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Multimedia Emissions Inventory of Polychlorinated Biphenyls for the U.S. Great Lakes States

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ABSTRACT

Polychlorinated biphenyls (PCBs) were banned in the United States in 1979, and since then a significant decline in their release to the environment has been observed. This decline has now reached a plateau. Several new regulatory programs have been put in place to further reduce PCB emissions/releases. However, our ability to measure the effectiveness of these regulatory/voluntary programs and to support regional fate/transport and source/receptor modeling efforts depend on reliable emission information. In this study, we attempt to improve the emission inventory for PCBs by compiling and analyzing the multimedia total PCB emission/release data reported for the U.S Great Lakes states for each year from 1990 to 2000. Although Toxic Release Inventory (TRI), National Emissions Inventory (NEI), Great Lakes Regional Air Toxic Emissions Inventory (GLRATEI), and Integrated Atmospheric Deposition Network (IADN) data formed the basis of estimating air emissions, we used the TRI, National Response Center (NRC), and PCB transformer inventory data to estimate PCB releases to land. We used the Permit Compliance System and NRC data to obtain estimates of PCB discharges to water systems in the Great Lakes states. The Remedial Action Plans for each area of concern were the primary source for estimating PCB loads of dredged sediments. On the basis of the NEI, IADN, and GLRATEI data, the total air emissions within the decade were approximately 126 t. The regionwide discharges to water systems and releases to land in the form of landfills and accidental spills in 1990–2000 were estimated as approximately 170 and 3225 t, respectively. We estimated that approximately 1.3 million t of PCB-contaminated sediment were removed or targeted for removal in five lakes of the U.S. portion of the Great Lakes basin. We stress that these estimates were based on reported amounts and the unreported PCB releases/emissions could result in significantly higher estimates.

IMPLICATIONS

There have been large-scale efforts by European countries to develop emission inventories for persistent organic pollutants. However, such comprehensive efforts have not been undertaken in the United States. This study addresses this critical need for the Great Lakes states. The PCB emission inventory presented should be instrumental in linking emissions with fate/transport/deposition models and aiding in understanding source/receptor relationships in the region. Knowledge about the size of releases and spatial/temporal patterns across the Great Lakes states should guide the efforts of regulatory agencies in assessing the effectiveness of the existing control programs and in devising future strategies for PCB reduction.

INTRODUCTION

Polychlorinated biphenyls (PCBs) were commercially manufactured in the United States beginning in 1929. They are also byproducts of other processes such as waste combustion. The principal producer of PCBs in the United States was Monsanto Industrial Chemicals Company, which made the products under the registered trademark of Aroclor and produced 700,000 t of PCBs from 1929 to 1977, with approximately 75,000 t exported from the United States. The global production of PCBs from 1930 to 1993 was estimated as ranging from 1.1 to 1.5 million t, with 97% of the total historical consumption occurring in the northern hemisphere and about one-third of this amount circulating in the global environment.[1–5] PCBs were primarily used for insulating and cooling of electrical equipment (e.g., transformers, capacitors, voltage regulators, and fluorescent light ballasts) because of their low flammability, high heat capacity, low electrical conductivity, and low chemical reactivity. Until 1971, 61% of PCBs were used in closed electrical systems (e.g., transformers, capacitors, and other electrical insulating/cooling applications), 13% was used in nominally closed systems (hydraulic fluids, heat transfer fluids, and lubricants), and 26% was used in open-end applications

(e.g., plasticizers in rubbers and resins, surface coatings, sealant materials in buildings, ink and dye carriers, paints, varnishes, adhesives, waxes, additives in cement and plaster, casting, dedusting, laminating agents, pesticide extenders, immersion oils, and carbonless copy paper).[6,7] After 1971, 100% of all PCBs produced were used in closed electrical systems. Large-scale U.S. production of PCBs was stopped voluntarily in 1977 because of persistence and accumulation of PCBs in the environment and demonstrated adverse effects on ecosystem and human health.[6,8]

In 1979, the U.S. Environmental Protection Agency (EPA) issued the PCB Ban Rule and lowered the cutoff point for inclusion in the regulation from 500 to 50 ppm PCBs for any item, mixture, or substance.[6] This rule, amended by EPA several times, banned all uses of PCBs under the Toxic Substances Control Act unless they fall into one of the following categories of allowable uses: "totally enclosed" activity (e.g., small capacitors used in home appliances); authorized uses (e.g., dielectric fluid in electrical equipment, use of PCBs in certain kinds of microscopy, and research and development); or exemptions obtained through a specific petition.[9,10]

Despite regulatory efforts, PCBs continue to pose a risk for the environment because of their continued release from several different sources,[9,11,12] which are presented in Table 1. In addition, PCBs may still be used in carbonless copy paper at any concentration and in inks at concentrations less than 50 ppm. Thus, paper mills and de-inking operations handling recycled fiber may discharge PCBs in their effluent.[9]

The PCB emission inventories developed so far primarily focused on estimating emissions into air. Because of continued environmental threats posed by PCBs and their global distribution, several international organizations such as the United Nations Environmental Program, the United Nations Economic Commission for Europe, and the International Forum for Chemical Safety have taken initiatives (e.g., Aarhus Protocol, Stockholm Convention) to control, reduce, or eliminate discharges, emissions, and losses of persistent organic pollutants (POPs) including PCBs. These bodies recommended studies to elicit fate and distribution of POPs in different regions using compartment mass balance models.[13] However, mass-balance models rely on detailed emission inventory data, which are often incomplete or lacking. Recently, there has been progress made in this regard, especially by European countries, by compiling air emission inventories. Some recent major air emission inventories for POPs in Europe are the POPCYCLING-BALTIC project; the European emission inventory for selected POPs and related compounds for 1990; the U.K. National Atmospheric Emissions Inventory for polycyclic aromatic hydrocarbons (PAHs), PCBs, dioxins, hexachlorocyclohexane, hexachlorobenzene, and pentachlorophenol; and the Czech Republic emission inventory for PAHs, PCBs and dioxins.[1] A similar effort for the United States has not yet occurred in a comprehensive manner. This study addresses this critical need, specifically for PCBs in the U.S Great Lakes (GL) region, with a multimedia focus. PCBs remain a cause for concern in the U.S. GL states, as evidenced by well-documented adverse effects on human health and ecosystems.[8] Although PCB concentrations in the environment have declined, PCBs are still present in all environmental compartments and most of the GLs, including many inland lakes, have fish consumption advisories as a result of PCB contamination.[9]

Table 1. Summary of current PCB sources to the environment.

1. Spills, leaks, fires, or improper disposal of electrical equipment that contain PCBs (e.g., transformers, capacitors, heat exchangers, etc.).
2. Combustion or incineration of waste containing PCBs, including used oil that contains <50 ppm PCBs.
3. Use or processing of excluded products containing <50 ppm PCBs.
4. Inadvertent generation of PCBs during certain production processes (up to 200 chemical processes may inadvertently generate PCBs, many of which involve the production of chlorinated solvents).
5. Storage and disposal facilities, landfills, and contaminated sites.
6. Application of sewage sludge in accordance with the Clean Water Act and the RCRA.
7. PCB-contaminated sediments.
8. Manufacture of small quantities of PCBs for research and development.

STUDY METHODOLOGY

The multimedia total PCB emissions and releases reported for the eight U.S. GL states were compiled and analyzed for each year from 1990 to 2000. These states are Illinois, Indiana, Michigan, Minnesota, Ohio, New York, Pennsylvania, and Wisconsin. The data sources utilized to estimate PCB load in air, water, land (i.e., soil), and sediment are listed below. The missing or incomplete data in a given database within the study period of 1990–2000 were noted.

Toxic Release Inventory (TRI)

TRI is a publicly available online database managed by EPA that can be queried to obtain information on toxic chemical releases and other waste management activities for approximately 650 chemicals reported annually by certain covered industry groups as well as federal facilities.[14] Air emissions data include stack and fugitive emissions. The main categories under on-site and off-site releases to land include underground injection to Class I Wells, Resource Conservation and Recovery Act (RCRA) Subtitle C landfills, other landfills, land treatment/application farming, surface impoundments, and other land disposal. Multimedia PCB emissions data (air, water, land) were obtained from the TRI database for each year from 1990 to 2000 for the GL states.

National Emissions Inventory (NEI)

The Office of Air Quality Planning and Standards (OAQPS) of EPA prepares a national database of air emissions, which includes information on major point and area sources along with mobile sources of 188 hazardous air pollutants (HAPs) listed in the 1990 Clean Air Act (CAA). The 1999 NEI for PCBs was obtained from the OAQPS.[15] The NEI HAP emission estimates are, in general, compiled from the following five primary sources: (1) state and local HAP inventories; (2) existing databases related to EPA's Maximum Achievable Control Technology (MACT) programs to reduce HAP emissions; (3) TRI data; (4) mobile source emissions estimates of EPA's Office

of Transportation and Air Quality; and (5) stationary nonpoint source emission estimates generated using emission factors and activity data. The major stationary point sources are those that emit or have the potential to emit 10 t/yr of a single HAP or 25 t/yr of a mixture of HAPs.[15] The major area sources are those that emit less than 10 t/yr of a single HAP, or less than 25 t/yr of a combination of HAPs. The 1999 data pertaining to PCB air emissions from major point and area sources for each GL state were summarized. No emissions of PCBs were reported for the mobile sources.

Great Lakes Regional Air Toxic Emissions Inventory (GLRATEI)

Under the auspices of the GL Commission, regulatory agencies charged with controlling air pollution in the GL states have collaborated to compile this regional air toxics emissions inventory in software named RAPIDS (Regional Air Pollutant Inventory Development System). The GLRATEI data for the years of 1996, 1997, 1998, 1999, and 2001 were available. Air emissions data associated with point and area sources for total PCBs for each GL state for each of these years were compiled.[16]

Integrated Atmospheric Deposition Network (IADN)

The IADN network monitors persistent and toxic chemicals under the U.S. and Canada Binational GL Toxic Strategy, which currently consists of 20 air and precipitation monitoring stations across five lakes within the basin. IADN data contain wet and dry deposition measurements, along with estimates of gas exchange of semi-volatile organic compounds (SVOCs), including PCBs, with the lake surfaces by using the air concentration measurements of the SVOCs at these sites in combination with water concentration measurements of the same chemicals. Available IADN data from 1992 to 2000 for each of the five lakes in the basin (i.e., Lakes Erie, Huron Michigan, Ontario, and Superior) were compiled and analyzed.[17]

Permit Compliance System (PCS)

EPA's Office of Enforcement and Compliance Assurance maintains the online PCS database, which tracks permitting, compliance, and enforcement status of facilities subject to National Pollutant Discharge and Elimination System (NPDES) permits under the Clean Water Act. The permitted facilities submit to EPA effluent monitoring data for all regulated pollutants discharged into waters of the United States in the form of a Discharge Monitoring Report (DMR). The PCS data extracted from the DMRs were compiled to estimate reported PCB discharges to the lakes and tributaries within the GL states. For many of the entries, the date of permit violation and thus release was not reported in the database, which prevented us from specifically obtaining data pertaining to the time period of 1990–2000. A close examination of compiled data revealed that the data for the GL states represented post-1992 PCB discharges to GL water systems.[18]

National Response Center (NRC)

The NRC includes all accidental releases and spills of hazardous substances and oil that exceed federal notification requirements in an online national database maintained by the U.S. Coast Guard. EPA established 0.454 kg (1 lb) as the reportable quantity (RQ) for PCBs under the comprehensive Environmental Response, Compensation, and Liability Act of 1980, Section 102(a). Thus, the NRC database includes any PCB releases or spills exceeding this amount. The PCB release data for the GL states from 1990 to 2000 were compiled using the NRC database. The majority of reported release entries were to land, with a minor fraction to water.[19]

PCB Transformer Inventory

EPA maintains an inventory of all in-use PCB transformers in the United States to inform emergency or fire response personnel and building owners. The inventory includes the amount of PCBs in listed transformers. EPA's PCB transformer inventory for 1999 and 2001 was examined and the 2001 data were used to validate the 1999 data. PCB amounts in transformers located in each GL state in 1999 and 2001 were estimated and PCB releases to the environment were predicted based on published leak rates for transformers.[20–22]

Summary of Contaminated Sediment Remediation Activities in GL Areas of Concern

There are 43 Areas of Concern (AOCs) across the GLs, out of which 25 are located in the United States and 3 are shared between the United States and Canada. Remedial Action Plans (RAPs) were developed for each of these AOCs to address impairments to any one of 14 beneficial uses (e.g., restrictions on fish and wildlife consumption, drinking water consumption) associated with these areas. Sediment contamination is the primary concern in AOCs. Available EPA Great Lakes National Program Office (GLNPO) documentation pertaining to each lake was examined to estimate total PCB loads of contaminated sediments removed or targeted for removal from the U.S GL AOCs.[23]

Although TRI, NEI, GLRATEI, and IADN data formed the basis of estimating regional air emissions, the TRI, NRC, and PCB transformer inventory data were used to estimate PCB releases to land. The PCS and NRC data were used to obtain estimates of PCB discharges to water systems in the GL states. The EPA GLNPO data formed the basis of PCB-contaminated sediment volume estimates. The emission and release estimates are for total PCBs, except in the case of IADN, in which a suite of 56 PCB congeners forms the basis of our estimates.

RESULTS

TRI

Total PCB air emissions from 1990 to 2000 were 167 kg (see Table 2). Air emissions mostly occurred between 1996 and 2000, with 74% as stack and 26% as fugitive emissions. Approximately 70% of total PCB air emissions were associated with a safety product facility in Illinois in 1996. Approximately 19% of total air emissions originated as fugitive emissions from a waste disposal facility in Michigan from 1998 to 2000.[14]

Table 2. Summary of the TRI PCB releases (kg) for the GL states (1990–2000).

GL State	Air	Land On-Site	Land Off-Site
IL	115.7	–	369
IN	12.4	–	61
MI	31.8	101,962	388,197
MN	–	–	731
NY	6.6	1,366,810	465,241
OH	–	8	10,059
PA	–	28	49,428
WI	0.1	–	36
Total	166.5	1,468,808	914,122

Total 1990–2000 on-site and off-site releases to land were 1469 and 914 t, respectively (see Table 2). For total on-site land releases, 93.1% of occurred in New York from 1997 to 2000, and 6.9% occurred in Michigan from 1998 to 2000, both in the form of RCRA Subtitle C landfills. These two states also had the largest share of the total off-site PCB releases, with 42.5 and 51% of the total for Michigan and New York, respectively. For total off-site land releases, 68.5% were to landfill and surface impoundments. The majority of the on-site releases (52%) occurred in 1999, with decreasing amounts in 1998 (29%) and 2000 (19%). On the other hand, a significant portion of the total (48.5%) off-site releases occurred in New York in 1997. Although 1990 (13%) and 1992 (20.4%) emissions were also significant contributors to the total, off-site PCB land releases were negligible later in the decade (from 1998 to 2000).[14]

The TRI data also revealed that 3325 t of PCBs were transferred off-site for further waste management from 1990 to 2000, with 70 and 27% occurring in Michigan and New York, respectively. The GL states also produced 25,101 t of PCB-containing production- (95% of total) and nonproduction- (5% of total) related waste from 1990 to 2000. The PCB-containing total production waste amounted to 23,912 t, with 91% in Indiana and 7.5% in New York. The regionwide PCB-containing nonproduction related waste was approximately 1189 t, with 57% in Indiana and 42% in New York.[14]

NEI

Total major point source PCB emissions were 8.7 kg in 1999, with New York sources contributing 90% to the total (see Table 3). Total major area source PCB emissions amounted to 8762 kg, with a nearly homogeneous distribution across the states ranging from 19.6% in Pennsylvania to 7.7% in Minnesota. The majority of air emissions (99.9%) were associated with area sources in the GL region in the form of open burning of residential waste. The GL states accounted for 0.1 and 28.5% of total national major point and area NEI PCB emission estimates, respectively.[15]

GLRATEI

PCB emissions inventory as a total of point and area emissions from GLRATEI is only available from 1996 to 1999, for 2001, and only for a limited number of states

Table 3. Summary of the 1999 NEI PCB air emissions (kg) for the GL states.

GL State	Point Source	Area Source
IL	0.2	707.1
IN	–	966.0
MI	–	1,298.1
MN	0.5	673.6
NY	7.8	1,173.4
OH	0.1	1,298.8
PA	0.1	1,716.3
WI	–	928.5
GL total	8.7	8,761.8
U.S. total	6,795.5	30,755.5

(see Table 4). The States of New York, Ohio, and Pennsylvania did not include PCBs in their inventories. Only Minnesota reported PCB emissions from area sources. Thus, 99.9% of total PCB emissions were associated with point sources in Illinois, Indiana, Michigan, Minnesota, and Wisconsin. The point source PCB emissions totaled 245 kg in 1996–1999 and 2001, with wet cement manufacturing sources in Minnesota contributing 91% to this amount.[16]

IADN

Wet deposition via precipitation and net gas exchange of a suite of PCBs in the five lakes of the GL basin from 1992 to 2000 were estimated (see Table 5). The loss of PCBs from the atmosphere occurs by chemical and photochemical degradation and by wet and dry deposition processes.[21] Dry deposition estimates for PCBs were not made because of insignificance of measured particulate concentrations.[14] Approximately 2 t of PCBs entered the lakes in the form of wet deposition, with Lake Superior (43%), and Lake Michigan (22%) contributing significantly to this total. The net gas exchange was estimated by summing the estimates of absorption and volatilization flux of PCBs through the lakes' surface. Volatilization of PCBs from the lakes' surface was the dominant term exceeding deposition. The net gas exchange of PCBs was approximately 31 t of PCBs. Examination of the total deposition estimates (i.e., wet deposition + net gas exchange) shown in Figure 1 reveals that the lakes have been a source of PCBs to the atmosphere, with a decreasing trend during the monitoring period. A total of approximately 29 t of PCBs was released from the lakes' surface to

Table 4. Summary of the GLRATEI PCB air emissions (kg) for 1996–1999 and 2001.

Year	Point Source	Area Source	Total
1996	11	0.04	11
1997	110	0.04	110
1998	55	0.04	55
1999	60	0.05	60
2001	8	0.05	8
Total	245	0.22	245

Note: No PCB emissions data for NY, OH, and PA were available.

Table 5. Summary of IADN PCB wet deposition (WD) and net gas exchange (NGE) estimates (kg) for the GLs.

	Superior		Michigan		Huron		Erie		Ontario	
Year	WD	NEG	WD	NEG	WD	NEG	WD	NEG	WD	NEG
1992	110	−2,000	34	−3,000			20	−450		−500
1993	84	−1,400	44	−3,200	130	−700	13	−300	37	−510
1994	45	−1,900	82	−3,400	200	−680	36	−620	35	−480
1995	96	−560	44	−800		−350	36	−670	25	−290
1996	93	−660	32	−770		−310	29	−770	24	−270
1997	78	−370	23	−190		−430	16	−460		−210
1998	110	−510	34	−80		−500	14	−430		−230
1999	100	−270	59	−50		−430	16	−460		−240
2000	69	−150	49	−250		−470	21	−550		−280
Total	785	−7,820	401	−11,740	330	−3,870	201	−4,710	121	−3,010

Note: WD = wet deposition; NEG = net gas exchange.

air in 1992–2000. Lake Michigan (39%), Lake Superior (24%), Lake Erie (15%), Lake Huron (12%), and Lake Ontario (10%) contributed to this total in decreasing order.[17] Although there is an annual 20% contribution to the total from 1992 to 1994, the contribution to the total remained around 5% annually starting in 1997. The decrease in PCBs emitted to the air appears to have reached a plateau in 1997, which suggests that air-water exchange in the lakes may currently be in equilibrium in terms of PCB flux.

PCS

The compilation of post-1992 average and maximum effluent PCB releases to GL states' water systems (i.e., lakes, rivers, tributaries, etc.) from industrial sources revealed that there is a discrepancy between average and maximum figures that stems from data entry problems (see Table 6). Although Wisconsin only reported the maximum amount of effluent releases, New York and Michigan only reported the average PCB release estimates for some of the incidents. Thus, the regionwide post-1992 effluent release containing PCBs is likely to be in the range of 367–2933 kg, although no quantitative data in the form of average or maximum amount of discharge were available for Illinois, Minnesota, Ohio, and Pennsylvania. Paper mills in Wisconsin contributed 89% to the total PCB maximum effluent estimate. In addition, secondary smelting and refining of nonferrous metals along with refuse systems in Indiana, motor vehicle parts and sewerage systems in New York, and motor vehicle parts in Michigan were additional industrial sources reported as discharging PCB-containing effluent to receiving waters in their respective states.[18]

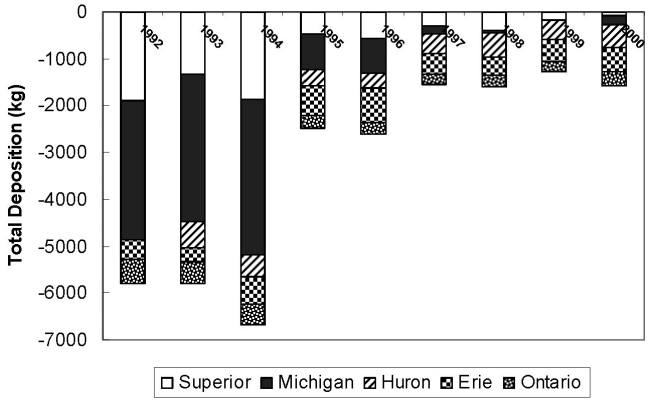

Figure 1. Summary of total deposition of a suite of PCBs for the GLs from 1992 to 2000.

NRC

The total estimates for PCB releases/spills to water (Table 7) and land (Table 8) should be construed as lower-end estimates because some of the releases could not be quantified and were thus entered into the NRC database as "uncertain amount" released. This was particularly the case in the post-1997 NRC data. Although the number of incidents was significantly reduced in the second half of the decade, the amount released was not known in many post-1997 incidents or was not entered into the NRC database. For example, although there were only five releases with unknown amounts out of 63 incidents in Illinois in 1990–1992, 6 of 13 did not provide released amount estimates in 1997–2000.[19]

Total 1990–2000 PCB release to water was estimated as approximately 166 t, with Indiana and New York contributing 80.5 and 15% to this total, respectively (see Table 7). Approximately 105 t of PCBs contaminated the land with releases across all GL states, with Ohio and Pennsylvania contributing 27 and 24% to the total, respectively (see Table 8). Although a decreasing trend has been observed within the decade, a lack of quantitative data for some of the incidents in the post-1997 period

Table 6. Summary of PCS PCB discharges (kg) to the GL waters.

GL State	PCS Average	PCS Maximum
IN	132	187
MI	194	127
NY	41	1
WI	N/A	2618
Total	367	2933

Note: N/A = not available.

Table 7. Summary of the NRC accidental PCB releases to water (kg) for the GL states.

GL State	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000
IL	1,304		UA		159					UA	14
IN	132,489			477	531						–
MI	UA	91	UA	371	UA	13	238	254			–
MN											UA
NY			8,612	1	5	583	371	9,889	231	1,305	3,284
OH	3	UA		323	138	154	291	3,185	265		1
PA	32	UA	UA	530	UA	172	477			24	3
WI	1		UA								–
Total	133,829	91	8,612	1,702	833	922	1,378	13,328	496	1,329	3,302

Note: UA = unknown amount.

prevents a definitive finding in this regard. The NRC database did not provide any information on the industrial or area sources of these accidental releases and/or spills.[19]

A total of 12.5 t of PCBs was estimated as the released amount into the environment in 1990–2000, for which the affected medium was specified as "unknown" in the NRC database. The majority of these releases occurred in 1993 (45%) and 2000 (27.5%) in Illinois (48%) and New York (47%).[19]

PCB Transformer Inventory

The entire 1999 and 2001 PCB transformer inventory databases for the United States were reviewed to ensure that PCB transformers owned in one state but operating in another were not overlooked. For example, transformers owned by companies in Iowa could actually be located in Illinois or Minnesota. The converse situation was also encountered. Where necessary, transformer entries were moved to the appropriate portion of the database to document those transformers operating in the GL states. As shown in Table 9, 5860 transformers containing approximately 18,800 t of dielectric fluid existed in the GL states in 1991. In 2001, 6118 transformers contained approximately 20,000 t of dielectric fluid. The increase in the number of transformers in 2001 is due to better record-keeping measures put in place by the regulatory agencies.

Some discrepancies were discovered within both the 1999 and 2001 databases, which could impact the estimates attributed to the GL states. Transformer owners did not complete some of the PCB transformer inventory forms correctly. The most common mistakes encountered were not specifying the location of the transformer but specifying the state of ownership, reporting one or more transformers without reporting weights of dielectric fluid, and not reporting the number of transformers owned. Also, in a few instances an owner reported values for dielectric fluid weight in both pounds and kilograms, with an inappropriate conversion used. To compensate for these errors, the state of ownership of the transformer was discerned by street location and relationship to other transformer locations within the area. The amount of PCB containing fluid in transformers varies with transformer size and the quantity changes from 40 to 500 gal (i.e., 235 to 2932 kg).[6] Where no weights were recorded for dielectric fluid in the EPA transformer database, a value of zero was assumed because the information on transformer size was not available. When the number of transformers owned was not reported, a value of 1 was assigned. In general, our adjustments and assumptions would underestimate the inventory for PCB transformers in the GL states.

The dielectric fluids in transformers typically contain 60–70% PCBs by weight (assumed 65%).[6] The electric utility industry estimated that average failure percentage for PCB transformers was 0.06%.[20–22] On the basis of this data, approximately 7 t of PCBs were estimated to be released to the environment from leaking transformers in 1999. In contrast, EPA's estimate for total amount of PCBs leaked or spilled from transformers was 228 t in 1987.[6] For

Table 8. Summary of the NRC accidental PCB releases to land (kg) for the GL states.

GL State	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000
IL	5,494	2,055	1,818	1,378	203	254	164	155	8		66
IN	1,241	5,751	966	191	2,284	315	8	107	2	4	2
MI	876	3,207	3,616	3,161	306	311	20	564	UA	14	25
MN	310	179	297	722	7,472	233	53	233	UA	3	34
NY	13	1,949	386	1,138	200	789	984	138	11	53	203
OH	4,003	4,460	1,253	742	1,808	15,910	11	11		105	53
PA	20,973	311	58	109	662	786	500	5	1,500	4	48
WI	898	344	5	12	53	26	UA	1	69		–
Total	33,808	18,255	8,399	7,452	12,990	18,626	1,740	1,215	1,590	183	432

Note: UA = unknown amount.

Table 9. Summary of number of transformers in the GL states and corresponding weight of dielectric fluid (kg).

	1999		2001	
GL State	**Number**	**Weight**	**Number**	**Weight**
IL	521	917,695	563	968,070
IN	524	840,274	681	1,299,139
MI	1,224	10,876,727	1,325	11,495,355
MN	382	242,006	391	242,162
NY	402	514,945	461	574,114
OH	1,361	3,263,512	1,383	3,279,640
PA	738	1,899,312	656	1,954,450
WI	708	211309	658	212,692
Total	5,860	18,765,780	6,118	20,025,621

1990–2000, a total PCB release of 77 t into the environment from leaking transformers was estimated, assuming an annual rate of 7 t/yr. These emissions were assumed to contaminate the soil in the vicinity of transformers and are regarded as PCB emissions to land. Because more transformers should have existed before 1999, the use of the 1999 release rate for each year from 1990 to 2000 should underestimate total PCB releases from the transformers during the decade.

Contaminated Sediment Remediation Activities

Of the 43 AOCs, 31 cited PCBs as a contaminant of concern; 25 of these 31 sites are within the United States and three are shared between the United States and Canada. However, apart from a few areas such as Waukegan Harbor, Green Bay, or the Saginaw River, the extent of PCB contamination could not be fully determined. EPA has compiled a list of sites that are in various stages of sediment remediation; 26 sites are awaiting sediment remediation and 19 sites have had some remediation but require more.

Table 10 provides a summary of the available EPA information on contaminated sediments in the GLs. A total of approximately 2 million m^3 of contaminated sediment has been removed or targeted for removal in the GL AOCs as of 2001.[23] Lake Erie and Lake Michigan AOCs accounted for 37 and 22.5% of this total, respectively. Our review of AOC reports for each of the AOCs in the United States revealed that over half of the AOCs cite PCBs as a contaminant in sediments. On the basis of the assumption that 40% of sediments removed or targeted for removal were contaminated with PCBs, this would yield a total volume of approximately 865,000 m^3 of PCB-contaminated sediments. Approximately 1.3 million t of PCB-contaminated sediments were estimated as removed or targeted for removal in the GL AOCs, assuming a mean sediment density of 1500 kg/m^3.[24]

DISCUSSION

In 1987, EPA estimated 340,000 t of PCBs remained in use (60%): 132,000 t in landfills and dumps (23%), 68,000 t released to the environment (12%), and 25,000 t were destroyed (5%) out of a total of 565,000 t of PCBs sold between 1930 and 1977.[6] Since that time, total PCB releases into the environment have significantly declined, as this multimedia assessment for the GL states validates. However, there is concern that the PCB levels have leveled off since the early 1990s. Although our analysis only examines PCB emissions inventory data for a decade, our media-specific PCB emissions estimates, findings, and observations, as derived from aforementioned databases and documented below, serve as a reference and/or benchmark and should guide future work in this area.

Air

Large differences in reported air emission estimates for both point and area sources were observed using different emission inventory databases (see Tables 2–4). A more relevant comparison of the data contained within these databases can be performed by a close examination of the 1999 data. The comparison of 1999 PCB air emissions inventory data between NEI and GLRATEI databases shows that although area source emissions were significantly underestimated in the GLRATEI inventory (8761 kg for NEI vs. 0.05 kg for GLRATEI), NEI underestimated the point source PCB emissions into the air (8.7 kg for NEI vs. 245 kg for GLRATEI). This discrepancy partly stems from different definitions used for area sources by NEI and GLRATEI. Whereas the NEI considers small point sources (emitting less than 10 t/yr of a single HAP, or less than 25 t/yr of a combination of HAPs) and nonpoint sources as area sources, the GLRATEI only treats nonpoint sources as area sources. Some other causes for these observed discrepancies are differences in reporting requirements, sources of emissions considered, and methods and approaches utilized in emissions estimations. This comparative analysis demonstrates discrepancies among various existing inventories for PCB air emissions and the need for improved cross-agency data quality control protocols and verification across databases. If the 1999 GLRATEI and the NEI estimates are assumed to be representative of annual total point and area sources, respectively, a total of approximately 97 t of PCBs was estimated as emitted into air in the GL states in 1990–2000. These findings show the need for more accurate and representative estimates of PCB point and area air emissions in the GL region.

The five GLs continue to serve as PCB sources into the air rather than sinks, with a release of 29 t of PCBs in 1992–2000. Additional research in this area also demonstrated volatilization of PCBs from contaminated waters as a major source of the PCB flux to the local atmosphere near the GL.[25,26]

Table 10. Volume of contaminated sediment (m^3) removed or slated for removal from U.S. AOCs in the GLs.

Lake	**Sediment Volume**
Erie	813,386
Superior	308,156
Huron	267,630
Ontario	286,394
Michigan	487,621
Total	2,163,187

We should caution that the air emission estimates provided here for the GL states have significant uncertainties. In addition to the problems encountered in various databases that formed the basis of our estimates and uncertain emission factor data for many controlled sources, there are many unregulated sources of PCBs that are not accounted for in these estimates. These include releases from treatment/storage and disposal facilities, landfills, hazardous waste sites, accidental releases, and landfills. The release of PCBs from landfills can occur by leaching, co-volatilization with methane generated during the decomposition of waste, and by fires.[9,27,28] It has been reported that atmospheric loadings of PCBs from unmeasured or unknown sources are likely to be greater than those emanating from regulated sources.[28] PCB emissions into air, especially for mobile PCB congeners, can be high during large-scale remedial, dredging, land farming operations involving contaminated solids, and the use of contaminated sediments for fill or landfill cover material.[29] Preliminary data based on a study conducted in Chicago illustrate the potential importance of sludge drying beds as a significance source of PCBs into the atmosphere, with annual estimates up to 90 kg.[30] In addition, experiments with wet soils and sediments contaminated with PCBs have indicated that volatilization of PCBs from these contaminated media to the atmosphere might have significant implications for the PCB mass balance, making it necessary to develop multimedia emission inventories.[3] The volatilization of PCBs from contaminated land/water in the GL states, from dredged sediment piles and PCB-waste storage sites, and other unknown sources is expected to increase the atmospheric burden of PCBs.

Water

Although adsorption to sediments and organic matter is the primary fate and transport mechanism for PCBs, some PCB congeners are water-soluble.[31] The post-1992 PCS emission estimates for the GL region (0.4–2.9 t) are highly uncertain because many of the entries did not include quantitative figures or had missing information. On the other hand, the NRC accidental release estimate (166 t) is likely to be a lower-end estimate because many entries in the post-1997 period did not provide quantitative estimates of releases. Because there was no remedial action information in either the NRC or the PCS databases, it was not possible to quantify the PCB amount available for environmental circulation. The IADN precipitation measurements showed deposition of approximately 2 t of PCBs into the GLs in the form of rain and/or snow.

Land

In general, soil receives significant portions of the PCB emission inventory through direct inputs at the surface from spills and leaks, at the subsurface via landfilling, and through indirect inputs such as atmospheric deposition.[32] Because all TRI on-site releases to land (1469 t) were to RCRA Subtitle C landfills, available PCBs for environmental circulation were expected to be low. However, it was not possible to ascertain the fraction of off-site PCB load (914 t) remaining in the GL states. The total accidental PCB land release for the GL states was approximately 105 t on the basis of the NRC estimates. A release of approximately 77 t of PCBs from leaking transformers was calculated.

Leaking capacitors can also be a significant PCB source. In the absence of publicly available data for an inventory of large capacitors operational in the region, we relied on EPA estimates for this important source. EPA estimated 1,473,000 PCB capacitors remained in service in the United States in 1994.[9,10] At the end of 2002, the estimated number of PCB capacitors remaining in service was 1,330,000.[33] In 1999, 1,383,625 capacitors were estimated to be remaining in service on the basis of linear interpolation of 1994 and 2002 data. In 1998, EPA estimated that 27% of nationally registered PCB transformers were found in the GL states.[10] If the same fraction of capacitors existed in the GL states, 373,579 capacitors would have been located in the GL region in 1999. Large capacitors typically contain 10 kg of PCBs and are estimated to have a leakage/spillage rate of 1.6%/yr.[20,21,27] Approximately 60 t of PCBs from large capacitors were predicted to be released in 1999 and 660 t of PCBs would have been released into the environment from capacitors in 1990–2000 on the basis of a 60-t/yr annual emission rate assumption. The failure rates developed by the utility industry for large capacitors and transformers may underestimate releases to the environment from these important sources because 20% of the transformers in inspected government buildings developed new leaks between quarterly inspection periods, and 19% of the PCB-transformers in the chemical industry in the United States started to leak within a year.[7,20]

There are also significant uncertainties in our estimates of PCB releases to soil. The amount of PCBs present in hazardous waste sites can be very high, which is not included in our estimates. In 1991, EPA found that PCBs were the predominant waste type at approximately 20% of the 1218 National Priorities List (NPL) sites and approximately 7% of the 29,461 Comprehensive Environmental Response and Liability Information System (CERLIS) sites nationally. The NPL sites alone were estimated to contain approximately 26 million m^3 of material contaminated with PCBs and other substances.[9] The portion attributable to the GL states could not be estimated. However, nine permitted PCB disposal facilities in the GL states were identified by examination of the national database maintained by EPA.[34] These data show that there are PCB-contaminated hazardous waste/disposal sites with the potential to serve as future sources of PCB releases to the environment.

Because of difficulty in predicting the total soil PCB burden, there are few attempts in this regard. Approximately 21,000 t of PCBs are estimated to be present in background surface soils (0–5 cm) globally, excluding the subsurface PCB burden and the added contribution from urban soils.[32,35]

Sediments

The reliable quantitative data concerning PCB removal and remediation activities in the GL AOCs are scarce. Thus, our estimate of approximately 1.3 million t of PCB-contaminated sediment removed or targeted for removal in the U.S. portion of the GL basin should be regarded

with a high degree of uncertainty. However, it is useful in terms of demonstrating the importance of the sediments as the most significant PCB reservoir in the GL region. It was not possible to ascertain the amount available for environmental circulation resulting from various sediment remediation processes. However, because the mean residence times of more chlorinated PCB congeners, PCB-153 and PCB-180, are on the order of 110 and 70 yr, respectively, sediments are expected to serve as sources of PCBs into the GL environment for decades to come.[36]

CONCLUSIONS

We emphasize the need to periodically develop the multimedia emission inventory for PCBs and critically track and evaluate the progress in terms of observed reductions. Although a consistent decline in PCB emissions/releases with the TRI and GLRATEI data from 1990 to 2000 was not observed, a significant reduction was observed with the NRC and the IADN data. Additional voluntary and regulatory initiatives recently undertaken by EPA, industry, and various stakeholders in the GL region are expected to further reduce the PCB levels and loads in the future. Some of these new initiatives include:

(1) The Canada/United States Strategy for the Virtual Elimination of Persistent Toxic Substances in the GL Basin (the Binational Toxics Strategy), which calls for a 90% reduction of high-level PCBs (>1% PCB) that were once or are currently in service in Canada by 2000, and a 90% reduction of high-level PCBs (>500 ppm) used in electrical equipment in the United States by 2006.
(2) The 1998 Persistent, Bioaccumulative, and Toxic (PBT) Pollutants Initiative of EPA, which aims to prevent additional PBTs from entering commerce.
(3) The Polychlorinated Biphenyl Phasedown Program, initiated in 1999 by EPA Region 5 and the region's major utilities to eliminate PCB-containing transformers and capacitors.

In addition, PCBs are managed and controlled under the Clean Water Action Plan, Contaminated Sediment Management Strategy, and various air program activities (e.g., MACT, Urban Area Source).[37,38] Although these regulatory programs and policy measures should continue to reduce the environmental PCB burden, there is evidence that decreases in PCB concentrations in air along with stable and persistent levels in soil/water/sediment will continue to cause re-emission from these terrestrial and aquatic compartments to air for a long period of time. Modeling studies in Europe show that this phenomenon will be most pronounced for heavy PCB congeners.[13] The time frame over which the atmospheric pool of PCBs is maintained due to repeated cycles of deposition from air to water/land and evaporation from soil/sediment/water (called the "grasshopper effect") is a function several of factors, such as the amount and concentration of PCBs in the soil/sediment/water compartments, persistence in these compartments, physicochemical properties of the PCB congeners, temperature, and whether there is free exchange of the PCBs that have been deposited in the past.[26,39] This repeated cycling is expected to continue until either the rate of supply from the PCB stocks in the terrestrial or aquatic compartments becomes limited or exhausted. IADN data indicate that air-water exchange is approaching equilibrium in the GLs. Other studies that use the fugacity concept or variants of it suggest the air-surface equilibrium may be beginning to be approached for the stock of PCBs that have escaped into the environment from past open uses.[39] Thus, it is important to continue making progress towards reducing releases of PCBs to the environment and to upgrade the PCB emission inventory in regular intervals to assess whether there is evidence that steady-state or multimedia equilibrium is being achieved. There is also a critical need to develop consistent methodology and harmonized instruments for development of emission inventories across agencies and interested bodies, as evidenced by the discrepancy shown between the NEI and GLRATEI data for the GL states. Therefore, caution must be exercised in comparing emissions inventory data across various inventory sources.

Table 11. Summary of 1990–2000 multimedia PCB emission inventory estimated for the GL states.

Affected Environmental Medium	Emission/Release Data Source	Released Amount (t)
Air	NEI/RAPIDS	97
	IADN	29
Water	IADN	1.8
	PCS	0.4–2.9
	NRC	166
Land	TRI on-site	1469
	TRI off-site	914
	NRC	105
	Transformer leaks	77
	Capacitor leaks	660
Unknown	NRC	12.5

Future emission inventories of PCBs in the United States should focus on resolution on the PCB congener level. This is especially important because PCB congeners have different toxicities and the dioxin-like toxicity of 3 non-ortho-, 8 mono-ortho-, and 2 di-ortho-substituted PCB congeners has been widely reported.[40] Inventories for total PCBs could be misleading in terms of understanding the relative impact of sources because of differences in the toxicity of PCB congeners constituting PCB mixtures. Thus, development of a speciated emissions inventory for the United States is important. This requires a renewed effort to develop congener-specific emission data for discharges from potential sources. Elsewhere, there has been limited effort to construct congener-specific emission inventories for PCBs. In the United Kingdom, PCB speciation has been incorporated into the air emission inventory since 1998.[41–43] The congener-specific emission inventory for dioxin-like PCBs is available for Japan[44,45] and for the Netherlands.[40] Breivik et al.[27,46] attempted to estimate the congener-specific global air PCB emission inventory for 22 PCB congeners.

As the emission inventory presented here demonstrates, PCB releases into the environment continue to occur in the GL states because of PCBs in electrical transformers and capacitors that are still in use, accidental releases to land/water, and releases/emissions from industrial processes. Table 11 provides a summary of the PCB

emission/release data for air, water, and land compartments of the GL states from 1990 to 2000. The total air emissions within the decade were 126 t on the basis of the NEI/GLRATEI and IADN data. The regionwide discharges to water systems and releases to land in the form of landfills and accidental spills in 1990–2000 were estimated as approximately 170 and 3225 t, respectively. If 10% of the water/land releases were assumed to evaporate into the air this would increase the atmospheric burden by approximately 340 t. These estimates do not account for potential multimedia contamination during PCB-contaminated sediment treatment, disposal, and storage. Given the significance of sediment as the PCB reservoir and extent of sediment contamination by PCBs in the GLs, this could be a very important source. We stress that most PCB emission estimates presented in this study are uncertain because valid and available data for many sources are scarce. This study clearly illustrates the need for improving the PCB emission inventory for the United States, including the GL region. There is a critical need to develop emission factors and source fingerprints for various sources and to develop inventory using newer techniques such as geographic information systems. However, our multimedia emission inventory for the GL region show that the large reservoir of PCBs present in soil and sediment will continue to feed the cycling of PCBs among air/water/land compartments for many years. The PCB emission inventory presented here should be instrumental in linking emissions with fate, transport, and deposition models and in aiding understanding source/receptor relationships in the GL region. Knowledge about the size of releases and spatial and temporal patterns across the GL states should guide the efforts of regulatory agencies in further controlling and reducing PCB releases to the environment.

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