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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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1. Introduction

Combustion of fossil fuels and biofuels during energy intensive production processes emits pollutants into the atmosphere [1], which threatens human and ecological health [2–5]. 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 [6]. In recent decades, stringent standards were developed around the world to protect human and environmental health [7,8]. 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 [9]. 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 [10–12]. 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 [13].

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) [14,15]. 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 [14,16]. 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 [17]. The creation of the NPRI program resulted in a decrease in overall emissions [15,18], although the toxicity of the releases did not decrease [15]. 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 1 for the period 2020 to 2021, must be reported to the NPRI program [19]. These thresholds are the lower limit trigger for reporting annual emissions; however, there are no regulations for exceedances.

Table 1. NPRI reporting thresholds for 2020 to 2021 (retrieved from [17]).

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

Desirable concentrations of ambient air pollutants, including fine particles ($PM_{2.5}$), ozone (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 $\mu g/m^3$ or ppb [20], 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 [5]. 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 [21,22]. 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) [23]. 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.

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 [6,24]. 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 [4,25]. Anthropogenic emissions, mainly from industrial activities and fossil fuel combustion, produce the majority of hazardous air pollutants [2,6,24–27]. Pulp and paper (P&P) industries emit large amounts of atmospheric pollutants and greenhouse gases [1,28], although there are few studies that quantify the spectrum of emissions [29]. Numerous studies have been conducted on water pollution impacts from wastewater effluent due to P&P mills [8,30–35], but there have been comparatively few studies to evaluate

the contribution of the industry to the concentrations of hazardous air emissions [36]. Air pollution from P&P mills is mainly caused by the production of various volatile sulfur compounds (from sulfite pulping process), odour emissions (from reduced sulfur), SO_2, and nitrogen oxides (NO_x) [37,38]. Stack emissions can also contain carbon monoxide (CO) [24]. Biomass combustion can increase the emissions of particulate matter (PM) and of gaseous and semi volatile organic compounds (VOCs) [24,29]. Exposure to these pollutants can affect human health, causing cancer or lung diseases [9,39,40], and may result in depression or psychological problems [2]. Exposure to particulates is associated with an increased risk of mortality from stroke, heart, or pulmonary disease, among others [6,41,42]. 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 [15,18].

In Canada, there are currently 102 pulp, paper, board, and/or tissue mills in operation or which have been temporarily closed [43]. 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 [44]. While managers of the facility report the best practices with respect to regulatory compliance [45], environmental monitoring revealed emission exceedances and locals remain concerned about impacts to human health [29,46–50]. Currently, Nova Scotia does not legislate requirements for emitters to assess human health risks from emissions [50]. 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 [50]. However, ambient air pollution concentrations in nearby communities were below the Canadian Council of Minister of the Environment guideline limits. Hoffman et al. [49] 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 [51] 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 [14,17].

2. Materials and Methods

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 1 shows the location of each facility in Atlantic Canada and the Appendix A presents background information about each one. Table 2 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 [52]. The sixth digit designates national industries (it is zero when there are no additional details) [52]. Pulp, paper, and paperboard mills are assigned the code 3221 [53]. A further classification including lower levels for those industries is presented in Table 2 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.

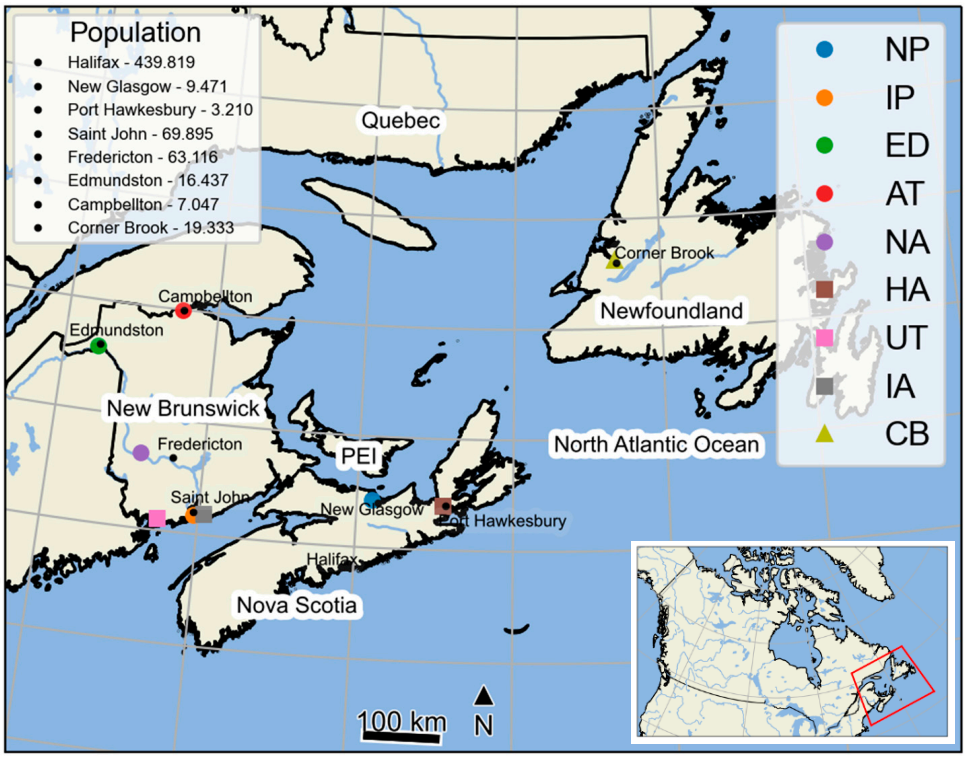

Figure 1. Location of mills and major populated centres. Pulp mills are represented with a circle, paper mills with a square, and the pulp and paper mill with a triangle.

Table 2. 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
Irving P & P Limited (−66.09°, 45.26°)	Saint John, NB	IP	PU	Bleached kraft pulp	335,000	335 [2]	2604	322112
Twin 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
Port Hawkesbury Paper (−61.36°, 45.60°)	Hawkesbury, NS	HA	PA	Paper	400,000	350 [3]	2221	322122
Lake Utopia Paper (−66.77°, 45.16°)	Utopia, NB	UT	PA	Corrugate medium	185,000	140 [2]	1572	322130
Irving 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 2022)). [4] Total annual capacity. Mills do not necessarily operate at maximum capacity every year.

2.2. 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 2. The difference in SO_2 and $PM_{2.5}$ CPMAEPPF ARL was calculated for each mill (Table 3). 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 3. Annual recommended limits (ARL) for TPM and SO_2 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. Results

3.1. 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 2). 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).

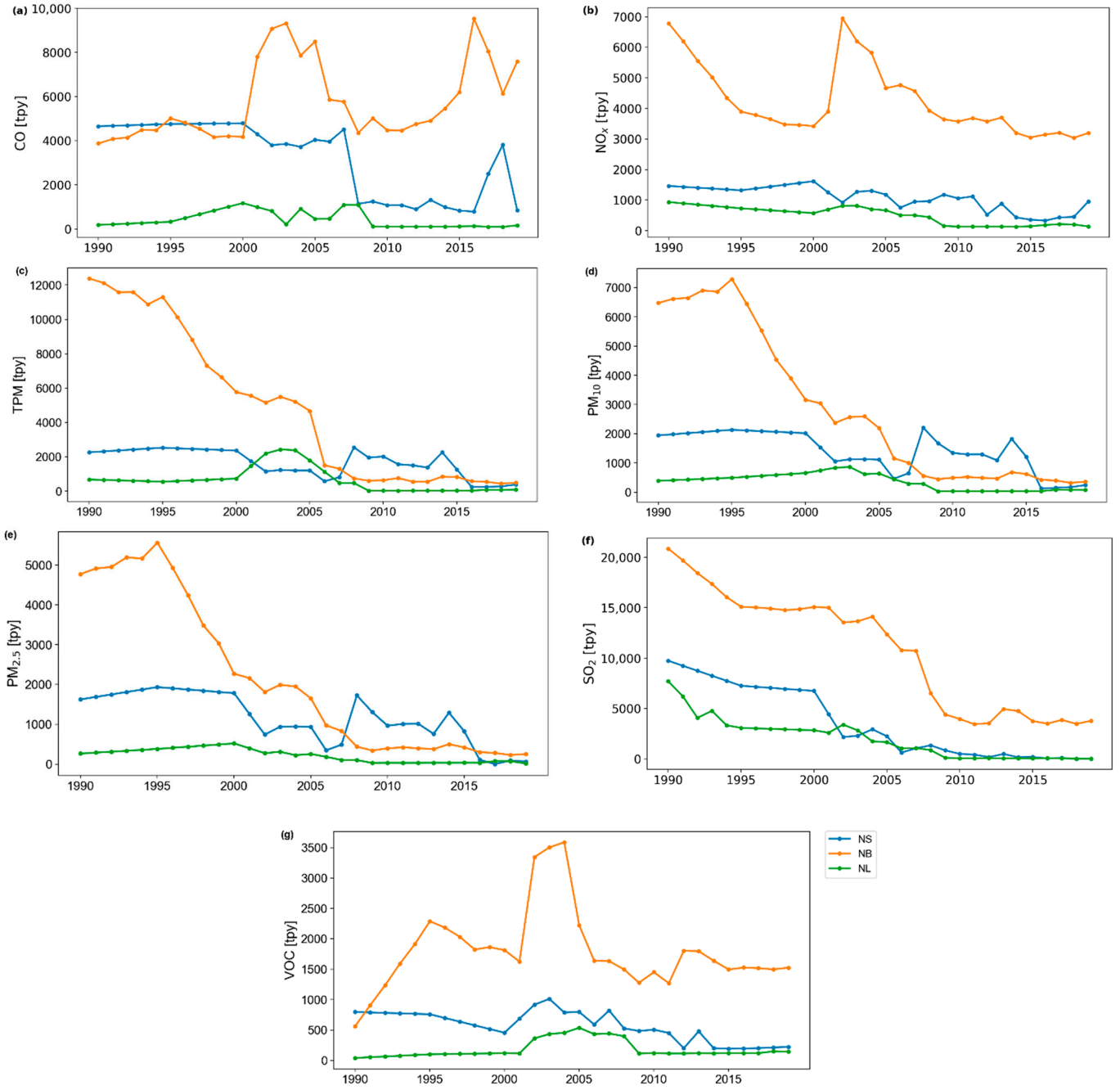

Figure 2. Air emissions registered in the APEI for Nova Scotia, New Brunswick and Newfoundland, and Labrador for pollutants (**a**) CO, (**b**) NO_x, (**c**) TPM, (**d**) PM_{10}, (**e**) $PM_{2.5}$, (**f**) SO_2, and (**g**) VOC.

3.2. Air Emissions Reported by Pollutant by Each Facility

Annual releases for all nine mills from 2002 to 2019 are shown in Figure 3 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 3a 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 4). 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 3b, 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.

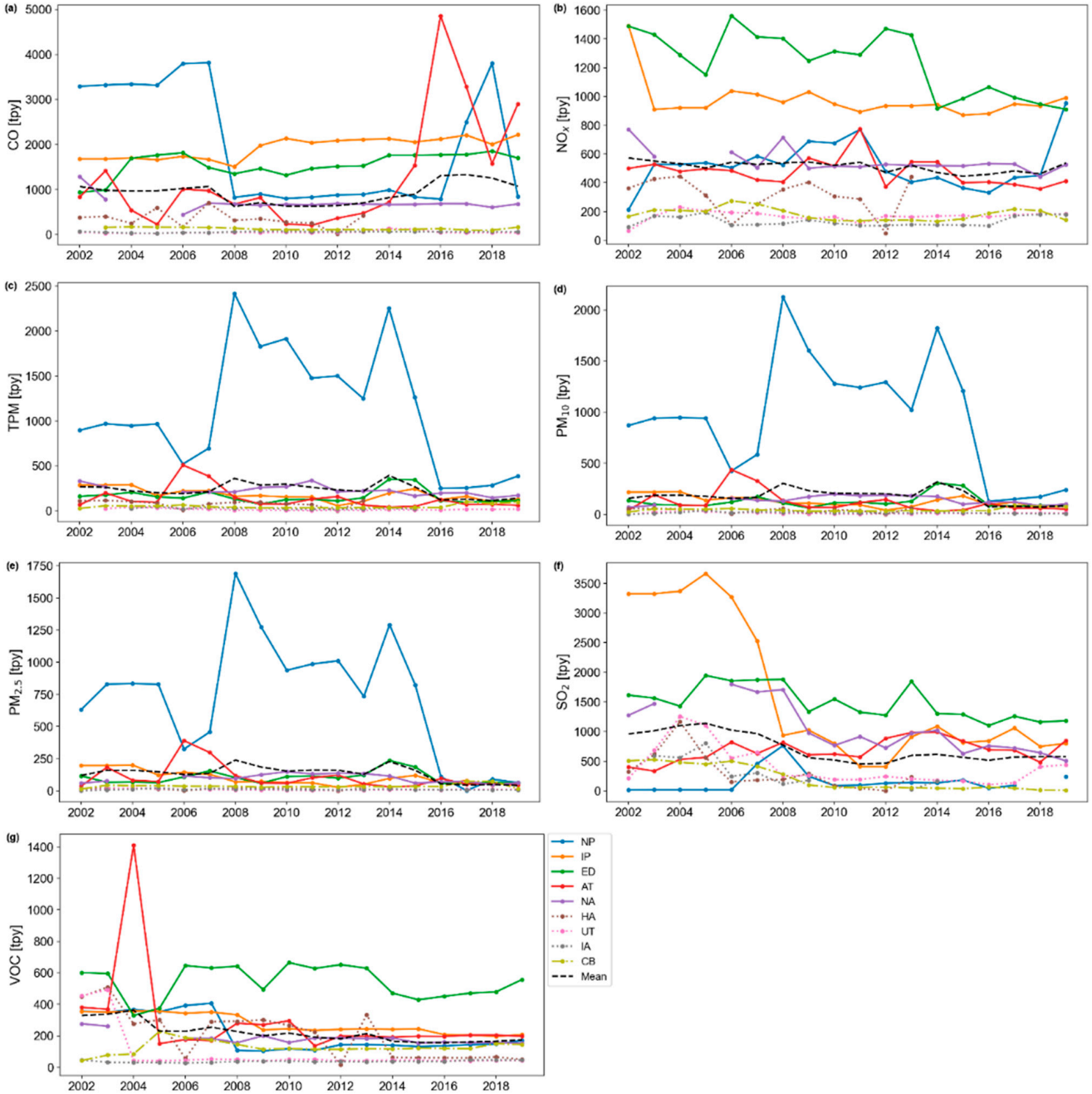

Figure 3. Air pollutant emissions from 2002 to 2019 for each facility for pollutants (**a**) CO, (**b**) NO_x, (**c**) TPM, (**d**) PM_{10}, (**e**) $PM_{2.5}$, (**f**) SO_2, and (**g**) VOC.

Table 4. *p*-values for the NPRI annual trends.

p-Value [1]	CO	NO_x	TPM	PM_{10}	$PM_{2.5}$	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.

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 3c). 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.

Similar characteristics were observed for PM_{10} emissions in Figure 3d, as previously shown by Hoffman et al. [49]. 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 3e, 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 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 3f) 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 3g 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.

Figure 4 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 [5]) are presented in Table 5. 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.

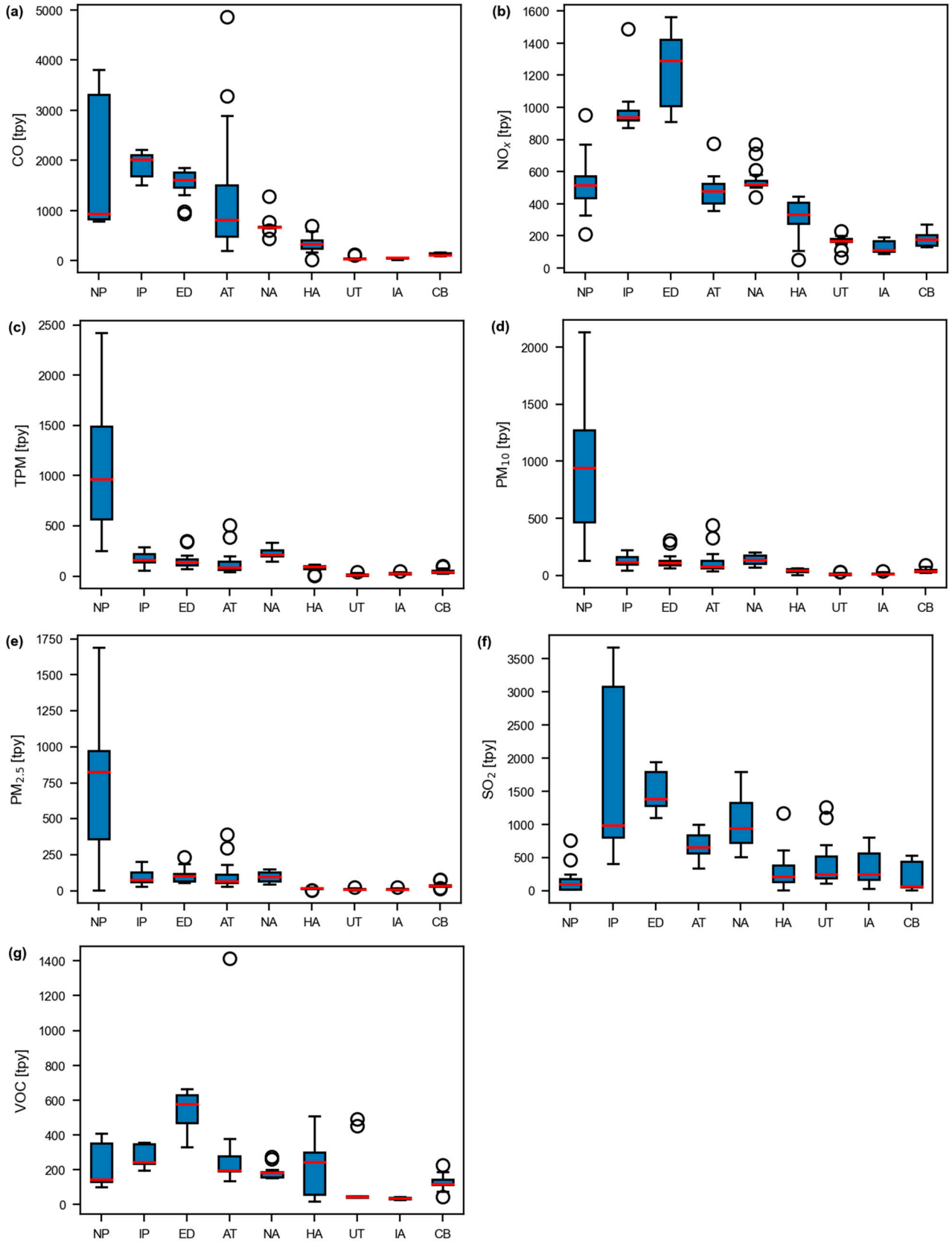

Figure 4. Distribution of air pollutant emissions from 2002 to 2019 for each facility for the pollutants (**a**) CO, (**b**) NO_x, (**c**) TPM, (**d**) PM_{10}, (**e**) $PM_{2.5}$, (**f**) SO_2, and (**g**) VOC.

Table 5. Mean and standard deviation (±) of air releases in each mill from 2002 to 2019.

Tonnes/Year	CO	NO_x	TPM	PM_{10}	$PM_{2.5}$	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.

3.3. Comparison with Thresholds

Figure 5a shows the mean DRT (Table 1) 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 3c,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 [29,45,47]. 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 5b. 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 5c 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%.

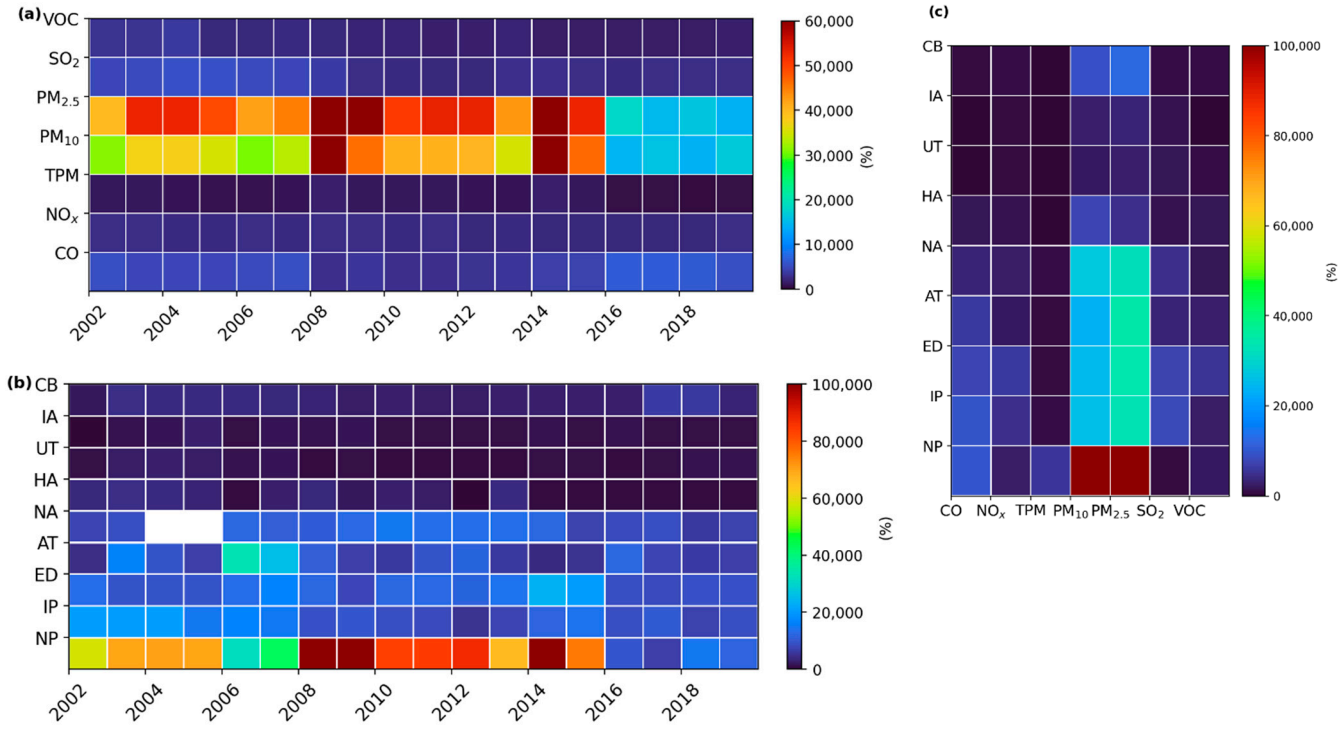

Figure 5. Mean difference from reporting thresholds (DRT) over (**a**) the sites, (**b**) the pollutants, and (**c**) the entire period. White represents no data.

The difference from the ARL on the CPMAEPFF for TPM and SO_2 is presented in Figure 6. 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.

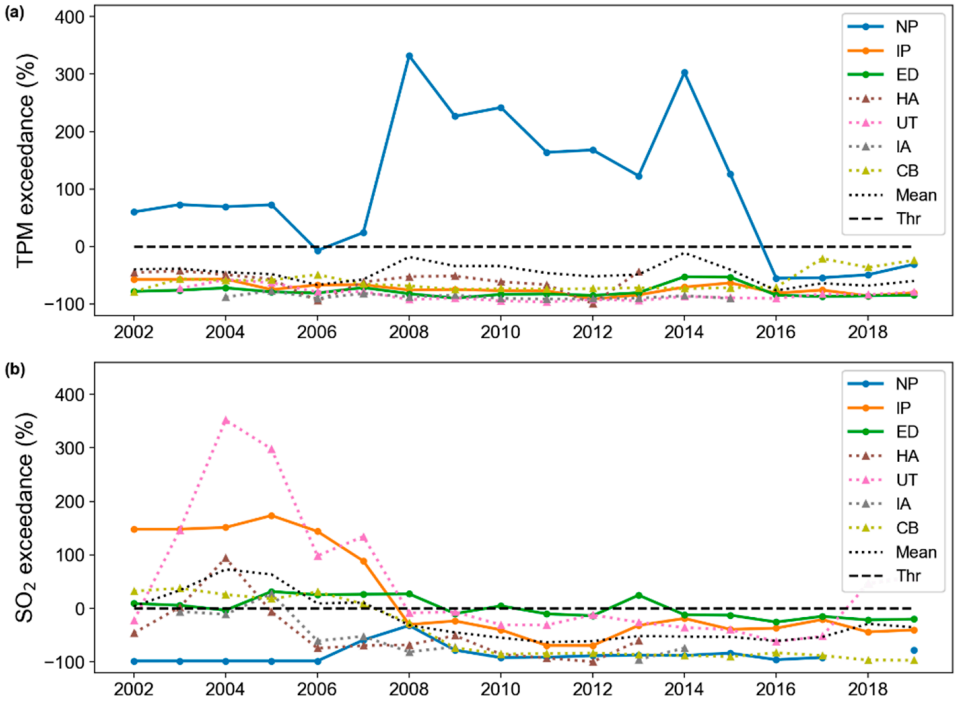

Figure 6. Percentual difference from the ARL on the CPMAEPPF for (**a**) TPM and (**b**) SO_2. Pulp mills are represented by a solid line and paper mills by a dotted line. The black dotted line represents the mean on all mills and the black slashed line represents when a release equals the threshold suggested by CPMAEPPF.

4. 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 [15,18]. 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 5). Techniques to reduce emissions can be hard for industries to implement due to high costs [28] and the challenge of adequately planning the improvements, as they depend both on the source of energy used and technology for upgrades [38]. In this case, NP could effectively reduce PM emissions with the incorporation of a recovery boiler [29].

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 [14].

Among the limitations that prevent NPRI from being widely used are issues with its completeness, data quality, and inventory comprehensiveness [5,14,15,18]. Most NPRI users are interested in air emissions, geospatial analysis of emissions, and environmental monitoring [14]. 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 [18,49]. 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 [54]). 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 [5], 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 5) show that all $PM_{2.5}$ releases in the P&P mills are four times higher in comparison to the findings of [5] and of similar a magnitude when only major emitters are considered. Conversely, SO_2 mean releases from P&P in Atlantic Canada (Table 5) are similar to those reported in [5], 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) [36], 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 [5] was previously introduced by [55], 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 [49]. 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" [33] and that to "achieve emissions reductions, threats of monitoring and actual enforcement have proven to be most effective" [15]. 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 [33] 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 exceedance in the emission rate regulation is not enforced [56].

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.

5. 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.

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Conflicts of Interest: The authors declare no conflict of interest.

Appendix A

Appendix A.1. Pulp Mills

Appendix A.1.1. 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 [57]. 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 [45,58]. 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 renamed 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 [61]. Before its closure, NP employed 330 fulltime workers and around 2000 indirect jobs across NS [62]. 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 [63]. Ten kilometres south of NP is New Glasgow, a town of 9471 people [63].

Appendix A.1.2. Irving Pulp & Paper Limited

Irving Pulp & Paper, a kraft pulp mill in Saint John, NB, is managed by Irving Ltd. [64]. 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 [64]. 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 [63].

Appendix A.1.3. 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 [65]. 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 [63]. 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 [66].

Appendix A.1.4. 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 [67]. 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 [68]. The Nackawic mill is located at northeast in the town of Nackawic, which has a population of 962 [63]. The Atholville mill is on the shore of Atholville village, within the town of Campbellton with a population of 7047 [63].

Appendix A.2. Paper Mills

Appendix A.2.1. 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 [69,70]. Located 2 km south of Port Hawkesbury, a town of 3210 inhabitants [63], 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 [71].

Appendix A.2.2. 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 [72]. 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 [73]. Irving paper produces specialty grade papers for flyers and magazines. They also supply rotogravure for commercial printing [74]. 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 [63].

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 [75]. 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 [76]. The mill is located in the north coast of the city Corner Brook, a city of 19,333 inhabitants [63].

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