

Chronic Disease and Early Exposure to Air-Borne Mixtures. 2. Exposure Assessment

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This work is part of a larger study of the impact of early exposure to releases from industry on the etiology of cancer. Releases from all kraft and sulfite mills, coke ovens, oil refineries, copper, nickel, and lead/zinc smelters operating in Canada during the exposure period of 1967–1970 have been determined. All plumes have been expressed in μg BaP eq/d using the RASH methodology. The releases have been divided into process, boiler fuel, dioxin, and SO_2 emissions. Combustion sources have been defined with FIREv6.23. Dioxin congeners are expected in all source types when the boiler fuel is heavy fuel oil, wood or wood bark, or coal. All ~ 90 communities examined have an inverted sex ratio.

Introduction

This work is part of a larger study examining the impact of early exposure to air-borne mixtures of industrial point sources. The end point is one of 18 rare and/or poorly understood cancers. In earlier work the origin of the study, the characteristics of the Environmental Quality Database (EQDB), and the approach used were discussed (1).

Two types of output from the EQDB are possible: a survey examining all sources of one industry type extant in Canada during the exposure period or an examination of one source. This work will address exposure assessment in the first case, for all kraft and sulfite mills, coke ovens, oil refineries, and copper, nickel, and lead/zinc smelters. The work supports the conclusion that, after solving the confounders of mobility and latency, the risk associated with early cancers may be considerable. An estimate was made attributing 5–15% of cancers to a cause of chronic chemical exposure to the physical environment (2).

Plumes from processes, boilers, and external generators may all contribute to the toxic burden released from a source to the adjacent community. The solution adopted for the EQDB to accommodate the multiplicity of mixtures is to use a relative potency method, called RASH, based upon benzo[a]pyrene (BaP) (3–8). The RASH approximation describes a mixture of chemical species emitted from any source, with a toxicologically weighted index, the relative potency, in terms of a reference compound, BaP. The mixture then is in units of BaP equivalents (BaP eq) and can be dispersed, added, or subtracted as a single substance.

$$rp_{\text{test}} = \frac{\text{dose}_{\text{BaP}}}{\text{dose}_{\text{test}}} \quad (1)$$

The relative potency of a test compound compared to BaP is [1], with the rp of BaP defined equal to 1. The relative potency of an individual chemical in a biological test is the ratio of the dose for the reference chemical divided by the dose for the test chemical at the same end point eq 1. Equation 1 is rearranged for the i th chemical in a mixture of n chemicals in amounts estimated with an emission factor, EF_i , and relative potency, RP_i , when fuel is consumed, to obtain eq 2.

$$\text{dose}_{\text{BaP}} = \text{fuel} \times \sum_{i=1}^{i=n} RP_i \times EF_i \quad (2)$$

The considerable advantage of RASH is that it involves a relatively simple process to form a quantitative estimate of dose in a mixture of chemicals. Plumes from several subsources gathered physically around a point source can be added together algebraically, as vectors, and dispersed as one.

Analysis of the point source is to identify process emissions and how the subsources are fueled. FIREv.6.23 for a defined Standard Combustion Code (SCC) provides a list of releases with emission factors. The reader can refer to ref 14 for relative potencies. Boiler fuels are important sources of chemical risk.

Methods

Cases and controls reside, at least 1 year between 1967 and 1970, within 25 km of kraft and sulfite process pulp mills (SIC = 2611), coke oven operations (SIC = 3312), oil refineries (SIC = 2911), primary copper (SIC = 3331), primary nickel (SIC = 3339), and primary lead/zinc operations (SIC = 3332) and were identified in the EQDB. Cases developed a cancer by 1993–1995. The case:control ratio is 3.902. The study area is 25 km from the source.

Exposure is to a surrogate source consisting of a mill or smelter or refinery that has a capacity equal to the 95th percentile of the industry during the exposure period. It is assumed to operate at that capacity. Auxiliary power sources, boilers, etc. correspond to a source combustion code (SCC) defined in FIREv.6.23 (9). They are fueled as defined and operate at the 95th percentile of fuel consumption. The surrogate has one boiler for each fuel type used in the industry. All oil refineries use at least one flare, and this is considered with the auxiliary power sources as a source of exposure. When the types of fuel and their amounts have been determined, the likely releases are estimated using data from FIRE for typical SCC types. Spreadsheets were prepared for several fuels named with the SCC code (no. 6 heavy oil, wood/bark, coal, flares) to calculate reported emissions in terms of MT/d and in reduced format, μg BaP eq/d.

All chemicals identified by FIRE are assumed released from the stack and treated with the RASH methodology (3–8) to reduce the plume to one in BaP equivalents. It is usual that ~ 40 to $\sim 90\%$ of the emissions from a source have a relative potency defined. The plumes from each source are the process stream; sulfur oxides; combustion products from fueling boilers; and combustion products from oil refinery flares.

Plumes are dispersed with a Gaussian plume model, using 15 kph winds and 75 m stack. According to Environment Canada 15 kph winds represent the 85th percentile of winds in all 480 long-term wind stations across Canada (10). Plume ground-level concentration is in μg BaP eq/m³. Cases are

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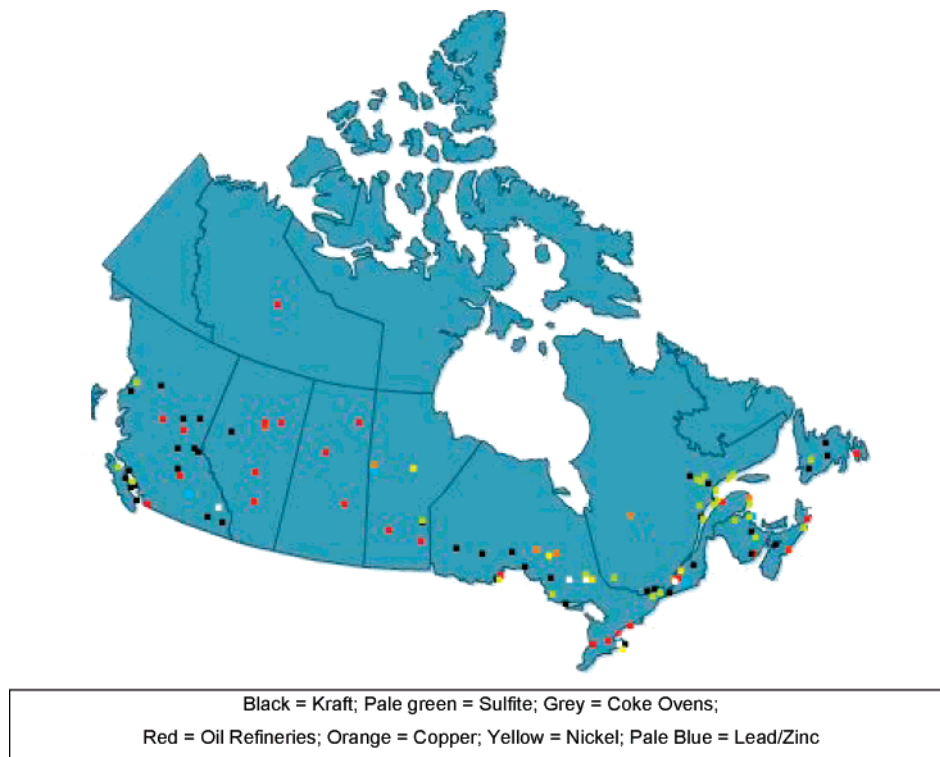


FIGURE 1. Distribution of source-types in 1967–1970.

identified every kilometer from 1 to 25 km. Figure 1 shows a distribution of source types in 1967–1970.

Kraft and Sulfite Process Pulp Mills. Inventories of kraft and sulfite pulp mills, in 1968, are from the Pulp and Paper Canada annual directory (11). The median value of kraft mill capacity was 1291 MT/d. The maximum capacity of sulfite mills was 1094 MT/d. The 95th percentile of sulfite mill capacity was 834 MT/d. Kraft process pulp mills digest wood chips in an acidic solution of sodium sulfate to separate the wood-fiber from lignin. Sulfite process pulp mills digest wood chips in a strongly alkaline solution of sodium, potassium, or ammonium bisulfite to separate the wood-fiber from the lignin. Kraft process mills are chemically intensive in recovering the sulfur, have very strong odors from organosulfur compounds, and are perceived as presenting significantly more health risks than sulfite process mills.

This study excludes kraft and sulfite mills in Quebec and New Brunswick as noted in part 1 (1). A study of boiler fuel use within the industry found the following: 22 of 27 kraft mills used an average of 646.95 bbl/d heavy fuel oil, each; 3 of 27 mills used an average of 242.43 MT/d coal, each; 13 of 27 mills used 7.14 Mcf/d natural gas, each; and 23 or 27 mills used 225.41 MT/d wood waste, each to fuel their boilers.

The same study found the following: 2 of 16 sulfite mills used 269 MT/d of coal, 13 of 16 mills used, on average, 1034.08 bbl/d heavy fuel oil, 4 of 16 mills used 12.44 Mcf/d natural gas, and 13 of 16 used 178.29 MT/d of wood waste each, to fuel the boilers (12).

Emissions for all kraft stages (blackstock, brownstock, recovery, smelt, DCE, NDCE) are from industry (13) and/or FIREv.6.23. Lime kiln releases could not be generalized. A separate survey (22) found the average particulate emission from lime kiln is = 0.725 kg/MTADP (metric tonnes air died pulp); the average Total Reduced Sulfur (TRS) emission is 0.12 kg/MTADP; and the average SO₂ emission was 0.255 kg/MTADP. The TRS is not speciated, is very complex, and is one of the main sources of odors in Kraft Process mills. Process releases for the surrogate kraft and sulfite mills are in Table 1.

SCC 10100401 was used to evaluate combustion product releases from heavy oil (no. 6) fueled equipment. SCC 10100901 was used to evaluate combustion product releases from wood/bark fueled equipment. SCC 10200701 was used to evaluate combustion product releases from natural gas fueled equipment. SCC 10200202 was used to evaluate combustion products from coal fueled equipment. In Table 1, boiler fuel types and amounts have been estimated from a survey of industry by Environment Canada in 1972 (12). The NCASI report (13) confirms the fuel used in the boilers is no. 6 fuel oil or wood/bark waste.

Coke Ovens. The inventory of coke oven operations in Canada in 1967–1970 is from government publications (14–20). The majority of the coke produced during the exposure period was used in blast furnaces. In two locations, in Quebec, municipal carbonization of coal produced coke as a byproduct. Total capacity of the industry in 1968 was 9 778 000 MT per year of coal. Coke ovens heat bituminous coal at about 800+ °C for about 17–18 h in the absence of air to drive off the volatiles, leaving a carboniferous residue that is used in the production of pig iron from iron ore. The environment within the coke oven is chemically neutral with SO₂ present with H₂S, NH₃ and NO_x, CO₂ with CH₄.

Emissions from coking contain extensive amounts of polycyclic aromatic hydrocarbons (PAHs). A partial list of constituents in coke oven emissions has been prepared for the Ontario Ministry of Labor (21). Data do not include emission factors, and the release from a coke oven battery is approximated for this work with the combustion of coal using an external combustion boiler, pulverized bituminous coal with a dry bottom (SCC = 10200202).

The maximum capacity of coke oven batteries in the inventory is 2 973 000 MT/y of coal. The median capacity is 1 952 000 MT/y (5346 MT/d). It takes on average 1.4 tons of coal to produce 1 ton of coke (22). Using this multiplier the median amount of coal used during the exposure period in the surrogate is 8246 MT/d.

Coal, freshly mined or aged and weathered, contains chloride as HCl or metal salts (23). Under these conditions

TABLE 1. Surrogate Capacity, Releases in μg BaP eq/d from Seven Source Types, 1968–1970

	kraft pulp	sulfite pulp	coke ovens	refineries oil	copper smelter	nickel smelter	lead+zinc smelter
maximum cap. MT/d	1200	1094	7397	19713 m3/d	1828	1797	471+898
95th pct. cap. MT/d	686	357	1836 coke	13101 m3/d	243	239	212+254
Process Emissions $\mu\text{g}/\text{BaP}$ eq/d Exclude CO, NO_x, and SO_x							
process μg BaP eq/d	1.72E+14	5.10E+09	3.1E+14	5.68E+13	9.21E+11	6.15E+11	2.91E+11
95th hydrocarbon MT/d	3.406	0.182	741	284.6			
95th SO ₂ release MT/d	3.773	10.37	2.22	222.3	5572	4994	131.5
SO ₂ release μg BaP eq	7.92E+11	2.18E+12	4.66E+11	267E+13	5.3E+14	4.76E+14	1.26E+13
95th NO _x release MT/d				214.4			
Fuel Used and Emissions							
no. 6 fuel oil L/d	102852	164398		90072	89993	34731	172085
μg BaP eq/d Oil/d	1.51E+12	2.41E+12		4.86E+12	4.87E+12	1.88E+12	9.31E+12
natural gas Mcf/d	6200	8350		2387	4517	1008	7986
μg BaP eq/d Gas	8.06E+13	1.09E+14		3.10E+13	5.87E+13	1.31E8+13	1.04E+14
wood bark MT/d	225.41	178.29					
μg BaP eq/d wood	4.73E+09	3.74E+09					
coal MT/d	242.43	269.09	2577 coal				
μg BaP eq/d coal	2.70E+12	5.67E+12	1.06E+14				
loss to flare (5%S)M3/d				235620			
μg BaP eq/d flare				5.34E+10			
CO Mt/y oil use	20.29	32.59		19.71	19.70	7.60	37.67
NO _x MT/y oil use	190.71	306.34		185.18	185.18	71.47	354.10
NO MT/y oil use	44.63	71.70		43.34	43.34	16.23	82.87
SO _x MT/y oil use	9.57	15.35		9.29	9.29	3.59	17.78
Dioxin Congeners μg BaP eq/d							
octachloro (oil)	2.25E+03	2.93E+04	1.34E+05	8.37E+03	8.37E+03	3.23E+03	1.60E+04
heptachloro (coal)	2.55E+03	2.81E+03	2.69E+04				
pentachloro (coal)	1.36E+03	1.50E+03	1.44E+04				
tetrachloro (coal)	2.81E+03	3.12E+03	2.94E+04				
TCDD (wood/coal)	5.72E+03	6.55E+03	6.08E+04				
polychloro (wood/coal)	5.34E+.04	5.92E+04	5.65E+05				
total	6.81E+04	7.61E+04	8.31E+05	8.37E+03	8.37E+03	3.23E+03	1.60E+04

any coke oven will release dioxin congeners. The presence of chloride in the burning organic matter is a necessary and sufficient condition to ensure the production of a range of dioxins, including 2,3,7,8-TCDD (24–27). The emission rate of dioxins from coking during the exposure period is in Table 1.

Petroleum Refineries. Source inventories of petroleum refineries in 1967–1970 are from the industry weekly newsletter, *Oilweek* (28). We are concerned here only with oil refineries. Refineries in Regina SK, Calgary AB, Brandon MB, Saskatoon SK, St. Boniface MB, and Kamsack SK stopped operations in the late 1960s and early 1970s and have been included in this study when they operated at least 1 year during the exposure period. They have been evaluated with their flare.

Refinery capacity is the amount of crude processed in a calendar day. There were 41 refineries operating in 1968 (28), with a total capacity of ~193 000 m³/d and 37 operating in 1978, with a capacity of 383 323 m³/d. The maximum capacity in 1968 was 19 713 m³/d. The 95th percentile capacity in 1968 is 13 101 m³/d.

The Petroleum Association for Canadian Environment (PACE) (29) reported the earliest inventory of releases. Release figures for 1968 are prorated from the 1978 report, based on the ratio of the crude capacity of the refinery in each year. The report estimated total annual release of sulfur oxides in 1978 was 163 833 MT/y SO₂, referred to as an “improvement”. This prorates to 651 529 MT/y in 1968. The report estimated total annual release of nitrogen oxides of 39 398 MT/y NO_x, prorated to 78 290 MT/y NO_x in 1968. The report estimated hydrocarbon releases at 52 281 MT/y hydrocarbons in 1978, prorated to 103 891 MT/y hydrocarbons in 1968. Reported hydrocarbon releases for process heaters are 366 MT/y in 1978, prorated to 728 MT/y fuel for process heaters in 1968. This equates to 1 171 500 L/y.

PACE reported on the volume lost to flaring for the first time in 1983 (30). In that year there were 28 refineries operating in Canada. The following is from the report (p 13): *Eight of the 28 refineries were unable to report their flare losses. Fourteen had significant losses relative to their throughput. Total losses to flare from 15 refineries were 123.69 million m³ annually. Assuming a molecular weight of 20 for this gas its weight is 110 302 tonnes. Feed charged to these 14 refineries was 41.2 million m³ in 1983. The weight of the feed charged was 34.6 million tonnes. The ratio of flared loss to feed input is 0.0032, i.e., 0.32%.*

The efficiency of a flare burning hydrocarbons in the open was not investigated with any rigor or quality control until 1996 (31). Approximately 150 chemical species were identified in either sweet or sour flares. The study determined the combustion efficiency (CE) ranged downward toward ~65% or less and was strongly and inversely dependent on wind-speed and dilution (32, 33). Raw fuel is released in progressively larger amounts as the CE falls. This is due to the basic process of combustion. Combustion takes time, as much as tens or hundreds of seconds. During this time the droplet of fuel can diffuse away from the combustion zone and escape the flame altogether (34, 35).

A stochastic analysis was conducted in the PACE 83 report ((30) Figure 4, hydrocarbons burned in flare) and found that the percent of feed flared in these refineries in 1983 had a median value of 0.551% and a 95th value of 1.658%. The value observed in the PACE 83 report, 0.32%, corresponds closely to the mode, or most probable value, of the distribution. The deterministic, single-valued approach of the PACE 83 report used a molecular weight (MW) of 20, where the stochastic approach, used here, infers a MW of 35 for the median and a MW of 103 for the upper limit. The median and 95th percentile values were applied to estimate the volume lost daily to flaring in the 41 refineries operating in

TABLE 2. Emission Rates $\mu\text{g BaP eq/s}$ for Release Streams - All Sources^a

	$\mu\text{g BaP eq/d}$						
	kraft pulp	sulfite pulp	coke ovens	refineries oil	copper smelter	nickel smelter	lead-zinc smelter
process	1.99E+09	5.90E+04	3.59E+09	6.84E+06	1.07E+07	7.12E+06	3.37E+06
burners - oil	1.07E+10	1.86E+10		5.63E+07	5.64E+07	2.18E+07	1.08E+08
burners - gas	3.41E+08	2.45E+09		3.59E+08	6.79E+08	1.52E+08	1.20E+09
burners - wood	3.33E+04	2.24E+04					
burners - coal	8.45E+06	1.16E+07	1.23E+09				
all burners	1.10E+10	1.89E+10	1.23E+09	4.15E+08	7.36E+08	1.73E+08	1.31E+09
SO ₂	9.17E+06	2.52E+07	5.39E+06	2.55E+09	6.13E+09	5.51E+09	1.46E+08
dioxin congeners	6.95E+00	3.27E+00	9.61E+00	9.69E-02	9.69E-02	3.74E-02	1.85E-01

^a Dioxin congeners included in oil, coal, and wood streams.

TABLE 3. Heavy Metals in Smelter Air Emissions Prorated to 1970 kg/d

smelter	As air	Cd air	Cr air	Hg air	Ni air	Pb air	total air $\mu\text{g BaP eq/d}$	SO ₂ air MT/d
lead	55.17	52.51	0.80	11.50	0.80	403.98	6.51E+10	131
zinc	134.69	174.53	18.30	56.04	10.80	860.48	2.26E+11	incl. Pb
copper	674.81	243.54	20.69	43.37	2409.81	3884.97	9.21E+11	5572
nickel	238.18	5.50	5.74	0	3824.56	641.42	6.15E+11	4994

1968. This estimate relies on the published volume capacity of the refineries for feedstock, available from the industry newsletter, and is described in the Supporting Information.

Strosher (31) quantitatively identified ~150 chemicals in the plume from a sour flare, in units of mg/m³. When the percent of feed flared is 0.551%, the average volume loss to flare is 28 747 m³/d with a 95th C.I. of 78 924 m³/d. When the percent of feed flared is 1.624%, the average volume loss to flare is 84 728 m³/d, and the upper 95th C.I. is 235 620 m³/d. The worksheet (provided in the Supporting Information) calculates the estimated amounts of the known chemical species and the dose, in $\mu\text{g BaP eq/d}$, released by flaring daily into the surrounding community for these two limits. When the volume loss to flaring is 78 924 m³/d, the plume transports at least 7.18E+10 $\mu\text{g BaP eq/d}$. When the volume loss to flaring is 235 620 m³/d, the plume transports at least 2.12E+11 $\mu\text{g BaP eq/d}$ into the community.

Copper, Nickel, and Lead Smelters. The inventory of smelters in 1970 is available in government of Canada publications (36–39). In this study exposure from smelting lead-zinc ore affects residents of the Belledune NB, Flin Flon MB, and Trail BC. The smelting of copper-nickel ores affects residents of Flin Flon MB, Thomson MB, Sudbury ON, Murdochville QC, and Noranda QC. Base metal refineries are in Port Colbourne ON (Ni), Montreal (Zn), Valleyfield QC (Cu), and Fort Saskatchewan AB (Ni). The Fort Saskatchewan refinery uses a hydrometallurgical process using sludges and effluents and has no emissions to air, using sludges and effluents. A smelter in Timmins ON started a few years after the exposure period. Production and releases are available (40).

During the 1970s and early 1980s a Priority Substance List (PSL) consisting of releases As, Cd, Cr, Ni, Pb, and Hg was prepared to provide a more complete picture of the type and amount of chemical releases for smelters (41). An assessment of heavy metal releases by pathway including air, water, etc. clearly demonstrates emissions from all the smelters are absorbed by all living matter lying in the path of the plume (42, 43).

PSL metals in wastes from smelters are available for 1988 (41). Pathways include releases to air, liquid effluents, sludges, and solid waste excluding slag and tailings. Because exposure conditions in this study include exposure up to 25 km, an

air path is required. PSL releases of heavy metals to air, prorated to 1970, based on the metal production amounts in 1970 and 1988 are in Table 3. These releases represent the toxic effect of the process stream.

Sulfur dioxide, from roasting the ore, is recovered to produce either sulfuric acid or liquid SO₂ or both, but there are sulfur dioxide releases to air over and above these streams and these are in Table 3. As an example of local contamination, a study in Trail, BC found heavy metals deposited from the lead/zinc smelter in town, in residential, park, and sandbox soils. There was a significant correlation between lead and arsenic, copper, cadmium, mercury, silver, and zinc levels in all three soils (44).

Fuel use within the smelting industry is available for 1970 from Statistics Canada (45). To break down the data, the total fuel has been prorated according to annual production of metal within the sector compared to all smelters to determine the amount of each fuel used in each sector. Within each sector the fuel has been prorated according to capacity for heavy fuel oil and natural gas. The estimated quantity of no. 6 fuel oil (L/d) and natural gas (Mcf/d) attributable to copper-nickel and lead-zinc smelters is in Table 1. SCC 10100402 was used for heavy fuel oils, and SCC 10200601 was used for natural gas to estimate the release in Table 1.

Results

The results of the calculations for each source-type are in Table 1. Emission rates, in $\mu\text{g BaP eq/s}$, in Table 2 are for the corresponding term in Table 1. Rates for kraft and sulfite pulp have been adjusted to account for the prevalence of burner fuels. Emission rates for all the burners have been summed into a composite “all burners” to simplify dispersion. Dioxin congeners are found when the fuel is coal, oil, or wood.

Discussion

The population in each of the ~90 communities where these source-types are located has been reviewed with the 1991 census. The male sex ratio, $M/(M+F)$, in all cases is inverted from ~0.51. This is a sign of chronic exposure to dioxins in these communities (46–49) and provides an internal marker. A spectrum of dioxin congeners form when organic material is burned in the presence of chloride. Corollary effects

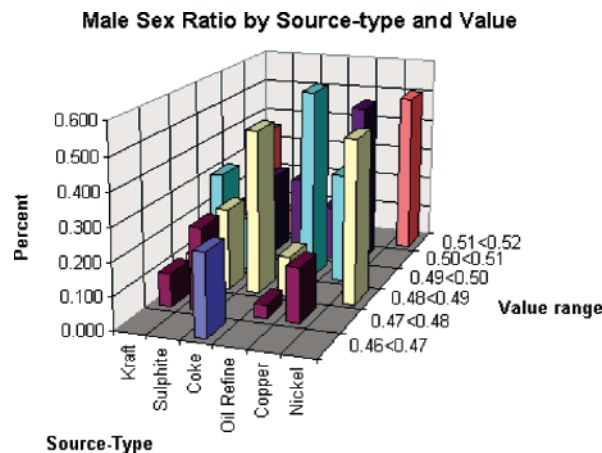


FIGURE 2. Male sex ratio by source-type and value.

associated with chronic exposure to dioxins include elevated cancer rates, elevated rates of diabetes, selected heart diseases, and kidney disease.

Figure 2 illustrates the distribution of male sex ratio, $M/(M+F)$, as a function of the source-type. The normal, biological value, established by genetics, is 0.51 ± 0.01 . Inverted (<0.50) sex ratios are widely observed in the 90 communities of this study. This infers chronic exposure to dioxins. There were insufficient communities with lead/zinc smelters to be included: the single source had a sex ratio of 0.4961.

Looking at all instances of a source is valuable when a factor that is consistently present from one location to another is not presenting a dramatic effect in any one location. The example is DMS, formed within the plume itself (50–52). It is potentially present in a small amount. Its effect may not have been observed before, because the ability to look at all instances of a source-type was not available. Statistically, the introduction of a new chemical species, that is also a carcinogen, into the plume introduces the potential for “tail generation probability effects” in the distribution of cases (53), including more risk at greater distances from the new source.

The operations of each source-type can be generalized with the RASH process. RASH is an ideal procedure, to examine, rigorously, how emissions from these seven industry types interact with the surrounding population (see Figure 3). The reduced plume in $\mu\text{g BaP eq}$ is an exact measure of dose. RASH can be extended to other pathways, including water treatment, potentially providing a basis for cumulative exposure assessment. This ability is extended to all of the communities where the industries operate and potentially affect several million persons.

Multiple exposures to comparably sized source-types, both the same and different, can be identified within the EQDB when considering one example of a source-type. The subject of multiple exposure has been ignored when considering all examples of a source-type. In general different source-types in proximity involve the following: 1. Kraft and sulfite process streams in the same physical establishment are widespread in the industry. 2. Pulp mill and oil refinery sources in proximity are found in Western Canada viz. Kamloops, Prince George. 3. Coke oven and coal-fired thermal generation in proximity is found in Sydney NS. In Nanticoke ON, the same two sources include, in addition, a very large oil refinery. 4. Refineries with thermal generating station nearby: This is found in Alberta viz. Edmonton. 5. Multiple sources of the same type in close proximity, viz. three steel mills in Calgary, AB, two kraft pulp mills in Welland, ON, and three pulp mills (2 kraft and 1 sulfite) in Thunder Bay.

CALCULATE RELATIVE POTENCY (RP) OF A TEST CHEMICAL

Search all available Toxicology literature viz.

RTECS, ATSDR, Toxline, Medline, DART, HSDB, IRIS etc.

for records of bio-tests using the test chemical and records using BaP in the same bio-test. BaP is used as a reference in about 200+ different bio-tests.

$$\text{Eqn A } \text{RP} = \text{dose BaP} / \text{dose test chemical}$$

Repeat this procedure for all the bio-test result found from the literature search for the test chemical

Apply stochastic analysis to the distribution of RP to

find the 25th, 50th, and 75th percentiles

Use 50th as a conservative estimate of RP

From Eqn A above

$$\text{Eqn B Dose BaP} = \text{Dose test chemical} \times \text{RP}$$

When the dose of test chemical is determined with an emission factor, EF, applied to a fuel used

then

$$\text{Eqn C Dose test chemical} = \text{EF} \times \text{Fuel used}$$

And

$$\text{Eqn D Dose BaP} = \text{Fuel used} \times \text{RP} \times \text{EF}$$

For a plume of chemicals from the same release and fuel

$$\text{Dose BaP} = \text{Fuel used} \times \sum \text{RP} \times \text{EF}$$

FIGURE 3. Schematic of the process of getting a relative potency for RASH.

TABLE 4. Distribution of Source Types Geographically Includes Copper, Nickel, and Lead/Zinc Refineries

province	kraft	sulfite	coke	refineries	copper	nickel	lead
Newfoundland		2		1			
Nova Scotia	1	1	1	2			
New Brunswick	3	5		1			2
Quebec	11	11	2	6	3	1	1
Ontario	8	9	3	8	3	5	
Manitoba	1			2	1		
Saskatchewan	1		1	7			
Alberta	1			6			
British Columbia	15	3	1	7			1
total	41	31	8	41	7	6	4

These all represent areas where subjects will have multiple exposures that cannot be addressed in a survey approach but are easily addressed with a single primary source and secondary sources of varying source-type.

Table 4 shows the distribution of these seven source-types throughout Canada in 1968–1970. Sources in Quebec and New Brunswick were not included at the request of their separate governments in the early 1990s when the EQDB was under construction.

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Supporting Information Available

Spreadsheets of SCC 10100401 heavy oil; SCC 10100901 wood/bark; SCC 10200202 bituminous coal; SCC 10200601 natural

gas; PACE 83.xls; and Flares.xls for all the known releases, except particulates, associated with a known combustion code (SCC) and fuel in FIRE and selected as potentially representative for the purposes of this study. [The amount of each fuel type is entered at the top and is converted into the units demanded by the emission factor, to produce an estimated release of each of the chemical species, in mg/d or kg/d. For the chemicals that have been evaluated for RASH, the value of the median relative potency, rp, appears as a column heading, and finally an estimate of the $\mu\text{g BaP eq/d}$ released. These are collected at the top and appear in bold red type. Two spreadsheets, PACE 83 and Flares, complete the calculation of the estimated loss to flare for each refinery in 1968.] This material is available free of charge via the Internet at <http://pubs.acs.org>.

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