

Sources of Dioxins and Furans in British Columbia

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A recent Canadian study of the levels of polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) in adipose tissue suggested higher levels in British Columbians than in other Canadians. This led to concerns about the sources of these compounds in B.C. In this paper, we review scientific publications to generate a list of possible sources of PCDDs and PCDFs. We outline which of these are likely to be found in British Columbia, and develop hypotheses concerning their potential contribution to an individual's "PCDD/PCDF load". Further sampling and analysis of British Columbians' adipose tissue could test hypotheses about occupational and environmental exposures to dioxins and furans.

Une récente étude canadienne portant sur les niveaux de polychlorodibenzo-paradiioxines (PCDD) et de polychlorodibenzofurannes (PCDF) dans les tissus adipeux suggérait des teneurs plus élevées chez les gens de la Colombie Britannique que chez les autres canadiens. Ceci souleva des inquiétudes quant à la provenance de ces composés en Colombie Britannique. Dans cet article, nous faisons une revue des publications scientifiques afin de dresser une liste des sources possibles de PCDD et de PCDF. Nous soulignons lesquelles de ces sources sont susceptibles de se retrouver en Colombie Britannique et développons des hypothèses sur leur contribution potentielle à la "teneur de PCDD/PCDF" d'un individu. D'autres échantillonnages et analyses des tissus adipeux des gens de la Colombie Britannique permettront de vérifier ces hypothèses portant sur l'exposition aux dioxines dans l'environnement et dans le milieu du travail.

Chlorophenols, phenoxy acid herbicides, polychlorinated biphenyls, and related chlorinated aromatic hydrocarbons are widely used industrial chemicals. Many of these chemicals are contaminated with polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs).

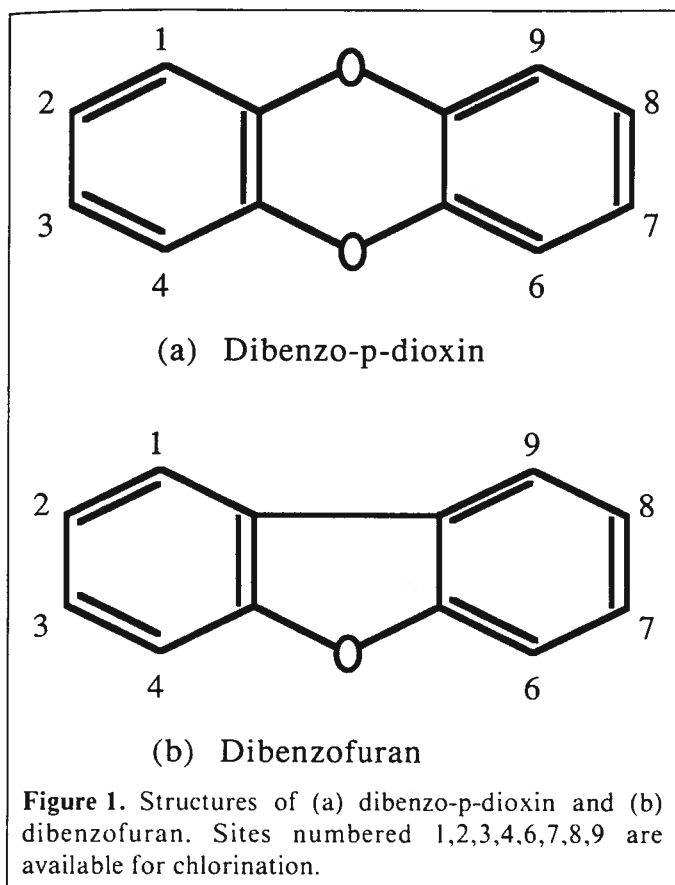
PCDDs and PCDFs are tricyclic aromatic compounds exhibiting similar chemical and physical properties¹ (Figure 1). From one to eight chlorine atoms can be substituted onto the rings, making a total of 75 dioxin and 135 furan congeners possible. Dioxins and furans with one to three chlorine atoms are generally considered to be relatively non-

toxic, while those with four chlorines in the lateral (2,3,7,8) positions are most toxic. Toxicity then decreases with increasing chlorine substitution.²

Based on animal studies, 2,3,7,8-tetrachlorodibenzo-p-dioxin has been suggested to be "the most toxic synthetic substance known to man".³ Since epidemiologic evidence to support this contention has not yet emerged, this compound can best be considered "the most toxic substance known to guinea pig", with an oral LD₅₀ for females of 0.0006 mg/kg of body weight.⁴ LD₅₀s for rats are higher, at 0.022 mg/kg for males and 0.045 for females.⁴ Some PCDDs and PCDFs are known to be carcinogenic to animals.^{5,6}

Human health effects found after exposure to chemicals contaminated with PCDDs and PCDFs include anorexia, chloracne, skin pigmentation changes, liver disease, psychological abnormalities, neurologic signs, hypertension, hyperlipidemia, hypercholesterolemia, alopecia and hirsutism.^{7,8} There have also been reports of reproductive

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disfunction, birth defects such as cleft palate and kidney malformations,⁹ and cancers such as soft tissue sarcoma.¹⁰

Concerns about adverse health outcomes have led to surveys of PCDD and PCDF concentrations in human tissues.¹¹⁻¹³ A Canadian study¹¹ reported PCDD and PCDF levels in fat samples taken from 66 deceased Canadians ranging in age from 14 to 88 years; 14 of these were taken from residents of British Columbia. The results suggested that the levels in British Columbia residents might be higher than the Canadian norm, and prompted this investigation.

Contaminated Chemicals and By-products of Production

1. Chlorophenols

Chlorophenols have been widely used since the 1930s as fungicides, slimicides, insecticides, and bactericides.⁵ They are produced industrially either by the direct chlorination of phenol, or by hydrolysis of chlorobenzene. PCDDs in commercial chlorophenols have ranged in concentration from 0.2 ppm tetraCDD to 690 ppm of octaCDD.²

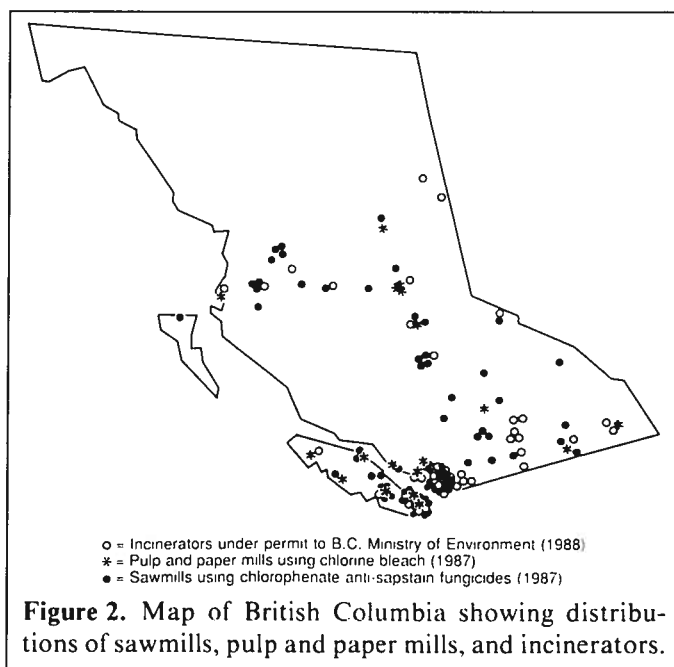
Until 1988, chlorophenols were the fungicide of choice for wood treatment in B.C. where an estimated 1100 tonnes per year were used.² A 1987 survey conducted by Environment Canada reported 95 sawmills throughout British Columbia using chlorophenols for anti-sapstain treatments (Figure

2).^{14,15} These chemicals are applied by hand-spraying, drive-in dip tanks, and low and high pressure spraying. Occupational exposures of sawmill employees are likely to be the most significant human exposures to PCDDs and PCDFs from contaminated chlorophenols. The exact dose would depend on the extent of contamination of the chlorophenol formulations used, the wood-treatment methods, and site industrial hygiene practices.

Other B.C. residents may be exposed by contact with treated lumber (approximately 25% of treated wood is sold to local markets, the rest is exported); stormwater runoff;^{14,15} air emissions from incinerators;¹⁶ and wood waste sent to pulp mills. Chlorophenols have also been applied to many other materials including leather goods, masonry, cooling-tower water, rope, paints, adhesives and textiles. B.C. residents may be exposed to PCDDs and PCDFs through industrial and domestic use of such products.

2. Other Pesticides

Phenoxy herbicides such as 2,4-dichlorophenoxyacetic acid (2,4-D), and 2,4,5-trichlorophenoxyacetic acid (2,4,5-T) are another significant source of dioxin input to the environment. TetraCDD and other PCDDs are formed during their production from chlorophenols. Current allowable levels of 2,3,7,8-tetraCDD in 2,4,5-T are 0.1 ppm of the technical grade, a standard instituted in 1971 in the U.S. and in 1975 in Canada. Gross contamination of up to 100 ppm probably existed prior to 1970.¹⁷ In 2,4-D, di-, tri-, and tetra-dioxin isomers have been found, but the toxic 2,3,7,8-tetraCDD has not. A 1980 sample of 2,4-D used in



Canada contained 66 ppt tetraCDD, 180 ppt triCDD, and 68 ppt diCDD.¹⁸ The current maximum permissible level of any isomers of PCDD in 2,4-D is 10 ppm.¹⁶

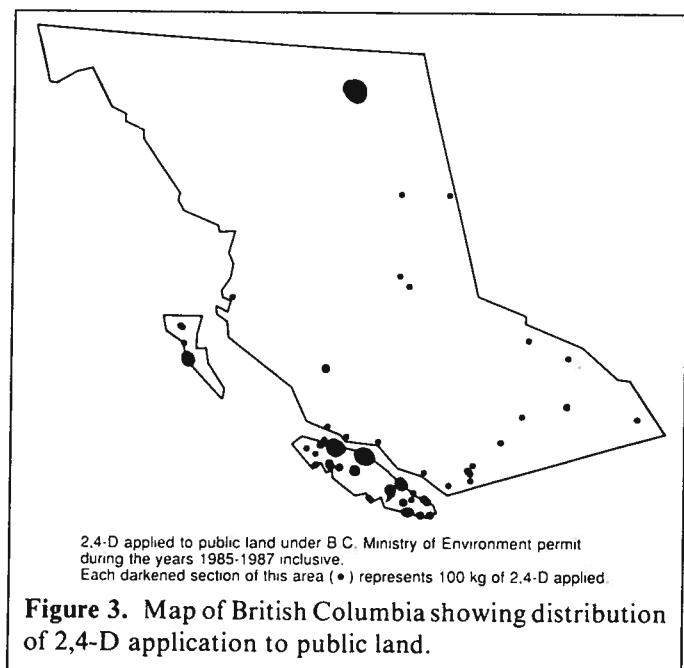
The Pesticide Control Branch of the B.C. Ministry of Environment and Parks regulates pesticide applications on Crown land. Currently 95 chemicals are in use, of which tetrachlorophenols, pentachlorophenols, and 2,4-D are very likely to contain dioxins. Many other pesticides are suspected to be contaminated, for example Dicamba² and MCPA. Still others, though probably not contaminated, contain chlorine and benzene rings and may therefore be PCDD and PCDF precursors in the event of fire.

Approximately 16,000 kg of 2,4-D active ingredient were applied to crown land from 1985 to 1987 inclusive (Figure 3). The B.C. Ministry of Environment does not regulate or record the use of pesticides on private land (including agricultural land) which accounts for an estimated 75% of the total pesticide volume (other than wood preservatives) sold in B.C.

B.C. residents may be exposed to PCDDs and PCDFs in herbicides and pesticides through occupational uses, domestic uses, direct ingestion of sprayed plants, and ingestion of contaminated organisms higher in the food chain.

3. By-products of pulp and paper production

A pulp mill's wood supply may be contaminated with chlorophenols used in the sawmill industry. Wood supplied via ocean transport may be salty, thus contributing an increased chlorine load. Wood itself contains lignins, which provide hydrocarbon structures that can act as PCDD/PCDF precursors.



The bleach plant has been identified as the major source of PCDDs and PCDFs in the Kraft pulp mill, specifically the chlorination stage of the bleaching process.¹⁹ Oil-based defoamers added to unbleached pulp during washing and screening contain unchlorinated dioxins and furans in concentrations sufficient to account for some of the observed pulp mill PCDD/PCDF.²⁰ The bleaching stage chlorine can also react with other organic compounds, including lignins in the pulp, to produce PCDDs and PCDFs.

There are 21 pulp mills currently operating in B.C., 18 of which use chlorine (Cl₂) to bleach their product (Figure 2). To minimize PCDD/PCDF formation, many B.C. mills have been considering bleaching methods which reduce chlorine use, including oxygen bleaching and increased substitution of chlorine dioxide.

Dioxins and furans are carried out of pulp mills as part of the liquid effluent. Effluent treatment practiced by pulp mills ranges from none to secondary treatment, depending on the requirements of provincial waste disposal permits. The effect of these traditional treatments on PCDDs and PCDFs is largely unknown. Armendola¹⁹ reported that treated pulp mill effluent in the U.S. had levels of 2,3,7,8-tetraCDD that ranged from < 0.003 to 0.12 ppt and levels of 2,3,7,8-tetraCDF from < 0.007 to 2.2 ppt.

Pulp mills are thus potential sources of occupational exposures as well as general population exposures, since PCDDs and PCDFs from the effluent enter the aquatic food chain. Direct contact with contaminated paper products probably poses a very minor risk of dioxin exposure. Marketplace²¹ tested five Canadian paper products for PCDD and PCDF levels. They reported values ranging from 0.39 ppt total PCDD/PCDF in coffee filters to 19.57 ppt in paper towels.

4. Polychlorinated biphenyls

Polychlorinated biphenyl (PCB) preparations were once widely used in capacitors and transformers, casting waxes, heat transfer fluids, and plasticizers in paints, hydraulic oils, sealants, etc. PCB mixtures have been found to contain PCDF with four to seven chlorine atoms, primarily the tetra- and pentaCDF isomers. For example, Arochlor 1248 was found to contain 330 ppb 2,3,7,8-tetraCDF and 830 ppb pentaCDF, which are among the most toxic isomers.⁹ PCDDs have not been detected in PCB mixtures,²² but are produced when PCBs are heated or burned,²³ as in transformer fires.

An inventory of PCB use is maintained by the Environmental Protection Service of Environment Canada. In B.C. in 1985, about 28% of PCBs were used by utilities, 43% by the forest industry, and 15% in mining.²⁴ About 22% were in capacitors, 63% in transformers, and 13% in storage for disposal.²⁴

When environmental concerns about polychlorinated biphenyls became widespread in the late 1970s, their use was

limited (Environmental Contaminants Act, 1977). The use patterns at B.C. Hydro, the major provincial electric utility company, provide an example. As part of a 10-year program to remove PCBs, all PCB transformers maintained by B.C. Hydro have been removed from service, and are being stored at the company's storage sites throughout the province²⁵ until a disposal method is approved. About 10% of B.C. Hydro mineral oil transformers are believed to be contaminated with from 50-250 ppm PCB. Many B.C. Hydro capacitors still contain PCBs. However they use a more easily biodegraded form, and most capacitors are in environmentally less risky substation locations.

Because PCB use in B.C. is supervised and decreasing, it is not likely a major current environmental source of furans, although it may have been in the past. PCBs are an even less likely source of dioxins. Occupational exposures of capacitor maintenance crews, substation employees, electricians, and others to PCBs and their contaminants may still occur.

5. *Pharmaceuticals*

Many medical, dental, and cosmetic products have used the trichlorophenol derivative hexachlorophene as a bactericide. It has been shown to contain 200 to 500 ppm 2,3,7,8-tetraCDD¹⁷ plus other PCDD and PCDF isomers. The hexachlorophene-containing products were often applied directly to the skin, through which PCDDs and PCDFs are easily absorbed. Hexachlorophene has been linked to birth defects in children of hospital workers,²⁶ with PCDDs the suspected cause. As a result, its use has now been almost eliminated.

Products of Combustion

Various combustion processes have been suggested as significant sources of PCDD and PCDF, with three possible mechanisms. PCDDs and PCDFs may exist as contaminants in fuel, and may be released as the material burns. PCDDs and PCDFs might be formed by thermally initiated reactions such as condensation or dechlorination of compounds present in the material. Finally, PCDDs and PCDFs may be synthesized "de novo" whenever the appropriate elements, such as carbon, hydrogen, chlorine, and copper or other metal catalysts, are combined under appropriate conditions.²⁷ The variables thought to affect the rate of formation of PCDDs and PCDFs during combustion are: molecular structure of the fuel; chlorine content of the fuel; presence of precursors and contaminants; reaction temperature; residence time; efficiency of air-fuel mixing; air-fuel ratio; and fuel feed size.²⁸ Combustion temperatures in the range of 140 to 400°C favour PCDD and PCDF production, while two seconds residence time at 800 to 1000°C will destroy most PCDDs and PCDFs.²⁹

1. *Incineration*

Approximately 0.6 tonnes of garbage per person per year are incinerated in Canada.¹⁶ Fly ash precipitated from

municipal incinerators in Ontario has been found to contain 10-170 ppb of adsorbed PCDDs.^{2,30} The most toxic 2,3,7,8-tetraCDD generally represents less than 5% of the total PCDDs.²⁷ Plastics such as polyvinyl chloride are a major source of organochlorine compounds in municipal waste and may provide precursors for PCDD and PCDF formation.²³

In British Columbia, there are 45 incinerators monitored directly by the Waste Management Branch, Ministry of Environment and Parks, including sewage and solid municipal waste incinerators, pathological incinerators at hospitals, and some industrial facilities (Figure 2). This does not include incinerators regulated by the Greater Vancouver Regional District. Information is not available on other incinerators, such as "beehive" burners used to dispose of sawmill waste, and apartment waste incinerators. It is estimated that there are 300 apartment-based incinerators in the Vancouver area alone. The majority of these units are of old or poor design, use no supplementary fuel, and are very likely sources of PCDD and PCDF emissions.

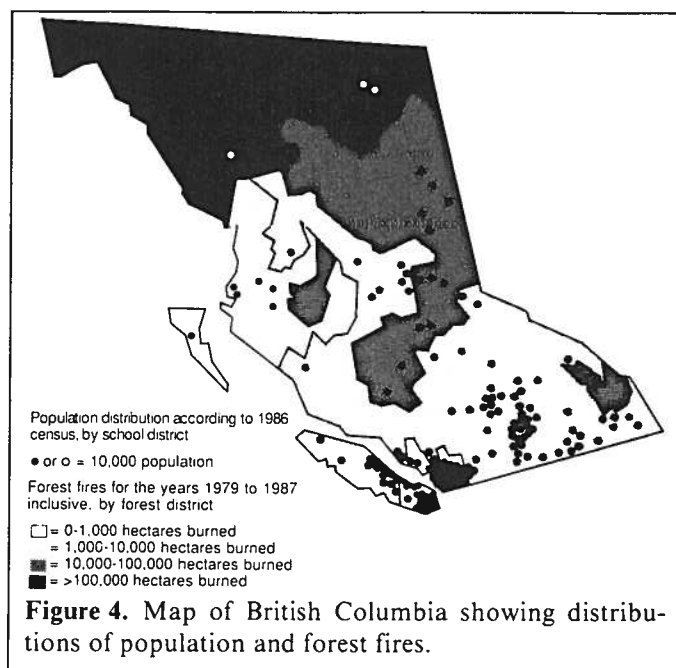
In March 1988, the Greater Vancouver Regional District started using a state-of-the-art mass burning incinerator designed to burn 800 tonnes of municipal waste per day. These incinerators employ supplementary fuel and appear to minimize PCDD/PCDF production because they operate at high temperatures with high efficiency burning.²³

Without specific data on the whereabouts of most incinerators, it has been assumed that their numbers are directly related to population density. The population of B.C. as of the 1986 census, classified by school districts, was obtained from the Ministry of Economic Development (Figure 4).

2. *Wood Burning*

It has been suggested that the trace amounts of chlorine found naturally in wood may be sufficient to allow dioxin and furan formation to occur. If this does happen, it would suggest that PCDDs and PCDFs are ubiquitous contaminants in the environment. This "trace chemistries of fire" hypothesis originated with scientists from the Dow Chemical Company in 1978. Efforts to test the hypothesis have had mixed results, however overall the data suggests that PCDDs and PCDFs are, indeed, formed during wood burning.³¹

Since forest fires represent large scale combustion of wood under uncontrolled inefficient conditions, it is conceivable that they might represent a significant input of dioxins and furans to the environment. The combustion of 2,4,5-T^{32,33} and other 2,4,5-trichlorophenoxy compounds³² has been shown to produce dioxins, an important consideration when forest fires occur in areas which have recently been sprayed with herbicides. However, the temperatures of forest fires can reach 1200 to 1400°C, temperatures sufficient to degrade a large percentage of any PCDDs and PCDFs formed.



Forest fire records were obtained from the B.C. Ministry of Forests and Lands for the years 1979 to 1987. The total area burned per region is shown schematically on Figure 4. It is apparent from the map that forest fires have been most extensive in the less populated regions, that is, in areas lacking many other probable sources of PCDDs and PCDFs.

3. Automobile Exhaust

Marklund et al.,³⁴ in co-operation with the Automobile Emission Laboratory of the National Swedish Council for Environmental Protection, found that automobiles burning leaded gas produced 0.05 to 0.3 nanograms of 2,3,7,8-tetraCDD/kilometer. Other tetra and penta isomers were also produced. PCDDs and PCDFs were not detected in the exhaust of automobiles burning unleaded gas. It is believed that the dioxin production is linked to the use of halogenated scavengers such as dichloroethane in leaded gasoline.³⁴ The impact of this source of dioxins and furans in B.C. is, like incinerators, expected to be directly related to population distribution.

Chemical Wastes

Sites containing chemical waste, such as chemical waste dumps, landfills, and contaminated industrial sites may serve as point sources for PCDD and PCDF emissions. No inventory of chemical waste sites is available for B.C. Without more detailed information about the number and location of contaminated waste sites, the contribution of these sources to a person's dioxin/furan load may be best estimated as secondary to that of the point sources and

population-based sources from which the waste is generated. For example, many sawmills have recently stopped using chlorophenols, but may not have cleaned up dip tank drip areas or treated lumber storage areas, nor disposed of remaining chlorophenols. Flyash precipitated from incinerators and boilers is almost exclusively sent to landfill. Transformer oils contaminated with PCDFs were probably also disposed of in landfills at one time.

Contaminated Food

Ingestion of PCDD/PCDF contaminated food likely represents the most significant exposure source for the general population. Canadian estimates suggest a daily intake of 1.52 pg of 2,3,7,8-tetraCDD equivalents per kg of body weight.³⁵

Due to their lipophilic nature and their slow metabolism and excretion, dioxins and furans are readily concentrated in the food chain. The above-ground parts of plants can be contaminated by chemical spraying, dry deposition of particulates from air, and by capturing PCDDs and PCDFs volatilized from the soil. Dioxins and furans tend to be absorbed by the waxy outer layer of plant leaves, and cannot simply be washed off with water. Plants such as potatoes, carrots, onions² and mushrooms¹⁶ have been shown to bioconcentrate PCDDs and PCDFs found in soil and water. PCDDs and PCDFs have also been found in cattle raised on rangeland sprayed with 2,4,5-T, and in poultry bedded on wood shavings from chlorophenol-treated wood.¹⁶ As a result, restrictions have been made regarding the use of contaminated chemicals and chlorophenol-contaminated materials for growing these products.

The bulk of available data regarding PCDD and PCDF levels in food is about aquatic life such as fish and crabs. Swedish data showed crabs with 2-124 ppt in hepatopancreas, and levels 50-100 times lower in the edible muscle.³⁶ Residues of 2,3,7,8-tetraCDD in crab hepatopancreas from B.C. ranged from none detected to 662 ppt.³⁷ Other less toxic dioxin congeners ranged from none detected to 24,968 ppt. No 2,3,7,8-tetraCDD was detected in 50% of the fin fish sampled and most levels were 3 ppt or less. Fish contamination may be a particular concern for persons consuming a great deal of this food source, for example native people for whom special fishing rights have been preserved. Health and Welfare Canada has proposed 20 ppt as the maximum allowable dioxin level in fish for human consumption.

Some cooking processes have also been suggested to produce dioxins, such as barbequing and smoking, but this has not been verified.³⁸

PCDDs and PCDFs have very low solubilities in water, 0.2 µg/L for 2,3,7,8-tetraCDD at 25°C,³⁹ but it is possible that suspended particles in unfiltered water could contain adsorbed dioxins and furans.

It has been estimated that adults passively ingest 10-20 mg of soil per day, and children 100 mg per day.⁴⁰ The

TABLE I
Occupational Exposures to Dioxins and Furans

Industry	Type of Job	Exposed to by
Forestry	Wood Treatment	spraying or dipping of lumber handling sorting chemical storage preparation equipment maintenance trimming and drying of wood shipping
	Pulp and Paper Production	handling treated wood waste mill effluent finished product handling
	Forest Maintenance	pesticide application chemical storage preparation equipment maintenance forest fire attack crews
Pesticide	Formulation	mixing, packaging
	Application	mixing, spraying, dusting
Agriculture	Farming	pesticide application chemical storage preparation equipment maintenance treated wood in fences
Miscellaneous	Firemen	combustion products
	Incinerator workers	combustion products
	Waste disposal site workers	contaminated waste
	Linemen, electricians	capacitors, transformers, coolants
	Construction workers	paints, stains, adhesives
	Healthcare workers	hexachlorophene disinfectants

strong tendency for dioxins and furans to bind to organic carbon in the soil makes this another potential exposure route for humans. Soil likely contains higher concentrations of PCDDs and PCDFs than air or water,¹⁶ but less soil is ingested.

CONCLUSIONS

Many occupations and industries in B.C. have the potential for dioxin and furan exposures which are higher than those in the general population (Table I). Occupational exposures would occur primarily through dermal contact and inhalation, and secondarily through ingestion (as a result of eating lunch or smoking with contaminated hands at the worksite). The potential environmental routes of exposure to dioxins and furans are listed in Table II. The general population of British Columbia is expected to be exposed to dioxins and furans primarily by eating contaminated foodstuffs. To a lesser extent, they may absorb these chemicals by inhaling PCDD- and PCDF-laden particulate matter from the air, and absorbing these chemicals through the skin by touching contaminated products.

It should be possible to test the validity of these sources by sampling tissue from B.C. residents, and comparing variations among population subgroups. The following

TABLE II
Environmental Exposures to Dioxins and Furans

Source	Main Exposure Route	Affected Population
Contaminated food: fish, shellfish, root vegetables, meat, fowl, fruits, other vegetables, breastmilk	Ingestion	All food consumers, with possible excesses for native people, fishermen, people liv- ing in coastal areas or near contamination sources (mainly pop- ulous areas)
Contaminated water	Ingestion	People living near con- tamination sources (mainly populous areas)
Point sources: incinerators sawmills pulp mills	Inhalation Ingestion	People living near con- tamination sources (populous areas for incinerators; specific locations for mills)
Area sources: pesticide spray areas automobile emissions forest fires	Inhalation Ingestion	People living near con- tamination sources (populous areas for auto emissions; agri- cultural and forestry areas for pesticides; northern B.C. for forest fires)
Household products: treated lumber pesticides paints/stains adhesives leather/textiles disinfectants paper products	Dermal absorption Inhalation Ingestion	Consumers of contami- nated products

examples illustrate how comparisons might test the predictions made in this paper:

- The role of occupational exposures can be verified by collecting work histories from tissue-sample subjects and determining if high PCDD and PCDF levels correspond to the predicted high exposure occupations.
- The impact of sawmills and pulp mills can be evaluated by examining differences in concentrations in residents of areas with similar population densities, with and without these point sources.
- The importance of incinerators, automobile emissions and forest fires can be verified by comparing PCDD and PCDF levels between residents of populous areas with residents of remote areas.

Once the sources of PCDD/PCDF in the B.C. population have been verified, the relationship between differing dioxin exposures and mortality or morbidity could be assessed using traditional epidemiologic methods, such as case-control studies.

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