

# Estimation of PCB Stocks, Emissions, and Urban Fate: Will our Policies Reduce Concentrations and Exposure?<sup>†</sup>

MIRIAM L. DIAMOND,<sup>\*,†,§</sup>  
LISA MELYUK,<sup>§</sup> SUSAN A. CSISZAR,<sup>§</sup>  
AND MATTHEW ROBSON<sup>†</sup>

*Department of Geography and Program in Planning, University of Toronto, Canada, Department of Chemical Engineering and Applied Chemistry, University of Toronto, Canada*

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PCBs, used to manage risks from the flammability of dielectric fluids and to increase the durability of elastic sealants, had declining environmental concentrations after legislation banning new production was passed during the 1970s and 1980s in Europe and North America. To answer why PCB temporal trends are now nearly stable and if current policies will further reduce concentrations and our exposure, we estimated PCB stocks in Toronto, Canada (population of ~2.5 million) of 437 (282–796) tonnes, of which 97 and 3% are in closed sources and building sealants, respectively. The greatest geographic density of PCBs is downtown, specifically in commercial, electricity-intensive skyscrapers. An unknown stock is within now-buried landfills and other waste-handling facilities as well as diffuse sources such as electrical wiring and paints. Using the Multimedia Urban Model, we estimated city-wide emissions of ~0.14–1.4 mg m<sup>-2</sup> y<sup>-1</sup> or 35–350 mg capita<sup>-1</sup> y<sup>-1</sup> of ΣPCB<sub>70</sub>, which is ~0.01–0.3% annually of total documented stocks. Canada, as one of 159 signatories of the Stockholm Convention and the 35 parties that have reported progress toward environmentally sound management of their PCB inventories by 2028, has passed national legislation with a timetable of inventory reductions. It is unclear whether this legislation will successfully reduce concentrations and exposures, however the analysis should inform our management of other contaminants.

## Introduction

PCBs are inextricably part of our urban infrastructure. As legacy Persistent Organic Pollutants (POPs), they have been slated for elimination under the Stockholm Convention (1). They were a technical success story after World War II, at which time the use of PCBs in dielectric fluids, elastic sealants, flame retardants, and other applications (Table S1 in the Supporting Information) facilitated the expansion of safe electrical service and new architectural building designs in rapidly expanding cities. Over the past decades, our appreciation of the advantages of PCBs has been outweighed

by our growing knowledge of their disadvantages. We know that PCBs are distributed worldwide and will remain so for decades because of their persistence. Our understanding of their toxicology has migrated from acute effects due to occupational exposure such as chloracne, headaches, and dizziness, to ever more subtle neurotoxicological, immune, reproductive, and metabolic effects (e.g., 2, 3).

PCBs remain a current concern, some 30 years after their manufacturing was banned by many countries. Concentrations in air, soil, sediment, and biota declined rapidly in the decade following this ban, but have remained stubbornly consistent or have declined slowly since then (e.g., 4). Today, PCBs remain ubiquitous in our food supply from terrestrial and aquatic sources (5, 6).

This paper asks whether policies aimed at PCBs will be effective at reducing concentrations and hence exposures. We address the question by examining the status of PCBs in the City of Toronto, Canada and policies promulgated at national and international levels. Cities, in general, have higher concentrations of PCBs than rural and remote locations (7, 8) and within cities “hotspots” in PCB air concentrations can be found near transformer storage yards and landfills (e.g., 9–11). Whereas earlier research ascribed the global persistence of PCB concentrations in air to secondary releases from sediment and soil reservoirs (12, 13), more recent evidence points to current releases of “fresh Aroclor” PCBs from urban sources supporting local and global air concentrations (14, 15). Most recently, “non-aroclor” PCBs such as PCB 11 have been measured in air and water where their source has been traced to production of diarylide yellow and other pigments (16). These observations are consistent with the relationship between PCB air concentrations in North America to population density (17).

Canadian legislation in 1977 banned production and importation of PCBs, and their use in any nonelectrical products manufactured after 1977. The legislation banned PCB use in any electrical products manufactured after 1980, but permitted their continued use in electrical equipment manufactured prior to 1980. Although PCBs were not manufactured in Canada, 40,000 tonnes were imported between 1929 and 1977. Environment Canada has identified numerous types of materials containing PCBs that were used in Canada between 1929 and 1977, primarily in the form of lubricants, plasticizers, and dielectric fluids (18) (Table S1 in the SI).

Below we briefly review the history and uses of PCBs in Canada focusing on “Aroclor PCBs”. We quantify known stocks and aggregate emissions in addition to quantifying fate in Toronto using the Multimedia Urban Model or MUM-Fate (19). We end by discussing the implications of our present situation—daily emissions of PCBs, policy and legislative progress, and whether our policies will successfully reduce PCBs in our environment.

## PCB Inventory for Toronto, Canada

**Methods.** *PCBs in Closed Sources.* A PCB inventory for Toronto was compiled from data contained in Canada’s national inventory of current PCBs in closed use and storage that was mandated as part of the 1977 legislation (Table S2). The national inventory lists sites of PCB use or storage from voluntary reporting by the PCB owners supplemented by limited inspections from federal compliance officers. The inventory is incomplete because of the mostly voluntary nature of reporting and the limited categories for PCB classification. The five categories for classification are: in-use askarel, waste askarel, in-use PCB-contaminated mineral

\* Corresponding author email: miriam.diamond@utoronto.ca; tel: +1 416 978 1586; fax: +1 416 946 5992.

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<sup>§</sup> Department of Geography and Program in Planning.

<sup>§</sup> Department of Chemical Engineering and Applied Chemistry.

oil, waste PCB-contaminated mineral oil, and other PCB wastes. Askarels are insulating fluids containing PCBs that were commonly used in electrical transformers. Mineral oil is a petroleum byproduct that was also used as an insulating fluid in transformers that would come into contact with PCBs in the askarel fluid during transformer use. PCBs compose 40–80% of askarel fluid and ~0.1% of mineral oils (20). For the Toronto inventory, the mass of pure PCBs was calculated from the masses of PCB-containing fluids in the five categories listed in the 2006 national inventory at the above concentrations (Table S3). High and low inventory estimates of pure PCBs were calculated using high and low ends of the range of askarel concentrations. Further details are in the SI.

**PCBs in Sealants.** Our inventory comprised polysulfide sealants that were used in building construction and not other open sources (Table S1) because sealants are the second major current use of PCBs, with PCBs in electrical fluids being the first (21, 22). PCBs were added to joint sealant materials to improve their flexibility, increase their resistance to mechanical erosion, and improve adherence to other building materials (23). Reported concentrations at which PCBs were added to sealants range from 50,000–300,000 mg kg<sup>-1</sup> (24–26). Concentrations in which PCBs have been found in aged sealants range from 0.56–550,000 mg kg<sup>-1</sup> and many exceed the regulatory limit of 50 mg kg<sup>-1</sup> (22, 27, 28).

We sampled and analyzed sealants in 80 Toronto buildings constructed from 1945 to 1980 (29). The geometric mean concentration was 4630 mg kg<sup>-1</sup>, with a maximum concentration of 82,100 mg kg<sup>-1</sup> detected in one building. Fourteen percent (10 out of 70) of the buildings had sealants that exceeded 50 mg kg<sup>-1</sup>.

These measured concentrations were used to estimate the stock of PCBs in sealants in Toronto based on the number and size of buildings constructed from 1945 to 1980. Building volumes were estimated for each of the 44 wards in the City of Toronto from building data, including construction date, number of storeys, and building type (30, 31). Single and semidetached residential properties were excluded because we found that they did not contain significant amounts of sealant compared with larger concrete buildings (29). The mass of pure PCBs in building sealants was calculated as the product of total building volume per ward, an average of 55 g of sealant per m<sup>3</sup> of building volume (22, 32), and the average PCB concentration of 4630 mg kg<sup>-1</sup>. Lower- and upper-bound estimates were calculated using PCB concentrations of 50 mg kg<sup>-1</sup> for buildings constructed 1945–1980, and 82,103 mg kg<sup>-1</sup> for buildings constructed 1950–1980. All estimates of sealant mass were multiplied by the 14% of buildings assumed to contain sealants.

**PCB Inventory. Closed Sources.** According to the 2006 national database, the City of Toronto, with an area of 635 km<sup>2</sup>, had 455 sites containing PCBs in closed sources, with a total of 850 tonnes of PCB material (>50 mg kg<sup>-1</sup> PCBs). The equivalent mass of pure PCBs is 424 (282–565) tonnes (Table 1). Seventy of the 455 sites of PCB use/storage were in the 24 km<sup>2</sup> south central downtown core which is the city's financial district (Figure 1). The estimated inventory downtown has a total gross weight of 364 tonnes PCB material and ~181 tonnes of pure PCBs.

The mass of in-use PCBs was proportional to the volume and electrical demand of a building hence the large downtown skyscrapers have the largest in-use volume. Other locations containing in-use PCBs were along the public transportation corridors and in public buildings including schools. PCBs in sensitive locations such as schools and hospitals accounted for 9% of the total closed stock in 2006.

The geographic concentration of 43% of city-wide PCBs in Toronto's downtown financial district runs contrary to our perception of PCBs as industrial chemicals. As Diamond and Hodge (33) commented, many first world cities have

**TABLE 1. Summary of PCB Stock in Toronto in Tonnes**

	gross weight of PCB material	mass of pure PCBs		
		best estimate	low estimate	high estimate
closed sources	692	345	230	460
closed storage	158	79	52	105
amount removed since January 2000	348	173	116	231
joint sealants*	2817	13	0.14	231
<b>total stock</b>	<b>3667</b>	<b>437</b>	<b>282</b>	<b>796</b>

\* This assumes that only 14% of the total joint sealants used in Toronto from 1950–1980 contain PCBs.

shifted their economies from industrial to commercial activities, thereby shifting PCB sources, at least in Toronto, from “smokestacks” to those associated with commercial and service enterprises. However, given that one skyscraper can house close to 10,000 people and consume several billion kWh, perhaps we should not be surprised at their PCB stocks.

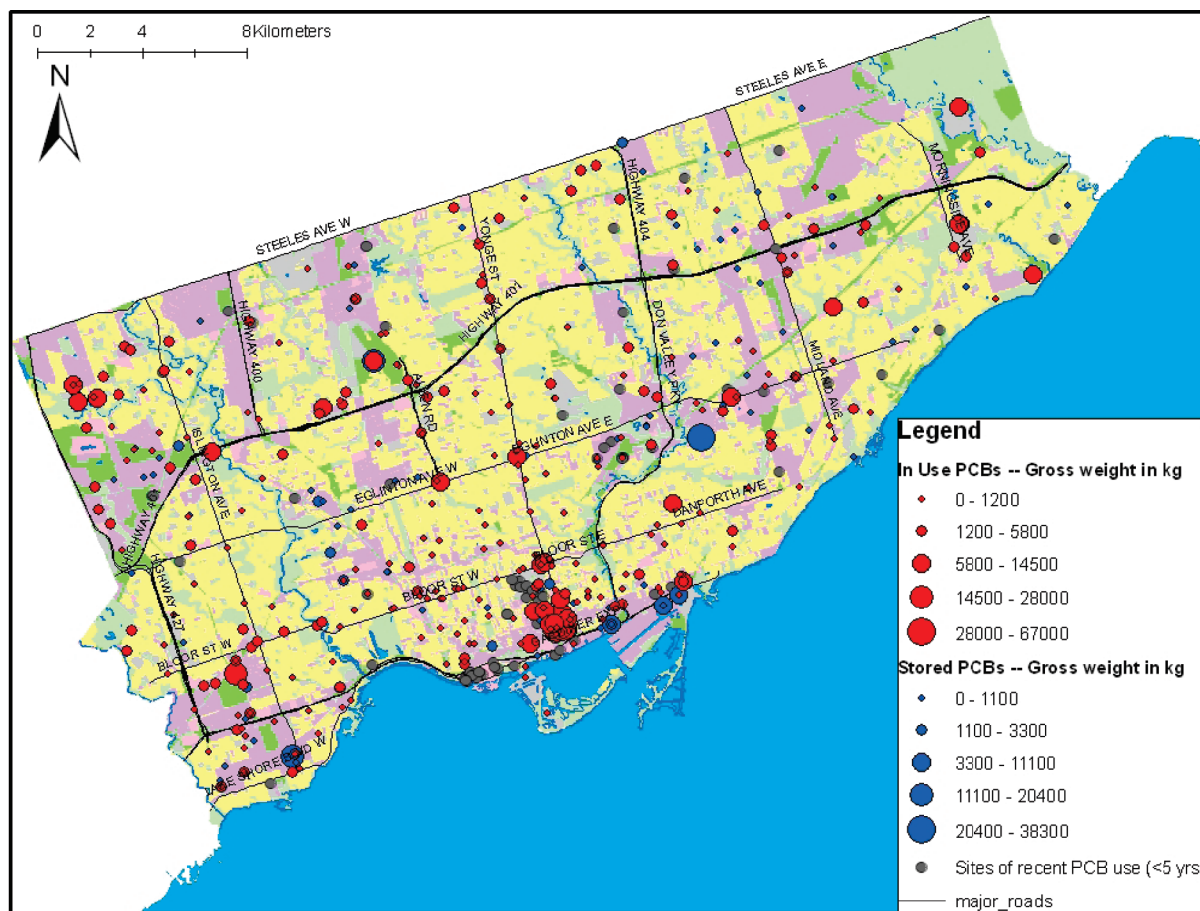
**Sealants.** The mass of PCBs in sealants in Toronto was estimated as 13 (0.14–231) tonnes (Table 1). We estimated that PCBs in sealants and caulking were geographically concentrated in residential areas within multiunit residential infrastructure and commercial buildings constructed during the post World War II boom-years from 1950 to 1970 (Figure S1).

**Total Inventory.** The total inventory of PCBs in Toronto of 437 (282–796) tonnes is the sum of the closed sources and sealants (Table 1). Of the inventory, 97% and 3% were in closed sources and building sealants, respectively. The spatial density of the total inventory was 687 (444–1253) mg m<sup>-2</sup> and 668 mg m<sup>-2</sup> for closed sources. The density in the downtown core was 7570 mg m<sup>-2</sup> and, as mentioned above, was greatest for the large office towers that define Toronto's skyline. For sealants, the city and downtown densities were 20.5 and 6.5 mg m<sup>-2</sup>, respectively where sealants in the numerous large buildings such as shopping centers and apartment buildings exceed those in the downtown skyscrapers. Overall, the downtown financial district contained >50% by mass of PCBs in closed sources whereas the Post World War II residential neighborhoods had proportionally more PCBs in building sealants.

## PCB Emissions and Fate

**Modeling Methods.** To estimate aggregate emissions and fate of PCBs from this inventory as well as unquantified stocks, we used the Multimedia Urban Model or MUM-Fate of Diamond et al. (19). Briefly, MUM-Fate is a seven-compartment mass-balance fugacity model that has two vertical air compartments (0–50 and 50–500 m), a compartment for films that coat impervious surfaces (100 nm depth), and compartments representing surface water (urban streams), soil (2 cm depth), sediment (2 cm depth), and vegetation (0.02 cm thickness). Definitions of fugacity capacities (Z-values) and D-values (transfer parameters) were from Diamond et al. (19) (Tables S4, S5). Further details are presented by Jones-Otazo et al. (34).

Two scenarios representing the City of Toronto and downtown Toronto (the latter represented by city wards 27 and 28) were modeled using an average steady-state scenario that spanned one year (i.e., representing four seasons), with an average temperature of 9.3 °C (35). The Toronto and downtown model areas were 636 and 19.6 km<sup>2</sup>, respectively. Land uses for these areas, which were extracted using GIS, were 58% impervious surface, 42% soil and vegetation, and 0.4% water for Toronto, and 61% impervious surface, 38% soil and vegetation, and 1% water for downtown (Table S6).



**FIGURE 1.** PCBs in use and in storage in closed electrical equipment superimposed on a land use map for Toronto. Also indicated are sites where PCBs have recently (<5 yrs) been removed from use.

MUM-Fate was run for five PCB congeners (CB-28, -52, -101, -153, -180). Their physical–chemical properties were obtained from Schenker et al. (36) and SPARC online calculator (37) (Table S7). Air reaction rates were calculated by assuming first order reactions with hydroxyl radicals with hydroxyl reaction rates obtained from EPI Suite (Version 3.20) and a hydroxyl concentration obtained from Rivett et al. (38). The remaining compartmental reaction rates were obtained from Wania and Daly (39) and Diamond et al. (19) (Table S6).

We considered two sources of PCBs to the model domain: gas- and particle-phase (together are bulk) PCBs from upwind air advection and bulk emissions to air from within the city. We neglected emissions directly to water (tributaries in Toronto) because they were assumed to be minimal. Emission rates of each of the five congeners from within the city to the lower air compartment were back-calculated by running the model iteratively with trial emissions until measured and modeled lower bulk air concentrations matched. Measured upwind and within-city congener-specific air concentrations were taken from Motelay-Massei et al. (35) who used passive samplers at rural and urban locations along an 80 km transect in Toronto. We assumed that reported concentrations represented only the gas phase, which were converted to bulk phase concentrations via the aerosol–air partition coefficient (40). This conversion method may have overestimated actual concentrations as passive samplers accumulate some particles (41). The measured concentrations represented the lower air compartment of 0–50 m; concentrations in the upper air compartment (50–500 m) were set to equal those of the lower compartment although this likely overestimated upper air concentrations and hence underestimated back-calculated emissions (42). A range of low and high emission rates was obtained by varying the

upwind air concentrations from that measured at urban and then rural locations (Toronto model run) or urban and then suburban locations (downtown model run), respectively (Table S8). Emissions of the five congeners were scaled to 70 congeners by multiplying by their fractional contributions to  $\Sigma\text{PCB}_{70}$ . The fractional contributions of the congeners were calculated using a PCB air concentration profile for Toronto (Diamond, unpublished data).

**Model Results.** Modeled soil, sediment, and film concentrations were within an order of magnitude of measured values (Table S9). Within-city emissions of  $\Sigma\text{PCB}_{70}$  ranged from  $\sim 0.4$  to  $4 \mu\text{g m}^{-2} \text{d}^{-1}$  or  $\sim 0.1$  to  $1 \text{ mg capita}^{-1} \text{d}^{-1}$  for Toronto and  $\sim 0.6$  to  $20 \mu\text{g m}^{-2} \text{d}^{-1}$  or  $\sim 0.1$  to  $3 \text{ mg capita}^{-1} \text{d}^{-1}$  for the downtown (Tables 2 and S10). Emissions accounted for 5–62% and 1–42% of total inputs for Toronto and its downtown, respectively, with the remainder coming from upwind advection, depending on the assumed inflow air concentration.  $\Sigma\text{PCB}_{70}$  emissions from downtown, both normalized on a per capita and area basis, were  $\sim 3$  and 5 times higher than city-wide estimates. The greater emissions from downtown than the entire city are consistent with, but less than the 10-fold greater PCB spatial density of the downtown inventory, where this discrepancy could arise because emissions are not directly proportion to the inventory and/or because of uncertainties associated with estimating the inventory and modeled emission rates. The emissions are well within the range of those estimated by researchers for other cities when expressed on a spatial or per capita basis (Table 2). This is notable as emission estimates from other studies were derived using a wide variety of methods. Although our model results can not be evaluated because the model is open-ended and the back-calculation of emissions from measured concentrations creates circularity,



TABLE 2.  $\Sigma$ PCB Emission Estimates

location	emission estimate			no. of congeners	reference
	kg yr <sup>-1</sup>	mg m <sup>-2</sup> yr <sup>-1</sup>	mg c <sup>-1</sup> yr <sup>-1</sup>		
City of Toronto	90–880	0.14–1.4	35–350	70	this paper
Downtown Toronto	4–140	0.2–7.3	33–1200		
City of Toronto	16–190	0.02–0.3	6–75	5 <sup>a</sup>	this paper
Downtown Toronto	1–31	0.04–2	6–250		
Canada	55–2100		1.8–72 <sup>b,c</sup>	5 <sup>a</sup>	(49)
Canada	240–9200		8–300 <sup>b,c</sup>	22	(49)
Chicago		~0.4–3.6 <sup>d</sup>		87	(63)
Chicago	~700 <sup>e</sup>			88	(64)
New York City	300	0.4 <sup>f</sup>		93	(65)
Wilrijk, Belgium	14.3 <sup>g</sup>		360 <sup>h</sup>		(66)
Zurich	600		1600 <sup>i</sup>	15	(67)

<sup>a</sup> The 5 congeners in the sum are 28, 52, 101, 153, and 180. <sup>b</sup> The range represents default to maximum estimates for the year 2000. <sup>c</sup> We used a population of ~30,000,000 to calculate per capita emissions (68). <sup>d</sup> This emission range was estimated by Eisenreich (63) by assuming a concentration of 1–10 ng m<sup>-3</sup> and assuming emissions are focused into a 5 × 5 km<sup>2</sup> area with a 1 km height. Their estimate is based on data from Simcik et al. (69) who reported results for air concentrations in Chicago for 87 congeners (55 peaks). <sup>e</sup> This emission estimate represents the difference between atmospheric loadings to Lake Michigan with and without urban influences from Chicago. <sup>f</sup> Based on a model area of 785 km<sup>2</sup>. <sup>g</sup> Estimate includes open and closed PCB sources. <sup>h</sup> Per capita emission is based on a population of ~40,000 for Wilrijk. <sup>i</sup> Based on a population of 375,000.

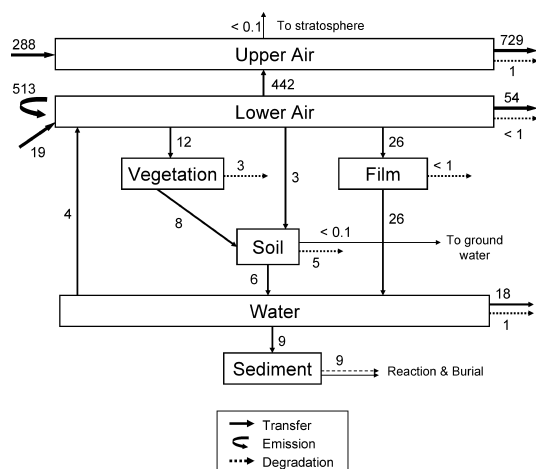


FIGURE 2. Fate of high emission scenario for  $\Sigma$ PCB<sub>5</sub> (28, 52, 101, 153, 180) in Toronto, expressed in g d<sup>-1</sup>. The solid arrows refer to chemical transfer and the dotted arrows refer to degradation.

some confidence in the results is gained by their concordance with those from other studies.

When considering  $\Sigma$ PCB<sub>5</sub>, ~95% of total inputs of 820 g d<sup>-1</sup> from upwind advection plus within city emissions is exported downwind (Figure 2). About 83% of the inputs to lower air are transferred to upper air with ~8% transferred to the other compartments and <1% lost via transformation. Since most of the transport of PCBs is predicted to occur in air, emission and fate estimates are very sensitive to assumed inflow concentrations.

Of the total inputs, only 26 g d<sup>-1</sup> or 3% is transferred to films on impervious surfaces and then to surface water which, in the model, is comprised of small urban streams and not nearshore Lake Ontario. Although this amount is low, air–film–water transport delivers 260 times more PCBs to the water than that from direct atmospheric deposition to the water surface of 0.1 g d<sup>-1</sup>, and 60 times more than that from soil runoff of 6 g d<sup>-1</sup>. In contrast, soil is an efficient chemical sink as only ~51% of PCB entering soil is lost via intermedia transport which is to water (43). In comparison, the film delivers nearly 100% of its atmospherically deposited burden to water. Water is a minor source of PCBs to air of 4 g d<sup>-1</sup> but delivers 9 g d<sup>-1</sup> to sediment.

For the individual congeners CB-28, -101, and -180, percentage loss downwind from the city is 99, 94, and 88%, respectively (Figure S2b–d). Conversely, transfer of PCB inputs and emissions ultimately to the aquatic system via film and vegetation–soil is ~1, 5 and 12%, respectively.

The residence time of PCBs in closed sources and sealants can be calculated as  $\tau = m/(A \times E)$ , where  $m$  (tonnes converted to mg) is PCB mass,  $A$  (m<sup>2</sup>) is the modeled area of the Toronto, and  $E$  (mg m<sup>-2</sup> y<sup>-1</sup>) is the emission rate. For the total PCB inventory of 282–796 tonnes and 636 km<sup>2</sup> area of the city, residence times range from ~300–9000 years or conversely, a loss of 0.01–0.3% of the inventory annually. If we consider losses only from sealants, then the residence time drops to 58 days to 2700 years, of which both extremes in the range are unreasonable.

Perhaps the most important point of this calculation is that emissions are only 0.01–0.3% of the total documented stock (the figure is probably toward the lower range because of the PCB stocks not included in our inventory). The reservoir is enormous and a loss of only a tiny fraction of the reservoir is sufficient to contribute to maintaining ambient PCB levels.

## Implications

### Have Policies Yielded Progress toward Controlling PCBs?

The answer is a guarded yes for the 159 parties that have signed the Stockholm Convention, including Canada, calling for the environmentally sound management (ESM) of PCBs by 2028 and particularly for the 35 parties (22% of all parties) who have reported progress towards this goal under Article 15 of the Convention (44). Progress refers to the signatories moving toward or passing national legislation that contains a plan for PCB elimination or ESM-specification of which stocks are subject to, and the timetable for, elimination or ESM. The timetable for elimination in Canada under legislation passed in 2008, is staged for 2009 for equipment with >500 mg/kg at all locations and 50–500 mg/kg at sensitive locations that include schools and hospitals. Equipment containing 50–500 mg kg<sup>-1</sup> PCBs at nonsensitive sites must be removed by December 2025. PCBs in equipment at <50 mg kg<sup>-1</sup> may remain in use until the end-of-life of that piece of equipment. This 2009 deadline may be extended on a case-by-case basis by the Minister until 2014. The Canadian legislation contains numerous exemptions such as equipment containing PCBs at <2 or 50 mg kg<sup>-1</sup>, continued sale and/or

use of PCBs in electrical capacitors that are an integral part of a consumer product, where the PCB-liquid is fusion sealed, etc. (45).

Declining concentrations seen in air and other media globally and locally, including our body burdens and food supply (e.g., 46, 47), after legislation was passed by jurisdictions in the 1970s and 1980s, point to the early control of PCBs as a science-policy success story!

**Will Policies Reduce Current and Near-Future Environmental Emissions?** Breivik et al. (48–50) estimated current global emissions at  $\sim 0.5\text{--}3$  tonnes  $\text{yr}^{-1}$  in 2010 which is easily realized given emissions estimated for Toronto and those of other cities (Table 2). Furthermore, Pozo et al. (51) have found that today's global air concentrations correlate well with the emission estimates of Breivik et al. (50). With these ongoing emissions, there is little question as to why temporal trends of concentrations in various media have been either stable or decreasing very slowly over the past decade. Looking into the future, Breivik et al. (50) predict gradual emission reductions until 2040–2050, depending on when countries remove remaining PCB stocks from usage. We expect faster reductions in emissions and local concentrations in the western countries that comprise most of the 35 parties to the Stockholm agreement that have or are destroying their PCB stocks.

**Why is the Timetable toward Elimination Taking so Long?** The “final target” of bringing global stocks of PCBs under ESM by 2028, if we achieve this goal, will have taken nearly 100 years since Monsanto first became aware of the health effects of PCBs in the late 1930s, 80 years since the first scientific evidence of the widespread distribution of PCBs became public, and  $\sim 55$  years since the first round of legislative action was taken in Europe and North America (52). Legislative progress under the Stockholm Convention is taking place in only 35 countries and 7 of these countries have reported a lack of capacity, resources, and technology to meet the Stockholm target.

In Canada, this progress should reduce emissions over 25 years by 83% to 1726 kg  $\text{yr}^{-1}$  but that will have taken almost 50 years after the first round of legislation was passed! It is disappointing that Canada, which was a lead country in creating the Stockholm Convention, has legislated a lengthy timetable that includes provisions for slowing the pace of elimination through Ministerial extensions. Legislative provisions for exemptions and extensions have worked to reduce the effectiveness of other pieces of environmental legislation in Canada. Further, it is a sad commentary that in Toronto, Canada's largest city, the greatest mass of in-use PCBs has been concentrated in the financial district which, until recently, was best equipped financially to deal with their removal.

The elimination of PCBs has been fraught with past and present obstacles to removal and destruction. Technically feasible, low-emission destruction methods have been available for over two decades. However, in some cases, public concern (“NIMBY” or not in my backyard) and/or the dubious nature of a facility have slowed the pace of PCB destruction (53). The Province of Ontario, in which Toronto is located, has four licensed PCB destruction facilities but high-level transformer waste ( $>500$  mg  $\text{kg}^{-1}$  PCBs) must be transported  $\sim 3700$  km to the major Canadian destruction facility located in Swan Hills, Alberta. This entails considerable cost and added risks.

**Will the Legislation Passed by Jurisdictions Take Us towards Eliminating PCBs?** First, we need to know what “elimination” means and how we measure it. We will arbitrarily define elimination as a benchmark that includes PCB concentrations in tissues (human, fish, etc.) reaching a low level, such as the fifth percentile of current values. Again turning to Canada, answering the question is complicated

because of the uncertain effect of exemptions allowed under the legislation and uncertainty with the extent of legislative compliance by PCB owners.

To answer this question from a science perspective, we need to know which PCB uses are driving tissue residues and hence nonoccupational exposure. We know that the main PCB exposure routes are indoor air and/or dust, and diet (6, 54). Which PCB emissions are contributing to elevated PCB concentrations indoors and in our diet? Again, the answer is complex. The sources of PCBs to indoor exposure via air and dust include PCBs used in building sealants, caulking, and floor coverings (e.g., 54–56). The contribution from electrical equipment such as building transformers, indoor wiring, and other uses is not clear. Whereas a few countries, including Germany and Sweden (57) have legislated the removal and management of PCB-containing building sealants, which will benefit building occupants and workers removing the sealants (e.g., 58), Canada's legislation does not contain such provisions.

Emissions driving exposure via diet are likely 2-fold. First, our food supply, particularly fish, meat, and dairy products, contain PCBs due to trophic transfer through food webs arising from “ambient” levels of PCBs that are supported by the burden of PCBs from past and present emissions. For example, our modeling results suggest the effective direct transport of PCBs (albeit a small fraction) emitted to the city's atmosphere, to surface waters and from there, fish. Could this explain, in part, why many nearshore fish in the Great Lakes have consumption advisories due to PCBs and that Great Lakes fish consumption can lead to elevated PCB exposure (59)? Second, dietary exposure may come through PCBs contained in animal and/or fish feed which is intended to enhance animal and fish growth but which inadvertently magnifies the trophic transfer of PCBs (e.g., 60, 61). One caveat here is that PCB exposure via fish consumption may be less harmful, due to the nutritional benefits of fish, than exposure via other exposure pathways (2).

This discussion could lead us to suggest that achieving PCB elimination requires a more complete understanding of which PCBs stocks are leading to emissions that, in turn, dominate our exposure. However, we argue that PCBs in in-use closed sources and stored waste, which likely contribute to overall emissions due to the shear mass of PCBs relative to PCBs in sealants, paint, etc., should be removed and destroyed swiftly; our lack of understanding of emission rates from these particular sources should not delay this action. Action has been taken and continues to be taken, to reduce PCB transfer through contaminated animal and fish feed in Europe and North America, thanks to numerous scientific studies and regulatory surveillance. Regrettably, the Secretariat of the Stockholm Convention (44) have found that some “PCB containing oils are being improperly recycled as skin and hair preparations, cooking oil, and for other inappropriate uses” (p 2). This speaks to the need to destroy existing stocks.

In closing, it is humbling to review the current situation with PCBs, and past and current scientific, health, and regulatory efforts. With total production estimated at 1.7 million tonnes from 1929–1989 (1), it appears difficult, if not impossible, to get the (Aroclor) PCB genie back into the bottle. Over 11,000 scientific papers have been devoted to PCBs (PubMed search Feb. 4, 2009). Enormous efforts and funds have been devoted to PCB management by setting up inventory programs, employing civil servants entrusted with regulatory surveillance, and developing new regulations to fulfill the terms of the Stockholm Convention. The cost to Canadian industry of destroying listed PCBs by 2025 is estimated at \$25.23 million (45). A scant \$2.4 million has been requested by the Secretariat to the Stockholm Convention (44) to assist developing nations with controlling their

stocks. If PCBs are, in part, linked to clinical or subclinical incidences of learning disabilities, reproductive, immune and/or other cancer and noncancer illness that could be occurring at population scales (e.g., 62), then the cost to society is in the billions if we do not expedite their safe elimination from present uses. This tale should serve as a warning about the widespread use of other chemicals including nanomaterials, even those being used to reduce risk, for which we have an incomplete understanding of their environmental and health consequences.

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## Supporting Information Available

Table S1, Uses of PCBs in Canada; Table S2, Summary of PCBs in Canada in-use and in waste up to December 31, 2006; Table S3 and text, Assumptions made in estimating mass of PCBs in joint federal/provincial database and building sealants; Table S4, Z values and D values for reaction and advection used in MUM-Fate for the City of Toronto and downtown Toronto; Table S5, D values for intercompartmental transport used in MUM-Fate for the City of Toronto and downtown Toronto; Table S6, Values used in MUM-Fate for the City of Toronto and downtown Toronto; Table S7, Physical-chemical properties of PCB congeners modeled using MUM-Fate; Table S8, Bulk air concentrations used as input values to calculate PCB emissions to air in MUM-Fate; Table S9, Comparison of measured versus modeled PCB concentrations for soil, sediment, and surface films; Table S10, Summary of PCB emissions for the City of Toronto and its downtown. This material is available free of charge via the Internet at <http://pubs.acs.org>.

## Literature Cited

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