Use of Pine Needles for the Identification of Polychlorinated Dibenzo-*p*-Dioxins and Polychlorinated Dibenzofurans Sources to Air in the Jonquière Area

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Executive Summary

In 1998. Environment Canada revealed the results of the polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) analyses conducted between 1987 and 1997 in Canada. The report showed that the average levels of PAHs and the 90th percentile value of PCDDs and PCDFs measured in Jonquière were high. The high levels of PAHs were not surprising, since the station was located close to an aluminum plant using the horizontal stud Söderberg process, but the high levels of PCDDs and PCDFs were surprising and gave rise to much concern among the population regarding air pollution in the region. This led the Environmental Protection Branch, Québec Region, to conduct a study of the results. During a meeting with the interested parties in the region and representatives of the population, the Ministry indicated what the department planned to do to identify the source or sources of PCDDs and PCDFs. Given that a study of the available data led to the conclusion that the source or sources involved were local, Environment Canada implemented, in the summer of 2000, a project using pine needles as bioindicators to verify whether or not there were more PCDDs and PCDFs in the air in certain sectors of the City of Jonquière, and to see if it would be possible to link these to specific sources. Control stations were also set up in Montreal and in Saint-Ignace-de-Loyola, close to Sorel. In addition, the regular collection of air samples was pursued in Jonquière, while an additional station was added in Chicoutimi.

The results of the pine needles collected at the end of August at different locations in Québec show that the PCDD and PCDF levels were higher in the needles collected in Saint-Ignace-de-Loyola, close to Sorel, in Montreal, and at the control station located in a rural area close to Jonquière. In December, the levels rose in all stations, except in Montreal where they remained the same as in the month of August. They are slightly higher in the stations located in the southern part of Jonquière, including the one located in the rural area. The period of exposure of the needles to air pollution could be one of the causes of the increase in levels when compared to those measured in August, but an increase was not noted that would corroborate this theory at the control station influenced by motor vehicle traffic in Montreal. Residential wood heating is probably an important source of PCDD and PCDF emissions in the Jonquière region. Motor vehicle traffic is also a source of PCDDs and PCDFs, but at a lesser level than in Montreal. Among the other sources, let us mention open fire burning and certain industrial activities, namely the metal recyclers and reclaimers who burn wiring or electric motors to rid them of their plastic sheath or varnish. In Saint-Ignace-de-Loyola, the industries in the steel and iron works sector explain in large part the levels of PCDDs and PCDFs found in the Sorel region.

The results of the campaign conducted in the summer of 2000 show that the levels of PCDDs and PCDFs present in the pine needles collected in Jonquière are similar to those measured in urban areas in the United States, and about 5 times lower than those measured in the vicinity of certain wood preservation plants in the States. They are up to 40 times lower than in Korea or Japan, where large quantities of PCDDs and PCDFs are present in the air.

There were no significant differences in PCDD and PCDF levels measured in ambient air in Jonquière, Chicoutimi and Montreal, and levels were similar to those found elsewhere in Canada between 1997 and 1999. On the other hand, these were approximately 10 times lower than those measured close to a municipal waste incinerator and a power PVC-coated cable reclaimer in the

United States, 20 times lower than those measured in Japan, or 40 times lower than those measured in Saxe, Germany.

Since 1995-1996, the average levels of PCDDs and PCDFs present in the air dropped by approximately 50% in Jonquière and Montreal, and are 2 to 3 times lower than the MENV's revised annual guideline.

A study of the PCDD and PCDF levels measured in the pine needles and in ambient air shows that the pine needles are capable of absorbing PCDDs and PCDFs, and of serving as bioindicators. The ratio of the levels measured in the air and in the pine needles is relatively constant, regardless of the levels present in the air.

Among the sources of PCDDs and PCDFs, let us mention wood burning, motor vehicle traffic, burning waste in open fires (or *back yard burning*), and certain industrial facilities. Given that the level of PCDD and PCDF contamination in ambient air in Chicoutimi and Jonquière is similar to that in Montreal, that it is lower than the MENV's guideline, and that regular sampling activities are pursued in Jonquière, we recommend the measurement of emissions stemming from the activities of industries recycling and reclaiming metal by heating cables and electric motors. An awareness campaign on a better use of wood stoves and on the effects of back yard burning would also help to reduce the levels of PCDDs and PCDFs in the air.

As regards PAHs and benzo(a)pyrene or B(a)P, the levels were similar in Jonquière and Chicoutimi, while they were lower in Montreal. Between 1989-1990 and 2000-2001, we noted a 96% reduction in B(a)P and 85% in PAHs in Jonquière; in Montreal, the reductions noted were 27% and 60% respectively. In Jonquière, the most marked reduction was noted between 1999-2000 and 2000-2001, when Alcan modified the composition of the anodic paste used in its electrolytic cells. In Chicoutimi, the levels measured between July 2000 and the end of March 2001 were similar to those obtained in 1993-1994 by the MENV.

The PCDD and PCDF levels varied slightly according to wind direction, but less than the PAH levels which were higher when the stations were downwind from the aluminum plant. In fact, the highest PAH value was measured in Jonquière when the station was downwind from the aluminum plant for 18 of the 24-hour sampling day.

Introduction

In 1998. Environment Canada's Analysis and Air Quality Division made public a report on the levels of polycyclic aromatic hydrocarbons (PAHs), of polychlorinated dibenzo-*p*-dioxins (PCDDs) and of polychlorinated dibenzofurans (PCDFs) measured between 1987 and 1997 at the stations of Canada's National Air Pollution Surveillance Network (NAPS) (Dann, 1998). This report indicated that the levels of PAHs in the air in Jonquière were the highest among 35 measuring stations in operation between 1994 and 1997. As regards PCDDs and PCDFs, a station located in Toronto held the highest average value in the country, that is 0.105 pg TEQ/m^3 (picogram toxic equivalent factor per cubic metre). The station located in Jonquière showed the highest value for the period from 1994 to 1997 once the 90th percentile value[1] was applied.

The presence of high levels of PAHs in Jonquière has been known for a long time (Allaire *et al*., 1993; Germain et Bisson, 1992), but the presence of PCDDs and PCDFs at the levels set out by Dann (1998) were surprising. The disclosure of these results gave rise to much concern among the population of the Jonquière region regarding air pollution in the area, and prompted the Environmental Protection Branch, Québec Region, to make a detailed study of the results. On February 2, 2000, a meeting took place in Jonquière and the 1998 report was presented to several regional players and various interested parties of the population. Environment Canada personnel took the opportunity to update the results (up to September 1999), which lessened the population's concern regarding the PCDD and PCDF levels measured in the area. Following this meeting, a study of the whole of the data available was conducted while taking weather conditions into account (Rousseau, 2000). We were able to note that the source or sources of PCDDs and PCDFs were local, and that the highest values were measured when winds blew from the south. A summary of the results was distributed in December, 2000, through the agency *Conseil régional de l'environnement Saguenay–Lac-Saint-Jean.*

Environment Canada developed a project that aimed to identify the source or sources of PCDDs and PCDFs. Given the scope of the region and the high cost of the analyses, it was decided to use bioindicators to verify whether or not there were any differences in PCDD and PCDF levels measured at several sites throughout the region. Documentation mentions that a number of bioindicators may serve to measure pollutants present in the air (Safe *et al*., 1992; GDG Environnement, 1995; Fiedler *et al*., 2000; Ok *et al.*, 2000; Viskari, 2000), and that these indicators afford the advantage of capturing the substances over relatively long periods of time. Among the bioindicators considered was peat moss, which we had already used in the Sorel region (Grenon et Lafortune, 1997), and pine needles (Myata *et al*., 2000; Ok *et al*., 2000). Safe *et al*. (1992) and Viskari (2000) report that the waxy section of the pine needles could serve as bioindicators of the deposition of pollutants, and be used to measure the lipophilic organic compounds in the air, namely PCBs, organochlorine pesticides such as DDT, or PCDDs and PCDFs. The collection of pine needles gathered among the year's shoots allows to determine and limit the period of exposure. A measuring program using Scots pine needles (*Pinus sylvestris*) as

[1] This value is obtained by classifying the values from the lowest to the highest, and by choosing the value that corresponds to the 90th largest value if the number of values is 100. If the number of values is not 100, the value retained is the value which represents the multiple of 90 in relation to 100 (example: the 9th value if there are 10 values, the 900th if there are 1000).

bioindicators was developed and implemented to identify the source or sources of PCDDs and PCDFs.

A “conventional” air sampling station using a device that pumps air through a system that captures the desired products over a determined period of time (generally 24 hours) was also set up in Chicoutimi to measure PAH, PCDD and PCDF levels in the ambient air and to compare these with levels measured in Jonquière. The Ministère de l’Environnement du Québec (MENV) increased the number of air samples by conducting PAHs, PCDDs and PCDFs analyses for a few samples in its laboratory.

Methodology

On July 11 and 12, 2000, Scots pines approximately 125 cm high were planted at 11 sites in the region to verify whether or not there were any differences between the levels of PCDDs and PCDFs (Figure 1). These sites were selected on the basis of prevailing wind direction and the region’s urban and industrial activity. Pines were also planted at the station used by Environment Canada since 1989 for air quality follow-up activities (station EC or station 1), on the property of Dominique-Racine school in Chicoutimi (station 2), in the field located behind homes on Dubuisson Street (station 3), on the property of the Abitibi-Consolidated Ltd. pulp and paper plant (station 4), at station 5 of Alcan’s air quality surveillance network (station 5), in an urban area at Poitras park (stations 7 and 14), in Chicoutimi’s industrial park (station 8), next to Jonquière’s industrial park (station 9), at the weather station located on Alcan’s property (station 10), close to the Saint-Jean-Eudes neighbourhood (station 11), and lastly, in a rural area south of Jonquière to serve as a control station (station 6). Pines were also planted at two control stations located outside the region. These were located at Saint-Ignace-de-Loyola (station 12), downwind from Sorel’s iron and steel works, and close to the Decarie and Metropolitain highways in Montreal (station 13).

In order to determine the levels of PCDDs and PCDFs initially present in the pine needles, we cut the ends of two or three branches of the year’s shoots on each of the trees used. These branches were then grouped and kept in a refrigerator until their analysis in the laboratory. The grouping of the branches and their analysis as one sole sample was justified since the trees all came from the same site.

A first series of pine needles was gathered at the end of August (August 29 to 31, 2000) and a second at the beginning of December (December 4 and 5, 2000). These periods were retained to determine the levels corresponding to the summer, when there is no wood burning activities, and to the beginning of winter, when temperatures are low and people have begun to heat their homes. At each site, the ends of 10 to 20 branches of this year’s shoots were cut and wrapped in aluminum foil previously cleaned at 550°C, and then placed in an airproof bag. The bags were then kept in an icebox at 4°C and sent to the laboratory where they were kept in a freezer until analysis. In the laboratory, the needles were separated from the branches and placed in a microwave extraction appliance. The extract was then purified before analysis by gas chromatography. The products were identified using a high-resolution mass spectrometer. This method of extraction was developed for the analysis of PCBs by Environment Canada’s Environmental Technology Center (Environment Canada, 1997). It has been recognized by the

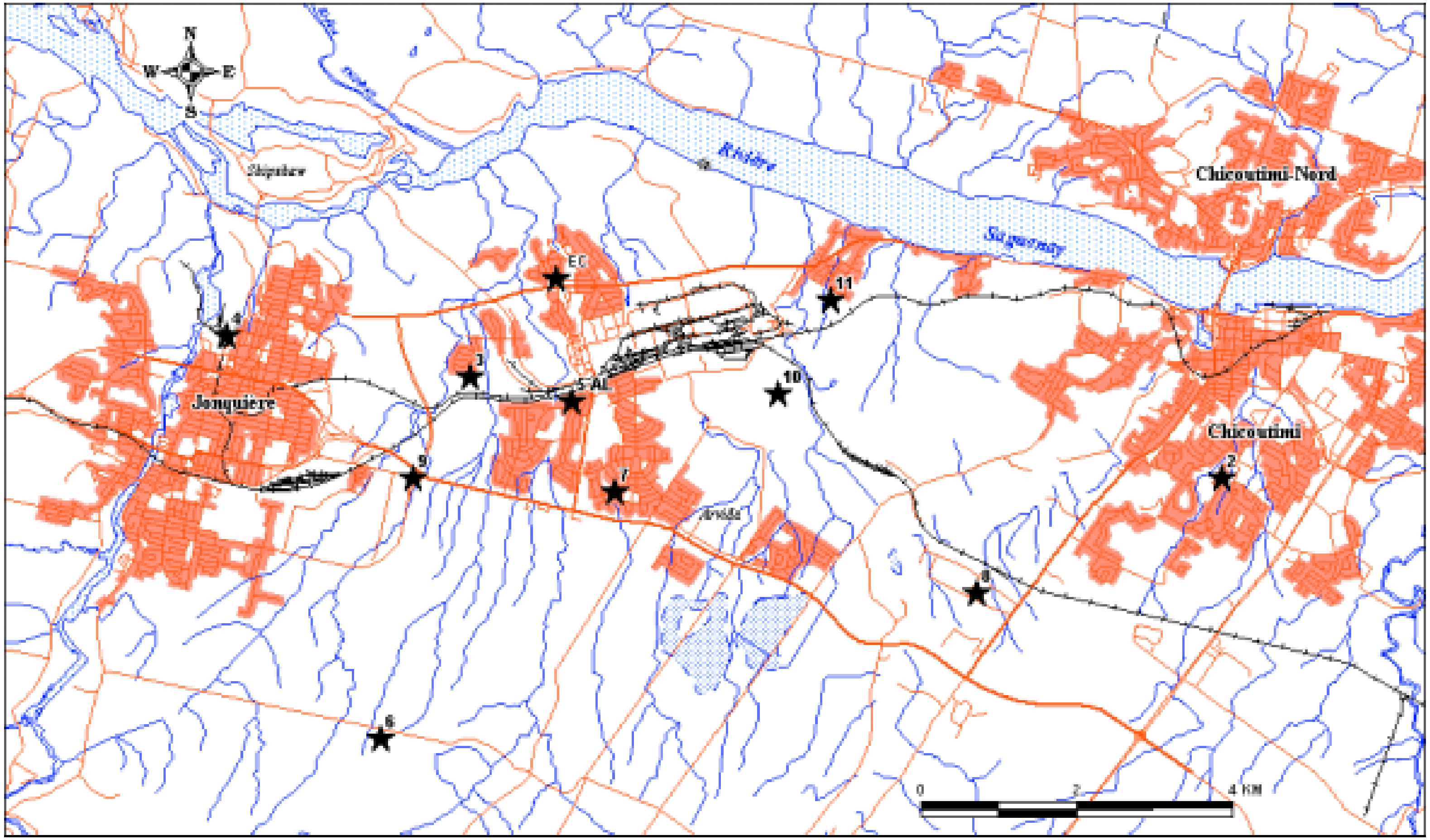

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Figure 1 Location of measurement stations in Jonquière and Chicoutimi

U.S. Environmental Protection Agency (US EPA) and serves as a reference method for biological material analysis. The moisture content of the needles was also determined in order to report results on a dry weight basis.

During the summer 2000 sampling campaign, a “conventional” air sample collection station was set up on the roof of Dominique-Racine school in Chicoutimi (station 2). Between August 2000 and the end of March 2001, nine air samples were taken every 28 days and analysed for PAHs, PCDDs and PCDFs by Environment Canada. In addition, three additional samples were taken and analysed at the MENV’s laboratory. Paralleling these activities, air sample collection continued at Environment Canada’s station in Jonquière (sampling dates provided in Annex 1). To verify the evolution of PAHs, PCDDs and PCDFs, we have grouped the results on the basis of the government’s financial exercise, that is from April 1 to March 31.

The analysis of the air samples was conducted following the methods currently used by the laboratories; quality control included a field blank (or reference sample), the analysis of lab blanks to verify the purity of the solvents and the cleanliness of the materials, and the spiking of each sample with reference standards including radioisotopes to check the efficiency of the method in recovering the targeted products. The lab results provided were adjusted while taking into account the recovery of each of the reference standards.

In order to facilitate the interpretation of the results, we used the meteorological data provided by the Alcan station, a station located in Jonquière and certified by the Canadian Meteorological Service which has integrated this station into its observation network. The results of the weather station located at the Bagotville Airport, consulted as needed, filled the gap left by missing data. Thus, we determined the wind rose for the whole of the project and for each day of air sampling activity. The wind directions noted for the periods July-end of August and July-beginning of December (see Annex 2) correspond to those generally noted in the region.

Results

PCDDs and PCDFs found in the pine needles

We found only two dioxins and no furans in the pine needles collected before the trees were planted (field blank): 1,2,3,7,8-pentachlorodibenzodioxin (or 1,2,3,7,8-P5CDD) and octachloro-dibenzodioxin (or T8CDD). Their total concentrations were 7.6 pg/g d.w. (dry weight) of PCDDs or 0.8 TEQ/g d.w. when expressed in terms of toxic equivalent (Table 1).

The PCDD and PCDF levels measured at the end of the summer were close to the limits of detection of various products, and the Chicoutmi-Jonquière area, station 6 (control station) showed the highest levels with 129 pg/g d.w. In general, the levels are lower in the region (overall average of 35.9 pg/g d.w.) than in Saint-Ignace-de-Loyola (91.7 pg/g d.w.) or Montreal (369 pg/g d.w.). Expressed in terms of toxic equivalent, the PCDD and PCDF levels are higher at the stations in Saint-Ignace-de-Loyola (0.79 pg TEQ/g. d.w.), Montreal (0.68 pg TEQ/g. d.w.), and at station 8 located in Chicoutimi’s industrial park (0.77 pg TEQ/g. d.w.). Regional results showed a slight variation depending on the location of the stations, but since the PCDD and PCDF levels and related TEQs were close to detection limits, it would be hazardous to draw a conclusion.

Table 1 Total concentrations and toxic equivalent (TEQ) of PCDDs and PCDFs measured in the pine needles collected at the end of August (dry weight)

Station	PCDD	PCDF	Total PCDD + PCDF	FET	FET corrigé
	pg/g	pg/g	pg/g	pg FET/g	pg FET/g
Station EC (1)	14.0	3.1	17.0	0.28	0.07
Station 2	5.4	1.5	6.9	0.34	0.02
Station 3	3.6	1.2	4.8	0.07	< l.d.
Station 4	16.4	8.2	24.2	0.35	0.20
Station 5	8.8	3.2	12.0	0.13	0.04
Station 6	116.0	13.7	129.7	0.44	0.14
Station 7	14.1	4.0	18.1	0.32	0.07
Station 7 (duplicate)	11.4	6.0	17.4	0.09	< l.d.
Station 7 (average)	12.8	5.0	17.8	0.20	0.03
Station 8	81.9	15.3	97.2	0.77*	0.39*
Station 9	24.6	8.3	32.9	0.49	0.21
Station 10	11.1	3.8	14.9	0.2	0.05
Station 11	33.5	3.5	37.0	0.18	0.03
Average (region)	29.8	6.1	35.9	0.31	0.11
Station 12 (SIL)	71.7	19.9	91.7	0.79	0.52
Station 13 (Montreal)	348.5	20.7	369.2	0.68	0.11
Control station	7.6	0.0	7.6	0.8	

* : values adjusted for a moisture content of 57% : 0.48 pg/m^3 et 0.27 pg TEQ/m^3

It should be emphasized that at station 8, the moisture content of the needles was higher (79.8%) than at the other stations (approx. 57%); it is possible that an error occured during the weighing process and that the sample's dry weight is higher. If we were to suppose that this sample's percentage of water content is 57%, then the PCDD and PCDF levels would drop to 0.27 pg TEQ/g d.w. In winter, the water content of the pine needles at this station (58.3%) is similar to that of the other stations (57.7%). The higher water content value (85.8%) of the needles collected in the trees at the time of planting in July can be explained by the young age of the needles. The fact is that young shoots contain more water and are more tender than at the end of August or at the beginning of December when they have aged.

When the TEQ concentrations are adjusted to take into account the two products initially found in the needles, the highest value is found at Saint-Ignace-de-Loyola, followed by the stations located at the two industrial parks. The TEQ value at the Montreal station drops heavily since the majority of the total PCDDs/PCDFs stem from the T8CDD, which has an equivalence factor of 0.001, and the P5CDD, which has an equivalence factor of 0.5.

The PCDD and PCDF levels in the pine needles collected at the beginning of December were higher at all stations in the region (Table 2) than those measured in August. Station 9, located next to Jonquière's industrial park, showed the highest value (265 pg/g d.w.), followed by the control station (6) located in a rural area (205 pg/g d.w.). They also increased at Saint-Ignace-de-Loyola (129 pg/g d.w.), but diminished slightly in Montreal (52 pg/g d.w.). The average level noted in the region (105.4 pg/g d.w.) is lower than the average level measured in Saint-Ignace-de-Loyola, but higher than that registered in Montreal. Results show that the levels were slightly higher in the south of the region. When the results are expressed in terms of toxic equivalent, the highest levels are found in the stations in or next to industrial parks, followed by those obtained at Environment Canada’s historic station and at Saint-Ignace-de-Loyola. The station located in a rural area also shows high levels. The adjustment of the results to take into account the levels of PCDDs and PCDFs initially present in the pine needles leads to the same conclusions, that is, higher levels at the stations in or next to industrial parks, followed by the historic station and the station at Saint-Ignace-de-Loyola.

Table 2 Total concentrations and toxic equivalent (TEQ) of PCDDs and PCDFs measured in the pine needles collected at the beginning of December (dry weight)

Station	PCDDs	PCDFs	Total PCDDs/PCDFs	TEQ	Adjusted TEQ
	pg/g	pg/g	pg/g	pg TEQ/g	pg TEQ/g
Station EC (1)	60.5	24.0	84.5	1.82	1.38
Station 2	37.3	12.5	49.8	1.26	0.69
Station 3	52.2	19.6	71.8	0.57	0.53
Station 4	42.5	30.0	72.5	1.35	0.86
Station 5	30.5	19.7	50.2	1.09	0.83
Station 6	158.1	46.7	204.8	1.55	1.25
Station 7	88.6	38.8	127.4	0.67	0.61
Station 7 (duplicate)	77.5	39.1	116.6	1.18	0.96
Station 7 (average)	83.1	38.9	122.0	0.92	0.78
Station 8	94.1	56.5	150.6	2.21	1.91
Station 9	222.5	42.1	264.6	2.2	1.55
Station 10	44.7	16.2	60.9	0.85	0.67
Station 11	19.7	8.3	28.0	0.46	0.45
Average (region)	76.8	28.6	105.4	1.29	0.99
Station 12 (SIL)	97.9	31.1	129.0	1.73	1.31
Station 13 (Montreal)	36.7	15.1	51.7	0.76	0.61
Control station	7.6	0.0	7.6	0.8	

Comparison with literature

The average PCDD and PCDF levels measured in the pine needles collected at the end of August and at the beginning of December 2000 in Québec, and in the Jonquière region in particular, compared with those measured in Texas (Figure 2). However, the average levels were lower than those measured close to wood preservation plants in Texas (5.4 pg TEQ/g d.w.) and in Montana (7.7 pg TEQ/g d.w.; Safe *et al*., 1992) or than those measured in Korea (urban area influenced by motor vehicle traffic; 4.5 to 8.2 pg TEQ/g d.w.; Ok *et al*., 2000) or in Japan (13.6 and 18.3 pg TEQ/g d.w.; Myata *et al*., 2000). The PCDD and PCDF content measured in the pine needles collected in the Kootenai National Park Forest and the Glacier-Waterton International Peace Park, the reference site used by Safe *et al.* (1992), reached less than 0.32 and less than 0.30 pg TEQ/g d.w. respectively, while in Cheju, the reference site used by Ok *et al.* (2000), the level was 0.5 pg TEQ/g d.w. In Germany, the PCDD and PCDF levels measured by Fiedler *et al.* (2000) in spruce needles varied from 0.31 to 1.12 pg TEQ/g d.w. in Bavaria, and from 1.2 to 2.67 pg TEQ/g p.s. in Hesse.

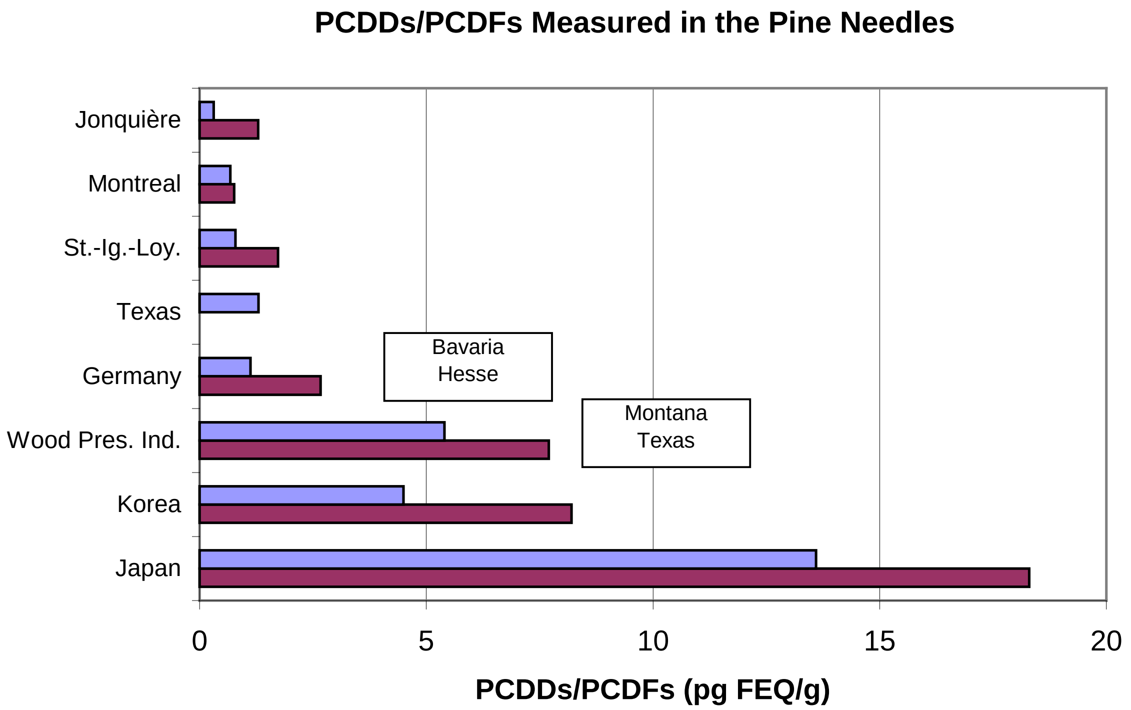

Figure 2 Comparison with documented levels of the average PCDD and PCDF levels measured in pine needles collected in Québec (*spruce needles)

PCDDs and PCDFs found in ambient air

Table 3 shows the statistical values of PCDDs and PCDFs measured between July 1, 2000, and March 31, 2001, in Chicoutimi, Jonquière and Montreal. Though the average, median and maximum values are higher in Chicoutimi during this period, the statistical analysis fails to show any significant differences ($p < 0.05$) between geometric averages and median values at the three stations.

Figure 3 shows that the average PCDD and PCDF levels recorded in the Saguenay and in Montreal compare with the calculated average (0.036 pg TEQ/m^3) for the whole of the stations where measurements were taken by Environment Canada between 1997 and 1999 (Dann, 2000). However, these were lower than those obtained in the Rivière-des-Prairies (RDP) neighborhood in Montreal, a sector influenced by wood heating activities (Germain *et al*., 2001). In addition, they are approximately 2 times lower than those noted by Fiedler *et al*. (2000) in Hesse (average of 0.058 pg TEQ/m^3), Germany, and approximately 20 times lower than those measured by Myata *et al*. (2000) in Hiraka, Japan (averages of 0.408 and 0.535 pg TEQ/m^3). The highest documented average value is of 0.900 pg TEQ/m^3, recorded by Fiedler *et al*. (2000) in Saxe, Germany. It is interesting to note that once the units are set aside, the air/needle ratio varies from 0.025 to 0.034 in Jonquière, Montreal, Hiraka (Japan) and Hesse (Germany), though the average PCDD and PCDF levels are higher, in both the air and the needles, in Hesse (approx. 2.5 times) and in Hiraka (approx. 20 times) than in Jonquière. The limited data used shows that the $PCDD_{air}/PCDF_{needles}$ ratio is relatively constant, regardless of the levels present in the air.

Table 3 Statistical summary of PCDD and PCDF levels measured between July 1, 2000, and March 31, 2001, in ambient air in Chicoutimi, Jonquière and Montreal (pg/m^3 and pg TEQ/m^3)

Location and parameters	Average		Median	Maximum
	Arithmetic	Geometric		
Chicoutimi (n* = 12)				
PCDDs (pg/m^3)	2.104	0.924	0.617	12.720
PCDFs (pg/m^3)	0.781	0.515	0.599	1.631
PCDDs + PCDFs (pg/m^3)	2.885	1.517	1.293	14.040
pg TEQ/m^3	0.039	0.021	0.024	0.102
Jonquière (n = 10)				
PCDDs (pg/m^3)	1.302	0.735	0.572	4.110
PCDFs (pg/m^3)	0.552	0.283	0.278	1.365
PCDDs + PCDFs (pg/m^3)	1.854	1.174	1.719	5.016
pg TEQ/m^3	0.029	0.015	0.019	0.071
Montreal (n = 12)				
PCDDs (pg/m^3)	0.962	0.726	0.831	2.058
PCDFs (pg/m^3)	0.426	0.355	0.376	1.057
PCDDs + PCDFs (pg/m^3)	1.388	1.110	1.207	2.912
pg TEQ/m^3	0.025	0.020	0.023	0.054

*number of samples

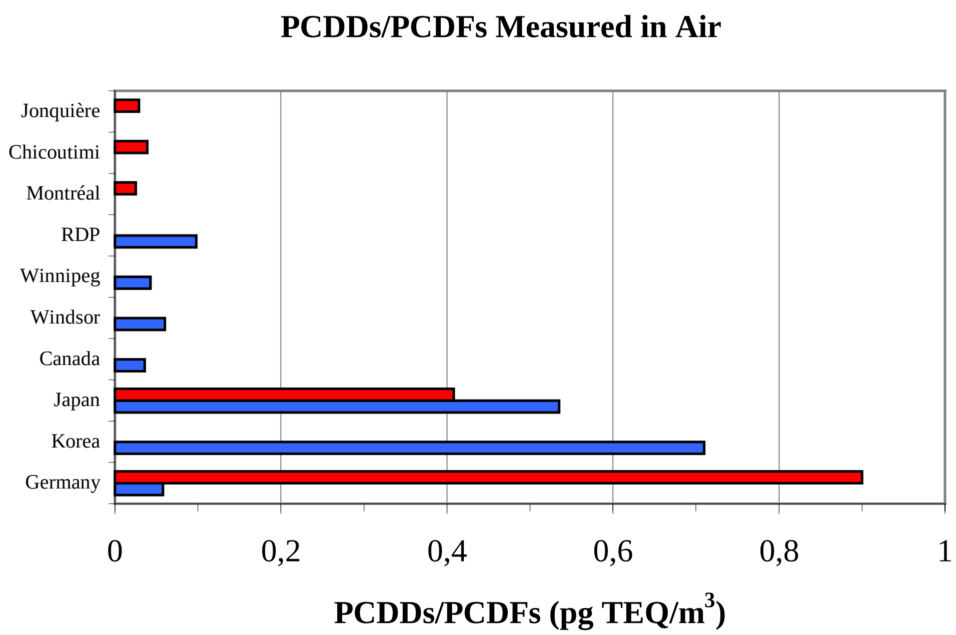

Figure 3 Comparison with documented levels of PCDD and PCDF levels measured in Chicoutimi, Jonquière and Montreal

PAHs and B(a)P found in ambient air

Table 4 shows the statistical values of benzo(a)pyrene or B(a)P, and of total PAHs measured between the beginning of July and the end of March in Chicoutimi, Jonquière and Montreal. As is the case for PCDDs and PCDFs, the median and average concentrations (geometric) are not significantly different between Chicoutimi and Jonquière, but are lower ($p < 0.05$) in Montreal. As regards the daily values, the highest value was measured in Jonquière and was approximately six times higher than the highest value recorded in Chicoutimi. The proximity of the aluminum plant and the prevailing wind direction are responsible for these results. The scope of the results is also greater in Jonquière and Chicoutimi than in Montreal. For purposes of statistical comparison, we have included the results of the B(a)P measured by the MENV, but did not do so for total PAHs, since the number of products measured by the two laboratories differed.

In Jonquière, the average and maximum levels of B(a)P and total PAHs measured between July 2000 and the end of March 2001 are lower than those measured between 1997 and 1999 (Dann, 2000). During this period (1997-1999), Jonquière had the highest levels of B(a)P and PAHs measured in the country, and these levels could be explained by the proximity of an aluminum plant using the horizontal stud Söderberg process. Results recorded in 2000-2001 in Jonquière cannot be compared to those recorded elsewhere in the country, since the data is unavailable for the period between July, 2000, and March 31, 2001, at the other sites. In Chicoutimi, the B(a)P levels are of the same order of magnitude as those recorded by the MENV between June 1993 and March 1994 (Bisson, 2001).

Table 4 Statistical summary of B(a)P and total PAHs measured between July 1, 2000, and March 31, 2001, in ambient air in Chicoutimi, Jonquière and Montreal (ng/m^3)

Product and location	Average		Median	Maximum
	Arithmetic	Geometric		
B(a)P				
Chicoutimi (n* = 12)	3.7	1.2	1.8	15.2
Jonquière (n = 16)	8.9	0.9	1.6	91.0
Montreal (n = 19)	0.3	0.2	0.2	1.2
PAHs				
Chicoutimi (n** = 9)	183.6	103.2	167.1	669.0
Jonquière (n = 16)	508.3	121.1	113.9	3834.3
Montreal (n = 19)	27.9	23.9	22.2	73.8

*number of data, including MENV data

**number of data, excluding MENV data given the different number of products measured by the laboratories

Evolution of PCDDs, PCDFs, PAHs and B(a)P measured in ambient air

Figure 4 shows the evolution of PCDD and PCDF levels measured in Jonquière and Montreal between 1995 and 2001. Reduced levels were noted in both places and the levels measured in 2000-2001 were approximately 2 times lower than those measured in 1995-1996. Except for 1996-1997, the PCDD and PCDF levels were similar at both sites. In 1996-1997, the discrepancy between the two stations can be explained by the 2 high values recorded in November and December 1996 in Jonquière. The average annual values calculated since 1995-1996 are approximately 2 to 3 times lower than the MENV's revised guideline of 0.06 pg TEQ/m^3, a value not to be exceeded on an annual basis.

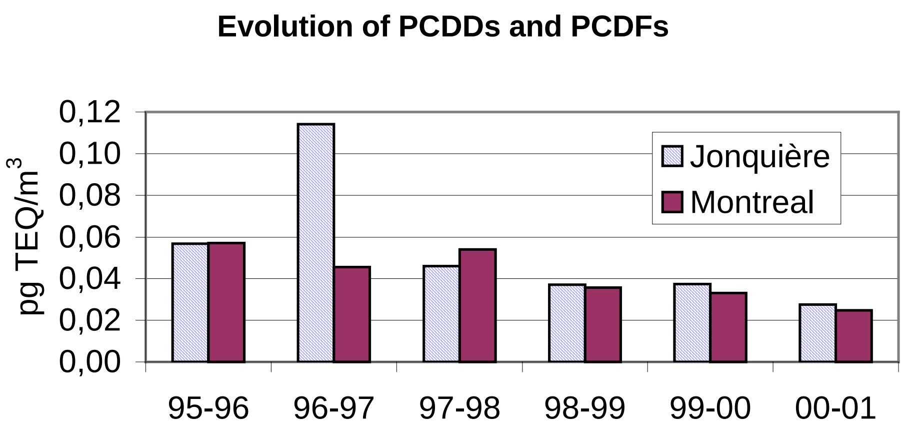

Figure 4 Evolution of PCDDs and PCDFs (in terms of toxic equivalent) in Jonquière and Montreal

Figure 5 illustrates the evolution of PAHs since the beginning of these measurements in 1989-1990. In Jonquière, the average value (geometric) of total PAHs occasionally increased and diminished during the period, but on the whole, there was an 85% reduction between 1989-1990 and 2000-2001. The most important reduction between two years was recorded between 1999-2000 and 2000-2001 when the reduction reached 60%. In Montreal, the average level of total PAHs varied less than in Jonquière, and the reduction noted since the beginning of the measurements reached 60%. While the average level measured in Jonquière was 10 times higher than in Montreal in 1989-1990, it was 4 times higher in 2000-2001.

Figure 6 shows that as is the case for total PAHs, B(a)P levels vary over the years in Jonquière and Montreal, and that on the whole, levels diminished between 1989-1990 and 2000-2001. There was a more important reduction in the geometric average in Jonquière (96%) than in Montreal (27%).

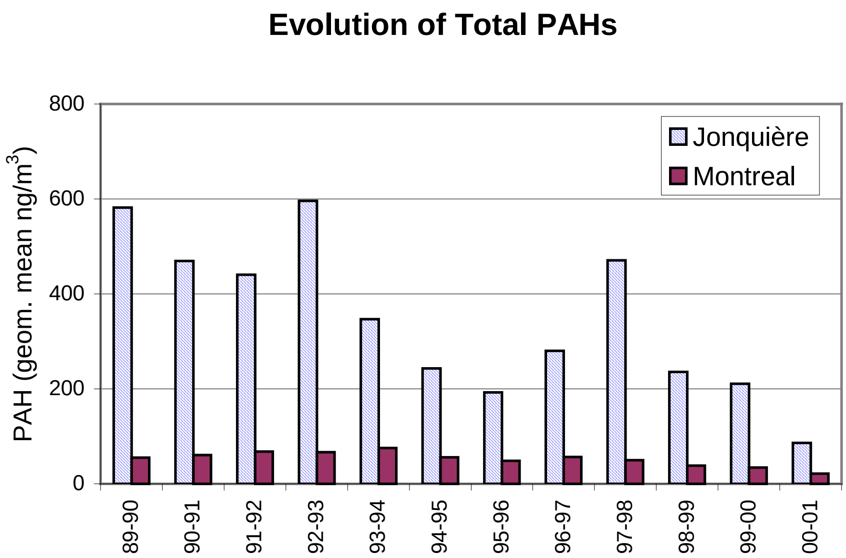

Figure 5 Evolution of total PAHs measured in Jonquière and Montreal

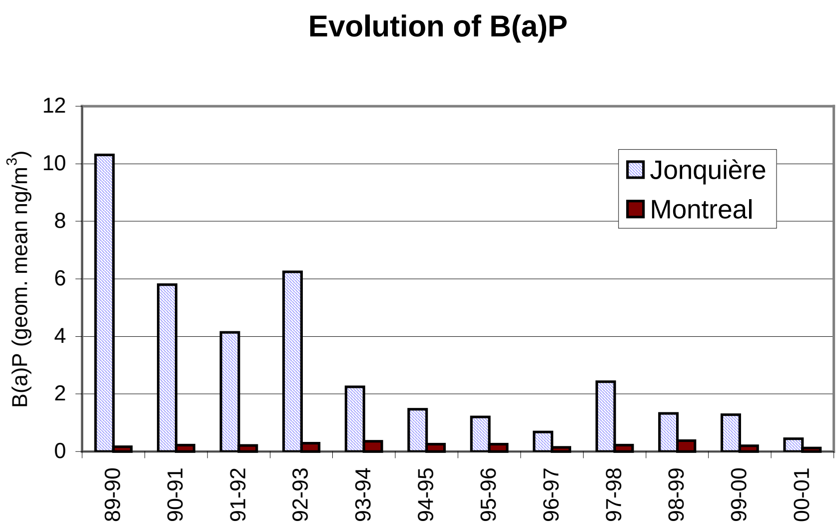

Figure 6 Annual evolution of B(a)P measured in Jonquière and Montreal

The wind factor

The levels of PCDDs, PCDFs and PAHs [including B(a)P] showed more important variations in Jonquière and Chicoutimi than in Montreal. Once statistics are calculated, we may tend to believe that certain samples were contaminated, given the important differences in levels; in fact, the statistical software flagged certain values as outliars. However, these results may be explained by studying weather conditions, including wind direction, on each day of sampling.

Table 5 shows the concentrations of PCDDs, PCDFs and B(a)P for the days when the wind blew mostly from one sector during the sampling procedure (except in one case, the days when the wind blew from several sectors are not taken into account in the following). When the wind blew from the west, that is from Jonquière towards Chicoutimi, we had a first block of high levels of PCDDs, PCDFs and B(a)P, and a second with lower levels. When the wind blew from the east, that is from Chicoutimi towards Jonquière, the PCDD and PCDF samples were taken once simultaneously at both locations. Since there was only one pair of results, we have chosen to include another pair of results recorded when the wind included an important northern component. It should be stressed here that ambient air sampling was more frequent in Jonquière than in Chicoutimi, and that on a number of occasions, samples were taken in Jonquière but not in Chicoutimi while the wind blew from the east. In a previous report, Rousseau (2000) indicated that the highest PCDD and PCDF values were noted when the wind blew from the south. Unfortunately, we have no samples taken when the wind blew mainly from this zone between July, 2000, and the end of March, 2001.

Table 5 Levels of PCDDs, PCDFs and B(a)P measured on days when the wind blew mainly from one sector

Date and wind direction	Jonquière	Chicoutimi	Jonquière	Chicoutimi	Chicoutimi/Jonquière ratio	
	PCDD+PCDF	PCDD+PCDF	B(a)P	B(a)P	PCDD+PCDF	B(a)P
	pg TEQ/m^3	pg TEQ/m^3	ng/m^3	ng/m^3		
West wind						
1st block						
December 8	0.071	0.073	1.5	4.8	1.0	3.2
February 6	0.017	0.098	3.4	2.8	5.6	0.7
February 25	0.049	0.079	1.7	15.2	1.6	10.6
March 26	0.001	0.102	0.02	5.4	146	270
2nd block						
September 3	0.014	0.008	0.02	1.8	0.6	90
January 1	0.002	0.002	0.06	0.36	1.3	6
East wind						
November 14	0.057	0.002	91	0.02	0.03	0.002
*March 2**	0.051	0.034	2.9	11.0	0.6	3.8

*: Important northern and northeastern components.

In the first block, the PCDD and PCDF values were quite high in Chicoutimi where they vary between 0.073 and 0.102 pg TEQ/m^3, while in Jonquière, there are two days with higher values and two with lower values. In addition, there is a day during which the PCDD and PCDF levels are quite similar in Jonquière and Chicoutimi. As regards the B(a)P, levels were generally higher in Chicoutimi. As to the block of low values, the PCDD and PCDF levels diminished upon approaching Chicoutimi in one case, and are similar in the other; an increase in B(a)P was apparent in both cases. The ratio of the concentrations noted at both locations indicates that the increase in B(a)P was greater in Chicoutimi than the increase in PCDDs and PCDFs when the wind blows west. When the wind blows from the east, the PCDD and PCDF levels were higher in Jonquière, as was the case for B(a)P. It is precisely when the wind blew mostly from the east, and that the station was downwind from the aluminum plant for 18 hours during the 24-hour sampling exercise, that we measured the highest values of B(a)P in Jonquière. The PAHs are not shown in Table 5, but present a behaviour similar to that of B(a)P.

Levels are also influenced by wind speed. Thus, on January 1, 2000, PCDD, PCDF and B(a)P levels were low at both locations, while the average wind speed was 23.6 km/h. Rousseau (2000) has illustrated that the PCDD, PCDF and PAH levels were inversely proportional to wind speed in Jonquière.

Discussion

We know that diesel trucks are responsible for 5% of PCDD and PCDF emissions in Canada, and the results obtained close to the Metropolitan highway in Montreal tend to prove the impact of motor vehicle traffic since levels there are high. We understand that no wood heating or industrial activities are known to release PCDDs and PCDFs in the vicinity of this station; in addition, the levels expressed in terms of TEQ did not increase between the end of the summer and the beginning of the winter. Motor vehicle traffic may be a source of PCDDs and PCDFs at stations 8 (Chicoutimi) and 9 (Jonquière), but its impact is not as important as that at the Montreal station since the volume of motor vehicle traffic is lower.

Wood heating, a popular activity in the Saguenay–Lac-Saint-Jean region, is certainly one source to be considered. We know for a fact that it is a source of PCDD and PCDF emissions, as shown by the results obtained in Jonquière's more residential neighborhood where levels increased between the end of August and the beginning of December. Bremmer *et al*. (1994) indicated that the PCDD and PCDF emissions from a wood stove may increase from 5 to 25 times if contaminated wood (painted, treated or other) is burned, rather than dry wood intended for wood heating. Launhardt *et al*. (1998) also mentioned that the combustion of painted wood or of a blend of plastic and wood generates up to 40 times more PCDDs and PCDFs than dry wood intended for wood heating. They also report that paper and cardboard combustion releases 2 to 20 times more PCDDs and PCDFs. The ashes also contain many more contaminants when waste or treated or painted wood is burned. Residential wood combustion represents 2% of the total PCDD and PCDF emissions in Canada.

The presence of metal reclaimers (or metal recycling firms) in Jonquière's industrial park may explain in part the higher values of PCDDs and PCDFS noted in the south section of Jonquière. Bremmer *et al*. (1994) reported that the heating or burning of electric cable to rid it of its plastic sheath releases approximately 40 µg TEQ of PCDDs and PCDFs per tonne (µg TEQ/t) of cable when conducted using the appropriate equipment. They believe that when this combustion is carried out illegally in makeshift installations, 500 µg TEQ/t, sometimes more, are released. Tiernan *et al*. (1989) measured the PCDDs and PCDFs at several locations in Dayton (Ohio), including one station located at approximately 300 m from a domestic waste incinerator (capacity of 150,000 tonnes/year) and at less than 1 kilometre from a plant that rid metals of PVC in a pyrolisis furnace. When only the incinerator was in operation, they measured an average of 3.36 pg/m^3 of PCDDs and PCDFs, while the level increased to 16.12 pg/m^3 when the pyrolisis furnace was in operation. The capacity of this plant's furnace was 182 kg per 9-hour period. Three metal reclaimers in Jonquière have already been convicted of having burned cable waste in makeshift installations, while another holds an authorization certificate issued by the MENV for a compliant installation intended for metal refusion (Labrecque, 2001).

In Jonquière, a firm repairing electric motors heats them in ovens to burn the varnish on the wiring. These ovens were installed without the MENV's authorization, and the MENV demanded that the company characterize its emissions and file a request for an authorization certificate in order to pursue its activities (Labrecque, 2001). Heating a motor in an oven to recover the metal compares with burning electric wires and Bremmer *et al*. (1994) identified this process as a source of PCDD and PCDF emissions to the air. This company's facilities are located south of the station used since 1989 by Environment Canada. In December, the PCDD and PCDF levels were similar in the pine needles planted at stations 3 and 5, located on either side of this recycler's facilities (while taking prevailing winds into account); however, levels were higher at station 5, closer to the motor recycler's facilities, when the levels were expressed in terms of TEQ. In August, the levels were too close to detection limits to draw a conclusion. Note that the winds blew from the south when we measured 2 high values in 1996, but that we didn't take any samples when the wind blew from this sector during the 2001 campaign.

Germain *et al*. (2000) showed that the station at Saint-Ignace-de-Loyola, located downwind from the iron and steel works in Sorel, was influenced by these works located at approximately 3.5 km. We know that the arc furnaces used in the production of steel release approximately 7% of the total PCDDs and PCDFs in Canada (Environment Canada, 2001). The levels found in the pine needles in Saint-Ignace-de-Loyola appeared to show the impact of Sorel's industrial activity on air quality in this region. The arc furnaces are the main source of PCDDs and PCDFs released by iron and steel works.

The high levels registered in rural areas in the summer may at first seem surprising, but burning waste in barrels or open fire burning is an important source of PCDD and PCDF emissions. Lemieux *et al*. (2000), from the US EPA, mentioned that burning waste in 220-litre (or 45-gallon) barrels releases between 6 and 264 µg of PCDDs and PCDFs per kg of waste, which is much more than the 0.0035 µg/kg released by a municipal waste incinerator burning 180,000 kg per day. They believe that between 40 to 50% of people living in rural areas in Illinois have adopted this practice and that burning waste in 2 to 40 barrels produces as many PCDDs and PCDFs in one day as does a municipal waste incinerator burning 180,000 kg. The Americans rate that burning waste in barrels, a procedure they call "backyard burning", produces close to 30% of all PCDDs and PCDFs released to the air in one year. In Switzerland, the federal Department of the Environment, Transport, Energy and Communication (DETEC, 2000) believes that the illegal burning of household waste represents 1 to 2% of incinerated waste, and that this type of burning causes the release of more than twice the total emissions released by household waste and special waste incinerators combined. These observations have prompted Environment Canada to include this source in the Canadian inventory; it represents slightly more than 10% of all PCDD and PCDF emissions (Environment Canada, 2001).

In Jonquière and Chicoutimi, the increase in PCDDs and PCDFs noted in the pine needles from the summer to the beginning of the winter could be explained in part by a longer exposure (145 days vs 50 days), but more particularly by the contribution of sources such as wood heating, open fire burning or "backyard burning", road transport and perhaps certain industrial sources, including metal recyclers and metal reclaimers. In Jonquière, the city's fire prevention service issues less than 20 permits per year for burning waste in barrels, but some people do so without requesting a permit (Laplante, 2001). When the results are studied on the basis of prevailing winds, the PCDD and PCDF levels in the pine needles appear to be slightly higher in the southern section of the Jonquière area.

Conclusion

In August, there are more PCDDs and PCDFs in the needles collected in Saint-Ignace-de-Loyola, in Montreal and at the pilot station located in a rural area. In December, the levels increase at all stations, except in Montreal where they remain similar to those recorded in the month of August. The period of exposure of the needles to air pollution may be the cause of the increase in levels over the levels measured in August, but no increases were noted at the pilot station located in Montreal, a station influenced by motor vehicle traffic. In the Saguenay, traffic is a probable source of PCDDs and PCDFs, but at a lesser level than in Montreal. Residential wood heating is probably an important source of PCDD and PCDF emissions to air in the Jonquière region. Open fire waste burning and certain industries, including metal reclaimers and metal recyclers that rid the wiring of electric motors of its plastic sheath or varnish may also release these substances during the combustion process. We know that three metal reclaimers in Jonquière have already been convicted of having illegally burned cables or electric wiring to recover the metal; another does it legally and an electric motor recycler has requested an authorization certificate from the MENV (Labrecque, 2001). The PCDD and PCDF levels were no higher in the pine needles taken from the pines planted on either side of this facility, but were higher at the closest station when the levels were expressed in terms of TEQ or toxic equivalent.

In this project, the levels of PCDDs and PCDFs present in the pine needles are similar to those measured in an urban environment in the United States and approximately 5 times lower than those measured close to wood preservation plants in the United States. They are up to 40 times lower than in Korea or in Japan where PCDDs and PCDFs are present in great quantity in the air.

In ambient air, there are no significant differences in PCDD and PCDF levels measured in Jonquière, Chicoutimi and Montreal, and the levels are similar to those recorded elsewhere in Canada between 1997 and 1999. On the other hand, the levels are approximately 10 times lower than those measured in the vicinity of a municipal waste incinerator and a reclaimer of PVC-coated electric wires in the United States, 20 times lower than those measured in Japan, or 40 times lower than those measured in Saxe, Germany.

Though PCDD and PCDF levels vary slightly depending on wind direction, they do not set out any significant trend since we measured high and low values for the same sector of origin. Since 1995-1996, the average levels diminished by approximately 50% in Jonquière and Montreal, and are 2 to 3 times lower than the MENV's revised annual guideline.

When we study the PCDD and PCDF levels measured in the needles and in ambient air, we notice that the pine needles are capable of absorbing PCDDs and PCDFs and of serving as bioindicators. The relationship between levels measured in the air and the needles is relatively constant, no matter the levels present in the air.

As regards the PAHs and the B(a)P, the levels are similar in Jonquière and Chicoutimi while they are lower in Montreal. In Jonquière, the values measured between July 2000 and March

2001 are lower than those reported by Dann in 1998, and since 1989, there has been a descending trend for PAHs and B(a)P, that is 96% for the B(a)P and 85% for the PAHs in ambient air in Jonquière. In Montreal, the reductions reach 27% and 60% respectively. In Jonquière, the most marked reduction was noted between 1999-2000 and 2000-2001, when Alcan modified the composition of the anodic paste used in its electrolytic cells. In Chicoutimi, the levels measured between July 2000 and the end of March 2001 are similar to those recorded in 1993-1994 by the MENV. Contrary to the PCDDs and PCDFs, the PAHs vary greatly depending on wind direction, since the highest values were measured when the stations were downwind from the aluminum plant. In fact, the highest value was recorded in Jonquière when the station was downwind from the aluminum plant for almost the whole day of sampling.

Recommendations

Among the probable sources of PCDDs and PCDFs in Jonquière, let us mention wood heating, open fire burning (or *back yard burning*), motor vehicle traffic, and certain industrial facilities. Given that the levels in Chicoutimi and Jonquière are similar to those in Montreal, that they are lower that the MENV criterion, and that regular sampling activities are pursued in Jonquière, we recommend the measurement of emissions stemming from the activities at the facilities of metal reclaimers and metal recyclers which recover metal by heating cables and electric motors. An awareness campaign urging better use of wood stoves and on the effects of backyard burning would also allow a reduction in the levels of PCDDs and PCDFs in the air.

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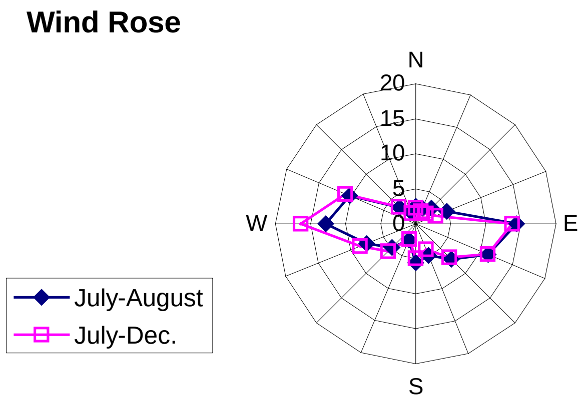
Annex 1 Sampling dates of PAHs, PCDDs and PCDFs in Jonquière, Chicoutimi and Montréal

	Jonquière		Chicoutimi		Montreal	
	PCDD/PCDF	HAP	PCDD/PCDF	HAP	PCDD/PCDF	HAP
00-07-05		X				
00-07-17						
00-07-29		X				X
00-08-10	X	X	X[2]	X[2]	X	X
00-08-22						X
00-09-03	X	X	X	X	X	X
00-09-15						
00-09-27			X	X		
00-10-09		X				X
00-10-21			X[3]	X[3]		
00-10-27			X	X		
00-11-02		X				X
00-11-14	X	X	X	X		X
00-11-26		X				X
00-12-08	X	X	X	X	X	X
00-12-20		X			X	X
01-12-26						X
01-01-01	X	X	X	X	X	X
01-01-13		X			X	X
01-01-25	X	X	X	X	X	X
01-02-06	X	X	Y[4]	Y	X	X
01-02-18	X[1]	X[1]	X	X		X
01-03-02	X	X	Y	Y	X	X
01-03-14	X	X	X	X	X	X
01-03-26	X	X	Y	Y	X	X

1 : Field blank
2 : results not available, problems during transport to laboratory
3 : sample lost following vandalism at the station, new sample on October 27
4 : Y : samples analysed by the MENV

Annex 2 Wind rose (%) noted in Jonquière for the periods July-end of August and July-December



Annex 3 Average recovery of products tagged with radioisotopes and added to the pine needles

Tagged Product	August		December	
	Average	Standard deviation	Average	Standard deviation
$^{13}C_{12}$-TCDD	74.2	7.6	63.2	8.0
$^{13}C_{12}$-TCDF	72.9	13.2	65.4	8.7
$^{13}C_{12}$-P5CDD	65.1	8.3	56.9	16.8
$^{13}C_{12}$-P5CDF	69.3	5.9	61.4	9.4
$^{13}C_{12}$-H6CDD	66.8	5.7	61.3	10.1
$^{13}C_{12}$-H6CDF	74.4	7.9	65.9	7.6
$^{13}C_{12}$-H7CDD	66.1	13.5	65.1	10.6
$^{13}C_{12}$-H7CDF	63.6	7.7	65.3	8.3
$^{13}C_{12}$-OCDD	49.2	9.5	59.6	8.3

Annex 4 Average recovery of products tagged with radioisotopes during the analysis of air samples

Tagged Product	Jonquière	Chicoutimi
$^{13}C_{12}$-TCDD	79.3	76.7
$^{13}C_{12}$-TCDF	86.3	79.3
$^{13}C_{12}$-P5CDD	80.1	76.2
$^{13}C_{12}$-P5CDF	79.1	75.7
$^{13}C_{12}$-H6CDD	84.1	76.9
$^{13}C_{12}$-H6CDF	86.9	80.3
$^{13}C_{12}$-H7CDD	77.2	71.7
$^{13}C_{12}$-H7CDF	79.0	73.3
$^{13}C_{12}$-OCDD	70.3	65.7

Annex 5 Percentage of moisture in pine needles

Station	Fall	Winter
	Percentage (%)	Percentage (%)
Station EC (1)	56.9	59.1
Station 2	55.5	55.9
Station 3	56.5	59.7
Station 4	54.9	58.0
Station 5	53.1	56.9
Station 6	57.9	57.2
Station 7	58.8	58.3
Station 7 (duplicate)	55.5	56.4
Station 7 (average)	57.2	57.4
Station 8	79.8	58.3
Station 9	57.0	58.4
Station 10	59.0	56.5
Station 11	58.5	57.7
Average (region)	58.8	57.7
Station 12 (SIL)	59.8	55.1
Station 13 (Montreal)	63.1	65.0
Control station	85.8	85.8

Annex 6 Average concentrations of PCDDs/PCDFs measured in the air in Jonquière, Chicoutimi and Montreal (pg/m^3)

	Chicoutimi				Jonquière				Montreal			
Congener	average	standard deviation	median	maximum	average	standard deviation	median	maximum	average	standard deviation	median	maximum
2378-TCDD	0.003	0.002	0.002	0.007	0.002	0.001	0.001	0.004	0.002	0.001	0.002	0.005
12378-P5CDD	0.013	0.013	0.006	0.040	0.008	0.008	0.006	0.026	0.010	0.006	0.009	0.019
123478-H6CDD	0.016	0.018	0.006	0.049	0.009	0.010	0.004	0.031	0.008	0.007	0.007	0.018
123678-H6CDD	0.031	0.047	0.012	0.170	0.014	0.017	0.005	0.053	0.014	0.011	0.013	0.030
123789-H6CDD	0.044	0.067	0.014	0.230	0.027	0.030	0.018	0.095	0.021	0.018	0.019	0.053
1234678-H7CDD	0.285	0.414	0.099	1.500	0.199	0.236	0.084	0.649	0.150	0.100	0.114	0.320
OCDD	0.869	1.472	0.363	5.400	0.547	0.646	0.213	1.931	0.426	0.278	0.370	0.862
2378-TCDF	0.062	0.044	0.044	0.140	0.050	0.064	0.019	0.210	0.030	0.018	0.026	0.076
12378-P5CDF	0.010	0.009	0.006	0.024	0.007	0.006	0.004	0.015	0.005	0.004	0.005	0.011
23478-P5CDF	0.015	0.013	0.012	0.039	0.013	0.013	0.006	0.035	0.009	0.006	0.008	0.019
123478-H6CDF	0.039	0.034	0.025	0.097	0.028	0.028	0.017	0.068	0.019	0.012	0.017	0.043
123678-H6CDF	0.015	0.014	0.009	0.045	0.011	0.010	0.007	0.027	0.007	0.004	0.007	0.015
234678-H6CDF	0.020	0.019	0.015	0.061	0.013	0.014	0.009	0.037	0.010	0.008	0.009	0.027
123789-H6CDF	0.004	0.005	0.002	0.016	0.002	0.001	0.002	0.004	0.002	0.001	0.002	0.004
1234678-H7CDF	0.065	0.057	0.047	0.190	0.052	0.042	0.039	0.118	0.041	0.023	0.036	0.080
1234789-H7CDF	0.008	0.009	0.004	0.029	0.007	0.006	0.004	0.020	0.005	0.004	0.003	0.014
OCDF	0.112	0.146	0.058	0.510	0.059	0.057	0.031	0.191	0.043	0.026	0.042	0.083
Total TCDD	0.045	0.046	0.026	0.140	0.037	0.048	0.011	0.124	0.022	0.029	0.010	0.086
Total P5CDD	0.125	0.146	0.053	0.400	0.071	0.100	0.024	0.314	0.063	0.066	0.038	0.182
Total H6CDD	0.427	0.693	0.125	2,400	0.210	0.253	0.111	0.812	0.152	0.137	0.141	0.378
Total H7CDD	0.687	1.247	0.180	4.500	0.439	0.547	0.163	1.489	0.300	0.202	0.237	0.593
OCDD	0.869	1.472	0.363	5.400	0.547	0.646	0.213	1.931	0.426	0.278	0.370	0.862
Total PCDDs	**2,104**	**3.501**	**0.617**	**12,720**	**1.302**	**1.455**	**0.572**	**4.110**	**0.962**	**0.685**	**0.831**	**2,058**
Total TCDF	0.265	0.205	0.185	0.713	0.179	0.210	0.062	0.609	0.149	0.121	0.104	0.476
Total P5CDF	0.165	0.131	0.127	0.380	0.125	0.134	0.052	0.346	0.091	0.063	0.086	0.216
Total H6CDF	0.159	0.142	0.127	0.410	0.104	0.101	0.070	0.271	0.080	0.050	0.068	0.186
Total H7CDF	0.102	0.077	0.097	0.240	0.085	0.068	0.077	0.217	0.065	0.036	0.060	0.123
OCDF	0.121	0.149	0.070	0.510	0.059	0.057	0.031	0.191	0.043	0.026	0.042	0.083
Total PCDFs	**0.781**	**0.585**	**0.599**	**1.631**	**0.552**	**0.538**	**0.278**	**1.365**	**0.426**	**0.270**	**0.376**	**1.057**
PCDDs + PCDFs	**2,885**	**3.840**	**1.293**	**14.040**	**1.854**	**1.593**	**1.719**	**5.016**	**1.388**	**0.927**	**1.207**	**2,912**
TEQ (equivalence factor)	**0.039**	**0.038**	**0.024**	**0.102**	**0.029**	**0.026**	**0.019**	**0.071**	**0.025**	**0.016**	**0.023**	**0.054**

Annex 7a Summary of the results of different PAHs measured in the air in Jonquière (n = 16) between July 2000 and March 2001 (ng/m^3)

	Detection frequency	Average arithmetic	Average geometric	Median	Minimum	Maximum
Acenaphthylene	100.0	4.897	1.825	2,875	0.161	21.843
Acenaphthene	100.0	14.144	2.706	3.117	0.088	68.907
Fluorene	100.0	12.933	4.291	4.996	0.312	69.111
2-Me-Fluorene	100.0	2.166	0.758	0.685	0.061	12.531
Phénanthrene	100.0	103.228	28.027	30.390	2.242	649.965
Anthracene	100.0	9.577	2.140	2.598	0.120	60.605
Fluoranthene	100.0	83.428	24.938	23.701	2.292	604.983
Pyrene	100.0	60.715	15.269	14.284	0.773	467.743
Benzo(a)Fluorene	100.0	5.120	1.077	1.210	0.031	43.954
Benzo(b)Fluorene	100.0	2.758	0.474	0.581	0.011	26.645
1-Me-Pyrene	100.0	1.736	0.400	0.555	0.009	13.105
Benzo(g,h,i)Fluoranthene	100.0	3.060	0.976	1.412	0.064	21.030
Benz(a)Anthracene	100.0	14.598	1.485	2.435	0.020	148.150
Chrysene	100.0	42.989	5.632	4.382	0.186	397.897
Triphenylene	100.0	9.288	1.923	1.222	0.098	78.291
7-Me-Benz(a)Anthracene	31.3	0.016	0.008	0.005	< l.d.	0.094
Benzo(b)Fluoranthene	100.0	61.246	7.678	5.680	0.179	514.584
Benzo(k)Fluoranthene	100.0	13.955	1.716	1.466	0.040	117.592
Benzo(e)Pyrene	100.0	30.012	3.495	2.668	0.077	253.674
Benzo(a)Pyrene	100.0	8.868	0.889	1.609	0.018	90.984
Perylene	81.3	1.316	0.254	0.321	0.009	10.953
3-Me-Cholanthrene	0.0	< l.d.	< l.d.	< l.d.	< l.d.	< l.d.
Indeno(1,2,3-cd)Pyrene	100.0	9.219	1.496	1.877	0.057	73.802
Dibenz(a,c)&(a,h)Anthracene	87.5	2.082	0.221	0.250	< l.d.	19.233
Benzo(b)Chrysene	62.5	0.637	0.077	0.101	< l.d.	5.365
Benzo(g,h,i)Perylene	100.0	10.273	1.552	2.095	0.048	84.993
Anthanthrene	56.3	0.238	0.060	0.105	< l.d.	1.684
TOTAL PAHs	100.0	508.290	121.119	113.905	8.772	3834.341

Annex 7b Summary of the results of different PAHs measured in the air in Chicoutimi (n = 12) between July 2000 and March 2001 (ng/m^3)

	Detection frequency	Average arithmetic	Average geometric	Median	Minimum	Maximum
Acenaphthylene	100.0	6.033	2.084	3.141	0.182	23.078
Acenaphthene	100.0	13.240	2.995	4.089	0.040	48.000
Fluorene	100.0	9.495	4.865	5.611	0.312	26.000
2-Me-Fluorene	100.0	0.939	0.538	0.572	0.054	3.432
Phenanthrene	100.0	41.960	25.087	26.945	1.363	123.388
Anthracene	100.0	2.715	1.560	1.887	0.094	9.728
Fluoranthene	100.0	34.739	23.967	29.451	2.698	97.236
Pyrene	100.0	24.799	16.149	19.833	1.792	73.481
Benzo(a)Fluorene	100.0	2.022	1.129	1.972	0.137	7.104
Benzo(b)Fluorene	100.0	0.916	0.473	0.868	0.054	3.505
1-Me-Pyrene	100.0	0.654	0.372	0.547	0.087	2.481
Benzo(g,h,i)Fluoranthene	100.0	1.597	1.026	1.356	0.190	4.184
Benz(a)Anthracene	100.0	5.730	2.470	4.286	0.145	20.567
Chrysene	100.0	3.625	2.112	3.004	0.250	12.689
Triphenylene	100.0	19.739	10.856	17.757	0.328	52.046
7-Me-Benz(a)Anthracene	11.1	0.009	0.006	0.006	< l.d.	0.044
Benzo(b)Fluoranthene	100.0	24.577	13.239	26.752	0.223	72.195
Benzo(k)Fluoranthene	100.0	7.540	3.625	6.589	0.053	22.000
Benzo(e)Pyrene	100.0	13.001	6.894	13.710	0.103	36.812
Benzo(a)Pyrene	100.0	4.357	1.730	3.179	0.019	15.210
Perylene	83.3	0.446	0.162	0.362	< l.d.	1.550
3-Me-Cholanthrene	0.0	< l.d.	< l.d.	< l.d.	< l.d.	< l.d.
Indeno(1,2,3-cd)Pyrene	100.0	4.180	2.364	3.840	0.074	11.468
Dibenz(a,c)&(a,h)Anthracene	83.3	0.923	0.379	0.835	< l.d.	2.672
Benzo(b)Chrysene	66.7	0.159	0.078	0.113	< l.d.	0.642
Benzo(g,h,i)Perylene	100.0	4.672	2.718	4.427	0.120	13.807
Anthanthrene	75.0	0.193	0.091	0.150	< l.d.	0.700
TOTAL PAHs	100.0	225.837	137.780	183.495	8.374	668.983

Annex 7c Summary of the results of different PAHs measured in the air in Montreal (Ontario and Amherst; n = 19) between July 2000 and March 2001 (ng/m^3)

	Detection frequency	Average		Median	Minimum	Maximum
		arithmetic	geometric			
Acenaphthylene	100.0	2.004	1.218	1.451	0.141	6.552
Acenaphthene	100.0	0.842	0.652	0.681	0.162	2.923
Fluorene	100.0	2.582	2.152	2.245	0.59	5.586
2-Me-Fluorene	100.0	0.912	0.758	0.706	0.235	2.334
Phenanthrene	100.0	9.543	8.223	7.827	3.66	23.218
Anthracene	100.0	0.670	0.549	0.556	0.204	1.881
Fluoranthene	100.0	2.963	2.593	2.499	1.212	7.085
Pyrene	100.0	2.090	1.805	1.778	0.786	5.698
Benzo(a)Fluorene	100.0	0.196	0.155	0.142	0.059	0.712
Benzo(b)Fluorene	100.0	0.093	0.074	0.074	0.024	0.302
1-Me-Pyrene	100.0	0.120	0.095	0.092	0.031	0.373
Benzo(g,h,i)Fluoranthene	100.0	0.348	0.279	0.237	0.117	0.962
Benz(a)Anthracene	100.0	0.304	0.203	0.192	0.036	1.297
Chrysene	100.0	0.878	0.627	0.781	0.123	3.502
Triphenylene	100.0	0.260	0.188	0.199	0.048	1.093
7-Me-Benz(a)Anthracene	5.3	0.005	0.004	0.005	< l.d.	0.011
Benzo(b)Fluoranthene	100.0	1.422	0.944	1.133	0.175	6.362
Benzo(k)Fluoranthene	100.0	0.365	0.254	0.263	0.049	1.519
Benzo(e)Pyrene	100.0	0.762	0.497	0.513	0.093	3.520
Benzo(a)Pyrene	100.0	0.299	0.185	0.203	0.037	1.174
Perylene	57.9	0.040	0.019	0.019	< l.d.	0.206
3-Me-Cholanthrene	0.0	< l.d.	< l.d.	< l.d.	< l.d.	< l.d.
Indeno(1,2,3-cd)Pyrene	100.0	0.467	0.349	0.420	0.075	1.749
Dibenz(a,c)&(a,h)Anthracene	84.2	0.059	0.038	0.044	< l.d.	0.292
Benzo(b)Chrysene	42.1	0.017	0.012	0.011	< l.d.	0.071
Benzo(g,h,i)Perylene	100.0	0.594	0.444	0.483	0.125	2.458
Anthanthrene	52.6	0.037	0.024	0.021	< l.d.	0.160
TOTAL PAHs	100.0	27.890	23.905	22.169	9.112	73.774