Health Canada in μg/m3) in the former Granton NAPS air quality station. The authors associated those emissions with NP (although there was no other conclusive study). Hoffman et al. (2015) found discordance between levels of pollution reported by NP, identifying PM and sulphur odours emissions two to three orders of magnitude higher in comparison to other Canadian kraft pulp mills and they supported the concern of the population as it was found that the industry was underreporting emissions. To comply with provincial regulations after the failure of the recovery boiler precipitator, in 2015 the mill installed new equipment to reduce PM emissions. It was expected that ambient air quality would improve after the installation of the precipitator, which was intended to remove PM from combustion discharge gases by attracting particulates to collection plates (Muralikrishna and Manickam 2017). Although (Hoffman, Guernsey, et al. 2017)recommend a follow-up research assessment to determine if air quality had improved, there has been no academic research to test whether the precipitator effectively removed pollutants as intended.

2.3.3 The pulp mill and the society, government, and industry

Scientific evidence is one of several components in the decision-making process. Social, political and economic considerations also play a role in elaborating a policy (Westwood et al. 2019). In the case of the mill in PC, the economic aspects seemed to be given more weight (Baxter 2017). Some aspects cannot be quantified, like the livelihood of a family that depends on a single source of income to eat and live, or the youth struggling to find a job. Some employers of the mill who have been working all their lives in the same facility, have little work experience in other areas or have a business that depends entirely on the mill activities such as forestry, and the closure of NP was detrimental to their survival (MacInnis 2018). The mill was an important source of employment (University of King College Journalism Students 2009) and NP has been considered an economic driver for the entire province as one of the largest customers of the Port of Halifax (Spicer 2019). Before the closure, the mill operated 330 full-time local jobs and 2050 indirect jobs from Nova Scotians (Paper Excellence 2021). Employers dependent on the mill defended their source of livelihood and did not agree on NP's closure (MacInnis 2018; Spicer 2019). Many of them do not work at the facility and are woodlot owners or operators.

The pulp mill has gone through minor changes during its more than 50 years of operation as further technology upgrading and process updates were not considered necessary (Baarda 2020). In recent years, when people claimed that PM emissions from the mill were high, the provincial minister of Health and Wellness said it was not an immediate threat (CBC News 2014b, 2014c). Some special events were considered anecdotal and some effects generated by the mill emissions were considered aesthetical as it was not found enough evidence of a higher incidence of hazardous pollution and health impact (Dalziel et al. 1993; Peer 1972; Scarratt 1969).

However, some events forced provincial authorities to act. In 2014, after an effluent spill, residents led a peaceful protest to claim a relocation of the lagoon discharge. An agreement was signed between PLFN and the minister of environment where BH was supposed to stop being used as a remediation site in January 2020 (Baxter 2021; Henderson 2014; Hoffman et al. 2015; Wood 2020). The authorities of NP had more than five years to plan a new remediation site, but they failed to do so, and the mill has been closed for more than

two years now (although it may reopen). Also in 2014, it was revealed that PM emissions were high above the safety standards and people were mobilized through the group Clean Pictou Air (Baxter, 2017; CBC News, 2014; Henderson, 2014; Hoffman et al., 2017; MacKinnon, 2018). In 2015, new industrial approvals were issued by the local government (Nova Scotia Environment 2015) and a new recovery boiler precipitator was installed the same year.

2.3.4 Pulp-making process in Northern Pulp and air releases

The chemical kraft process proposed to occur in NP (when the mill reopens) is schematized in Figure 6, where the best available technology standards for the P&P industry are presented by NP to Nova Scotia Environment and Climate Change. Following is the description of the three processes, steam generation, sodium liquor cycle and calcium liquor cycle, focusing on the discharges to the atmosphere (adapted from the Northern Pulp Nova Scotia Mill Transformation Project as a Class II (Northern Pulp Nova Scotia 2021).

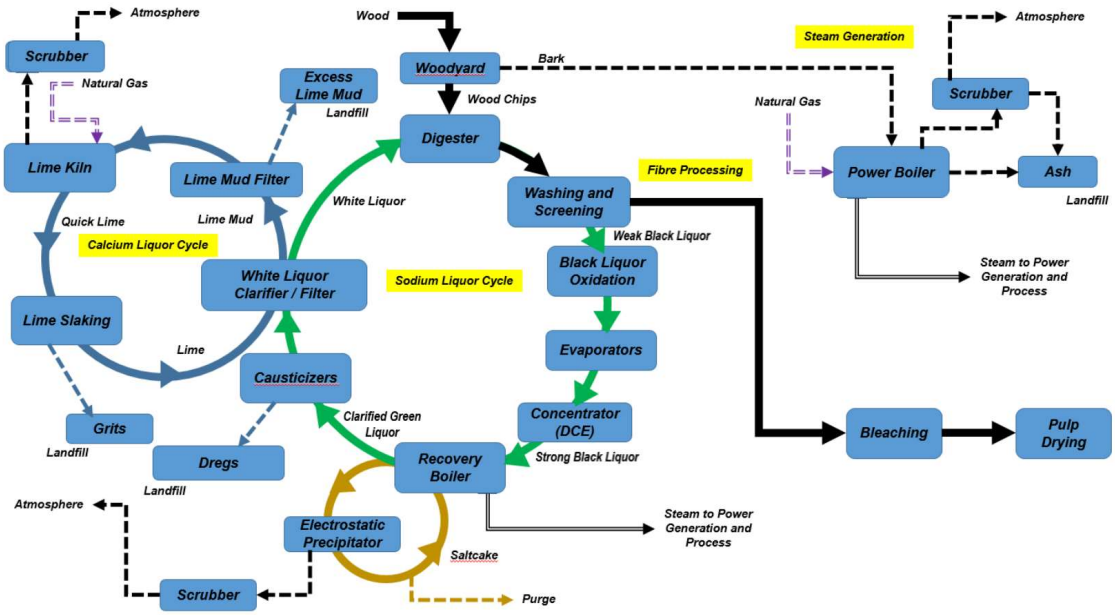

Figure 6. Northern Pulp process configuration (from Northern Pulp Nova Scotia, 2021).

1) Steam generation: Wood debris and bark are burned to generate high-pressure steam (complemented with natural gas). The resulting air is controlled by a wet

scrubber before being released into the atmosphere. The ash from the scrubber is dewatered before its ultimate disposal as a landfill.

2) Sodium liquor cycle: During the fibre processing, the washed and screened wood chips form the black liquor. The thickened black liquor (because of several evaporator stages) is burned in the recovery boiler generating steam. The resulting air is controlled by an electrostatic precipitator and a wet Modo scrubber prior to being released into the atmosphere.
3) Calcium liquor cycle: The recycling and reusing of the calcium carbonate (or lime mud) include cleaning, washing, and thickening. Then, it is sent to a lime kiln where combustion generates gases that pass through a scrubber before being released into the atmosphere. This process removes PM and residual sulphur compounds released from the lime mud.

2.4 Air quality regulations

In Canada, the Air Quality Management System (AQMS) implements a system for protecting and improving air quality collectively (CCME 2019). One of the mechanisms included in the AQMS is the CAAQS, which consists of standards of air concentration of pollutants to improve air quality. All provinces, except for Quebec which has its own Clean Air Regulation, have roles and responsibilities in the implementation of the CAAQS regulation (CCME 2021). Also, individual provinces have their own regulations, which seem to vary widely across Canada (Hoffman et al., 2015; Hoffman et al., 2017b). For example, Xiong et al. (2020) used the British Columbia's provincial acceptable risk level to test the carcinogenic risk of hazardous VOCs compounds. In New Brunswick, the Department of Environment and Local Government has its own Clean Air Act (Dionne and Walker 2021). NS air quality is regulated by the Regulatory Framework for Air Emissions (Nova Scotia 2005) and is currently undergoing a process of updating the ambient air quality standards by 2025 (Air Quality Unit Department of Environment and Climate Change 2022).

The four colours of the CAAQS management levels determine air quality management action depending on the $PM_{2.5}$, O_3, SO_2, and NO_2 concentrations measured in $\mu g/m^3$ for

$PM_{2.5}$ and in ppb for the other three pollutants (Table 2). While a green level indicates that air quality should proactively be kept clean, the yellow, orange and red levels indicate preventive and management action to prevent the further deterioration of air quality (CCME 2021). The red and orange levels were updated in 2020 from their initial levels in 2015 for $PM_{2.5}$ and O_3 management levels; and will be updated by 2025 for the SO_2 and NO_2 from the 2020 levels (CCME 2019).

Table 2. Canadian Ambient Air Quality Standards (CCME 2021).

CAAQS (2020)	$PM_{2.5}$ ($\mu g/m^3$)		O_3 (ppb)	SO_2 (ppb)		NO_2 (ppb)	
	24-hour	Annual	8-hour	1-hour	Annual	1-hour	Annual
Red	>8.8	>27	>62	>70	>5.0	>60	>17.0
Orange	6.5 - 8.8	20 - 27	57 - 62	51 - 70	3.1 - 5.0	32 - 60	7.1 - 17.0
Yellow	4.1 - 6.4	11 - 19	51 - 56	31 - 50	2.1 - 3.0	21 - 31	2.1 -7.0
Green	≤4.0	≤10	≤50	≤30	≤2.0	≤20	≤2.0

The Pollutant Release and Transfer Registers are programs aimed to provide control on air releases from industrial sources (Taylor et al., 2020). Worldwide, several programs exist for industries report air emissions into the atmosphere. In the US, the Toxics Release Inventory was introduced in 1986 and consist on air mass releases reports, providing useful information for the government about decision making on air pollution from industrial sources (Bui and Mayer 2003). In Canada, the National Pollutant Release Inventory (NPRI), consist of a similar program, also reporting emissions in mass, and it initiated in 1993 (Government of Canada 1999). Other programs include the Australia's National Pollutant Inventory initiated in 1998 and the European Union's European-Pollutant Release and Transfer Register initiated in 2007 (Sullivan and Gouldson 2007).

2.4.1 Summary

This chapter presents an overview of the air pollutants analyzed in the next two chapters, including a description of the sources and potential health and environmental impacts of PM, SO_2, NO_x, CO, and VOCs; it also briefly summarizes the process of pulp and paper making identifying the main stages where air pollutants are emitted into the atmosphere. As this research compares air emissions from the pulp mill in Pictou with other industrial facilities, a summary of the history of the pulp mill is presented based on diverse sources (including reports, academic research, and news articles). The next two chapters present the results of this thesis (first, a comparison between air emissions from P&P mills; then a comparison of $PM_{2.5}$ emissions by three industrial facilities in Pictou) and describe in more detail the industrial facilities considered and the methodology used in each case.

CHAPTER 3 CHARACTERIZATION OF ANNUAL AIR EMISSIONS REPORTED BY PULP AND PAPER MILLS IN ATLANTIC CANADA

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Abstract: The pulp and paper industry is a major contributor to water and air pollution globally. Pulp and paper processing is an intensive energy consuming process that produces multiple contaminants that pollute water, air, and affect ecological and human health. In Canada, the National Pollutant Release Inventory (NPRI) is used to assess the release of air pollutants into the atmosphere from industrial facilities (including pulp and paper mills) and provides a repository of annual emissions reported by individual facilities. This study compared annual air emissions of carbon monoxide, nitrogen oxides, total particulate matter (TPM), $PM_{2.5}$, PM_{10}, sulphur dioxide, and volatile organic compounds from nine different pulp and/or paper mills in Atlantic Canada from three provinces (Nova Scotia, New Brunswick, and Newfoundland and Labrador) between 2002 and 2019. Results revealed that annual releases were several orders of magnitude higher than federal reporting thresholds suggested by Environment and Climate Change Canada. Pulp mills emit higher pollutant loads than those producing paper. The highest exceedance of a reporting threshold was for particulate matter ($PM_{2.5}$) at Northern Pulp in Nova Scotia. The emissions of $PM_{2.5}$ were on average (over a 17-year period) about 100,000% above the reporting threshold of 0.3 tonnes per year.

Keywords: air pollution; pulp and paper; National Pollutant Release Inventory (NPRI); Atlantic Canadian provinces

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3.1 Introduction

Combustion of fossil fuels and biofuels during energy intensive production processes emits pollutants into the atmosphere (Ghosal et al. 2019), which threatens human and ecological health (Buteau et al. 2018; Kelly and Fussell 2015; Stewart et al. 2019; Strum and Scheffe 2016). However, atmospheric emissions from industrial facilities were once considered a symbol of economic growth, and their effects were not considered harmful until late in the last century (Stanek et al. 2011). In recent decades, stringent standards were developed around the world to protect human and environmental health (Coble et al. 2001; Kamali and Khodaparast 2015). In 1970, the *Clean Air Act* was established in the United States due to growing concerns over air pollution impacts. Even though current legislation in the United States has saved millions of lives, many premature deaths still occur because of breathing polluted air, principally in marginalized sectors such as poor Black or Latino communities (Gardiner 2020). In Canada, the *Canadian Environmental Protection Act*, 1999 provided legislative authority to protect human and environmental health but was highly criticized for its lack of scientific basis and is currently being amended (Greig and Duinker 2011; Taylor 2021). In 2006, the Canadian government published a Notice of Intent to regulate air emissions, and one year later, the Clean Air Regulatory Agenda was published to establish a framework on emissions reduction targets and enforceable regulations (Minister of Environment 2007).

3.1.1 Air Emission Regulation in Canada

In Canada, all companies and organizations that emit certain substances into air, water or land and meet specific threshold requirements must report annual emissions to the federal National Pollutant Release Inventory (NPRI) (Berthiaume 2021; Johnston Edwards and Walker 2020). The NPRI is a registry of annual estimations of emissions and disposal to the environment and was created with the purpose of making pollution data accessible to the Canadian public (Berthiaume 2021; Environment and Climate Change Canada 2020b). Due to the threat that some of the releases may pose to human and environmental health, this information was then published on the Environment and Climate Change Canada (ECCC) website (Environment and Climate Change Canada 2021a). The creation of the NPRI program resulted in a decrease in overall emissions (Johnston Edwards and Walker

2020; Taylor et al. 2020), although the toxicity of the releases did not decrease (Johnston Edwards and Walker 2020). The decrease in overall emissions may have been a direct result of increased public awareness and public pressure, which is the intent of the NPRI program. However, there may have been other reasons, such as improvements in equipment and technology or improvements in industry practices and activities. ECCC requires that releases above reporting thresholds, shown in Table 3 for the period 2020 to 2021, must be reported to the NPRI program (Environment and Climate Change Canada 2021c). These thresholds are the lower limit trigger for reporting annual emissions; however, there are no regulations for exceedances.

Desirable concentrations of ambient air pollutants, including fine particles ($PM_{2.5}$), O_3, sulphur dioxide (SO_2), and nitrogen dioxide (NO_2), are specified in the Canadian Ambient Air Quality Standards (CAAQS), a national guideline that considers an averaging period to report the concentration of the pollutant in µg/m3 or ppb (Government of Canada 2021b), not compatible with the units reported by NPRI. While there are few studies comparing thresholds in units of tonnes per year, a recent study relates 10-year NPRI industrial air emissions to childhood-onset asthma in Quebec characterizing 'major emitters' those industries, for which its $PM_{2.5}$ or SO_2 annual emissions exceeded 100 tonnes (Buteau et al. 2018). Emissions reported through the NPRI program and science-based estimation tools are combined to generate the Air Pollutant Emission Inventory (APEI), which reports air emissions for 17 pollutants since 1990 (Government of Canada 2021b, 2021a). ECCC recommended thresholds for annual releases (in tonnes) only for total particulate matter (TPM) and SO_2 in the Code of Practice for the Management of Air Emissions from Pulp and Paper Facilities (CPMAEPPF) (Environment and Climate Change Canada 2018). These limits are divided into two categories for both pollutants depending on the processes occurring in the facilities (chemical or mechanical) and should be calculated by considering the annual production of each mill. For chemical facilities, emission limits are 2 kg and 4 kg/tonne of production for TPM and SO_2, respectively; for mechanical facilities, those emission limits are 0.5 kg and 1.5 kg/tonne of production for TPM and SO_2, respectively.

Table 3. NPRI reporting thresholds for 2020 to 2021 (retrieved from NPRI (2021)).

Pollutant	CO	NO_x	SO_2	TPM	$PM_{2.5}$	PM_{10}	VOC
NPRI threshold (tonnes/year)	20	20	20	20	0.3	0.5	10

3.1.2 Pulp and Paper Mills and Air Pollution

Currently, mixtures of air pollutants are considered a management challenge due to the interaction between the chemical and physical components of substances that can increase potential toxicity of emissions (Nadadur et al. 2007; Stanek et al. 2011). While air pollutants exist naturally in the atmosphere from wildfires, volcanic eruptions and biological decomposition, these acid and particulate emissions are relatively small quantities on average and rarely pose threats to human or ecological health (Muralikrishna and Manickam 2017; Strum and Scheffe 2016). Anthropogenic emissions, mainly from industrial activities and fossil fuel combustion, produce the majority of hazardous air pollutants (Kelly and Fussell 2015; Muralikrishna and Manickam 2017; Nadadur et al. 2007; Stanek et al. 2011; Walker et al. 2003, 2006). Pulp and paper (P&P) industries emit large amounts of atmospheric pollutants and greenhouse gases (Cheremisinoff and Rosenfeld 2010; Ghosal et al. 2019), although there are few studies that quantify the spectrum of emissions (Hoffman, Guernsey, et al. 2017). Numerous studies have been conducted on water pollution impacts from wastewater effluent due to P&P mills (Adesida 2020; Couillard et al. 1999; Hocking 1991; Hoffman, Lyons, et al. 2017; Kamali and Khodaparast 2015; McMaster, Hewitt, and Parrott 2006; Owens 1991), but there have been comparatively few studies to evaluate the contribution of the industry to the concentrations of hazardous air emissions (Tong et al. 2018). Air pollution from P&P mills is mainly caused by the production of various volatile sulfur compounds (from sulfite pulping process), odour emissions (from TRS), SO_2, and nitrogen oxides (NO_x) (Bhander and Jozewicz 2017; Mandeep et al. 2019). Stack emissions can also contain carbon monoxide (CO) (Nadadur et al. 2007). Biomass combustion can increase the emissions of particulate matter (PM) and of gaseous and semi volatile organic compounds (VOCs) (Hoffman, Guernsey, et al. 2017; Nadadur et al. 2007). Exposure to these pollutants can affect human

health, causing cancer or lung diseases (Gardiner 2020; Soskolne and Sieswerda 2010; Torén et al. 1996),, and may result in depression or psychological problems (Kelly and Fussell 2015). Exposure to particulates is associated with an increased risk of mortality from stroke, heart, or pulmonary disease, among others (Krewski and Rainham 2007; Stanek et al. 2011; World Health Organization 2019). One method of reducing human health and environmental impacts of air pollutants is via continuous atmospheric monitoring of pollutant concentrations. Modeling exposures to these concentrations supports epidemiological studies to evaluate health risks and the development of management strategies and relevant health interventions (Johnston Edwards and Walker 2020; Taylor et al. 2020).

In Canada, there are currently 102 pulp, paper, board, and/or tissue mills in operation or which have been temporarily closed (Pulp and Paper Canada 2021). However, few studies have quantified their compliance with emission standards based on our literature review. One of the most controversial mills in Atlantic Canada is the Northern Pulp mill in Pictou, Nova Scotia (NS), where locals have long been concerned about health issues and unpleasant odours (Henderson 2014). While managers of the facility report the best practices with respect to regulatory compliance (Baarda 2020), environmental monitoring revealed emission exceedances and locals remain concerned about impacts to human health (Group Pictou Landing Native Women's (c/o Sheila Francis Past President) et al. 2016; Hoffman et al. 2015, 2019; Hoffman, Guernsey, et al. 2017; McDonald 2015; The Canadian Press 2014). Currently, Nova Scotia does not legislate requirements for emitters to assess human health risks from emissions (Group Pictou Landing Native Women's (c/o Sheila Francis Past President) et al. 2016). Hoffman et al. (2017) also revealed that sediment and water samples, specifically dioxins and furans and metals, from Boat Harbour (a tidal estuary designed to receive wastewater effluent from the mill) were above both the Canadian Sediment Quality Guidelines and Canadian Water Quality Guidelines (Group Pictou Landing Native Women's (c/o Sheila Francis Past President) et al. 2016). However, ambient air pollution concentrations in nearby communities were below the Canadian Council of Minister of the Environment guideline limits. Hoffman et al. (2015) found discordance between levels of pollution reported by the mill on NPRI, identifying Total

Reduced Sulfurs (TRS) to be three times higher in comparison to other Canadian Kraft mills, revealing that the main pollutants from the NP mill were sulfur odours and PM.

The current limitations of air pollution monitoring (e.g., the NPRI is a self-reported estimated inventory) and the absence of upper air emissions thresholds entangles the identification of harmful releases into the environment. This study, focused on the air emissions from the P&P industry in Atlantic Canada, compares annual releases among different facilities, identifies the main limitations of the existent reporting tools, and recommends how to improve environmental and human protection regarding air quality. The analysis includes air emissions by nine pulp and/or paper mills for 17 years using publicly available data from NPRI (Environment and Climate Change Canada 2021b) and for 30 years using data from the APEI inventory. The seven pollutants studied were CO, NO_x, TPM, $PM_{2.5}$, PM_{10}, SO_2, and VOCs, as these air pollutants are widely reported by industrial facilities as mandated by ECCC for the NPRI inventory and are known to increase risk to human health (Berthiaume 2021; Environment and Climate Change Canada 2021a).

3.2 Materials and Methods

3.2.1 Data Collection

There are nine pulp and/or paper mills in Atlantic Canada and all of them report to NPRI, although only eight of them are currently in operation. Six are in New Brunswick (NB), two in Nova Scotia (NS), and one in Newfoundland and Labrador (NL); five out the nine facilities are pulp mills. Figure 7 shows the location of each facility in Atlantic Canada and the Appendix A presents background information about each one. Table 4 provides details of each facility as pulp mill (PU), paper mill (PA), or pulp and paper mill (PP). The NPRI ID identifies each facility that reports, and the North America Industry Classification System (NAICS) code classifies the type of processes and production associated with each facility. The two first digits of the NAICS indicate the sector; the third digit indicates the subsector; and the fourth and fifth digit indicate the industry group and the industry, respectively (Statistics Canada, 2017b). The sixth digit designates national industries (it is zero when there are no additional details) (Statistics Canada, 2017b). Pulp, paper, and

paperboard mills are assigned the code 3221 (Statistics Canada, 2017a). A further classification including lower levels for those industries is presented in Table 4 where the following is the case: (i) 322112, indicates chemical pulp mills, (ii) 322121, indicates papermills producing paper except from newsprint, (iii) 322122, indicates papermills producing newsprint, and (iv) 322130, indicates paperboard mills.

Table 4. Location of Atlantic Canadian pulp and/or paper mills and their characteristics. PA: Paper; PU: Pulp; PP: Pulp and Paper

Mill	Location	ID	Type	Product	Production (tpy) [4]	Employees	NPRI ID	NAICS 6 Code
Northern Pulp -62.72°, 45.65°)	New Glasgow, NS	NP	PU	Kraft pulp	280,000	330 [2]	815	322112
ing P & P Limited -66.09°, 45.26°)	Saint John, NB	IP	PU	Bleached kraft pulp	335,000	335 [2]	2604	322112
win Rivers Paper -68.33°, 47.37°)	Edmundston, NB	ED	PU	Bleached pulp	370,000	280 [2]	1221	322112
Atholville, AV Group -66.72°, 47.99°)	Atholville, NB	AT	PU	Pulp	ND [1]	275 [3]	5008	322112
Nackawic, AV Group -67.23°, 46.01°)	Nackawic, NB	NA	PU	Pulp	ND [1]	365 [3]	2181	322112
ort Hawkesbury Paper -61.36°, 45.60°)	Hawkesbury, NS	HA	PA	Paper	400,000	350 [3]	2221	322122
ake Utopia Paper -66.77°, 45.16°)	Utopia, NB	UT	PA	Corrugate medium	185,000	140 [2]	1572	322130
ing Paper Limited -66.02°, 45.28°)	Saint John, NB	IA	PA	Paper	420,000	310 [2]	3394	322121
Corner Brook P & P (−57.95°, 48.95°)	Corner Brook, NL	CB	PP	Pulp and paper	255,500	304 [2]	4929	322122

[1] No Data. [2] Number of employees retrieved from individual mill websites. [3] Number of employees retrieved from ECCC website (National Release Inventory. Available online: https://www.canada.ca/en/environment-climate-change/services/national-pollutant-release-inventory/tools-resources-data/all-year-dashboard.html (accessed on 29 March 29, 2022).). [4] Total annual capacity. Mills do not necessarily operate at maximum capacity every year.

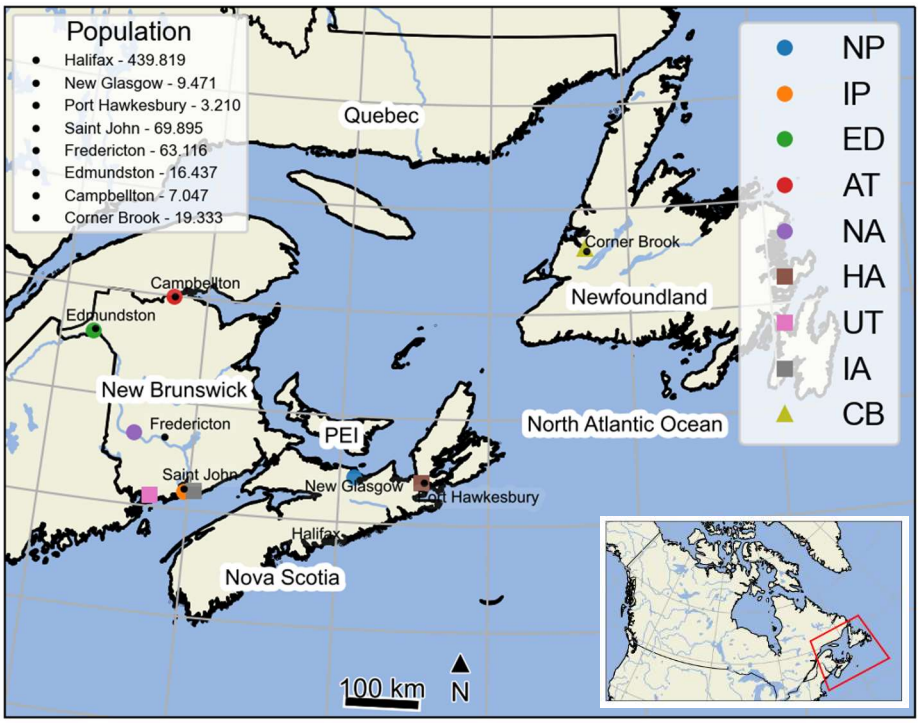

Figure 7. Location of mills and major populated centres. Pulp mills are represented with a circle, paper mills with a square, and the pulp and paper mill with a triangle.

3.3 Data Analysis

Pollutant release data were collected from the APEI inventory for the provinces of NS, NB, and NL between 1990 and 2019 and from the NPRI inventory for the nine mills from 2002 to 2019. The air pollutants analyzed were pollutants reported by the facilities: CO, NO_x, TPM, $PM_{2.5}$, PM_{10}, SO_2, and VOCs. Annual emissions reported in the NPRI were compared for each facility for each pollutant. Then, differences from the reporting threshold (DRT) for each pollutant in every mill were calculated using the following equation (Equation (1)):

$$DRT = ((NPRI_{reported} - Threshold) \times 100)/Threshold \quad (1)$$

where $NPRI_{reported}$ is each emission reported by the facility per annum and per pollutant. The threshold indicates the lower release that industries need to report to NPRI (it establishes the lower limit to report to NPRI) and the corresponding value for each

pollutant is in Table 1. For the SO_2 and $PM_{2.5}$ thresholds suggested in CPMAEPPF, the specific annual recommended limits, ARL, were calculated with the following equation (Equation (2)):

$$ARL = emi_{req} \times prd_{ann} \quad (2)$$

where emi_{req} is the base level emission requirements described in the previous section, and prd_{ann} is the annual capacity of the mill presented in Table 4. The difference in SO2 and PM2.5 CPMAEPPF ARL was calculated for each mill (Table 5). We compared emission trends in time for each site, as well as compliance with the reporting thresholds. A linear regression model was implemented to assess if there is significant change in the annual trends. The model was calculated with the emission as the dependent variable with the year as a predictor to obtain the p-value at a 95% confidence level. Emissions are considered statistically significant when the p-value is <0.05 for the individual site's and <0.001 (rounding the confidence level to the multiple models, 0.05/63) when all sites and contaminants are considered.

Table 5. Annual recommended limits (ARL) for TPM and SO2 releases according to the CPMAEPPF (retrieved from [54]).

Tonnes Per Year	NP	IP	ED	AT	NA	HA	UT	IA	CB
TPM	560	670	740	ND [1]	ND [1]	200	93	210	128
SO_2	1120	1340	1480	ND [1]	ND [1]	600	278	630	383

[1] ND indicates no data. Annual capacity production in the mill was unavailable to calculate the annual threshold emission.

3.4 Results

3.4.1 Air Emissions Reported by Province

Annual emissions of NO_x, TPM, PM_{10}, $PM_{2.5}$, SO_2, and VOCs registered in the APEI for each province show that the releases have decreased since 1990 (

Figure 8). NB, the province with more than half of the pulp and paper mills in Atlantic Canada, leads in air emissions for almost all pollutants and the entire 30-year period. However, mills from NS reported more TPM, PM_{10}, and $PM_{2.5}$ emissions than NB between 2008 and 2015 (panels c, d, e). NL, with only one mill, maintained the lowest emissions among the three provinces for the entire period for all pollutants, except for TPM (which was higher than NS between 2002 and 2006) and for SO_2 (which was of similar magnitude and sometimes above NS emissions since 2002).

3.4.1 Air Emissions Reported by Pollutant by Each Facility

Annual releases for all nine mills from 2002 to 2019 are shown in Figure 9 for the seven pollutants. Solid lines represent the pulp mills: NP in blue, Irving P & P (IP) in orange, Twin Rivers (ED) in green, Atholville (AT) in red, and Nackawic (NA) in purple; dotted lines represent the paper mills: Port Hawkesbury (HA) in brown, Lake Utopia (UT) in pink, and Irving Paper (IA) in grey; and dot-dashed lines represent the P&P mill, Corner Brook (CB) in yellow. The facility mean values are represented by the black dotted line. Figure 9a shows that, until 2007, NP was the major emitter of CO, which was above 3000 tonnes/year while all remaining mills were <2000 tonnes/year. Between 2008 and 2015, IP and ED were the major emitters with releases above 1700 tonnes/year. From 2016, the major emitters were NP and AT with emissions of more than 3000 tonnes/year and were significant at $p = 0.005$ (Table 6). UT and HA, both paper mills, emitted little across the period in comparison with the rest of the mills, although both were close to the threshold.

CB, the P&P mill, also emitted little CO during the period in comparison to the others, but its emissions were above thresholds for the entire period and significant at $p = 0.005$. According to Figure 9b, the major emitter of NO_x in the study period was ED with an average of 1238.8 tonnes/year for almost the entire period and significant at $p = 0.001$. IP was the second highest emitter for the entire period with an average of 975.7 tonnes/year. The rest of the mills were below 600 tonnes/year for almost the entire period, except for NP in 2019, which emitted 954 tonnes. None of the mills emitted below the threshold in any reporting year. The major emitter of TPM in the period was NP (Figure 9c). Before 2015, NP emitted more than 1000 tonnes/year and some years even more than 2000 tonnes/year, while the other mills remained below 300 tonnes/year. From 2016 to 2019,

NP emitted <400 tonnes/year (but still more than the remaining mills) with a slight increasing trend observed in the last 3 years.

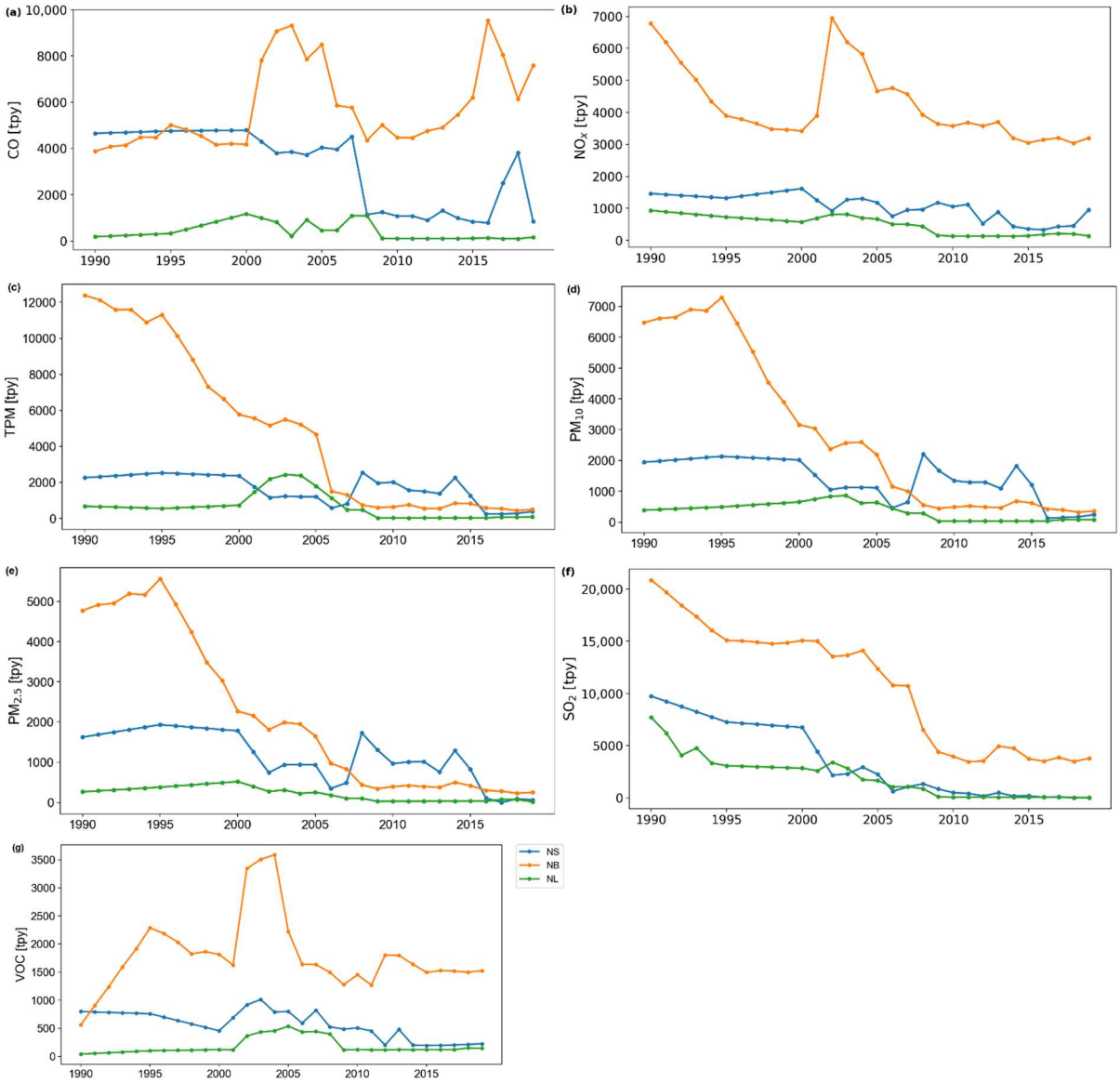

Figure 8. Air emissions registered in the APEI for Nova Scotia, New Brunswick and Newfoundland, and Labrador for pollutants (a) CO, (b) NOx, (c) TPM, (d) PM_{10}, (e) $PM_{2.5}$, (f) SO_2, and (g) VOC.

Similar characteristics were observed for PM_{10} emissions in Figure 9d, as previously shown by Hoffman et al. (2015). From 2016 to 2019, all mills emitted <120 tonnes/year, except

from NP, which emitted 127, 149, 172, and 240 tonnes, respectively, from 2016 to 2019. The releases of $PM_{2.5}$, presented in Figure 9e, show that, from 2002 to 2015, NP was the major emitter (except in 2006 where AT emitted 390 tonnes and NP 325 tonnes) with a

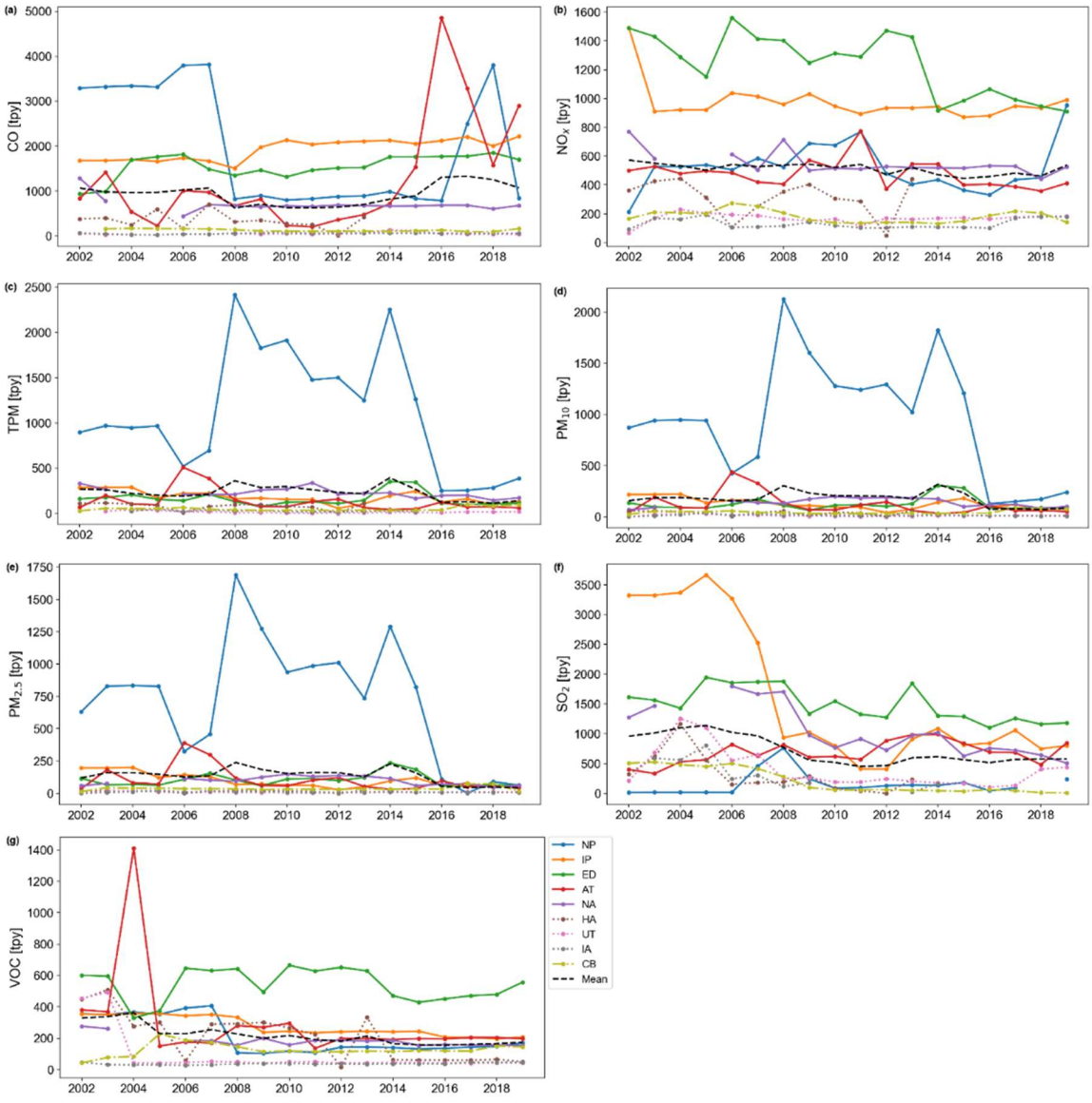

Figure 9. Air pollutant emissions from 2002 to 2019 for each facility for pollutants (a) CO, (b) NOx, (c) TPM, (d) PM10, (e) PM2.5, (f) SO2, and (g) VOC.

peak of 1689 tonnes in 2008. Mean emissions were 994.4 and 174.1 tonnes/year for the study period for NP and for all the mills, respectively. From 2016 to 2019, all mills emitted <110 tonnes/year. Sulphur oxides data reveal that from 2002 to 2007, IP was the major emitter that was consistently above 2500 tonnes/year (Figure 9f) with a maximum of 3665 tonnes in 2005. From 2008 to 2019, ED was the major emitter with ~1300 tonnes/year. The rest of the mills were below 1200 tonnes/year almost all the time but exceeded thresholds most of the time except for NP and CB, which were below the threshold five and two out of the seventeen years, respectively. Except for NP, all the mill's emissions were significant at least at p = 0.001. Figure 9g shows VOC emissions. In 2002 and 2003, ED was the largest emitter with 601 and 596 tonnes, respectively, and in 2004, the emission was 1412 tonnes in AT. From 2005 to 2019, ED emitted between 400 and 650 tonnes/year and was the major VOC emitter. The rest of the VOC emissions remained <400 tonnes/year for all mills.

Table 6. P-values for the NPRI annual trends.

***p*-Value** [1]	**CO**	**NO_x**	**TPM**	**$PM_{2.5}$**	**PM_{10}**	**SO_2**	**VOC**
Northern Pulp	0.037	0.566	0.410	0.219	0.095	0.721	0.001
Irving P & P Limited	0.000	0.074	0.002	0.003	0.000	0.000	0.000
Twin Rivers Paper	0.008	0.000	0.821	0.596	0.902	0.002	0.601
Atholville, AV Group	0.017	0.197	0.105	0.104	0.098	0.031	0.095
Nackawic, AV Group	0.098	0.006	0.006	0.971	0.388	0.000	0.001
Port Hawkesbury Paper	0.333	0.403	0.279	0.322	0.517	0.028	0.000
Lake Utopia Paper	0.208	0.665	0.044	0.178	0.510	0.019	0.019
Irving Paper Limited	0.030	0.728	0.124	0.191	0.354	0.015	0.017
Corner Brook P & P	0.013	0.130	0.091	0.109	0.234	0.000	0.623

[1] Significant values at p = 0.05 are in blue. Significant values at p = 0.001 are in red.

Figure 10 presents boxplots for all pollutants, where the red line represents the median, the extremes of the box represent the upper and the lower quartile, the whiskers the range of the data, and the empty circles the outliers. The mean releases and the standard deviation of each mill along with the mean releases of the major pollutants (considering the criteria in Buteau et al. (2018)) are presented in Table 7. The major spread for CO was in NP

releases, even though the highest mean release was in IP and the major release was in AT. Releases of NO_x had the highest spread and the highest mean in ED. For TPM, PM_{10}, and $PM_{2.5}$ the highest spread was in NP with 659.3, 567.8, and 457.7, respectively, vs. 119.4 to 5.0 tonnes in the other mills. The mean of TPM, PM_{10}, and $PM_{2.5}$ in NP is ~10 times higher for the three pollutants in comparison with the rest of the mills. It is interesting to note that the lowest release of TPM in NP is higher than the median release in the rest of the mills. The highest dispersion in SO_2 releases was observed at IP, but the highest mean occurred in ED. The highest spread among all pollutants is observed in SO_2. VOCs presented the highest spread and the highest release in AT, but the highest mean releases were in ED.

Table 7. Mean and standard deviation (±) of air releases in each mill from 2002 to 2019.

Tonnes/Year	**CO**	**NO_x**	**TPM**	**$PM_{2.5}$**	**PM_{10}**	**SO_2**	**VOC**
Northern Pulp	1985.8 ± 1292.7	522.8 ± 166.0	1115 ± 659.3	944.4 ± 567.8	717.5 ± 457.7	159.1 ± 186.9	211.9 ± 114.0
Irving P & P Limited	1927.1 ± 224.3	975.7 ± 133.1	179 ± 66.1	129.9 ± 52.4	100.2 ± 53.1	1628.8 ± 1173.5	274.7 ± 61.3
Twin Rivers Paper	1551.4 ± 263.0	1238.8 ± 213.5	159.9 ± 74.8	125.9 ± 64.0	102.7 ± 48.0	1488.2 ± 276.0	541.4 ± 102.3
Atholville, AV Group	1256.9 ± 1208.3	477.9 ± 96.7	134.9 ± 119.4	117.7 ± 103.3	105.3 ± 93.3	683.9 ± 185.6	290.2 ± 280.1
Nackawic, AV Group	698.9 ± 164.2	551.8 ± 80.7	226.8 ± 52.0	137.8 ± 41.1	97.1 ± 32.1	1034.4 ± 406.4	183.6 ± 35.4
Port Hawkesbury Paper	342.9 ± 174.0	312.2 ± 120.9	79.0 ± 35.8	38.2 ± 21.2	15.0 ± 6.9	320.8 ± 311.3	203.8 ± 148.3
Lake Utopia Paper	49.6 ± 27.9	167.9 ± 33.4	15.0 ± 9.9	12.0 ± 8.4	9.5 ± 5.2	401.7 ± 323.1	91.7 ± 135.2
Irving Paper Limited	56.1 ± 12.9	131.3 ± 33.2	26.4 ± 8.8	14.7 ± 8.5	10.3 ± 5.0	331.7 ± 245.7	36.9 ± 5.8
Corner Brook P & P	127.8 ± 27.1	181.2 ± 41.9	49.6 ± 21.9	45.9 ± 17.6	37.3 ± 15.1	206.2 ± 202.9	127.1 ± 40.5
Mean [1]	888.5	506.6	220.6	174.1	132.8	695.0	217.9
Major emitters [2]	1006.1	510.2	304.8	325.6	295.4	746.1	304.8
Percentage of major emissions [3]	22.8%	1.9%	42.0%	59.3%	67.3%	14.2%	27.8%

[1] Mean of all releases in each mill and in each pollutant. [2] Mean of all releases in each mill considering only those > 100 tonnes/year. [3] Percentage of all releases considering only those > 100 tonnes/year.

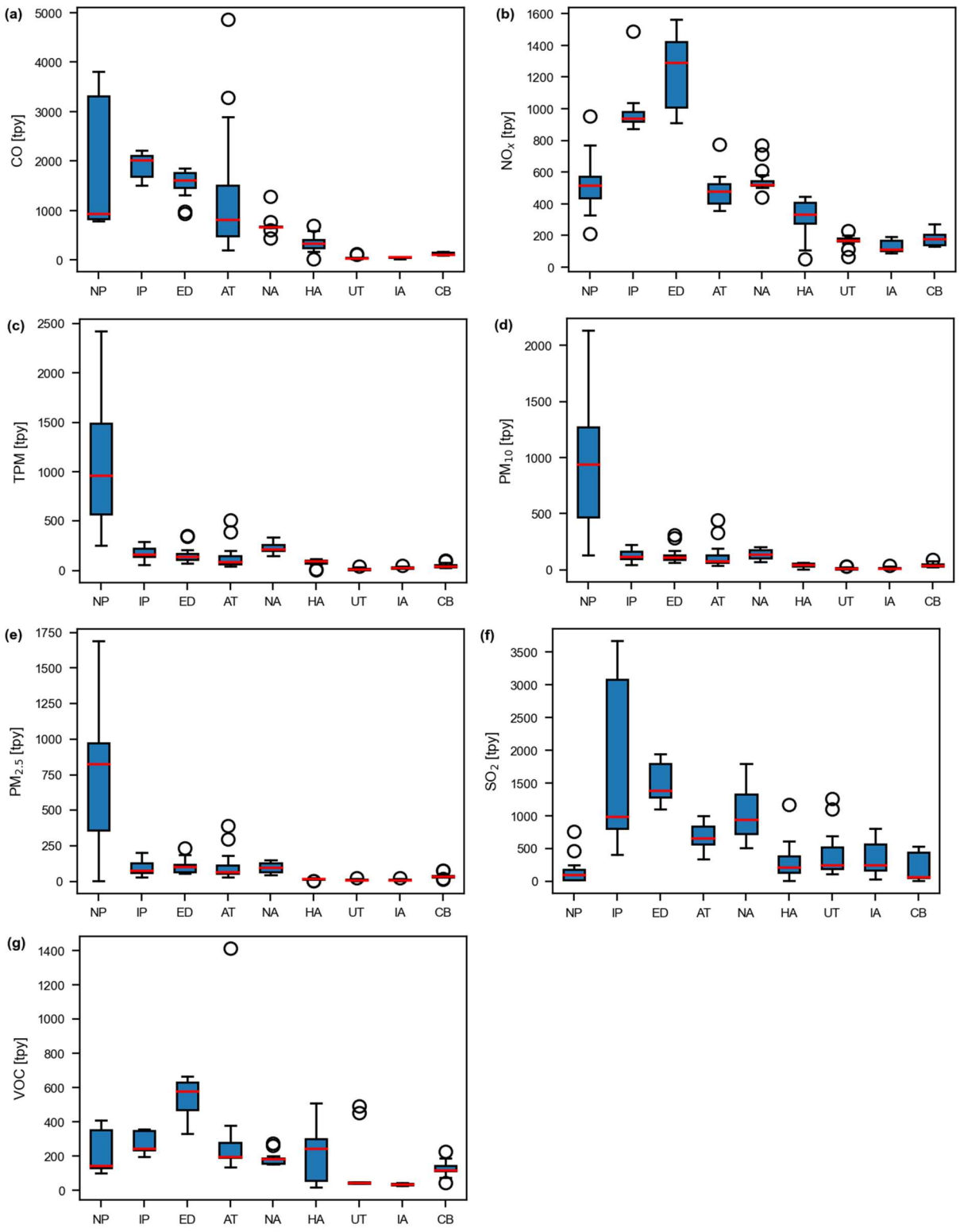

Figure 10. Distribution of air pollutant emissions from 2002 to 2019 for each facility for the pollutants (a) CO, (b) NOx, (c) TPM, (d) PM10, (e) PM2.5, (f) SO2, and (g) VOC.

3.4.2 Comparison with Thresholds

Figure 11a shows the mean DRT (Table 3) observed for every pollutant and for the entire period for all sites. $PM_{2.5}$ and PM_{10} were often orders of magnitude higher than thresholds, sometimes above 3000% and in some cases above 6000% prior to 2016. This decreased to <2000% after 2016. However, it is likely that this difference is heavily influenced by NP emissions, observed in Figure 9c,d. The dramatic reduction observed in 2017 at NP was likely due to a precipitator installation in 2016, which was intended to reduce PM emissions (Baarda 2020; Hoffman, Guernsey, et al. 2017; McDonald 2015). Other pollutants exhibited a more stable behavior, with values within 1000% of the reporting threshold for the study period. DRT for all mills for each year, averaged across all pollutants, is shown in Figure 11b. NP consistently reported higher thresholds for all pollutants, especially prior to 2017, which was as much as 100,000% above the reporting PM thresholds in some years. From 2017, NP emissions still exceeded reporting thresholds by ~20,000% on average across all pollutants. Those values are comparable to other pulp mills, which appear to release more pollutants compared to the three paper mills (HA, UT, and IA) and the P&P mill (CB), where releases were on average <10,000% above threshold values. Even though HA changed its operation from pulp to paper in 2012, it did not seem to have a substantial impact on overall pollutant releases. Figure 11c shows the DRT averaged over the period, discriminated by site and pollutant. Again, major pollutants were $PM_{2.5}$ and PM_{10} at NP, which were 100,000% above thresholds for the entire period. For pulp mills, $PM_{2.5}$ was the principal pollutant higher than the threshold, being ~4000% above, followed by PM_{10} at ~3000% over the threshold. At CB, $PM_{2.5}$ and PM_{10} releases were ~2000% above threshold, showing that releases from these P&P mills were not as high as other pulp mills but were higher than paper mills. For other pollutants and mills, differences from thresholds were <1000%.

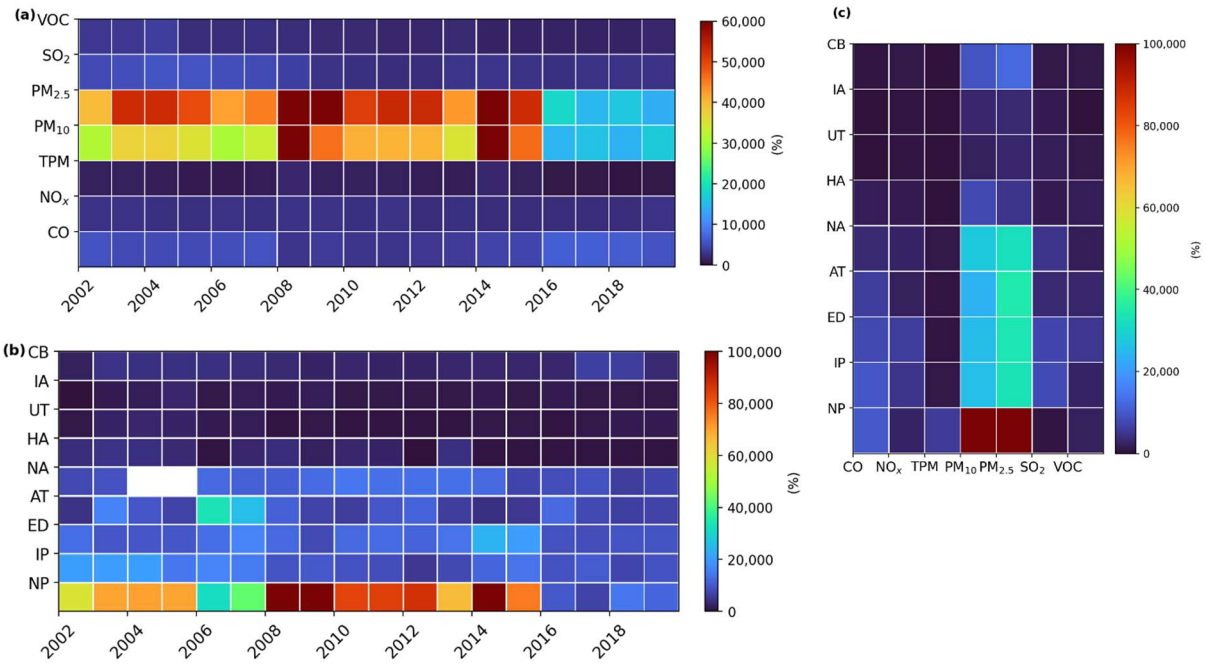

Figure 11. Mean difference from reporting thresholds (DRT) over (a) the sites, (b) the pollutants, and (c) the entire period. White represents no data.

The difference from the ARL on the CPMAEPFF for TPM and SO_2 is presented in Figure 12. For TPM, all mills emitted under the limit suggested (~70%) and with a low dispersion (~10%) for the entire period, except for NP. The TPM exceedance in NP was as high as ~300%, twice in the period, and only below ARL after 2016. NP had the highest mean exceedance of 99.1% and the highest standard deviation exceedance of 117.7% above ARL. In contrast, NP presented the lowest standard deviation of exceedance (16.7%) and the lowest mean exceedance (−85.7%) for SO_2 emissions. IP and UT presented the highest annual exceedance of ~200% and ~350%, respectively, and both were at the beginning of the study period. The highest standard deviation exceedance was in UT with 116.4%, followed by IP with 87.6%. ED had a mean of exceedance 0.6% and a standard deviation exceedance of 18.6%, which indicated that ED was close to the exceedance limit during the entire study period. After 2008, most mills emitted below the CPMAEPPF ARL or were comparable.

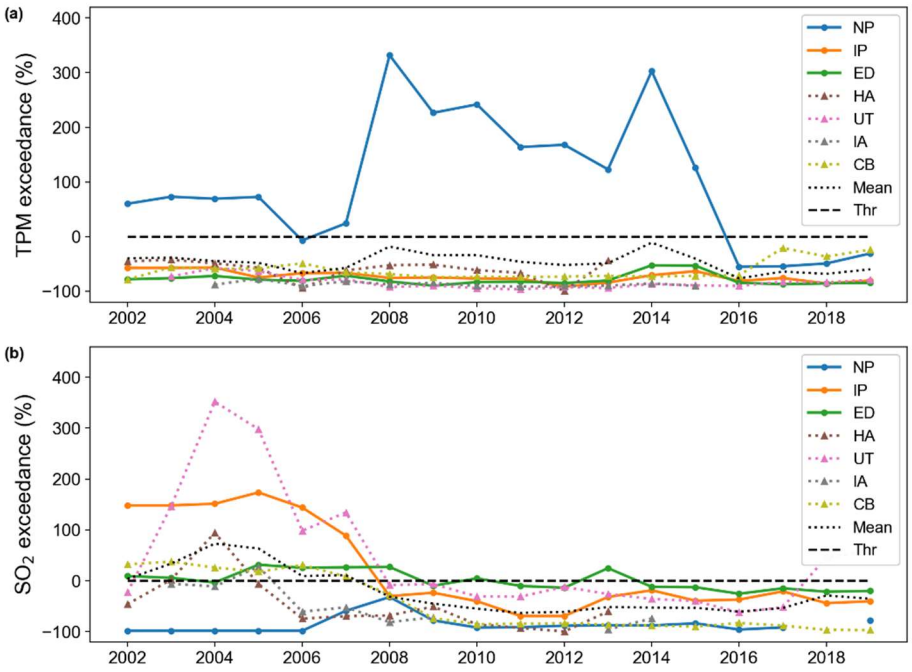

Figure 12. Percentual difference from the ARL on the CPMAEPPF for (a) TPM and (b) SO2. Pulp mills are represented by a solid line and paper mills by a dotted line. The black dotted line represents the mean on all mills and the black slashed line represents when a release equals the threshold suggested by CPMAEPPF.

3.5 Discussion

All mills in Atlantic Canada are located in or near urban areas (<5 km away, see Appendix A) and have air emissions well above NPRI reporting thresholds for a suite of pollutants, meaning that local populations (including mill workers) are regularly exposed to air pollutants emitted by mills regardless of prevailing winds. The intensity and duration of air pollutant exposure on populated centres distant from the mill depend on how frequently winds blow from the mill towards nearby towns, which cannot be determined by using only annual NPRI data. No P&P mill studied significantly reduced their mean annual air emissions (pollutant means) in the 17 years of analysis, as has been observed in other facilities (Johnston Edwards and Walker 2020; Taylor et al. 2020). The only exception (although still above reporting thresholds and higher than other pulp mill releases) was NP, which dramatically reduced PM emissions after installing a precipitator, which resulted in

emissions of similar magnitude compared to other pulp mills in Atlantic Canada (Figure 9c,d and e). Techniques to reduce emissions can be hard for industries to implement due to high costs (Cheremisinoff and Rosenfeld 2010) and the challenge of adequately planning the improvements, as they depend both on the source of energy used and technology for upgrades (Bhander and Jozewicz 2017). In this case, NP could effectively reduce PM emissions with the incorporation of a recovery boiler (Hoffman, Guernsey, et al. 2017).

While the use of NPRI has increased in the last decade, in part because of its open access and the addition of new pollutants, its limitations may undermine its usefulness (Berthiaume 2021). Among the limitations that prevent NPRI from being widely used are issues with its completeness, data quality, and inventory comprehensiveness (Berthiaume 2021; Buteau et al. 2018; Johnston Edwards and Walker 2020; Taylor et al. 2020). Most NPRI users are interested in air emissions, geospatial analysis of emissions, and environmental monitoring (Berthiaume 2021). Therefore, the completeness of the repository, where yearly releases are always included (regardless of whether they are below the NPRI lower threshold or not), is essential to generate a proper mapping of emissions and to compare how they evolve over time. Moreover, high-quality data where industries not only estimate but accurately report emissions, as well as enforced regulation, would allow better environmental assessments.

Reporting NPRI data alone is not enough to assure a decrease in pollutant emissions. Although NPRI annual release data are designed to protect human and environmental health, the releases are not regulated; therefore, the high emissions of potentially harmful pollutants are not reduced. Huge variations were observed in emission releases, with some reported just above NPRI release thresholds, whereas other emissions were reported >100,000% above the threshold (e.g., PM in NP). Reporting thresholds appear to have little to no influence on the adaptive management of P&P operations when PM emissions are 20, 1000, or even 50,000 times higher than the reporting threshold. This raise concerns over when releases pose potential threats to human or environmental health, as it is tremendously difficult to understand the contribution of these emissions to local or regional air quality and from there to understand human exposures. This information is not currently available, and a lower limit without a corresponding upper limit does not provide the

protection intended for human and environmental health. Even when data to identify potential harmful releases are publicly available, if no action is taken to reduce high emissions, it is unlikely that the problem will be addressed. Air emissions from all facilities together, especially when they are in close proximity to each other, are an accumulative source of air pollutants that can induce potential harm to ecosystems and human health.

There is a lack of standardization in how emissions are reported, due to differences in units of measurement, different permissions depending on the jurisdiction, and on the characteristics of the industry (Hoffman et al. 2015; Taylor et al. 2020). Air releases for P&P mills on NPRI are reported in tonnes/year, so a comparison with CAAQS is not simple nor direct as more information is needed to convert the release in tonnes to an emission rate (i.e., ambient temperature, pressure, and gas flow (Statistics Canada 2017)). Consequently, NPRI does not seem to be a tool that effectively informs industrial performance but is rather an accounting tool that is not linked to the enforcement of air quality standards or the enhancement of air quality for populations living in and around the mills. Considering that most of the time all mills exceeded the reporting thresholds, it would be helpful to compare how hazardous those releases are with the inclusion of an upper limit or threshold or change the unit of reporting to enable the comparison with other standards and countries. Moreover, it would be helpful to compare how much mills are emitting relative to their annual pulp or paper production. If it was possible to determine a ratio of production to release, such as tonnes of pulp produced as a function of emissions, it would be possible to identify an unsustainable or inefficient facility, which could be an indicator of outdated technology or poor management practices. Another issue of this reporting inventory is that the indicators are relatively crude measures of air pollution and do not provide insight into the toxicity of emissions. In addition, some meteorological conditions (such as inversions or the direction of the prevailing winds) or topographical features (such as a valley or hill) may increase the impact of air pollution, which could be accumulated not only by different sources but also by intrinsic characteristics of the location. Thus, exceeding a reporting threshold has an impact not only on the point location of the release but it also has the potential of affecting the surrounding environment (depending on the meteorological conditions and the topography) and of combining with

other mill's releases (which can increase the toxicity of emissions either by accumulation or because of chemical reactions).

In Buteau et al. (2018), the industries reporting NPRI within 2.5 km of children's houses had mean annual releases of 31.8 tonnes for $PM_{2.5}$ and 788.0 tonnes for SO_2. When they only considered the 'major emitters' (annual releases above 100 tonnes), mean releases increased to 290 tonnes/year for $PM_{2.5}$ and 1350 tonnes/year for SO_2. Mean releases from all P&P mills in Atlantic Canada (Table 7) show that all $PM_{2.5}$ releases in the P&P mills are four times higher in comparison to the findings of Buteau et al. (2018) and of similar a magnitude when only major emitters are considered. Conversely, SO_2 mean releases from P&P in Atlantic Canada (Table 7) are similar to those reported in Buteau et al. (2018), but when we consider the major emitters, the industries in Quebec are almost twice as high as P&P mills in Atlantic Canada.

If the criteria of 'major emitters' were applied to this study, all pulp mills would exceed this threshold. Only UT and IA (both paper mills) would not be considered major emitters for most air pollutants, and CB and HA may fall in the major emitters category dependent on the pollutant and on the year considered. Pulp mills seem to pollute more than paper mills, and the P&P facility falls in the middle. This result is consistent with Tong et al. (2018), who compared emissions from five mills in China and found that gaseous pollutants emitted from pulp mills are greater than those from paper mills. To reduce the pollution caused by pulping, they suggested recycling wastepaper. The threshold of 100 tonnes/year for $PM_{2.5}$ and SO_2 discussed in Buteau et al. (2018) was previously introduced by (Loyo-Berríos et al. 2007), although there is a lack of justification for using these criteria. Regardless, this threshold is used in this study to analyze P&P air emissions considering that there are no standards or regulations to compare with annual emissions in tonnes. This absence of regulation shows the need for a standard that could be used to inform how hazardous air emissions from different sources can be and to identify when action is needed to reduce the impact on human health and the environment.

When the releases are compared to the threshold suggested on CPMAPPF, the behaviors of pulp and paper mills do not follow a clear pattern. Instead, emissions depend on the total

production of the facility more than on the type of process carried out in the mill. Even though the CPMAPPF threshold is more permissive for SO_2 releases than for TPM, the mills are closer to the SO_2 threshold and surpass it more frequently. In contrast, most of the mills emit TPM well below the limit, with NP being the exception exceeding the threshold with a mean of ~100% over the threshold during the entire period. The emission limits and best practices recommended in CPMAPPF are still voluntary. As chemical facilities produce more pollution than the mechanical facilities due to the nature of the processes, they are 'allowed' to emit more, i.e., four times more for TPM and almost three times more for SO_2 emissions. However, the lack of enforcement can deter chemical facilities from adopting cleaner technologies, which can seem unfair to mechanical facilities that have a more stringent recommendation.

One strategy to regulate high emissions is applying fines or incentives. Incentives could include emission reductions through discounts on CO_2 taxes or the ability to draw from specific funds for upgrading technology. Although historically fines have not been consistently imposed on facilities that exceed thresholds, they can be an effective tool when implemented properly (Hoffman et al. 2015). It has been suggested that "fines must be higher than the cost to the most outdated mill of installing the technology necessary to reach compliance levels" (Hocking 1991) and that to "achieve emissions reductions, threats of monitoring and actual enforcement have proven to be most effective" (Johnston Edwards and Walker 2020). Both suggestions support the idea of strongly disincentivize high emissions (if not voluntary) by making it impossible financially; it would be more affordable to update the equipment and the facility's technology rather than paying fees for not complying with emission limits. Another important suggestion made by (Hocking 1991) is to increase the stringency on 'hot spots', sites where many mills are close to one another and were limiting each one independently may not be enough to protect human and environmental health. This type of regulation could be extremely important for some sites in Atlantic Canada where different industries are located close to each other and near populated centers. For example, in Saint John, New Brunswick, there are two P&P mills as well as a refinery within the city. Moreover, in the cities Nackawic-Madawaska, there are two P&P mills, one on either side of the Canada–United States border, located within

2 km of each other. As facilities are under different regulations, it seems that sometimes an excedance in the emission rate regulation is not enforced (Dionne and Walker 2021).

In summary, the existence of limits in air emissions is confusing and too flexible. Industries that do not follow recommendations or that are high emitters are not punished in most of the cases. The lack of annual compliance standards reduces the utility of NPRI, as this single database is not enough to determine how industries are performing towards health environmental protection.

3.6 Conclusions

The air releases of CO, NO_x, TPM, $PM_{2.5}$, PM_{10}, SO_2, and VOCs were compared for the three provinces in Atlantic Canada with pulp and paper mills in a 30-years period (the longest period of APEI data available) and from the nine mills operating in Atlantic Canada from 2002 to 2019 (the longest period of NPRI data available). Annual releases were compared to reporting thresholds established by ECCC, but neither mills nor ECCC provide a precise quantity released to the atmosphere, as the reports are annual estimates. As NPRI reports in tonnes/year, a regulation or threshold in this unit of measure seems necessary to compare with industrial emissions, as NPRI has a lower threshold for report emissions but lacks an upper threshold to limit emissions. While the lower threshold is surpassed most of the time (often by thousands of times), the lack of an upper limit results in misinformation about when those releases are harmful and may be considered a threat to human and environmental health. The recommended emission limit in the code for best practices for P&P, CPMAPPF, is not always followed by the mills as it is not compulsory. Thus, even when there are inventories accessible to the population and recommendations for industries to follow, there is no enforcement nor penalty for the facilities that emit above the safe recommended limit. A more detailed analysis using other sources of data with an hourly measurement, such as the National Air Pollution Surveillance program from ECCC, would be valuable to determine how the pollutants are emitted throughout the year. As the only air pollutants that are regulated by federal legislation through CAAQS are O_3, NO_2, SO_2, and $PM_{2.5}$, a future analysis could only evaluate compliance with regulation for those parameters.

CHAPTER 4 IDENTIFYING SOURCES OF INDUSTRIAL EMISSIONS OF FINE PARTICULATE MATTER (PM2.5) IN NOVA SCOTIA, CANADA

Abstract

Three industrial facilities (a kraft pulp mill, a tire manufacturing, and a coal-fired power generation plant), have operated in Pictou County, Nova Scotia, Canada for more than 50 years. The local population, including an Indigenous community, has raised concerns for several decades about the environmental and human health impacts of local air and effluent pollution. Numerous studies have reported negative air, water, sediment, and ecological and human health impacts in the region. However, previous studies mainly focused on wastewater effluent discharge from the kraft pulp mill, with only a few studies focused on air pollution. These limited air pollution studies assumed the pulp mill was primarily responsible for local emissions of fine particulate matter ($PM_{2.5}$), but with high levels of uncertainty. This study analyzes hourly and daily $PM_{2.5}$ concentrations measured at an air quality monitoring station located in Pictou (part of the National Air Pollution Surveillance [NAPS] network) between 2004 and 2021. For events of high $PM_{2.5}$ concentrations (which occurred predominantly in April and May in 2014 and 2015), air masses are tracked using the HYSPLIT model to evaluate if long-range transboundary pollution contributed to $PM_{2.5}$ concentration and to identify potential emission sources. Results suggest that the pulp mill was likely the primary source of high $PM_{2.5}$ concentrations recorded at the Pictou NAPS station. Measured $PM_{2.5}$ concentrations only fall into the Green air quality management level after the pulp mill installed a recovery boiler electrostatic precipitator in October 2015 to reduce $PM_{2.5}$ emissions.

Keywords: Particulate matter ($PM_{2.5}$); Industrial air pollution; National Air Pollution Surveillance (NAPS); HYSPLIT; National Pollutant Release Inventory (NPRI).

"* In preparation to be submitted to Atmospheric Pollution Research"

4.1 Introduction

Air pollution from particulate matter (PM) (a mixture of solid and liquid particles), was estimated to be responsible for more than four million deaths globally in 2016 (World Health Organization 2021). Environmental effects of PM include reduced visibility, acidification of lakes and streams (depending on PM chemical composition), and damage to vegetation, farm crops and buildings. Health effects linked to PM exposure primarily include cardiovascular and respiratory diseases, including pulmonary cancer (Environmental Protection Agency 2022a). Industrial and domestic fossil fuel combustion and the transport sector are primary sources of PM generation (Kelly and Fussell 2015; Stanek et al. 2011). Total particulate matter (TPM) includes solid and liquid suspended particles of all sizes, chemical compositions and origins (Pope and Dockery 2006). Particles with a diameter of ≤10 μm (PM_{10}), also known as the thoracic fraction, can penetrate past the bronchus and into the lungs leading to acute health response. The respirable fraction of TPM is particles less than 2.5 microns in diameter ($PM_{2.5}$) that can reach the alveoli and lead to chronic diseases such as lung cancer and cardiovascular disease (Environmental Protection Agency 2022b). Many studies have shown how $PM_{2.5}$ affects human health (Burnett et al. 2018; Buteau et al. 2018; Pope and Dockery 2006; Rajagopalan, Al-Kindi, and Brook 2018; Schwartz and Neas 2000) although there is little debate as to the role of fine particles in the etiology of cardiopulmonary disease.

In Nova Scotia (NS), Canada, air quality is affected not only by transportation, industrial and local fossil fuel combustion sources but also, because of its geographical location, the long-range air transport from the northeast United States (Dabek-Zlotorzynska et al. 2011; Gibson et al. 2009; Jeong et al. 2011; Mitchell et al. 2021). Three industrial facilities, a kraft pulp mill (KPM), a coal-fired power generation station (CFPGS), and a tire manufacturing facility (TMF), have been operating in Pictou County (PC), NS, Canada for more than 50 years (Figure 13). The KPM began operating in 1967 and was closed in January 2021 (Davidson et al. 2021; Quanz et al. 2021). The CFPGS in Trenton started its operation in 1969 and is planned to close by 2024 as part of the provincial plan to reduce dependency on fossil fuels (Hoffman, Guernsey, et al. 2017; Nova Scotia Power 2021). Michelin Tire Granton was established in 1969 and 1971 produced its first tire with a

generous economic incentive from the province of Nova Scotia (Miltenburg 2015). The local population situated around these facilities has been concerned about air quality in the region and how releases from nearby industrial facilities have impacted human and environmental health (Castleden et al. 2017; Hoffman et al. 2015; Lewis, Francis, et al. 2020; Reid 1989). Previous studies have shown that the pulp mill emits diverse PM size classes (e.g., TPM, PM_{10} and $PM_{2.5}$) and that emissions were several orders of magnitude higher than other similar mills (Giacosa et al. 2022; Hoffman, Guernsey, et al. 2017). However, there has been no study to investigate how other nearby facilities (i.e., the coal-fired power generation station and tire manufacturing facility), or long-range transboundary air movements have contributed to local air pollution in the area.

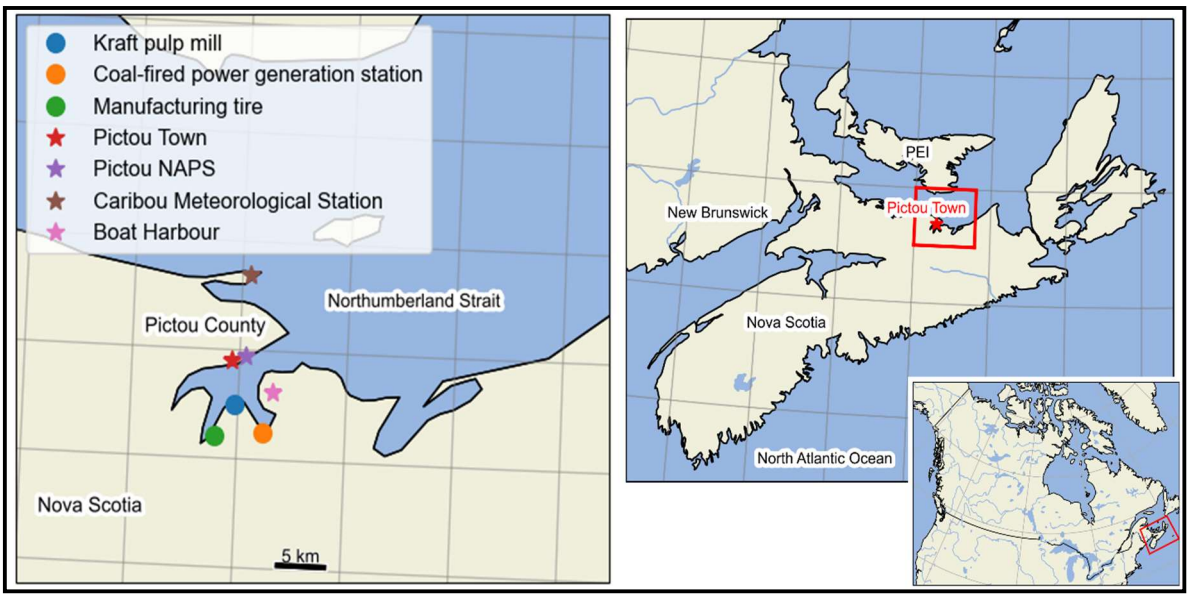

Figure 13. Location of industrial facilities in Pictou County (the pulp mill, the tire manufacturing, and the coal-fired generation plant), population centres and monitoring stations.

In this study, we investigate the combined influence of these facilities on air quality based on hourly and daily $PM_{2.5}$ concentration measurements over a 19 year period (from 2003 to 2021). Data are obtained from a local air quality monitoring station located in Pictou County, northeast of Pictou town (Figure 13). The air quality station provides publicly

available hourly air quality data for different air pollutants (including $PM_{2.5}$) and is operated by Environment and Climate Change Canada (ECCC) as part of the National Air Surveillance Program (NAPS). The primary objective of this study is to investigate how $PM_{2.5}$ concentrations measured at the NAPS monitoring station compare to Canadian Ambient Air Quality Standards (CAAQS). A secondary objective aims to assess how surrounding industrial facilities have contributed to the local $PM_{2.5}$ pollution concentrations in the last 19 years.

4.2 Industrial Facilities

At Abercrombie Point in PC, a KPM (currently owned by Paper Excellence and named Northern Pulp [NP]), operated from 1967 to 2020. Building of the mill began in 1965 and sparked the beginning of industrial development in the region (Castleden et al. 2017). Boat Harbour (BH) or A'se'k ("the other room" in the Indigenous Mi'kmaq language), is a tidal estuary connected to the Northumberland Strait and a sacred place for Pictou Landing First Nations (PLFN). The site was frequently used for hunting, fishing and for recreational and religious purposes (Group Pictou Landing Native Women's (c/o Sheila Francis Past President) et al. 2016; Hoffman, Lyons, et al. 2017) until it became a lagoon for handling more than 50 years of wastewater discharges from the mill. In 2014, a smoke plume and an effluent leak from the mill triggered a provincial response to enforce air emissions and disposal effluents regulations (Hoffman et al. 2015). The particulate emissions caused by the failure of a recovery boiler electrostatic precipitator resulted in the creation of a ministerial order that forced the mill to upgrade equipment and replace the precipitator. That upgrade was completed by October 2015 (Hoffman, Guernsey, et al. 2017). The effluent spill resulted in the enactment of the *Boat Harbour Act* (2015), an agreement between PLFN and the minister of environment to cease the discharge of industrial effluents in BH before remediation (Chaudhary et al. 2020; Hoffman et al. 2015; NSE 2015; Romo et al. 2019). The mill failed to secure approvals for an environmental impact assessment describing the alternative effluent treatment facility, resulting in the closure of the mill by January 31, 2020 (Quanz et al. 2021). NP is currently suing the provincial government of Nova Scotia for almost half a billion Canadian dollars alleging that they

were forced to close losing millions as a result (Beswick 2021; Gorman 2022), and it is uncertain if NP will resume operations.

Michelin Tire in Granton is one of three Michelin plants in Nova Scotia and its main activity consists of preparing rubber mixtures for use in tire manufacturing in the other two plants (Anningson et al. 1992). In the TMF, metal fabric for tires is made from steel cord (transported from the plant in Bridgewater) integrated with rubber (Langille 1981). When established in 1969, the TMF facility employed 1,050 people, and due to a high maintenance cost, it announced the closure in 2014 (Miltenburg 2015). However, it has remained open with reduced production employing 500 people since 2014 (CBC News 2014a). Although other Michelin plants upgraded their technology, no technology updates were made at the TMF plant (Miltenburg 2015).

The CFPGS in PC, Trenton, is one of the four currently operating coal plants in Nova Scotia (Pearre and Swan 2013). The CFPGS station has two generating units with a combined capacity of 307 MW. One generating unit in Trenton, knowns as unit 5, was commissioned in 1969 and in 2009 underwent a complete refurbishment to improve unit efficiency. The other generating unit in Trenton, known as unit 6, was commissioned in 1991 (Nova Scotia Power 2021). Part of the resulting ash in the facility is reused in the construction industry in applications such as cement and concrete; the remaining ash is dry stack stored and shipped to other locations for export (Nova Scotia Power 2022). As part of a provincial plan to move towards renewable electricity sources the generating unit Trenton 5 is slated to close by 2024 (Giacosa and Walker 2022).

4.3 Environmental impacts in the area

It has been suggested that the location of the KPM was deliberately chosen to be near an indigenous community (Castleden et al. 2017). The PLFN community feels they suffered negative psychological impacts because of the loss of A'se'k due to exposure to untreated effluent released for more than 50 years, and considered the placement of KPM a case of environmental racism (Castleden et al. 2017; Paul 2006; Waldron 2018).

Few studies of air pollution have been conducted in PC (Group Pictou Landing Native Women's (c/o Sheila Francis Past President) et al. 2016), and there is limited information about how the operation of local industries has impacted PC. While the three industrial facilities in PC have been operating for more than 50 years, few improvements have been made to the facilities to upgrade with technologies required to reduce atmospheric emissions, except for the precipitator upgraded at the KPM in 2015. Some studies have linked air pollution findings with the pulp mill (Giacosa et al. 2022; Hoffman et al. 2015; Hoffman, Guernsey, et al. 2017). Hoffman et al. (2017a) documented elevated concentrations of certain VOCs between 2006 and 2013 exceeded cancer risk thresholds in the former Granton NAPS air quality station. Hoffman et al. (2015) found discordance between levels of pollution reported by different mills, identifying PM and sulphur odours emissions two to three orders of magnitude higher in comparison to other Canadian kraft pulp mills and the study corroborated the concerns of the locals. A recent study shows how the KPM annual self-reported PM releases were reduced by two orders of magnitude after precipitator upgrading in 2015 (Giacosa et al. 2022). In 2013, a study evaluated concentrations of $PM_{2.5}$ around PLFN with 13 $PM_{2.5}$ samplers located in PC downwind between the KPM and the community (Group Pictou Landing Native Women's (c/o Sheila Francis Past President) et al. 2016). One of the samplers was located in Caribou Point (north of the KPM), another sampler next to the KPM, and eleven around BH (east-northeast from the KPM). The $PM_{2.5}$ concentrations were low and comparable to those found in another region of the province of NS. The authors suggest that further studies are required to better determine whether there could be other potential sources of $PM_{2.5}$ concentrations in the region.

Although PM emissions in PC remain a concern (Giacosa et al. 2022; Hoffman et al. 2015), there has not been a study to compare PM emissions from all three local facilities to assess which facilities are the major emitters of $PM_{2.5}$ in PC. It could be expected that emissions are greater when winds are coming from the CFPGS rather than from the KPM or the TMF, as the former directly emit PM during fossil fuel combustion (while the manufacturing processes emits PM from indirect sources). Also, there are other contributing PM sources,

such as local traffic, construction sites or biomass combustion (from wood burning or wildfires) that can be even more challenging to quantify.

Air emission limits

The CAAQS are the established national guidelines for criteria air contaminants to protect human and environmental health (CCME 2021) and describes different air quality management levels of concentrations of ambient air pollutants, including $PM_{2.5}$, ozone (O_3), sulphur dioxide (SO_2), and nitrogen dioxide (NO_2) averaged over specific periods (1-hr, 8-hr, 24-hr, or annual depending on the pollutant). The pollutant concentration is classified following a traffic light colour scheme. For $PM_{2.5}$, management levels depend on pollutant concentrations in $\mu g/m^3$ (Table 8). Management levels "were not developed as facility level regulatory standards. Rather, they are used by provinces and territories to guide air zone management actions intended to reduce ambient concentrations below the CAAQS and prevent CAAQS exceedances" (CCME 2019), so the different management levels guide actions regarding appropriate air quality management based on health risk. The lowest level, green, indicates air quality should be maintained; yellow, indicates air quality should be improved through continuous action; orange, indicates air quality should be improved by preventive action; and the highest level of air quality management, red, indicates CAAQS have been exceeded and advanced action should be taken (CCME 2019).

Table 8. Canadian Ambient Air Quality Standards for $PM_{2.5}$ (CCME 2021).

Air quality management levels	CAAQS Management Levels for the 24-hour $PM_{2.5}$ ($\mu g/m^3$)		CAAQS Management Levels for the Annual $PM_{2.5}$ ($\mu g/m^3$)	
	2015	2020	2015	2020
Red	>28	>27	>10.0	>8.8
Orange	20 to 28	20 to 27	6.5 to 10.0	6.5 to 8.8
Yellow	11 to 19	11 to 19	4.1 to 6.4	4.1 to 6.4
Green	≤10	≤10	≤4.0	≤4.0

The Nova Scotia provincial government does not have a specific limit for $PM_{2.5}$, but there is a daily and annual limit for TPM in the *Air Quality Regulations of the Environmental*

Act (Nova Scotia 2005). The 24-hour average limit for TPM is 120 µg/m^3 and the annual geometric average is 70 µg/m^3. When a facility in Nova Scotia emits above a certain threshold, there is a fee for each tonne of emission. The threshold for industrial air emissions is 30 tonnes per year, so when industrial facility releases are above that threshold, the facility must pay $7.35/tonne (Nova Scotia 2016). NSE plans to update air quality guidelines by 2025 (and every five years after that) considerably expanding the list of pollutants to meet the World Health Organization (WHO) standards (Air Quality Unit Department of Environment and Climate Change 2022).

4.4 Methodology

The study is divided into two sections. The first comprises an hourly analysis of $PM_{2.5}$ concentrations and wind direction between 2003 and 2021; the second section compares annual $PM_{2.5}$ emissions from the three industrial sources with ambient $PM_{2.5}$ concentrations for ten years. The data used for this study consist of publicly available $PM_{2.5}$ data from a stationary federal monitoring station, as part of the Canadian National Air Pollution Surveillance Program (NAPS), and from annual emissions reported to the National Pollutant Release Inventory (NPRI). To identify where the air emissions are coming from wind direction (WD) and wind speed (WS) are obtained from ECCC.

Hourly $PM_{2.5}$ concentrations were compared with the CAAQS at daily and hourly frequencies following the three-years average calculation level (CCME 2019). To determine $PM_{2.5}$ peak events, the 20th highest hourly and daily $PM_{2.5}$ measurements were identified with the corresponding wind direction associated with that date. To ensure that the highest $PM_{2.5}$ concentrations were not isolated high concentrations events, the dates from those 20th maximums when high concentrations were recorded both on a daily and hourly basis were considered a peak event and selected for further analysis. Those events identified as the highest peak dates in the entire period were further analyzed to infer if the peak events were caused by a local emission source or by transboundary transport. For that purpose, the criterion used was the one defined by Mitchell et al. (2021), who defined transboundary transport when several air quality stations record high concentrations of an air pollutant simultaneously. Although Mitchell et al. (2021) focused on ozone, which has

a longer lifetime than $PM_{2.5}$, the 24 hours of transport tracking of $PM_{2.5}$ are still within its expected lifetime in the atmosphere. The second part of the analysis includes a statistical comparison (described in further detail in subsection 4.4.3) of annual $PM_{2.5}$ releases reported by the three industrial facilities with different annual percentiles of the hourly $PM_{2.5}$ measured concentrations to identify the contribution of each facility.

In March 2020, the Nova Scotia provincial government declared a state of emergency due to the COVID-19 pandemic. This lockdown may have affected normal operations of these industrial facilities (due to reduced personnel numbers to maintain social distancing mandates). Also, other secondary sources that were not considered in this study, such as fine particles emitted from vehicular transportation, would have been considerably lower during this period (Arregocés, Rojano, and Restrepo 2021; Kumar et al. 2020).

4.4.1 $PM_{2.5}$ concentrations and meteorological data

The NAPS program consists of urban and rural air quality monitoring stations dispersed across Canada reporting hourly ambient air quality data (Environment and Climate Change Canada, 2021a). Although the program started in 1969, not all stations began operating at the same time, nor do they measure the same parameters. In Nova Scotia, there are seven NAPS stations currently in operation (Figure 14 and Table 9). Southeast of the Pictou station, there used to be a VOCs NAPS monitoring station in Granton (complementary to the Pictou station which does not measure VOCs), although it became inactive in April 2016 (Nova Scotia Environment, 2022). This study includes $PM_{2.5}$ from the Pictou station between 2004 and 2021. Hourly and daily wind speed (WS) and wind direction (WD) are from Caribou Point automatic meteorological station (location in Figure 13, 10 km from the NAPS station) as Pictou only began to record WS and WD in 2020. WD data is recorded in tens of degrees, so it was multiplied by ten to present it in the range (0 - 360 degrees). Additionally, $PM_{2.5}$ data from Aylesford, Halifax Johnston, and Lake Major between 2014 and 2015 were included in the analysis (locations in Figure 14).

Data on $PM_{2.5}$ concentrations were divided into three different periods to assess temporal variation: i) 2004-2015 (before the precipitator upgrade at NP; ii) 2016-2019 (during operations with precipitator before the closure); and iii) 2020-2021 (after the closure of NP). Peak event detection was conducted by selecting dates when a peak in $PM_{2.5}$ concentrations occurred both in hourly and daily data to ensure that the event captured represents a day with $PM_{2.5}$ concentrations considerably higher than the average concentrations. . To determine potential sources responsible for peak events the air mass path and the $PM_{2.5}$ concentration on each event were calculated for a 24-hour period before the hourly peak recorded in the Pictou NAPS station.

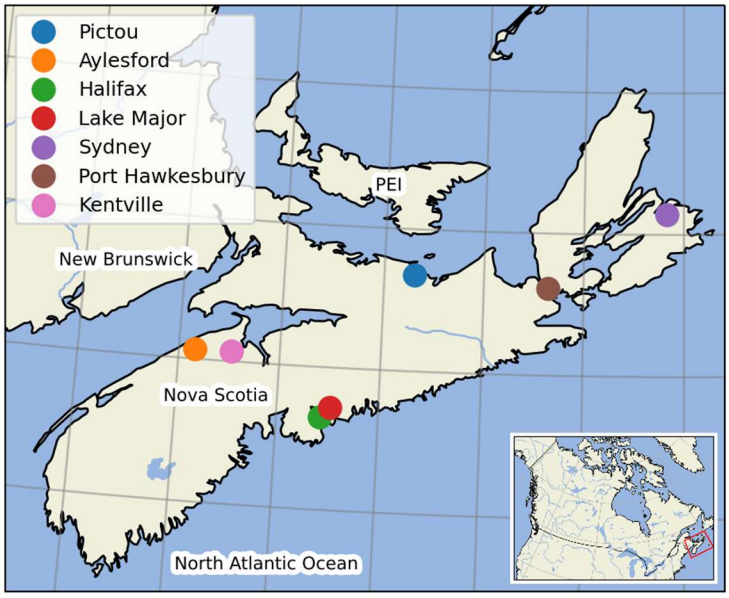

Figure 14. Location of currently operating air quality NAPS stations in Nova Scotia.

Table 9. Air quality NAPS stations currently operating in Nova Scotia.

Station (Latitude, Longitude)	NS Zone	NAPS ID	Established date[1]	Elevation (m)	Parameters[2]
Kentville (45.07, -64.48)	Western	31101	2016/08/01	60	NO_x, NO, NO_2, O_3, $PM_{2.5}$
Aylesford (45.07, -64.84)	Western	30701	1991/01/01	235	O_3, $PM_{2.5}$, WS, WD
Pictou (45.68, -62.70)	Northern	30901	2001/01/01	12	SO_2, NO_x, NO, NO_2, O_3, $PM_{2.5}$, TRS, WS, WD

Halifax Johnston (44.65, -63.57)	Centre	30113	2006/01/01	28	CO, SO_2, NO_x, NO, NO_2, O_3, $PM_{2.5}$, VOC, WS, WD
Lake Major (44.72, -63.48)	Centre	30120	2001/01/01	68	SO_2, NO_x, NO, NO_2, O_3, $PM_{2.5}$, WS, WD
Sydney (46.14, -60.17)	Eastern	30310	1974/01/01	27	CO, SO_2, NO_x, NO, NO_2, O_3, $PM_{2.5}$, WS, WD
Port Hawkesbury (45.61, -61.36)	Eastern	30201	1994/01/01	14	SO_2, NO_x, NO, NO_2, O_3, $PM_{2.5}$,WS, WD

[1] Indicates when the station began operation. Not all currently monitored pollutants or meteorological variables were initiated on the same date.
[2] CO: Carbon Monoxide. SO_2: Sulfur Dioxide. NO_x: Nitrogen Oxides. O_3: Ozone. $PM_{2.5}$: Particulate Matter. TRS: Total Reduce Sulfurs. VOC: Volatile Organic Compounds. WD: Wind Direction. WS: Wind Speed.

4.4.2 Industrial releases of $PM_{2.5}$

The NPRI is a publicly available self-reported database where facilities that reach a reporting threshold report to ECCC their annual air, water, and land releases into the environment (Berthiaume 2021; Environment and Climate Change Canada 2020b). The inventory "is a major starting point for identifying and monitoring sources of pollution in Canada, and in developing indicators for the quality of our air, land and water. The NPRI helps determine if regulatory or other action is necessary to ensure reductions, and if so, the form that action should take" (Environment and Climate Change Canada, 2020b, p.1). For this study, annual $PM_{2.5}$ releases from the TMF, CFPGS, and KPM were analyzed between 2013 to 2020. Although NPRI 2021 data are published on the ECCC website, the data are preliminary and not used in this study. The comparison between annual NPRI releases and NAPS data is implemented by calculating the annual $PM_{2.5}$ NAPS concentrations on the median (P50), percentile 90% (P90) and percentile 99% (P99) to capture the relationship between the variables when high $PM_{2.5}$ concentrations are recorded.

4.4.3 Metrics

The Mann-Whitney test (2-sided) was employed to explore if there is a statistically significant relationship between the NAPS annual concentrations and the NPRI annual

releases, with the null hypothesis that the two samples come from the same population. This test was selected as the samples analyzed are continuous, independent, and non-normal, and each data set has more than five samples. Linear regressions of NAPS percentiles on NPRI releases between 2011 and 2021 are also calculated where the predicted variables are the P50, P90 and P99 for the three industrial facilities. In total, nine linear regressions were calculated, and for each of the three facilities, three NAPS data annual modelling was considered by the P50, P90 and P99. For each facility, linear regression calculations considered that the independent variable was the NPRI releases and that each of the three annual NAPS percentiles was the dependent variable. To capture only the emissions that were predominantly coming from the three industrial facilities, the analysis included those $PM_{2.5}$ concentrations when 150°< WD < 255°.

The statistical indicators evaluated are the root mean square error (RMSE), the mean absolute error (MAE), the coefficient of determination (R^2) and the Pearson correlation (ρ). The indicators are presented in Eq. 1, 2, 3 and 4 respectively, where x represents the NPRI releases from the industrial facility, y represents the actual NAPS observations, $\hat{y}$ represents the estimated NAPS observations when implementing the linear regression, $\bar{x}$ represents the average of the NPRI releases, $\bar{y}$ represents the average of the NAPS observations, and N is the total observations (N=10 as the period for this calculations includes data from 2011 to 2020).

$$RMSE = \sqrt{\frac{\sum_{i=1}^{N}(y_i - \hat{y}_i)^2}{N}} \quad \text{Eq. 1}$$

$$MAE = \frac{\sum_{i=1}^{N}|y - \hat{y}_i|}{N} \quad \text{Eq. 2}$$

$$\rho = \frac{\sum_{i=1}^{N}(x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum_{i=1}^{N}(x_i - \bar{x})^2 (y_i - \bar{y})^2}} \quad \text{Eq. 3}$$

$$R^2 = 1 - \frac{\sum_{i=1}^{N}(y_i - \hat{y}_i)^2}{\sum_{i=1}^{N}(y_i - \bar{y})^2} \quad \text{Eq. 4}$$

4.4.4 Trajectory model

The Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) was implemented to understand where the air masses came from when high $PM_{2.5}$ concentrations were recorded. HYSPLIT integrates the meteorological wind fields to calculate air mass trajectories (Rolph, Stein, and Stunder 2017; Stein et al. 2015). The model was developed by the National Oceanic and Atmospheric Administration (NOAA), is publicly available and can be run within the website (https://www.ready.noaa.gov/HYSPLIT_traj.php). Following a recent study in Nova Scotia (Mitchell et al. 2021), the model was used to calculate trajectories using the North American Mesoscale (NAM) meteorological fields with 12 km of horizontal resolution. The backward trajectory ensemble was initiated at 500 m above ground level for 24 hours arriving at Pictou station to identify the estimated trajectory of the air mass arriving at that location.

4.5 Results

4.5.1 Local Winds and Annual $PM_{2.5}$ Concentration in Pictou

Hourly $PM_{2.5}$ measurements show ambient $PM_{2.5}$ concentrations increased until mid-2015 and then decreased by more than 50% after that time (Figure 15a). Annual $PM_{2.5}$ concentration cycles indicate that independently of the period assessed; the summer season exhibited the highest $PM_{2.5}$ concentrations (Figure 15b). $PM_{2.5}$ concentrations measured until 2015 show that the daily mean was above the mean for the total period (2004 to 2021). $PM_{2.5}$ concentrations measured during the 2016-2019 and 2020-2021 periods were more variable, where concentrations during the first period were slightly lower than the latter. The highest $PM_{2.5}$ concentrations recorded in Pictou coincided with air masses with origins predominantly from between the southeast and southwest (SW) and before 2016 (Figure 15c).

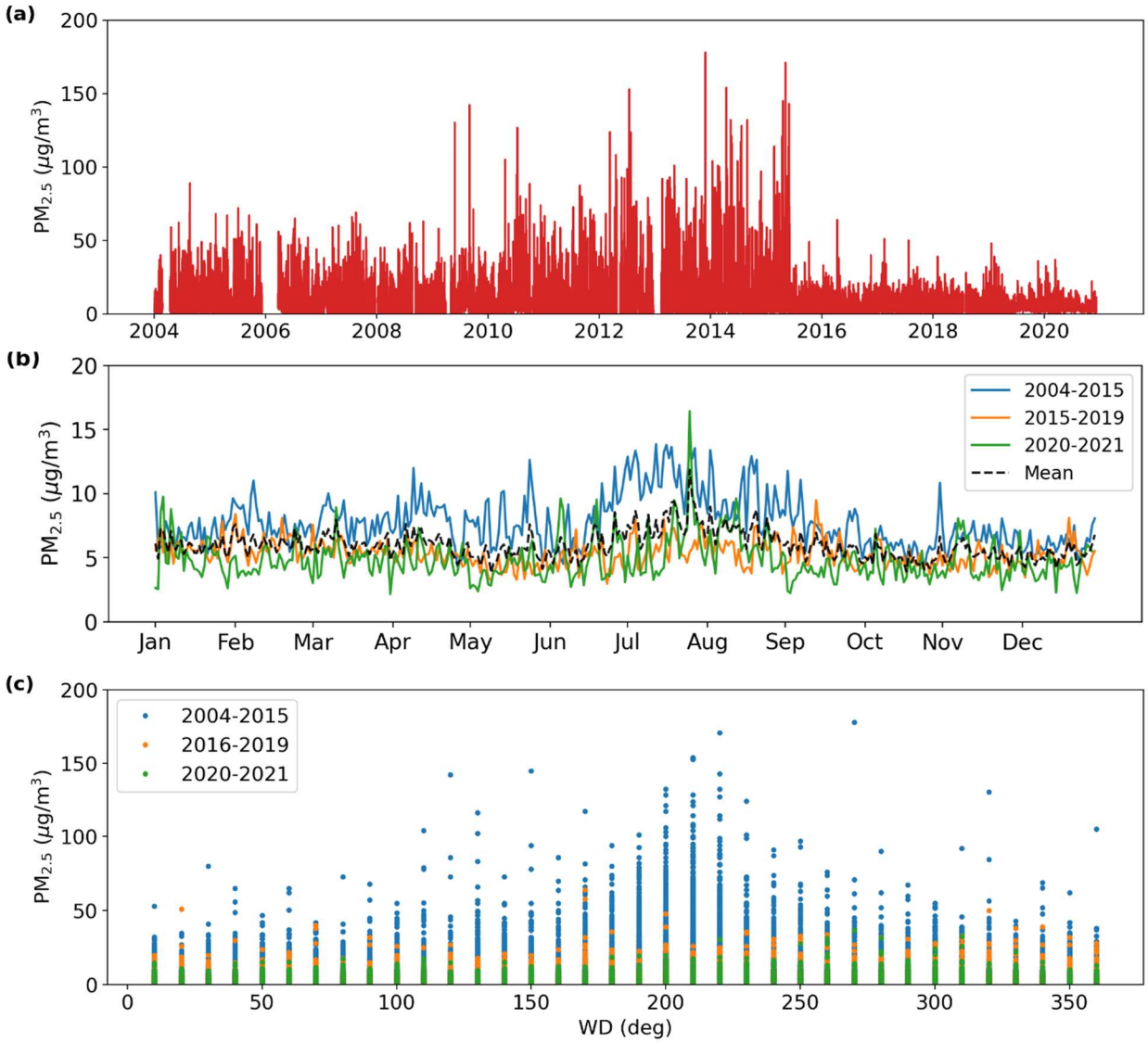

Figure 15. a) Hourly $PM_{2.5}$ in Pictou NAPS station; b) annual cycle of daily $PM_{2.5}$ averaged in three different periods with the mean of the period (2004-2021) dashed; and c) hourly $PM_{2.5}$ in Pictou NAPS stations discriminated by wind direction in Caribou meteorological station in three different periods (2004-2015 in blue, 2015-2019 in orange, 2020-2021 in green).

To assess how prevailing wind directions affected $PM_{2.5}$ measured at the Pictou NAPS station, bi-monthly pollutant roses were calculated for the entire period (Figure 16). Pollutant roses indicate WD and the $PM_{2.5}$ concentrations measured at that time (in $\mu g/m^3$). During the spring and summer months, prevailing winds were from the S and the SW, and $PM_{2.5}$ concentrations were higher than in the rest of the year (Figure 16c,d,e). The highest

average concentrations occurred during April and May and are > 50 µg/m^3 when the WD blows from the south-southwest (SSW). Although there was a secondary maximum of WD frequency from the west and west-northwest in the fall and winter months (Figure 16a,b,f), the $PM_{2.5}$ concentrations were lower in comparison to the concentration from the SW.

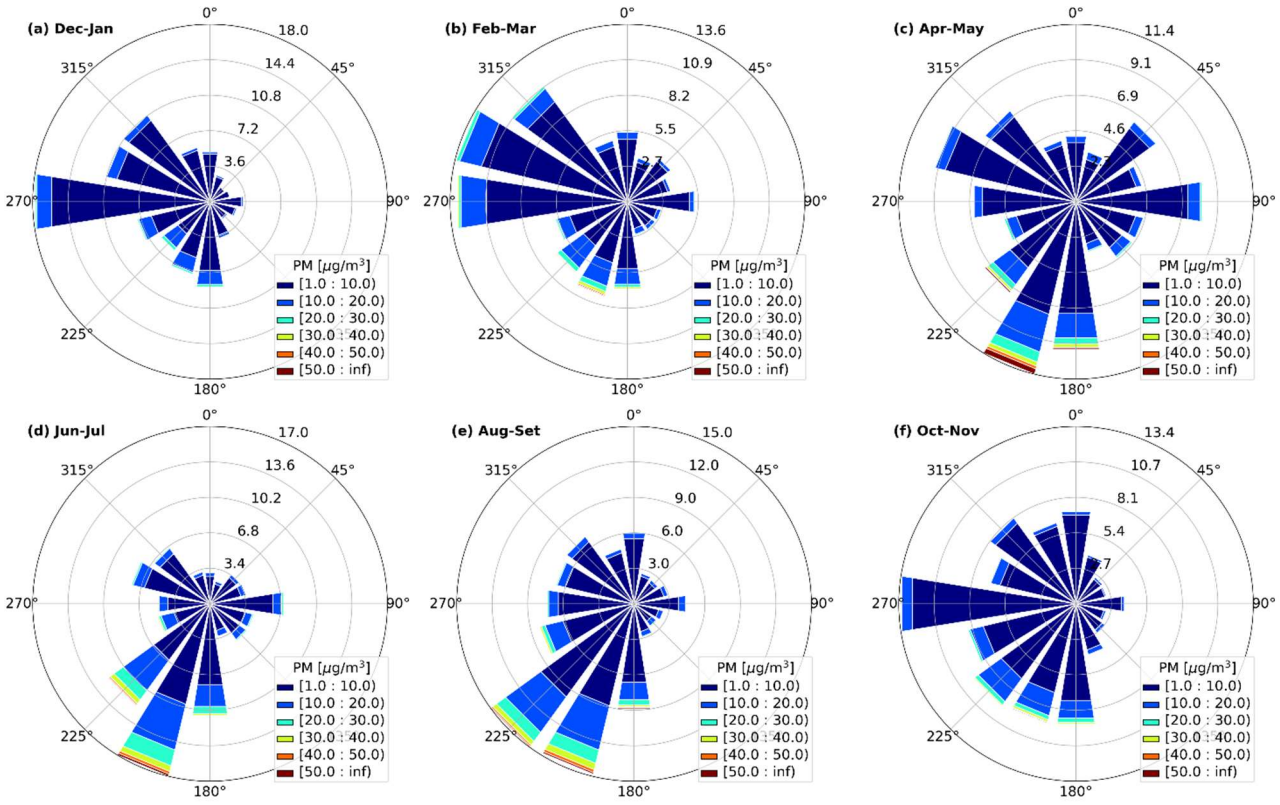

Figure 16. Bi-monthly pollutant roses for 2004-2021 for $PM_{2.5}$ concentrations in Pictou for a) December and January; b) February and March; c) April and May; d) June and July; e) August and September; f) October and November. The colour scale indicates the $PM_{2.5}$ hourly concentration in µg/m^3. Direction indicates where the wind is blowing from. The radial numbers indicate the frequency of occurrence.

Table 10 classifies the daily and annual $PM_{2.5}$ concentrations in Pictou according to the Air Quality Management Levels in Table 8. Even though management levels began in 2015, Table 3 classifies all $PM_{2.5}$ concentrations since 2003 to gauge air quality before the

implementation of CAAQS. Daily means were rounded to the nearest whole number to match the CAAQS levels (presented in Table 8). For example, for the 24-hour classification, the green level includes all $PM_{2.5}$ concentrations <10 µg/m^3 and the yellow level starts at 11 µg/m^3, so in 2017 and 2018 when $PM_{2.5}$ concentrations were 10.1 and 10.3 µg/m^3 respectively, and the management levels were selected by rounding to the nearest whole number, which is green in this example. Since 2016, $PM_{2.5}$ concentrations were classified at yellow and green levels. For the same period, the hourly mean was reduced by 35% (from an average of 7.9 µg/m^3 between 2003-2015 to an average of 5.2 µg/m^3 between 2016-2021). The standard deviation (STD) presented the same reduction for that period.

Table 10. $PM_{2.5}$ concentrations from Pictou. The hourly minimum, maximum, mean, and standard deviation were calculated by averaging the data on an hourly basis. The daily mean and the annual mean are calculated according to the CAAQS standards and coloured according to the management levels.

Year	Hourly min. (µg/m^3)	Hourly max. (µg/m^3)	Hourly mean (µg/m^3)	Hourly STD (µg/m^3)	Daily mean[1] (µg/m^3)	Annual mean[2] (µg/m^3)
2003	1.0	76.0	9.4	8.6	Nan[3]	Nan[3]
2004	1.0	89.0	8.4	7.1	24	7.7
2005	1.0	72.0	8.0	7.6	23	7.6
2006	1.0	65.0	8.9	7.6	23	7.0
2007	1.0	69.0	7.3	7.5	21	6.7
2008	1.0	63.0	6.9	6.4	19	6.1
2009	0.1	142.2	7.0	6.7	21	6.8
2010	0.2	126.8	8.5	8.8	21	7.2
2011	0.1	87.4	7.1	6.8	21	7.3
2012	0.1	152.9	7.2	8.7	20	6.9
2013	1.0	178.0	7.8	9.5	22	7.3
2014	1.0	154.0	8.6	10.4	23	7.4
2015	1.0	171.0	7.5	8.4	18	6.5
2016	1.0	64.0	5.1	3.5	14	5.6
2017	1.0	51.0	5.6	3.7	10	5.0
2018	1.0	39.0	5.5	3.3	10	5.2
2019	0.2	48.0	5.0	3.0	10	4.9
2020	0.1	36.8	4.8	2.6	10	4.9
2021	0.1	43.0	5.0	3.0	Nan[3]	Nan[3]

[1] The 3-year average of the annual average of the daily 24-hour average concentrations.

[2] The 3-year average of the annual 98th percentile of the daily 24-hour average concentrations.
[3] Not enough data to calculate the 3-year average (one year of data is unavailable).
Bi-annual pollutant roses for the period 2004-2021 (Figure 17) show that $PM_{2.5}$ concentrations > 30 µg/m^3 were only recorded from the south, SSW, and SW before 2016. The highest $PM_{2.5}$ concentrations (>50 µg/m^3) occurred between 2012 and 2015 (with a peak of 178 µg/m^3 in 2013) and were predominantly from the SSW and the south (~30% of the time). From 2004 to 2011 the highest concentrations were from the south and SSW, although in lower magnitude in comparison to the period 2012-2015. Between 2016 and 2019 around ~75% of the concentrations recorded came between the northwest and south and did not overpass the 20 µg/m^3 from any direction.

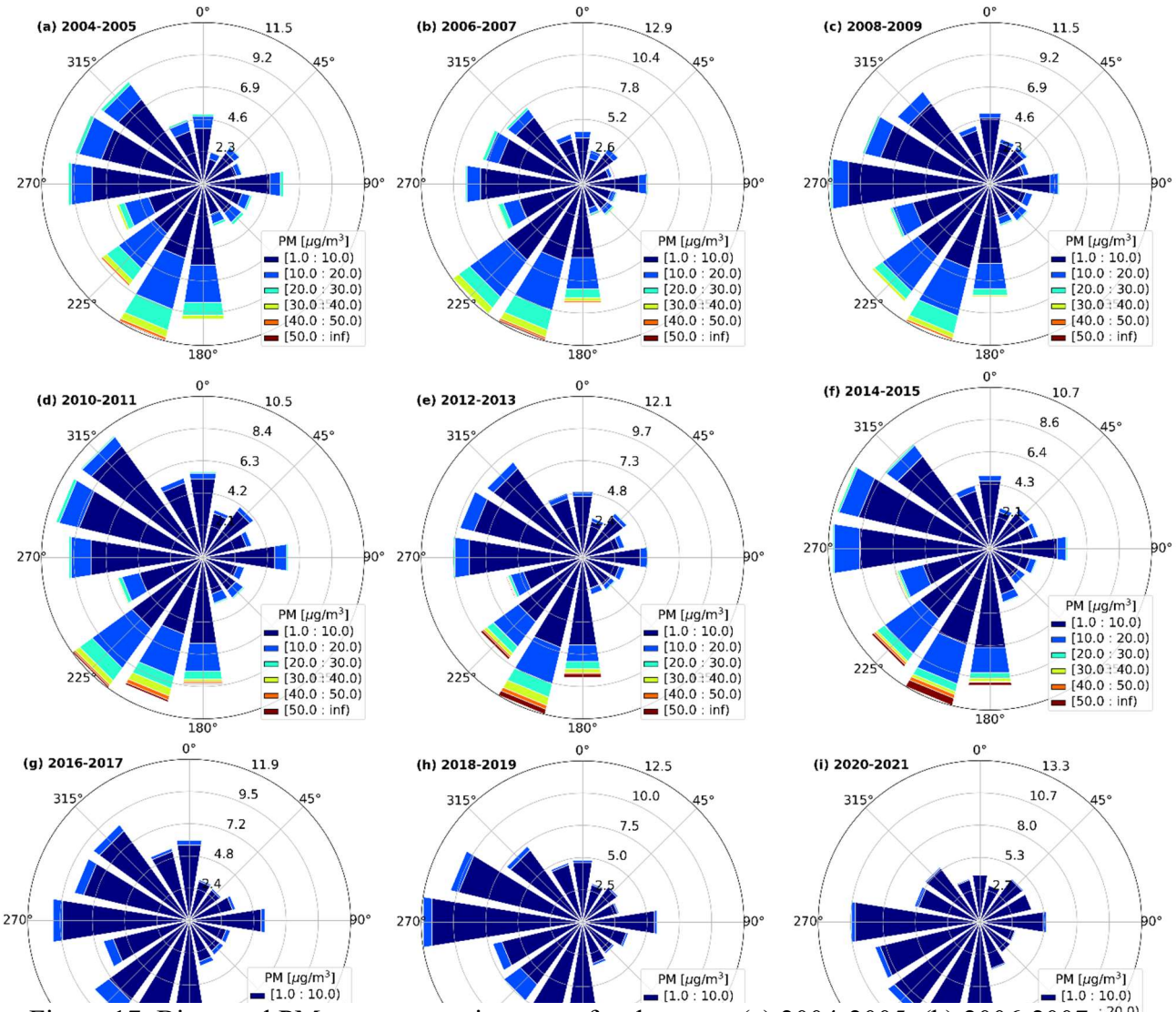

Figure 17. Bi-annual $PM_{2.5}$ concentration roses for the years (a) 2004-2005, (b) 2006-2007, (c) 2008-2009, (d) 2010-2011, (e) 2012-2013, (f) 2014-2015, (g) 2012016-2017, (h) 2018-2019, and (i) 2020-2021. Properties are same as Figure 16.

Between 2020 and 2021 the predominant WD was between SW and south with most of the concentrations below 10 $\mu g/m^3$, which can be considered the baseline $PM_{2.5}$ concentrations from all sources (excluding the KPM as it was closed). From the mean and maximum annual concentrations, it is evident that since 2016 $PM_{2.5}$ concentrations decreased in comparison to previous years (concurrent with the year the pulp mill installed the precipitator to reduce PM concentrations and that the MTF reduced its capacity to 50%). The STD decreased as well by at least two-fold, indicating a lower spread of $PM_{2.5}$ concentrations since 2016.

4.5.2 High $PM_{2.5}$ concentrations events

Twenty hourly and daily highest maximums of $PM_{2.5}$ concentrations between 2003 and 2021 were identified (Table 11). All of the 20 highest maximums (both hourly and daily) occurred before 2016. Ten of the hourly maximums occurred between April and May, and six of the daily maximums occurred in April or May. The highest $PM_{2.5}$ concentrations events (identified as the dates when maximum occurred both on a daily and hourly basis) resulted in five events identified using different colours in Table 11. All maximum $PM_{2.5}$ concentration events occurred in April or May of 2014 or 2015 and the WD was between 200-220° (between south and west SW).

For all the five major events the trajectories show that air mass origins were from the SW and SSW (Figure 18). In the western zone of Nova Scotia, there was only one station, Aylesford, that measured $PM_{2.5}$ in 2014 and 2015; and two stations in the southern zone, Halifax Johnston and Lake Major. **Error! Reference source not found.** shows how those three stations, compare with the Pictou station for high $PM_{2.5}$ concentration events during April and May in 2014 and 2015, respectively. All major events were recorded in Pictou (**Error! Reference source not found.**). The other stations did not record any peak events during the entire period. $PM_{2.5}$ concentrations at Aylesford, Halifax Johnston, and Lake Major were always <25 $\mu g/m^3$ (except for one observation at Halifax Johnston when there was a peak of 62.8 $\mu g/m^3$), while Pictou was often above that concentration. Following the criteria defined by Mitchell et al. (2021), regional pollution transport could be considered when several NAPS stations simultaneously record positive anomalies. In this case, the lack of high $PM_{2.5}$ concentrations measured at stations where the air mass is coming from

indicates that the likely source of $PM_{2.5}$ emissions should have been local and not due to long-range transport.

Table 11. Identification of the 20 highest $PM_{2.5}$ concentrations in Pictou on an hourly and daily basis. The different colours indicate different peak events. When a date was identified with a high $PM_{2.5}$ concentration both hourly and daily the date was identified as a peak event (and it corresponds with the coloured events in Figure 18 and **Error! Reference source not found.**).

	20 highest $PM_{2.5}$ hourly maximums					20 highest $PM_{2.5}$ daily maximums			
	$PM_{2.5}$ [µg/m^3]	WD [°]	WS [km/h]	Date	Hour	$PM_{2.5}$ [µg/m^3]	WD [°]	WS [km/h]	Date
1	178.0	270	30	2013/11/25	19	43.1	213	20.3	2010/9/3
2	171.0	220	18	2015/5/4	23	39.2	ND[1]	ND[1]	2003/8/22
3	154.0	210	24	2014/4/11	15	39.2	207	20.7	2010/7/29
4	152.9	210	22	2012/7/13	4	38.4	ND[1]	ND[1]	2003/6/28
5	145.0	150	13	2015/4/17	0	38.4	263	14.2	2010/9/2
6	143.0	220	20	2015/5/27	23	36.9	202	25.7	2014/5/10
7	142.2	120	44	2009/8/30	7	36.5	211	18.0	2014/4/11
8	132.0	200	27	2014/5/10	0	34.4	215	9.2	2015/5/27
9	132.0	220	9	2014/8/26	23	33.3	211	105	2013/11/1
10	130.1	320	22	2009/5/26	1	32.2	217	19.7	2010/9/29
11	128.0	210	22	2014/4/11	14	31.9	195	17.5	2012/5/25
12	128.0	200	6	2014/7/20	9	31.8	205	16.8	2004/8/21
13	126.8	220	13	2010/7/10	0	31.2	231	12.1	2004/6/9
14	123.8	230	24	2012/3/8	23	31.2	213	16.7	2005/7/27
15	123.6	210	19	2012/7/24	0	30.7	206	11.9	2007/9/8
16	121.0	210	14	2014/5/15	20	30.3	ND[1]	ND[1]	2003/6/27
17	121.0	200	29	2015/4/14	9	30.3	172	8.0	2015/4/13
18	117.0	170	23	2014/7/14	13	30.2	149	2.5	2010/7/7
19	117.0	200	17	2015/4/13	22	29.8	167	4.0	2010/9/1
20	116.0	130	9	2014/5/16	19	29.6	202	4.1	2015/5/4

[1] ND indicates No Data available.

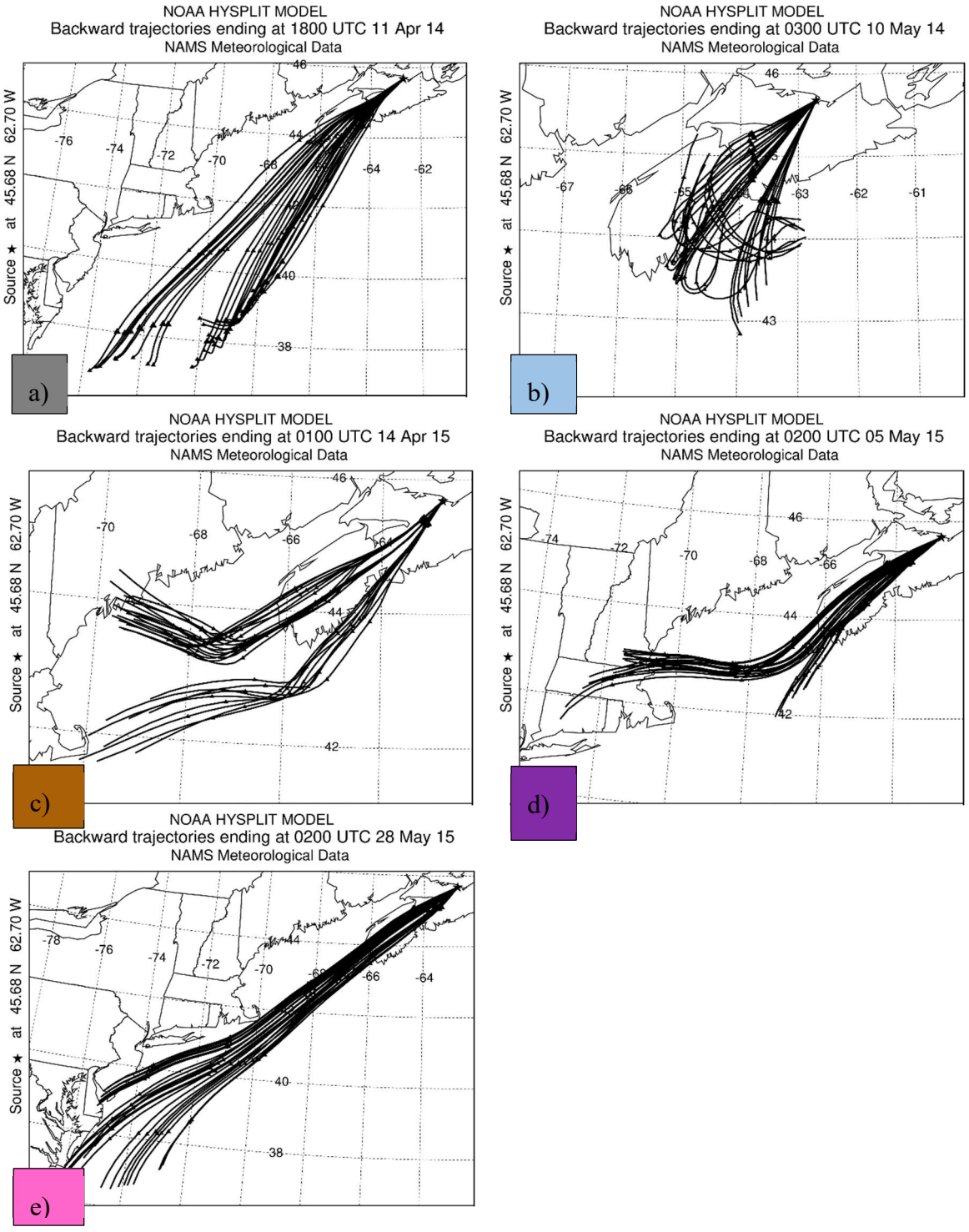

Figure 18. HYSPLIT 24 hours backward trajectories for the dates identified in Table 4 ending in Pictou station on a) 2014/04/11 18 UTC; b) 2014/05/10 3UTC; c) 2015/04/14 1UTC; d) 2015/05/05 3UTC; e) 2015/05/28 2UTC. The peak events are marked in corresponding colours with **Error! Reference source not found.** and Table 11.

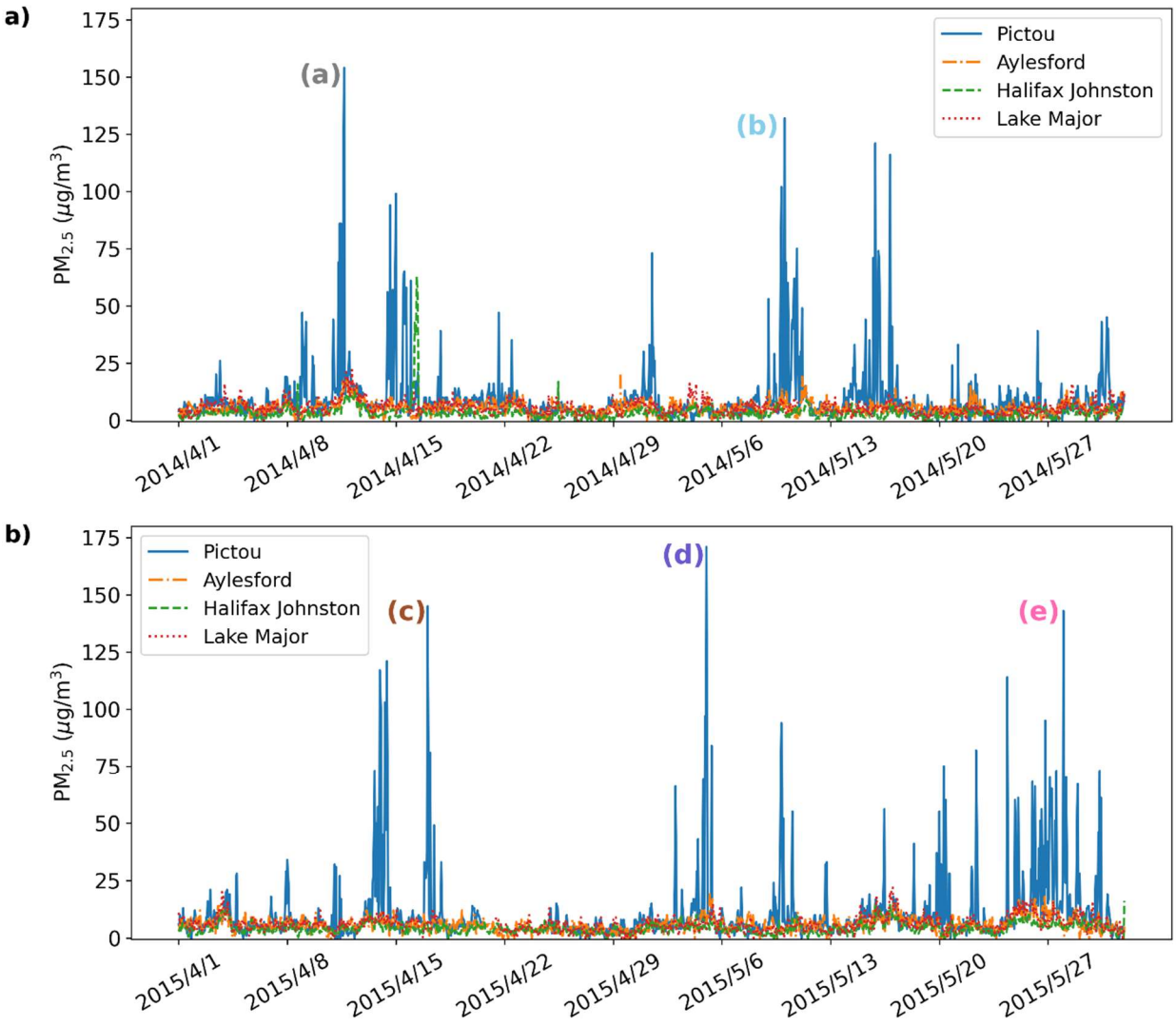

Figure 19. Hourly $PM_{2.5}$ concentrations measured in Nova Scotia NAPS stations during April and May in a) 2014 and b) 2015. The peak events are marked in corresponding colours with Table 11 and Figure 18.

4.5.3 Local Sources

NPRI is the only inventory that reports industrial releases and is publicly available, so the comparison among the different industries could only be implemented with annual releases. The annual $PM_{2.5}$ release from the TMF, the CFPGS and the KPM presented in Figure 20(panels b, d, f) show that the KPM releases between 2011 and 2020 were higher than the TMF and the CFPGS releases, except in 2017 and 2020 when the CFPGS releases were the highest and the KPM and TMF releases were of similar magnitude. The TMF and

the CFPGS reduced their releases after 2013 at least two-fold. In 2014 and 2015, the KPM releases (1291 tpy and 823 tpy) were three and two orders of magnitude higher than the TMF (5 tpy and 0.2 tpy) and the CFPGS (60 tpy and 46 tpy) releases, respectively. KPM releases were substantially reduced after 2016, from 1290 tpy in 2014 to 109 tpy in 2016 and 1.6 tpy in 2017.

For the three NAPS comparisons (P50, P90 and 99) the KPM is the facility with the best linear fit and correlation (Table 12). KPM presents a ρ and R^2 closer to 1 ($0.80 \geq R^2 \leq 0.86$, $0.89 \geq \rho \leq 0.93$) than the TMF and the CFPGS (for TMF, $0.43 \geq R^2 < 0.71$ and $0.66 \geq \rho \leq 0.84$; for the CFGTS, $0.49 \geq R^2 < 0.77$ and $0.70 \geq \rho \leq 0.88$). The RMSE and the MAE for the linear regression on the KPM were the smallest among all the facilities by nearly half. The p-value shows that the null hypothesis of similar populations is rejected for the MTF and NAPS P90 and P99, and the CFPGS and NAPS P50 and P90 at a confidence level of 0.01. For a confidence level of 0.05, the similarity between the KPM and the three NAPS percentiles, the MFT and NAPS P50 and the CFPGS and P99 is rejected

Table 12. Statistics for annual mean, P75, P90 and P95 $PM_{2.5}$ concentrations from Pictou NAPS station and NPRI annual releases from the TMF, CFPGS, and KPM.

	Manufacturing Tire			Coal-fired power generating station			Kraft pulp mill		
	P50	P90	P99	P50	P90	P99	P50	P90	P99
ρ	0.66	0.84	0.66	0.70	0.88	0.75	0.91	0.89	0.93
p-value	0.21	<0.01	<0.01	<0.01	<0.01	0.27	0.03	0.03	0.05
R^2	0.44	0.71	0.43	0.49	0.77	0.56	0.83	0.80	0.86
MAE	0.67	2.90	14.84	0.65	2.35	12.54	0.37	1.90	6.64
RMSE	0.82	3.48	18.07	0.79	3.11	15.92	0.46	2.92	9.12

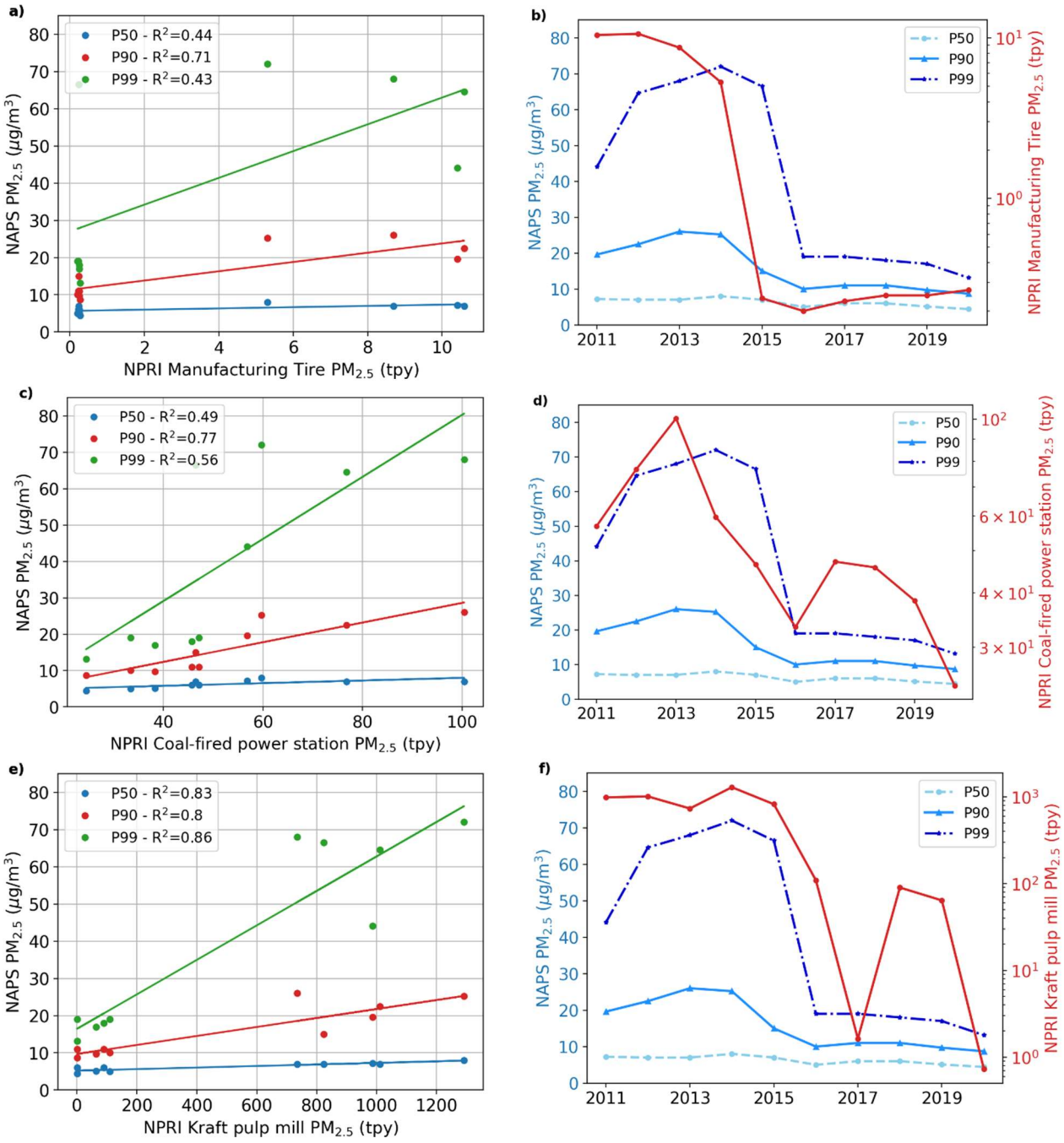

Figure 20. Linear regressions and R^2 between P50, P90 and P99 $PM_{2.5}$ concentrations from Pictou NAPS station and NPRI annual releases from a) the manufacturing tire, c) the coal-fired power generating station and e) the kraft pulp mill. Annual trend of P50, P90 and P99 PM2.5 concentrations from Pictou NAPS station and NPRI annual releases in logarithmic scale from b) TMF, d) CFPGS and f) KPM.

4.6 DISCUSSION

The objectives of this study were to analyze $PM_{2.5}$ concentrations recorded in Pictou NAPS station, and; to compare them with $PM_{2.5}$ annual NPRI releases from three industrial facilities in Pictou. Results suggest that between 2016 to 2021, the mean hourly $PM_{2.5}$ concentrations were reduced by 35% in comparison to the period between 2002-2015. The maximum hourly concentration for the 19-year period, recorded in 2013, was 178 $\mu g/m^3$. Hourly and daily $PM_{2.5}$ concentrations recorded in the air quality station in Pictou were compared with the wind direction recorded at the time of the highest concentrations and with other $PM_{2.5}$ air quality stations to analyze the potential emission sources of $PM_{2.5}$ recorded in Pictou. The evaluation suggested that during high concentration events, the prevailing winds were from the SSW, the direction downwind of three local industrial facilities; and that the main emission source is unlikely to be transboundary transport. Annual $PM_{2.5}$ releases from the KPM were higher than the annual releases from the TMF and the CFPGS.

4.6.1 $PM_{2.5}$ source identification

This study, apart from comparing annual means, also compared high emissions (identified as P90 and P99) under the assumption that peaks of $PM_{2.5}$ in Pictou were more likely to occur because of an industrial release rather than long-range transboundary transport. This assumption was based on the HYSPLIT model and $PM_{2.5}$ concentrations measured at four NAPS stations in Nova Scotia. The comparison revealed that when high $PM_{2.5}$ concentrations occurred in Pictou, the other air quality stations did not record the $PM_{2.5}$ peak. Previous studies have made quantitative comparisons between mean annual NAPS concentrations and annual self-reported releases for pollutants like nitrogen oxides, sulphur dioxide, VOCs, metal and metalloids (Galarneau et al. 2016; Mitchell et al. 2021; Walker 2018). While Mitchell et al. (2021) found similarities between annual trends of VOCs from NAPS and NPRI annual releases, Walker (2018) found no correlation between sulphur dioxides and nitrogen oxides NAPS concentrations and emissions from industrial sources.

Consistent with previous studies (Hoffman et al. 2015; Hoffman, Guernsey, et al. 2017), the tracking of $PM_{2.5}$ concentrations reveals that during high concentration events, the

prevailing winds were from the SW, SSW, and S, where the KPM, the CFPGS and the TMF are located. Based only on wind direction, it was difficult to determine the relationship between each facility and the $PM_{2.5}$ concentrations recorded at Pictou, as there are other sources (such as vehicular traffic, construction, and wood fires) that also contribute to $PM_{2.5}$ emissions. Thus, to evaluate the contribution from each facility, $PM_{2.5}$ NPRI annual releases were compared to P50, P90 and P99 of $PM_{2.5}$ concentrations (as the relationship between the variables could differ for annual median or high concentrations). This comparison between NPRI releases and NAPS percentiles suggests that the highest $PM_{2.5}$ concentrations could have been mostly from the KPM; the TMF and CFPGS releases were negligible compared to the KPM releases. Before 2016, NPRI releases from the KPM were one and two orders of magnitude higher than the NPRI releases from the TMF and the CFPGS, respectively. Eight out of the 20 hourly maximum $PM_{2.5}$ peaks between 2003 to 2020 occurred in 2014 (Table 11). That same year, $PM_{2.5}$ releases from the KPM, CFPGS and TMF were 1290.83, 59.7 and 5.30 tonnes, respectively, suggesting that the KPM could be a major source of high $PM_{2.5}$ concentrations in Pictou. The linear regressions and correlations between P99 and the NPRI releases (R^2=0.86, ρ=0.93 for KPM; R^2<0.43, ρ=0.66 for TMF; and R^2<0.46, ρ=0.75 for CFPGS) suggest that major peak events occurred predominantly due to emissions from the KPM. This was also corroborated when NAPS concentrations and NPRI releases from the KPM declined in 2016 following the precipitator installation. Pollutant roses revealed that hourly $PM_{2.5}$ concentrations before 2016 sometimes exceeded 50 $\mu g/m^3$ (and always recorded from the south, SSW, and SW). On the other hand, since 2016 hourly $PM_{2.5}$ concentrations never surpassed 20 $\mu g/m^3$ and the annual CAAQS management level has been green (

Table 10).

The major pollutants emitted from a coal plant are sulphur dioxide and nitrogen oxides, which form PM and VOCs, (Kelly and Fussell 2015). For example, in 2016, SO_2, NO_x and $PM_{2.5}$ emissions from coal-fired power generating stations accounted for around 25%, 10%, and 4.5%, respectively of the total global emissions (International Energy Agency 2016). Thus, it is possible that $PM_{2.5}$ is not the main air pollutant emitted from CFPGS, or that the concentrations are not fully captured due to the prevailing winds not coming from

the south. Also, after the complete refurbishment of CFPGS in 2009, $PM_{2.5}$ concentrations measured in Pictou remained relatively stable, so presumably, this upgrade did not affect $PM_{2.5}$ releases from the CFPGS. Alternatively, a reduction in the annual releases from the TMF is evident after the facility reduced its personnel in 2014 and 2015, although these releases are quite small in comparison to emissions from KPM. Therefore, although both facilities reduced their annual releases concurrently (5.3 to 0.2 tonnes in the MTF and 1291 to 110 tonnes in the KPM from 2014 to 2016, respectively) it is unlikely that the personnel reduction in the TMF could have affected the $PM_{2.5}$ concentrations as the installation of the precipitator in the KPM did. It is possible that the main source of $PM_{2.5}$ emissions for TMF is transport, as chemically and physically unstable materials must be in continuous movement from one plant to the other (Langille 1981). Also, there could be other secondary sources not considered in this study, such as local transportation or residential wood and coal burning. Between 2020 and 2021, after the KPM closure and during the COVID lockdown (which happened almost at the same time), the hourly concentrations remained below 20 $\mu g/m^3$ and the prevailing winds were from the SW. It could mean that the pandemic did not affect the air quality in the region (likely because there are towns and no major urban centres in the county) and that the baseline emissions remain approximately constant. Similar to the studies establishing a baseline in water pollution before BH remediation (Chaudhary et al. 2020; Quanz et al. 2021; Romo et al. 2019) the $PM_{2.5}$ concentration on Pictou could be considered the baseline for secondary sources of $PM_{2.5}$ when the KPM is not in operation (with the caveat that the COVID pandemic happened at the same time).

4.6.2 Management and monitoring

The NAPS network covers different regions of Nova Scotia, although not all the stations monitor the same pollutants. This could pose a challenge when trying to characterize and understand different sources of pollutants in the region, or to separate transboundary pollution from local sources (Buteau et al. 2018; Galarneau et al. 2016). Thus, it is important that WD and WS are measured along with the different pollutants, otherwise, it is not possible to identify where the main source of pollution may be located. For this study, 10 years of $PM_{2.5}$ concentrations could not be analyzed including the wind direction as the

pollutant monitoring began 10 years before the WD and WS monitoring. For example, between 2009 and 2012, there were elevated hourly and daily concentrations of $PM_{2.5}$ along with high peaks (Tables 3 and 4), although it was not possible to determine the WD associated with those events (and therefore to track possible sources of emission) as it was not being measured at that time. The NAPS stations which are closer to multiple industry sources, like Pictou, should measure WD and WS since their installation, along with all the air pollutants (for example, the province of Nova Scotia only monitors VOCs at one site).

4.6.3 Limitations and strengths of the study

This study provides an updated and long-term comparison (2003 to 2021) of $PM_{2.5}$ industrial releases and $PM_{2.5}$ measured concentration. This challenging comparison between two different databases, an annual self-reported release inventory in tonnes (NPRI) and hourly measurements recorded in $\mu g/m^3$, was presented by calculating different annual percentiles for the hourly measurements. The annual percentiles enabled the comparison with the annual NPRI releases, and also provided different alternatives to compare industrial releases to mean (P50) and maximum (P90, P99) $PM_{2.5}$ measured concentrations. This choice of this percentiles enables to assure that not only the average concentrations were considered but also to compare only the peaks emissions.

The comparison between both inventories has its limitations as the NAPS measurements include all the sources contributing to the $PM_{2.5}$ concentrations (not only from industrial facilities), which are not considered in this study. Those contributions, like vehicular traffic or wood burning, were considered to be consistent in time and therefore, not considered to add elevated annual variability to the measured concentrations. To more accurately understand how each facility contributes to the measured $PM_{2.5,}$ chemical speciation and source apportionment modelling could be implemented to identify the source types contributing to the high emissions. Another factor that influences the comparison between the NPRI and the NAPS is WD. The air quality station only records $PM_{2.5}$ emissions from the industrial facilities when the wind is between south and SW, so when the WD is blowing towards the industrial facilities (even if the emissions are high) the measured concentrations in the station are not going to reflect high $PM_{2.5}$ concentration. Also, due to the lack of WD and WS for the period of study in the same location where the $PM_{2.5}$ is

measured, wind data were retrieved from the closest automatic meteorological station (10 km away from the air quality station). Both stations are very close to each other, so it is expected that the analysis presented here accurately reflects the observed conditions in Pictou.

The initial aim of this study was to consider as a "natural experiment" the closure of the KPM to analyze $PM_{2.5}$ concentrations in Pictou when the mill was not operating. The analysis would consider that all the other sources (like wood burning, vehicle emission, and construction) contributions were consistent on time. As the COVID global pandemic occurred at the same time, it could no longer be assumed that all the other contributions remained exactly unchanged in that period. Still, the concentration for the period of 2020-2021 could be considered a baseline concentration as there are no big cities nor a large population in Pictou town that could have substantially impacted the measured $PM_{2.5}$ concentrations.

4.7 CONCLUSION

$PM_{2.5}$ in Pictou, Nova Scotia was analyzed between 2003 and 2020 to identify peaks of $PM_{2.5}$ concentrations and track air mass trajectories during those events to identify potential sources of emissions. Three industrial facilities were considered as possible sources of $PM_{2.5}$ concentrations. Although previous studies have revealed high $PM_{2.5}$ concentrations in Pictou and implicated KPM as a major point source emitter, there has been no assessment of other industries in the region to identify potential sources of $PM_{2.5}$ concentrations. This study demonstrates that during high $PM_{2.5}$ concentration events the prevailing winds are downwind of the industrial facilities. Self-reported annual releases of $PM_{2.5}$ by the three facilities suggest that KPM was the primary source for high $PM_{2.5}$ concentrations, as KPM releases are 10 and 100 times higher than CFPGS and TMF releases, and also KPM NPRI releases have the best linear fit with NAPS P99. Also, $PM_{2.5}$ concentrations and KPM releases were substantially reduced since 2016 following the installation of a recovery boiler precipitator on KPM.

CHAPTER 5 CONCLUSION AND FINAL COMMENTS

This research aimed to i) investigate the effectiveness of environmental mitigation strategies to improve air quality by emissions from P&P facilities in Atlantic Canada, with a special focus on the pulp facility in Pictou, NS; and ii) to analyze how air pollutant emissions from the different facilities compare among each other and how they comply with regulations. Both objectives were achieved by comparing annual self-reported releases and air quality measurements from publicly available data from ECCC and NSE. This chapter provides an overview of the main findings, provides management recommendations, discusses future research that could continue from this study, and presents a personal reflection derived from the research.

5.1 Main findings

CHAPTER 3 compared annual air releases from the nine P&P facilities in Atlantic Canada with the lower reporting threshold established by the NPRI guidelines and with the recommended air emissions limits for P&P facilities from ECCC. The air pollutants analyzed were CO, NO_x, TPM, PM_{10}, $PM_{2.5}$, SO_2 and VOCs for the period 2002-2019. This comparison revealed that all the facilities' releases were several orders of magnitude above the lower NPRI reporting thresholds. When a facility exceeds this lower reporting thresholds, it must report the annual release to the NPRI. The highest emissions were TPM from the pulp mill in Pictou, NS (currently owned by Paper Excellence and named NP), for TPM, PM_{10} and $PM_{2.5}$. Until 2015, TPM releases were > 1000 tpy (and some years > 2000 tpy) while the other mills' releases were around 300 tpy. After 2015 (when the electrostatic precipitator was installed in NP to reduce PM emissions), NP's TPM was still higher than the rest of the facilities (around 400 tpy) but of similar magnitude. Between 2002 to 2019, NP's $PM_{2.5}$ mean releases were 994.4 tonnes, while the mean releases for all the mills were 174.1 tonnes. The recommended air emissions limits for the P&P industry for SO_2 and $PM_{2.5}$ emissions were not always followed. In particular, the pulp mill in Pictou reported $PM_{2.5}$ releases above the recommended limit before 2015.

CHAPTER 4 focused on $PM_{2.5}$ concentrations recorded in Pictou, NS, and $PM_{2.5}$ emissions from three local industrial facilities, the kraft pulp mill, a tire manufacturing facility and a coal-fired power generating station. The main objective of this part of the research was to understand how each of the three facilities contributed to the $PM_{2.5}$ concentrations recorded in Pictou, especially when high emissions were recorded; and to evaluate if the high emissions could have been a result of transboundary transport. When high $PM_{2.5}$ concentrations were recorded, the prevailing winds came generally between the south and the southwest, the direction where the three facilities are located. A comparison with $PM_{2.5}$ recorded on the air quality stations in the province of NS downwind the prevailing winds during the peak emissions suggested that the high emissions events were not caused by transboundary transport of $PM_{2.5}$, but from a local source in PC. All the high concentrations events occurred before 2016. The comparison with air releases from the three facilities suggests that the kraft pulp mill, now NP, is the facility that contributed the most to the $PM_{2.5}$ high emissions.

The specific purpose of this research was to answer three questions, which are discussed following.

1) Did the precipitator installed at NP in 2015 effectively remove the intended atmospheric pollutants?

The research revealed that after 2015, TPM, PM_{10} and $PM_{2.5}$ annual NPRI releases from NP substantially decreased. In 2014, $PM_{2.5}$ releases reached 1290 tpy, while two and three years later they were reduced to 109 tpy and 1.6 tpy, respectively. This decrease in the particulate emissions could only be explained by the installation of the precipitator, as it was the only technological upgrade in the mill aiming to reduce those emissions.

2) How was air emission compliance regulation in NP in comparison to similar pulp and/or paper mills in the region?

Most of the P&P mills in Atlantic Canada emit air pollutants in a similar quantity, except for TPM, PM_{10} and $PM_{2.5}$ from NP; these emissions were above 1000 tpy (in some years even above 2000 tonnes) until 2015, while the rest of the facilities released were around

300 tpy. In NS, industrial emissions sources are regulated based on each facility's Industrial Approval, where limits on air emissions of various pollutants are determined on a case-by-case basis. However, no federal or provincial enforced air emission limit that applies to all industrial sources, only the code of best practices for the P&P industry (published in 2017) that recommends annual emission limits for SO_2 and $PM_{2.5}$. For SO_2, some facilities emitted above the suggested threshold until 2008, after that the emissions on all the facilities were above or close to the suggested limit. For the $PM_{2.5}$, all the facilities emitted below the suggested limit, except NP that before 2016 two-folded and three-folded the suggested limit.

3) How much did NP contribute to the PM monitored in the Pictou air quality station?

Since 2016, $PM_{2.5}$ concentrations recorded in Pictou air quality station were reduced by ~35% in comparison to the previous years. The air quality management levels, which were yellow in the previous years, has become green since 2016. The most noticeable change in 2015, that could have influenced decrease in the $PM_{2.5}$ concentration, was the precipitator installation in NP. However, it is not straightforward to assume that NP's upgrade was the only explanation for the $PM_{2.5}$ substantial reduction after 2016, as there are two other local industrial facilities (the TMF and the CFPGS) plus there are other contributions to particulates formation such as wood burning or transport. Regardless, when only the three industrial facilities are considered, releases from the KPM before 2016 were 100 and 10 times higher than the TMF and the CFPGS, respectively. Since 2014, $PM_{2.5}$ releases from the MTF and the CFPGS never surpassed 6 tpy and 60 tpy, respectively; while the KPM releases decreased progressively from 1290, 823 and 109 tpy in 2014, 2015 and 2016, respectively. This suggests that the precipitator installation could partly explain the decrease in the $PM_{2.5}$ concentrations in measured Pictou and that the high $PM_{2.5}$ concentrations measured before 2016 could have been a result of NP releases.

5.2 Management implications and recommendations

Canada a monitoring programs, NAPS, and two inventories, the APEI and the NPRI, which are publicly available and relatively up to date. These features make the databases very valuable. This research used data from the NPRI and the NAPS program. While the former

is an inventory of annual releases self-reported by the facilities that emit above a specific reporting threshold, the latter is a database of air quality monitoring station concentration measurements. This section discusses the advantages and limitations of both inventories and makes recommendations to improve the usefulness of the inventories.

The NPRI requires that facilities report different air pollutants (along with land and effluent releases). This inventory is a powerful tool that gathers industrial annual releases from facilities in Canada, and it is publicly available. However, some of the limitations of this inventory include the reporting threshold for each pollutant, which is not clear how it is established. The reporting threshold only considers mass releases without considering toxicity, and it is only a lower reporting threshold. Some disincentive tools that control the quantities of pollutants released into the atmosphere could help to protect human and environmental health from harmful industrial releases. For example, imposing large fines when elevated quantities are released and air quality is affected promotes technology upgrades rather than paying the fine.

The NAPS database is a useful tool that provides air quality data in real-time (for the province of NS through the NSE portal). Monitoring stations are distributed across Canada and monitor several pollutants and, in some cases, meteorological variables. These stations record hourly measurements, which is valuable when comparing emissions in different locations. However, as not all the same pollutants are recorded on all stations (as it may not always be necessary), it may be challenging to identify potential sources of emission or transboundary transport of pollution when only a couple of stations recorded the pollutants. For example, VOCs are recorded in only one station in NS, so if high concentrations are detected, it would not be possible to know if high concentrations were also detected in other parts of the province. So, harmful pollutants, such as VOCs, could be monitored in more than one location to improve the spatial coverage where potential high concentrations could occur. Another limitation of the NAPS program is that not all the stations started to record wind speed and wind direction (which are essential for air quality monitoring) at the same time as air pollutants. In some cases, like in Pictou station, the wind direction and speed began to be recorded in 2020, although the air pollutants' monitoring started almost 20 years before. So, to combine air quality data with wind data

it is necessary to use other sources such as a meteorological station. In the case of Pictou, the closest automatic meteorological station is close (9 km away from the NAPS station) to use its wind speed and direction.

5.3 Future research

The first part of this study compared air emissions from the P&P sector in Atlantic Canada. This study included only ~10% of the total P&P facilities in Canada. Further research could include a comparison between the rest of the mills in the country, to obtain a more comprehensive comparison between pulp, paper, and pulp and paper. This research suggested that pulp mills emit more air pollutants in comparison to paper mills. To understand if this is a characteristic of the sector (and not a localized finding for the mills in NS, NB and NL), a comparison of air emission from all the pulp mills versus all the paper mills, for example, could be conducted. In addition, a qualitative study through a literature review could be helpful to compare how the kraft pulp mills' air emissions are in the rest of the world. While P&P mills in Atlantic Canada have been operating for many decades, this industry recently began operating in South America. It could be interesting to evaluate how air emissions from new facilities (for similar annual production) compared to air emissions from old facilities with little equipment upgrade to identify if the installation of new technology is a critical factor when aiming to reduce air emissions.

The second part of this research focused on $PM_{2.5}$ from three industrial facilities in Pictou, including the kraft pulp mill, as this pollutant from this facility was the highest release identified in the previous analysis. This study could be extended to compare $PM_{2.5}$ concentrations from all the NAPS stations in the rest of the province. The preliminary comparison during the months of the highest $PM_{2.5}$ with the other three stations showed that Pictou was the only station to record those peak events. A study analyzing the $PM_{2.5}$ concentrations in the rest of the province could help to understand if the magnitude of the high events recorded on Pictou was also registered in other locations (although probably on different dates as they may have different local sources contributing). Also, it could be worth analyzing hourly measurements for the rest of the air pollutants to investigate how each facility contributes to the concentrations recorded in Pictou. This research suggested

that the pulp mill was the main contributor to the high $PM_{2.5}$ concentrations recorded in Pictou; although it could be that for other pollutants, such as SO_2 and NO_x, the coal-fired power generating station was the most contributing. It is known that fossil fuel burning emits significant amounts of SO_2 and NO_x. So, those hourly air pollutants recorded in Pictou could be compared with the NPRI releases from each of the three industrial facilities. Also, chemical speciation and source appointment modelling could contribute to identifying the main sources contributing to the high $PM_{2.5}$ concentrations recorded in Pictou.

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Appendix A Supplementary Material

Appendix A.1. Pulp Mills

Northern Pulp

In New Glasgow, NS, a Kraft pulp mill operated from 1967 to 2020. The closure occurred on January 2020 due to a lack of Ministerial approval of the Environmental Assessment for a proposed new effluent treatment facility (NSE 2021). The mill operator claimed that they did not have enough time to compile an adequate environmental assessment prior to the closure, as the measures they initially proposed were not accepted by the provincial government (Baarda 2020; Spicer 2019). Scott Paper first owned and operated the mill in 1967. In 1995, it was acquired for Kimberley-Clark and nine years later for Neenah Paper. In 2008, the mill was acquired by Northern Resources and was re-named Northern Pulp. Finally, it has been a property of Paper Excellence since 2011 [59,60]. Even though technology advances very quickly, the basis in elaborating paper have not changed significantly in the last century (Bernard et al. 2020). Before its closure, NP employed 330 fulltime workers and around 2000 indirect jobs across NS (Paper Excellence 2021). Annually, it produced 280,000 tonnes of Bleached softwood Kraft. On the north shore of the Pictou Harbour and 1 km away, NP is Pictou, a town of 3107 inhabitants (Statistics Canada 2022). Ten kilometres south of NP is New Glasgow, a town of 9,471 people (Statistics Canada 2022).

Irving Pulp & Paper Limited

Irving Pulp & Paper, a kraft pulp mill in Saint John, NB, is managed by Irving Ltd. (Irving Pulp and Paper 2016). The products are bleached softwood and hardwood kraft for premium tissue products. The mill opened in 1851 as The Cushing Sulphite Fibre Company, and in 1910, it became the Edward Partington Pulp and Paper Company. In 1932, the mill became Port Royal Pulp. Finally, 14 years later, in 1946, K.C. Irving purchased the mill, making some modernizations to increase the capacity of production (Irving Pulp and Paper 2016). The mill employs more than 330 workers and produces 335,000 tonnes per year. It is located near the Bay of Fundy within the city of Saint John of 69,895 inhabitants (Statistics Canada 2022).

Twin Rivers

The pulp mill located in Edmundston, NB, built in 1916, has operated for more than 100 years. Twin Rivers was acquired in 2013 by Atlas Holdings and Blue Wolf Capital Partners. Apart from the pulp facility, the company owns a lumber mill in NB and three paper mills in the United States. The pulp mill in Edmundston produces bleached softwood sulphite and bleached groundwood pulp as well as 45 MK of biomass cogeneration of electricity sold to New Brunswick Power (Twin Rivers Paper Company 2021). Its production capacity is 370,000 tonnes per year and employs more than 280 workers. The mill is located in the shore of the St John River in the city of Edmundston, of 16,437inhabitants (Statistics Canada 2022). Across the river and the United States–Canada border, a paper mill also operated by Twin Rivers company is installed in Madawaska, a city with a population estimated at 3735 for 2019 (United States Census Bureau 2019).

Nackawic and Atholville

Both pulp mills have operated in NB for around 19 years and are owned by AV Group NB, a company established in 1977. The cellulose pulp produced in both mills is used in the manufacture of textile. The final products are home and apparel textile made of viscose staple fibre as well as non-woven textiles (AV Group 2021b). The company manages 1.6 million acres of company-owned and Crown land. In total, they employed around 1200 people directly and many more indirectly (AV Group 2021a). The Nackawic mill is located at northeast in the town of Nackawic, which has a population of 962 (Statistics Canada 2022). The Atholville mill is on the shore of Atholville village, within the town of Campbellton with a population of 7047 (Statistics Canada 2022).

Appendix A.2. Paper Mills

Port Hawkesbury Paper

Initiated in 1962 in NS as a sulphate pulp mill owned by Stora, it was sold to NewPage in 2007. After its closure four years later, it reopened as the property of Stern Partners and Wayne Nystrom as a paper mill in 2012 (Boudrot 2020; CBC News 2011). Located 2 km south of Port Hawkesbury, a town of 3,210inhabitants (Statistics Canada

2022), it employs around 300 people directly and near 400 as contractors across the province and manages around 523,000 ha of public land. The mill can produce 400,000 tonnes of paper annually to be used in catalogs, magazines, retail inserts, and wrapping paper (Port Hawkesbury Paper 2018).

Lake Utopia Paper and Irving Pulp and Paper

Both paper mills are operated by Irving Ltd. Lake Utopia Paper in NB produces high-quality corrugated medium since 1972. Their product comprises around two-thirds of virgin hardwood fibre and one-third of recycled cardboard (J.D. Irving Limited 2016). The final consumers for box packaging are food, beverage, agriculture, and electronic sector. Its annual capacity is 185,000 tonnes, and it employs around 140 workers directly. They designed a biomass boiler project and a new effluent treatment plant to improve environmental impact due to emissions (Lake Utopia Paper 2021). Irving paper produces specialty grade papers for flyers and magazines. They also supply rotogravure for commercial printing (Irving Paper 2021). Irving paper can produce 420,000 tonnes per year and has about 310 employees. Only 5 km away from Irving Paper, Irving Pulp and Paper is also located within the city near the shore of the Bay of Fundy, but on the west side of the St John River. Lake Utopia Paper is around 65 km east from Saint John city. The nearest towns are Pennfield, of 2222 inhabitants, around 5 km south of the mill and Saint George with a population of 2495 and around 7 km southeast of the mill (Statistics Canada 2022).

Appendix A.3. Pulp and Paper Mill

Corner Brook Pulp & Paper Limited

In NL, paper began to be produced almost a century ago in 1923. After a couple of owners at the end of 1984, the Kruger organization acquired the mill, naming it as Corner Brook (Kruger Industrial 2021). The company manages 1.4 ha of forest land but only half of it is a productive forest. They are constantly performing surveillance audits, have made some upgrades in the technology, and have a detailed environmental and commitment plan. Around 1750 workers are employed in the woodland department (Corner Brook Pulp and Paper Ltd 2019). The mill is located in the north coast of the city Corner Brook, a city of 19,333 inhabitants (Statistics Canada 2022).

Appendix B Copyright Agreement Letters for Co-Authorship

September 16, 2022

Codey Barnett
Nova Scotia Department of Environment and Climate Change
Halifax, NS, B3J 2P8, Canada

I am preparing my Master in Environmental Studies (MES) thesis for submission to the Faculty of Graduate Studies at Dalhousie University, Halifax, Nova Scotia, Canada. I am seeking your permission to include a manuscript version of the following paper(s) as a chapter in the thesis:

Characterization of Annual Air Emissions Reported by Pulp and Paper Mills in Atlantic Canada
Gianina Giacosa, Codey Barnett, Daniel G. Rainham and Tony R. Walker
Published in Pollutants, 2022, 2 (2), 135-155.

Identifying Sources of Industrial Emissions of Fine Particulate Matter ($PM_{2.5}$) in Nova Scotia, Canada
Gianina Giacosa, Codey Barnett, Daniel G. Rainham and Tony R. Walker

Canadian graduate theses are collected and stored online by the Library and Archives of Canada. I am also seeking your permission for the material described above to be stored online with the LAC. Further details about the LAC thesis program are available on the LAC website (www.bac-lac.gc.ca).

Full publication details and a copy of this permission letter will be included in the thesis.

Yours sincerely,

Gianina Giacosa

Permission is granted for:

a) the inclusion of the material described above in your thesis.

b) for the material described above to be included in the copy of your thesis that is sent to the Library and Archives of Canada (formerly National Library of Canada) for online storage.

Name: Codey Barnett Title: Adjunct (Scholar)

Signature: Codey Barnett Date: 2022-09-20

September 16, 2022

Dr. Daniel Rainham

School of Health and Human Performance

Dalhousie University

Halifax, NS, B3H 4R2, Canada

I am preparing my Master in Environmental Studies (MES) thesis for submission to the Faculty of Graduate Studies at Dalhousie University, Halifax, Nova Scotia, Canada. I am seeking your permission to include a manuscript version of the following paper(s) as a chapter in the thesis:

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Yours sincerely,

Gianina Giacosa

Permission is granted for:

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b) for the material described above to be included in the copy of your thesis that is sent to the Library and Archives of Canada (formerly National Library of Canada) for online storage.

Name:	Daniel Rainham	Title:	Professor
Signature:		Date:	September 20, 2022

September 16, 2022

Dr. Tony R. Walker

School for Resource and Environmental Studies

Dalhousie University

Halifax, NS, B3H 4R2, Canada

I am preparing my Master in Environmental Studies (MES) thesis for submission to the Faculty of Graduate Studies at Dalhousie University, Halifax, Nova Scotia, Canada. I am seeking your permission to include a manuscript version of the following paper(s) as a chapter in the thesis:

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Gianina Giacosa

Permission is granted for:

c) the inclusion of the material described above in your thesis.

d) for the material described above to be included in the copy of your thesis that is sent to the Library and Archives of Canada (formerly National Library of Canada) for online storage.

Name:	Tony Walker	Title:	Associate Professor
Signature:	T. Walker	Date:	September 20, 2022