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Article

Persistent Problem: Global Challenges to Managing PCBs

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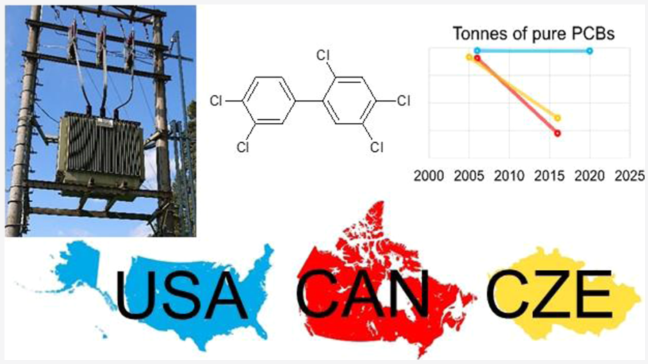
ABSTRACT: Polychlorinated biphenyls (PCBs), "famous" as persistent organic pollutants (POPs), have been managed nationally since the 1970s and globally under the Stockholm Convention on POPs since 2004, requiring environmentally sound management (ESM) of PCBs by 2028. At most, 30% of countries are on track to achieve ESM by 2028. Globally over 10 million tonnes of PCB-containing materials remain, mostly in countries lacking the ability to manage PCB waste. Canada (Ontario) and Czechia, both parties to the Stockholm Convention, are close to achieving the 2028 goal, having reduced their stocks of pure PCBs by 99% in the past 10 years. In contrast, the USA, not a party to the Stockholm Convention, continues to have a substantial but poorly inventoried stock of PCBs and only ∼3% decrease in mass of PCBs since 2006. PCB management, which depends on Stockholm Convention support and national compliance, portends major challenges for POP management. The failure to manage global PCB stocks >30 years after the end of production highlights the urgent need to prioritize reducing production and use of newer, more widely distributed POPs such as chlorinated paraffins and per- and polyfluorinated alkyl substances, as these management challenges are unlikely to be resolved in the coming decades.



KEYWORDS: *polychlorinated biphenyls, Stockholm Convention, chemicals management, persistent organic pollutants, PCB stocks, environmentally sound management, Canada, Czechia, USA*

■ INTRODUCTION

Polychlorinated biphenyls (PCBs) are the epitome of a persistent organic pollutant (POP) because of their persistence, bioaccumulative potential, and toxicity. Owing to their environmental mobility and persistence, they are distributed globally, from the high Arctic and Antarctic to the Mariana Trench in the deep Pacific Ocean.[1] PCBs pose risks to ecosystems as they potently bioaccumulate through the food web to reach levels of concern among top trophic level animals. In utero exposures are associated with neurodevelopmental toxicity, manifesting as learning, behavioral, or intellectual impairment in children.[2−6] PCB exposures are also associated with impaired immunological function, auditory deficits, and central nervous system disorders such as Parkinson-like symptoms.[7−9]

PCBs were introduced for use in dielectric fluids to reduce the risk of explosion in capacitors and transformers and saw widespread use as plasticizers and flame retardants in products such as building materials and paints.[10] Breivik et al.[11] estimated that more than 1.3 million tonnes of pure PCBs were manufactured between 1930 and 1993 in at least 10 countries, primarily in the USA, followed by West Germany, the USSR, and France (Figure S1). However, PCBs were widely exported from manufacturing countries, resulting in use in at least 114 countries.[11] The ∼1.3 million tonnes of pure PCBs, through dilution for use and subsequent poor management, expanded to 17 million tonnes of PCB-contaminated materials and waste (Figure 1), with an estimated 20−35% of PCBs already released in the environment.[17] Monsanto, in the USA, produced more than 50% of global PCBs and recognized the toxicity of PCBs shortly after the start of large-scale commercial production in the 1930s.[13] According to Monsanto's documentation, the company argued that PCBs were of minimal risk in closed systems such as capacitors and transformers, but the company intended to use PCBs in a very wide array of products that would result in environmental release and human exposure.[12] Clear evidence of their widespread environmental distribution came in the 1960s and 1970s, first in Baltic Sea biota,[13] along with indications of their toxicity.[14]

Global production of PCBs decreased with the introduction of restrictions in the 1970s in Western countries (Figures 1 and

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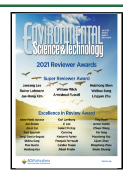

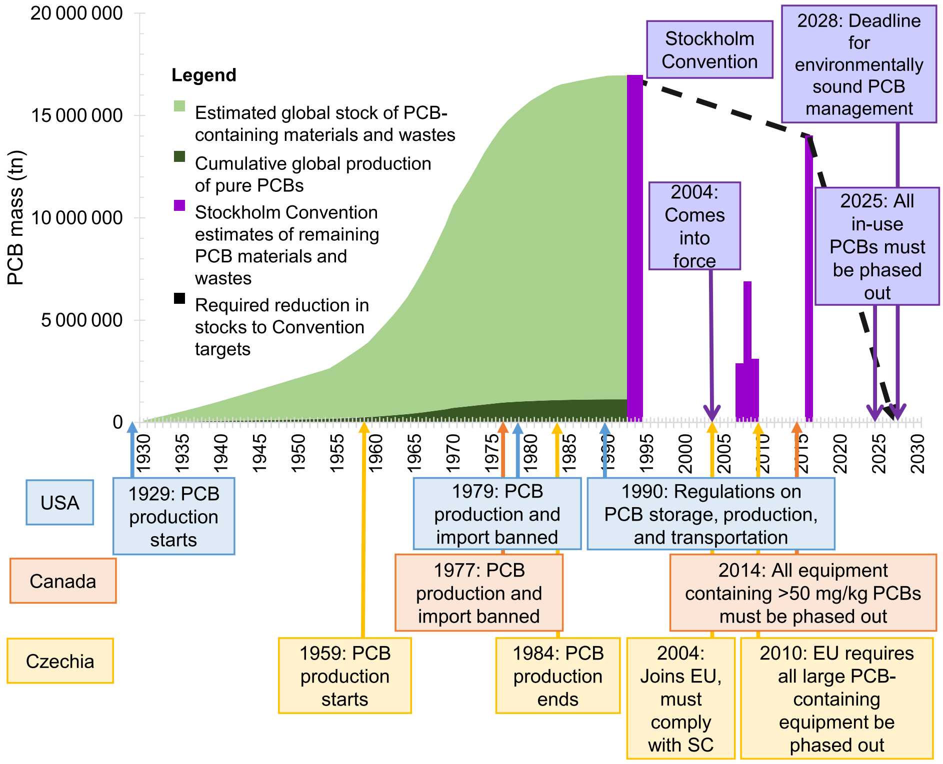

Figure 1. Timeline of major policy actions on PCBs in the USA, Canada, and Czechia and in the Stockholm Convention. Rate of PCB production and associated increase in stocks is estimated based on the study by Breivik et al.[11] The Stockholm Convention estimates of remaining stocks are obtained from refs 17−181920. The total stock of PCB-containing materials and waste (up to 20 million t) reflects how pure PCBs are diluted to create this mass and how mismanagement spreads pure PCBs to create a larger contaminated mass. The variation in the estimated stock over 1994−2016 reflects how reported global inventories changed over time due to uncertainties in reported PCB stocks. Additional details on the timeline of policies impacting PCBs are given in Figure S2.

S2). These were followed by restrictions implemented through international agreements, notably the Stockholm Convention on POPs, which entered into force in 2004 and currently has 185 parties (184 countries plus the European Union, chm.pops.int). The Stockholm Convention bans the production of PCBs and aims to phase out in-use PCBs by 2025 and ensure environmentally sound management (ESM) of materials with >0.005% (50 mg/kg) PCB content by 2028. ESM largely constitutes chemical destruction by high-temperature combustion methods where the PCB content of the waste is destroyed, with exceptions for low PCB content wastes with large volumes, in which case specially engineered landfills or permanent storage in underground mines/rock formations can serve as reasonable substitutes (Table S1).[15] Each party implements the Stockholm Convention through the enactment of binding legislation. Parties have devoted considerable efforts over decades to eliminating PCB stocks and controlling further primary and major secondary releases. Much has been accomplished. However, as of 2016, UNEP identified that only 17% of PCB-containing materials have been eliminated, at the rate of about 200,000 t/y since 2000. Addressing the remaining 83% would require the elimination of ~1 million tonnes of PCB-containing oils and contaminated equipment per year to reach the 2028 target.[16]

Today, despite restrictions, primary PCB emissions continue from on-going use in products and materials: closed applications (e.g., transformers, capacitors, electric motors, and light ballasts), partially open applications (e.g., hydraulic fluid, heat transfer fluid, switches, electrical cables, and vacuum pumps), and open applications (e.g., paints, sealants, inks, lubricants, flame retardants, insulation, dyes, and pesticides) (see Table S2).[21,22] This diversity of uses, combined with their poor documentation, creates a global challenge for managing primary PCB sources.

Although the greatest mass of PCB use has been in closed applications, open applications of PCBs have received increasing attention, particularly in relation to sensitive indoor environments such as schools.[23−25] Open applications have been shown to result in direct exposure, particularly in schools, and secondary exposure from emissions to the surrounding environment; open applications were estimated to be the primary contributors to global emissions up to 1980.[29] Building materials have received the most attention, notably joint sealants and paints, but >15 types of open applications have been identified by UNEP,[26] and previously undocumented open source uses (e.g., floor waxes,[27] book bindings[28]) continue to be identified. Open applications were the first to receive international regulatory attention through OECD restriction on open applications in 1973.[30] While reports frequently state that approximately 21−26% of PCBs were used in open applications,[26,29] this is a rough global average, and the type and amount of open PCB use varied substantially by region: in Japan, the majority of open use was in carbonless copy paper, while in Western Europe, it was in building sealants.[26,29] Open applications of PCBs present a unique challenge as they are not typically included in national inventories and are frequently not even recognized as PCB wastes.[26] Countries that have estimated stocks of PCBs in open applications (e.g., Germany, Finland, Norway, Sweden, and Switzerland) have identified amounts from hundreds to

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thousands of tonnes.[26] For example, Germany had an estimated 12,000 t remaining in open applications as of 2013, contributing 7−12 t of PCBs to the environment annually.[31]

Global PCB contamination has many facets, from the legacy use of PCBs in industrial and consumer products, to unintentional production and releases from industries and combustion[32,33] and to emissions of unintentionally produced nonlegacy PCB congeners from modern materials such as paints and cabinetry.[34,35] Here, we focus on the challenge of managing primary legacy PCB stocks through a review and analysis of current PCB management status globally. We assess progress toward ESM of PCBs through a two-part analysis: (1) challenges of PCB management on a global scale and (2) a detailed case study comparison of the current status of PCB management in three economically developed countries (Canada, Czechia, and USA) with differing histories of Stockholm Convention participation, PCB production and use, and management capabilities.

The motivation for this analysis is two-fold. First, it is critical to understand the scale of the future threat posed by PCBs to human and ecosystem health. After countries enacted controls in the 1970s and early 1980s, concentrations in air, water, and relatively short-lived biota dropped rapidly.[36] However, decreases have slowed in recent years.[37,38] Today, 40 years after major production ceased, PCBs may cause the demise of over 50% of the world's killer whale populations.[39] As a neurotoxicant, PCBs contribute to the significant global burden of disease attributable to widespread human exposure to hazardous pollutants.[40] Second, the analysis of successes and failures in managing PCBs provides a clear cautionary lesson on the long-term impacts of producing and widely using persistent compounds and the inability of even wealthy countries to manage and eliminate their on-going use.

METHODS

Global PCB Management. Historical PCB consumption was adapted from Breivik et al.[11] to reflect the total PCB mass used (see Text S1). The status of PCB use and management of all UN-registered countries was classified based on the most recent information (Table S3). In most cases, this information was the most recent Stockholm Convention status document (National Implementation Plan or Conference of the Parties update) and the responses to a 2018 questionnaire given to Stockholm Convention parties on PCB management. Where available, other reports were also used, particularly Global Environment Facility (GEF) project reports (see references in Table S3). Based on this information, we placed countries into eight categories according to their PCB management, ranging from no existing PCB management plan or inventory, to full ESM. We also included countries that are not parties to the Stockholm Convention.

Case Study: Canada. Data from Ontario, containing nearly 40% of the Canadian population,[41] were used as an indicator of Canadian performance. Details of the Ontario PCB inventory are provided in the Supporting Information (Text S2). Briefly, the PCB stock in Ontario was estimated by combining data from the Canadian federal "ePCB" database and a provincial-level PCB Waste database maintained by the Ontario Ministry of the Environment, Conservation and Parks. The federal ePCB reporting system lists locations of PCB holdings in-use and in-storage with concentrations >50 mg/kg. Data as of December 31, 2016 for closed sources were used. The Ontario database separately lists PCB waste storage sites in Ontario; data used were as of 2013. Combined, the databases reported a total of 270 unique sites with PCBs in use, stored, or classified as wastes not yet subject to ESM.

Any values given in units of volume were converted to mass based on assumptions of the density of the PCB-containing material. The masses of PCB-containing materials were then converted to estimates of pure PCB mass assuming average concentrations per category of application.[42] Askarel fluid was assumed to have 600,000 mg/kg PCBs (range 400,000−800,000 mg/kg) and mineral oil 250 mg/kg PCBs (range 50−500 mg/kg).[43,44] Concentrations in waste categories were estimated based on their classification as either low- or high-level waste. Assumptions are detailed in Table S4.

We also compared these 2013−2016 data with an inventory of closed sources from 2006 before the enactment of revised Canadian Federal PCB regulations in 2008. This comparison was restricted to the Toronto (largest city in Canada) area because of data availability.[42]

Case Study: Czechia. Data for Czechia were compiled from the current inventory of PCB-containing products and materials, maintained by the CENIA—the Czech Environmental Information Agency. Since the itemized database is not publicly available, our inventory relied on the totals of individual application categories, which have been reported to the Stockholm Convention[45] and the European Union.[46]

Where available, exact reported masses of PCB-contaminated material were used, and when missing, the mass was estimated based on the number of items and the median mass of PCB-containing fluid per item in the category (Table S5).

The masses of PCB-containing fluids were converted to estimates of pure PCB mass using assumptions specific to Czech/EU regulations (Table S5). Czechia complies with the European Council Directive 96/59/EC, which required elimination of all materials containing >500 mg/kg PCBs by 2010; thus, all remaining large PCB equipment (>5 dm^3) should be <500 ppm PCBs. However, there is some ambiguity in the reported information; thus, we have used a 10,000 ppm upper threshold, based on Czech reporting to UNEP,[45] to account for a worst-case scenario of instances of non-compliance.[47]

To evaluate progress since the ratification of the Stockholm Convention, we compared the most recent inventory with totals from the initial Czech National Implementation Plan reflecting 2002−2004.[48] As the original inventory preceded EU legislation, we assumed higher concentrations of PCBs in materials using the values of 20,000 mg/kg (range 10,000−30,000 mg/kg).[47]

Case Study: USA. To estimate the current stock of PCBs in use and waste in USA, we utilized publicly available information from the US Environmental Protection Agency (US EPA), specifically the PCB transformer registration database[49] (Table S6, as of Jan 2020) and the PCB Cleanup and Disposal Program[50] (up to 2020). The US EPA tracks transformers and regulated PCB waste pursuant to regulations under the Toxic Substances Control Act (TSCA). Owners of PCB transformers must register details on transformer location, ownership, and mass in the PCB transformer registration database. PCB transformers that are removed from use may be optionally deregistered from the database. We extracted information on the number, location, and mass of transformers to estimate the stock of PCBs currently held in in-use and stored transformers. Incomplete records were assigned

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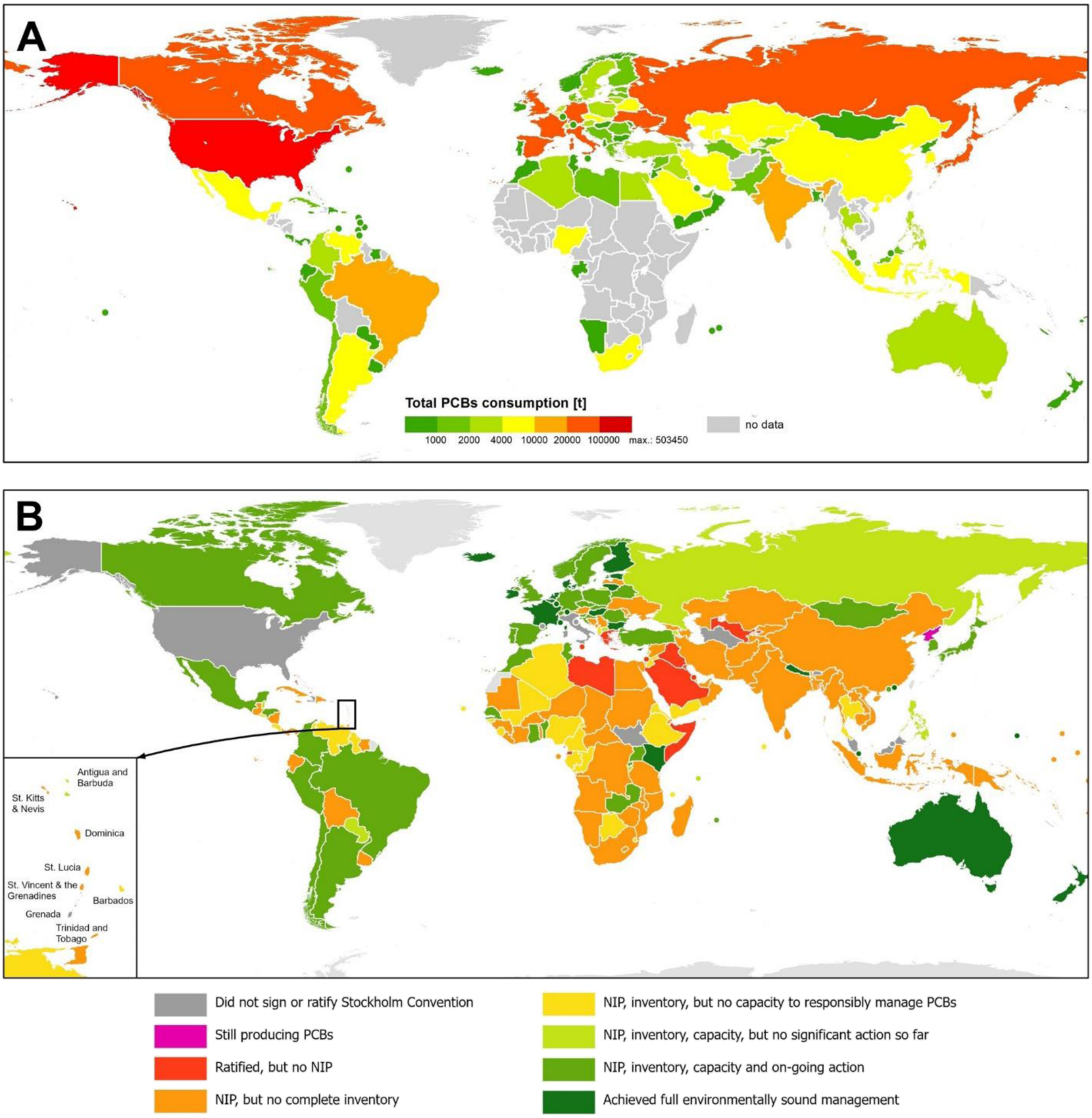

Figure 2. Global PCB use and management. (A) Total PCB consumption by country throughout the period 1930−2000 based on data from Breivik et al.[11] extrapolated to total PCB mass consumption according to Text S1. Figure S6 presents the same data presented as per capita consumption. (B) Current status of PCB management according to the latest reported status for each country, compiled from Stockholm Convention reporting and other sources. Sources used are given in Table S3. NIP indicates Stockholm Convention National Implementation Plan. A color-blind accessible version of this figure can be found in Figure S5.

the median value of the complete records (median two transformers per site; median mass 782 kg/transformer). A transformer was considered to be 30% fluid by mass;[51] this factor was used to convert total transformer masses to masses of PCB-containing fluids. Fluid masses were converted to estimates of pure PCB mass using assumptions about the concentrations in a typical transformer, as for the Canadian inventory. We assumed that before Jan 1, 2000, all transformers reported were askarel transformers, and after that date, all were mineral oil, based on the assumption that transformer owners would be aware of high-level PCB content and report in compliance with the TSCA regulations. However, PCB-contaminated mineral oil may be less thoroughly documented, leading to delayed reporting.[52] Average concentrations used for askarel transformers were 600,000 mg/kg PCBs (range 400,000−800,000 mg/kg) and those for mineral oil transformers were 1000 mg/kg PCBs (range 500−5000 mg/kg).[43,44,52] To evaluate the completeness of the transformer registration database, we compared the total number and mass of transformers reported to the PCB Cleanup and Disposal

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Program with the total number of transformers in the registration database and the mass of deregistered transformers from 1998 to 2018.

To evaluate changes in the USA stock over time, we compared the current remaining stock of transformers to records of PCB transformer deregistrations to calculate the number of transformers and mass of PCBs that would have remained in use/stored in 2006.

Uncertainty Analyses. Monte Carlo analysis was performed for the Canadian PCB databases, which identified that concentration levels (i.e., mg/kg PCBs per equipment/waste category), particularly the concentration selected for the high-level equipment/wastes, had the greatest impact on the estimate of the total stock (Text S3, Figures S3, S4). Density and other assumptions made to complete missing data had negligible impact. Therefore, we addressed the uncertainties in the conversion of database entries to pure PCB masses through the inclusion of upper and lower concentration thresholds for individual PCB equipment and waste categories according to the regulations and database thresholds for each country, as described above.

RESULTS

Global Management of PCB Stocks. Global use of PCBs varies widely, and the current PCB management capacities of countries also vary widely (Figure 2B, Table S3). One country (Democratic People's Republic of Korea) continues to produce PCBs.[17] 185 parties (184 countries plus the European Union) have ratified the Stockholm Convention, while 13 countries have not, notably the USA, Italy, Malaysia, Haiti, Israel, and Turkmenistan; all other nonratifying countries have <1 million population or are recently established (e.g., South Sudan). We note that while Italy has not ratified the Stockholm Convention, the European Union, of which Italy is a member, is a party to the Convention, and the EU has stricter regulations on PCB management than the Stockholm Convention. Of the 184 ratifying parties, 10 have not submitted any implementation plan. Greece and Malta, as EU member states, should also follow EU PCB management regulations, despite not yet submitting documentation to the Stockholm Convention.

For the 174 Parties that have submitted reports, our analysis highlights that 72 national PCB inventories (42%) are partial or preliminary. Many inventories are limited to transformers and/or only to the public electricity sector, which may capture only half of the uses of PCBs (considering 48% of the PCBs produced were used in transformers[22]). An additional 23 countries (13%) reported complete PCB inventories but no capacity to achieve ESM, while 11 countries had inventories and capacity to manage PCBs but had made no significant progress toward ESM. The number of countries achieving or progressing toward ESM was small; 34 countries (18%) are progressing toward ESM through removal from use and destruction of PCB materials. Only 23 countries (13%) have achieved ESM of PCBs. With three exceptions (Nepal, Kenya, and Micronesia), all of the 23 countries that have achieved ESM are classified as "very high" in the UNDP Human Development Index, or "high income" under by the World Bank (Table S3). Only three countries classified as "low development"/"low income" are currently making substantial progress toward ESM of PCBs: Benin, Rwanda, and Uganda.

National reporting to the Stockholm Convention contains a wealth of information about PCB management, but the quality and quantity of information provided by individual countries vary widely. A questionnaire from the Convention was distributed to 182 parties to evaluate progress toward ESM of PCBs. Fewer than 60 parties provided responses.[53] Based on these responses and additional Stockholm Convention information sources, estimates of current PCB stocks could be determined for 52 countries (see references in Table S3). Many countries lacked recent information, with some countries not submitting documentation since 2004. Therefore, our analysis is uncertain for two reasons. First, we may be presenting a worst-case scenario since countries may have made unreported progress toward PCB elimination. Second, and conversely, many countries reported incomplete inventories (e.g., only inventories of transformers owned by a national electricity provider), and thus most recent reports under-record true stocks.

Many countries are challenged by weak institutions, corruption, and mismanagement, making tracking PCB stocks and limiting their misuse extremely difficult. There are multiple reports of transformers being improperly recycled. For example, Sri Lanka identified that PCB-containing transformers were transferred to informal recyclers and that spilled PCB-containing oils were cleaned up with sawdust of which some were disposed of through burning.[54] Nauru reported that a transformer confirmed to contain PCBs was slated to be shipped to Australia but was instead collected by a scrap metal recycling company with an undetermined fate.[55] In other instances, transformer owners were reported to have actively drained and disposed of PCB contents to avoid responsibility for PCB materials. For example, in Malawi, numerous pieces of equipment suspected to contain PCBs had their contents poured directly onto the ground before they could be tested.[56] The Dominican Republic reported that owners of transformer shops, to avoid PCB disposal regulations, diluted the PCB concentration in the fluids by continually removing them and adding more mineral oils and sold the removed PCB oils to illegal foundries.[57] Ghana reported the use of PCB oils to create beauty creams and to lubricate domestic sewing machines,[58] while in Montenegro, factory workers were reported to have used PCB oils for handwashing and to heat homes.[59]

Several initiatives, such as those funded by the Global Environment Fund (GEF) and implemented by UNEP, have made progress in addressing some of the challenges low-income countries face in managing PCBs. For example, a project, harmonizing efforts in Southern Africa to centralize dismantling, draining, and accumulation of PCB oils/equipment for disposal,[60] was among more than 40 GEF-funded projects on ESM of PCBs. Together, these projects have eliminated 23,000 tonnes of PCBs.[17] However, even these specific projects can be hindered by unreliable national reports, delays in laboratory analysis of suspected PCB materials, and, most crucially, incomplete inventories. The technological and financial capacities required to eliminate PCBs are not available in many regions. GEF-funded projects on PCBs have received ~$450 million USD to support the elimination of 88,000 tonnes of PCB-containing materials and waste (23,000 tonnes eliminated and 65,000 tonnes planned), averaging USD 5,000 per tonne of PCB waste eliminated. It is noted that these project costs include items not directly related to elimination (i.e., capacity building and education).[17] Currently, the cost burden of managing PCBs lies with national governments or international agencies (e.g., national environment agencies,

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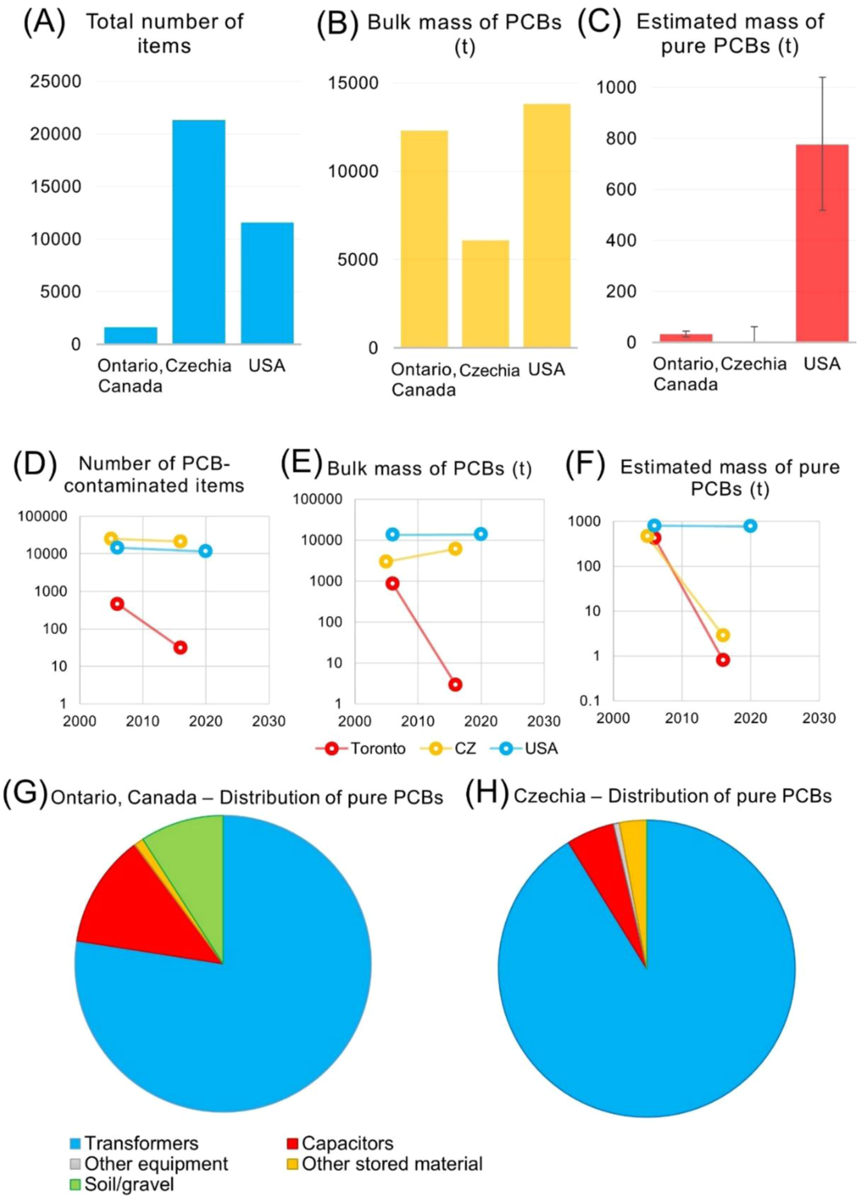

Figure 3. Inventories of PCBs for USA, Czechia, and Ontario, Canada. (A−C) Estimated stocks as of mid 2010s by (A) number of items, (B) bulk mass of PCB-containing materials, and (C) pure PCBs, with error bars indicating the uncertainty of the estimates of mass. (D−F) Change over time in PCB-contaminated items, bulk mass of PCBs, and pure PCBs. (G,H) Distribution of current stocks of pure PCBs in Ontario and Czechia according to item categories. USA is not shown because only transformers are included in the inventory.

UNEP, and GEF). Producer financial responsibility to date has only been in the form of legal settlements. Funding PCB elimination is clearly a problem, with the estimated need to eliminate 1 million tonnes of PCB-containing materials and waste per year to achieve Stockholm Convention compliance, mostly in countries with minimal financial and/or technical capacity. Yet despite the costs of elimination, there is also a clear public health cost of inaction: toxic chemicals, including PCBs, are neurotoxicants contributing to the "pandemic of developmental neurotoxicity," placing a significant burden on societal resources.[6,10]

At most, 30% of countries are on track to achieve ESM by 2028, and the lack of capacities for PCB management is a barrier to achieving this goal. With the specific examples of Canada, Czechia, and the USA, we demonstrate successes and barriers to ESM even in high-income/highly developed countries.

Case Studies. *Inventory Results: Canada.* Based on the combined provincial and federal databases as of 2013 and 2016, we identified a total of 12,200 tonnes of material in Ontario with PCB content >50 mg/kg. These materials were estimated to contain 32 t of pure PCBs (Figure 3, Table S11) with a range of 21−44 t. The PCB sites were widely distributed across heavily populated Southern Ontario and within or close to urban areas. The stock of PCB materials in Ontario was dominated by PCB-contaminated soil/gravel held at two sites, making up 97% of the bulk mass of PCB materials but <10% of the pure PCBs, which was largely held in transformers (77%) and capacitors (12%) (Figure 3G).

According to a 2006 PCB inventory,[42] Toronto had 455 sites containing 850 t of PCB-containing equipment and materials, equivalent to 424 t (range 282−565 t) of pure PCBs. A large proportion of this was located in Toronto's central business district in electrical transformers in large office

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towers.[42] In 2016, Toronto had only 7 sites holding 31 separate PCB items, all located outside of the city center, of which 3 sites had PCBs remaining in use (all others were stored for disposal), constituting only ∼40 g of pure PCBs. The remaining four sites stored 2.9 t of bulk PCB material, equivalent to 0.8 t (range 0.6−1.0 t) of pure PCBs. Thus, >99% of the 2006 stock of PCBs in Toronto was removed in the 10 years between inventories and after 2008 legislation mandating ESM, clearly showing progress toward elimination of PCB stocks in Toronto (Figure 3F). This agrees with Canada's reporting to the Stockholm Convention[61] which states that 0.02% of the 2008 PCB stock remains today.

Inventory Results: Czechia. Based on the most recent Czech records, we estimated a total stock of 6092 tonnes of material with PCB content >50 mg/kg in 2016. This bulk mass was estimated to contain 2.84 t (0.30−62 t) of pure PCBs (Table S11). Most of the mass of PCB materials (74% of bulk material and 79% of pure PCBs) was in transformers held by a large electricity production and distribution company responsible for more than 14,000 pieces of PCB-containing materials, including close to 10,000 transformers (Figure 3A). However, despite the large number of transformers, Czechia has prioritized the removal of equipment containing high concentrations of PCBs in compliance with European Union and Stockholm Convention regulations. This has resulted in a relatively low level of pure PCBs (∼2 tonnes) in these ∼10,000 transformers.

In 2005, 25,000 items contained ∼3000 t of PCB-contaminated fluid/materials,[48] equivalent to an estimated 460 tonnes of pure PCBs. An inventory in the intervening years (from 2009) reported 9193 t of known PCB materials and 3228 t of possible PCB materials.[62] Later inventories (2009, 2016) were more comprehensive, which resulted in a higher reported bulk mass of PCBs (Figure 3E). The prioritized removal of high concentration items has led to the large decrease of pure PCBs since the 2005 inventory was compiled (Figure 3F).[48]

Inventory Results: USA. The USA inventory was based only on the US EPA transformer registration database[49] (Table S6) and, consequently, was incomplete in two respects. First, it contained only records of transformers—no inventories exist in the USA for any other PCB-containing materials. Data from the PCB Cleanup and Disposal program[50] (Tables S7 and S8) indicate the disposal of millions of kilograms of large low- and high-voltage capacitors and bulk waste from 1998 to 2018, none of which has been included in any inventory. Second, the transformer registration database likely does not include all PCB transformers. From 1998 to 2018, the registration database listed 20,130 total transformers, but the PCB disposal program data indicated that over 180,000 transformers were disposed of over the same period, strongly suggesting that the registration database did not, and likely still does not, include all PCB-containing transformers. Therefore, our stock estimates in Table S11 are a clear underestimate of closed-source PCBs in USA.

As of 2020, the USA transformer database contained records of 11,577 transformers, estimated to contain 13,755 tonnes of PCB material with 776 (517−1040) tonnes of pure PCBs. To provide a temporal comparison similar to Canada and Czechia, the database was re-evaluated considering the stock of transformers existing in 2006. In 2006, the USA had 14,457 transformers containing 47,500 tonnes of PCB material and 770 tonnes of pure PCBs. This suggests a 20% reduction in the number of transformers and a 71% reduction in the mass of bulk PCB material, but only a 3% reduction in pure PCBs over ∼15 years (Figure 3D−F).

■ DISCUSSION

Canada. PCBs were never manufactured in Canada, but an estimated 40,000 t of PCBs were imported up to 1980,[44] leading to the second-highest per capita use in the world (1.2 kg/person), behind only the USA (Figure S5). New uses of PCBs were banned in 1977. Canada signed the Stockholm Convention in 2001 and enacted legislation to comply with the Convention in 2008. The purpose of the 2008 Canadian PCB regulations was to accelerate the elimination of PCBs in concentrations greater than 50 mg/kg by 2025 by stipulating end-of-use deadlines, especially for PCBs near sensitive sites (schools, daycares, hospitals, etc.).[63]

Of the three profiled countries, Canada has most successfully managed PCBs. The mass of pure PCBs in Toronto, Canada's largest city, decreased by 3 orders of magnitude within 10 years, indicating that regulations were successful in phasing out PCBs in Toronto. PCB material has been removed from all transformers in large skyscrapers built in the 1960s and 1970s in the downtown core and from sensitive sites such as schools.[42] The enforcement of PCB regulations in Canada includes compliance strategies and environmental officers that inspect PCB facilities. For example, in 2015, inspections were conducted at 44 companies that were set to remove and destroy their PCB equipment, finding 89% compliance.[64] Some inventory data indicated a lack of compliance in a small number of cases; three entries in Ontario databases reported concentrations over 500 mg/kg for equipment that should have been removed by 2009.

Canada's database has only limited inclusion of open sources of PCBs, although these were used in Ontario, for example, as joint sealants in buildings constructed from the 1950s to 1970s.[65] Past inventories estimated that the contributions of PCB-containing building sealants were low relative to the total amount of PCBs held in transformers,[42,65] but with the prioritized removal of closed PCB equipment and little attention given to open sources, their relative importance may now be greater. In countries that consider open sources in their PCB inventories, they typically account for more than a third of the remaining PCB bulk mass, for example, 37% in Germany[66] and 39% in Switzerland.[67]

Committing to national and international PCB agreements has helped expedite Canada's progress on phasing out PCBs. The 2008 regulation was proposed to meet the targets and commitments of the Stockholm Convention.[68] If the current trend continues across Canada, we suspect that Canada will be well on track to meet the 2025 target set by the Stockholm Convention.

Czechia. At the time of PCB production, Czechia was Czechoslovakia, and PCBs were produced by the Chemko Strážske factory in the east of the country (now Slovakia). Chemko produced 21,481 tonnes of PCBs from 1959 to 1984, of which 11,613 tonnes were used in Czechoslovakia, and the rest was exported to other Eastern Bloc countries, primarily East Germany.[47] Per capita use in Czechoslovakia was 580 g/person, the 20th highest in the world (Figure S6). It is estimated that 7000−8500 t of pure PCBs were used in the area that is now Czechia.[48] Most PCB use was by three state companies manufacturing PCB-containing paints and coatings, electrical capacitors, and electrical equipment for transport and

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industry.[46] These materials were then distributed to over 200 other companies/state agencies. PCB manufacturing ceased in 1984, and use restrictions were introduced in stages over the 1990s as Czechia moved toward compliance with EU regulations with the goal of eventually entering the EU. These late restrictions are the reason why the breast milk of Czech and Slovak mothers contains, on average, the highest concentration of PCBs among industrialized countries.[69,70]

The Czech PCB stockpile and waste management plan covering 2003 to 2013 dictates that all PCB materials exceeding 50 ppm be managed according to ESM provisions of the Stockholm Convention. Czechia, as a member of the EU, additionally follows the EU PCB regulations, which are stricter than those of the Stockholm Convention for large equipment, requiring that equipment with PCB volumes >5 dm^3 was decontaminated or disposed of by 2010.

As with Canada, open sources were not considered in the Czech inventory. PCB-containing building sealants had limited use in Czechia and other former Eastern Bloc countries, unlike in Western Europe and North America, due to their higher cost. The major open use of PCBs was in paints, particularly on bridges and in military applications, constituting approximately 21% of total use.[45,47] PCBs released from paints have caused significant environmental contamination, including of Elbe River sediments with up to 6 mg/kg PCBs due to renovations to a railway bridge that had PCB-containing paint.[71]

The biggest challenge in Czechia is not the documented inventory of PCBs but rather the numerous abandoned industrial/contaminated sites. This number is larger than typical in Western countries due to the country's transition from a communist economy in the early 1990s, dissolution of state-owned companies, and subsequent bankruptcies and abandonment of these sites. Numerous facilities now remain without responsible ownership and contain abandoned industrial infrastructure, with possible PCB contamination, as well as contaminated soils and other materials. The database of contaminated sites maintained by the Czech Ministry of Environment listed 387 sites with PCB contamination as of 2016; however, 88% remain only as suspected contamination, still lacking a proper site survey.[45]

Since the early 2000s, the Czech PCB inventory appears to have grown because of the "discovery" of many of these sites and reporting from companies that had not initially disclosed their stocks. The 2002−2005 database reported 25,000 contaminated or potentially contaminated items, with ∼3000 t of bulk PCB fluid/materials.[48] By 2016, there were slightly fewer PCB-containing items (21,300) but a much higher bulk mass of PCBs (6093 t) compared to 2005. This discrepancy arose because of the inclusion of small PCB items that were not included in the original inventory. Most importantly, the mass of pure PCBs has dramatically decreased: only 1% (0.1−17%) of the 2005 stock of pure PCBs was estimated to remain in 2016 (Figure 3). This large decrease in pure PCBs is attributed to progress toward compliance with EU regulations, with priority given to ESM of high-level and large-volume PCB equipment.

Czechia has sufficient capacity for ESM of PCBs (e.g., annual hazardous/POP waste incineration capacity of >21,000 t[46]); however, the country requires action from both private entities and the state, in case of the abandoned stock, and a significant effort to remove PCBs from use if it is to achieve ESM by 2028.

USA. The USA was the world's largest producer and consumer of PCB products with an estimated use of 500,000 tonnes of PCBs, or 1.9 kg per capita, the highest in the world (Figures 2A and S5). The USA passed regulations in 1979 under the TSCA to prohibit the manufacture, processing, distribution in commerce, and new use of PCBs. However, the USA has not ratified the Stockholm Convention and does not have national legislation that sets deadlines for PCB elimination.[72] This lack of national legislation is also reflected in its fragmented and incomplete PCB data. While it is clear from the PCB Cleanup and Disposal program[50] that the USA has removed a large stock of PCBs from use, the incomplete current inventories prohibit the evaluation of the remaining burden of PCBs in the USA. Our analysis strongly suggested that the transformer registration database is missing a substantial number of transformers, and no inventory exists for other PCB materials (capacitors, ballasts, other electrical equipment, and contaminated soil). In Canada, these materials accounted for ∼25% of the total mass of pure PCBs and 99% of the mass of bulk PCB materials; we expect similar proportions in the USA. The US transformer database requires responsible parties to self-report information, with limited enforcement, and the concentration of PCBs is not reported. The incomplete or erroneous information greatly limits the accuracy of the inventory.

Our assessment of the US records as incomplete contrasts with a recent UNEP report suggesting that US PCB records are more comprehensive than those for other countries.[17] While this may be the case in comparison to developing countries, we argue that the fragmented and inconsistent nature of reporting, limited mainly to transformers, coupled with the highest global PCB manufacturing and use, presents the USA as a worst-case scenario for PCB management in countries with a capacity to do so.

Even based on incomplete information, the stock of PCBs in the USA remains large compared to Canada and Czechia. While the USA has removed 100,000 t of PCBs from use, the impact of this removal on total PCB stocks is highly uncertain because of poor record-keeping. Moreover, the USA did not show a significant decrease in the mass of pure PCBs between 2005 and 2020 as was seen for Canada and Czechia (Figure 3F), which have prioritized removal of high-level PCB materials (e.g., askarel transformers). Further, PCBs removed from use in the USA are legally allowed to be disposed of by methods not considered ESM by the Stockholm Convention, such as landfilling. This is a major concern as landfills may act as secondary PCB sources by contaminating surrounding ecosystems with resulting ongoing human and environmental exposures.[73] The environmental and societal burden due to the ongoing use and non-ESM disposal of PCBs in the USA is a clear concern, given the country's history as the world's largest producer and user of PCBs.

The per capita mass of pure PCBs in the USA inventory (2.40 g/person for 2020) is comparable to Ontario, Canada (2.41 g/person for 2013−2016), while Czechia is lower, at 0.27 g/person for 2016. However, of all three countries, Canada has the most complete inventory, which includes large masses of contaminated soil and gravel that are not included in inventories in either Czechia or USA. The per capita stock in the USA would be significantly higher if the inventory included the additional categories of capacitors, other electrical equipment, and contaminated waste materials that have been significant portions of the Canadian stock (Figure 3G). It is

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estimated that USA has at least 26 million cubic meters of soils contaminated with PCBs[17] and 350 "Superfund" sites with reported PCB contamination.

■ IMPLICATIONS AND LESSONS LEARNED FROM PCB FAILURES

In 2016, UNEP completed an assessment of global efforts to eliminate PCBs, with an update in 2019.[17,53] The report highlighted similar challenges as identified here: incomplete inventories of PCBs in many regions and large uncertainties in current stocks and extent of ESM. The report identified that "the majority of countries (with some notable exceptions) are currently not on track to achieve the ESM of PCBs by 2028." Our analysis of the progress, available infrastructure, and the challenges within individual countries in managing PCBs confirm the implausibility of achieving this goal without a rapid change in actions. UNEP also reported that the PCBs eliminated so far were likely "low-hanging fruit," and further elimination will present more logistical and technical problems, challenging our ability to achieve the rates of elimination needed to reach Stockholm Convention goals.[17]

One substantial challenge is PCBs remaining in open applications, which are poorly documented even in countries that report ESM. To date, Sweden has given the most comprehensive attention to open applications through a program of identification and decontamination of buildings with PCBs in building materials.[74] Also, a small number of other countries have developed general estimates and addressed contaminated buildings on a case-by-case basis. The difficulty in managing open applications of PCBs is the lack of documentation of use combined with their importance to human exposure[25,75] as well as the lack of coherent strategies to manage PCB-containing/PCB-contaminated building materials without increasing emissions of PCBs.

In addition to the challenge of ESM for the remaining global stock of PCBs, we are faced with the challenge that some large fraction of PCBs is no longer "manageable." These "unmanageable" stocks have been released to the environment, landfilled without documentation, or, in arguably worst cases, are "lost" due to a lack of labelling and documentation and have entered the commercial sector as oils without identified PCB content. The longer the use and improper storage of PCBs persists, the greater the potential for environmental releases and human exposures, particularly considering the aging infrastructure that houses PCBs.

Strong regulation combined with financial and technological capacity and enforcement, as demonstrated in Canada, can successfully reduce stocks of PCBs and advance ESM. Many countries face substantial challenges due to historical structures (e.g., as in Czechia), and even with sufficient financial and technological capacity, their progress toward achieving the Stockholm Convention deadlines is hindered by legacies of poor record keeping, environmental practices, and site ownership. However, most troubling is the prevalent lack of capacity to manage PCB stocks. The Stockholm Convention plays an important role in education and capacity building toward PCB elimination, motivating countries, particularly those with the ability to implement the Convention, to inventory and dispose of PCBs. Nonetheless, the inventory quality is generally poor in many countries, compounding the challenge due to the lack of resources and competing national pressures.

The USA is absent from the Stockholm Convention and lacks effective federal policies to remove and safely dispose of PCBs despite being the largest producer, user, and likely holder of the largest stock of PCBs. This is a clear challenge to the global goal of achieving ESM of PCBs because, by their properties of persistence and long-range transport, PCBs are a global threat.

The global distribution of PCB use and stocks is not uniform. The USA, Canada, Soviet Union, Japan, and Western European countries dominated the use of PCBs (Figure 2A). In contrast, today that legacy is shifting globally due to transboundary transport of PCB-containing equipment and wastes. While such movement of hazardous substances is under the purview of the Basel and Rotterdam Conventions, compliance remains a challenge. This global shift is particularly problematic considering the lack of PCB management capacities in lower income/lower development countries. Global inequality is a major challenge in the implementation of the Stockholm Convention objective of achieving ESM of PCBs and POPs in general.

The Stockholm Convention set a deadline to phase out PCBs some 40 years after cessation of production and more than 50 years after many highly developed countries banned their manufacturing, import, and new use. Yet, this deadline appears unachievable due to the resources (financial and technical) and the political will required to address the problem. As the effort required to eliminate PCBs has been seriously underestimated,[17] we question the feasibility of removing other newer POPs that have entered widespread use. In the context of POPs, it could be argued that PCBs are one of the simpler problems: most use was in large, closed items, global trade in PCB-containing materials was limited, and most PCBs were held by large industries which were compelled to inventory and report them. This is a sharp contrast to two classes of POPs which have received recent attention—chlorinated paraffins and per- and polyfluoroalkyl substances, such as PFOS and PFOA. The major use of these chemicals is fundamentally different from PCBs; they are primarily used in open sources and held by millions of individuals, which stymies efforts to inventory and manage removal, with major consequences for the environmental and human health of future generations.[76] Chlorinated paraffins are a clear case of regrettable substitution as the short-chained chlorinated paraffins replaced PCBs in many open applications[26] and, since 2018, are also restricted under the Stockholm Convention. This highlights the critical need and urgency to curtail production and use of chemicals with POP characteristics as complex management challenges will not soon be solved, and the consequences are likely to fall disproportionately on lower-income/lower-development countries.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.2c01204.

Additional methods details, sensitivity analysis, and additional figures and tables (PDF)

Complete data sources for global analysis (XLSX)

Complete data sources for USA analysis (XLSX)

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Notes

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