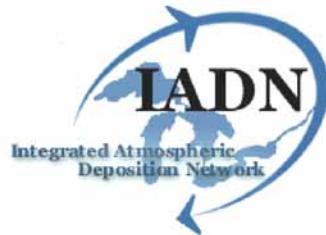


ATMOSPHERIC DEPOSITION OF TOXIC SUBSTANCES TO THE GREAT LAKES: IADN RESULTS TO 1996



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ATMOSPHERIC DEPOSITION OF TOXIC SUBSTANCES TO THE GREAT LAKES: IADN RESULTS TO 1996

Executive Summary

The primary objective of this document is to report biennial loadings estimates for the atmospheric deposition of toxic substances to the Great Lakes for the years 1995 to 1996. Atmospheric deposition is the phenomenon by which airborne substances are transferred to water, soil or vegetation at ground level. In IADN, three deposition processes to the Great Lakes are considered: wet deposition by precipitation, dry particle deposition by sedimentation, and net diffusive gas exchange that combines the effects of absorption from air to water with volatilisation from water to air.

The loading estimates presented herein are based on measurements taken largely at the IADN Master Stations, of which there is one per lake. Substances considered include those traditionally tracked by IADN,

- α - and γ -hexachlorocyclohexane, dieldrin, *p,p'*-DDE, *p,p'*-DDT, and *p,p'*-DDD
- hexachlorobenzene (HCB) and polychlorinated biphenyls expressed as Σ PCB and four individual PCBs: 18, 44, 52 and 101
- four polycyclic aromatic hydrocarbons (PAHs): phenanthrene, pyrene, benzo(k)fluoranthene, and benzo(a)pyrene
- four trace elements: lead, arsenic, selenium and cadmium, as well as others reported here for the first time,
- *trans*- and *cis*-chlordane and *trans*-nonachlor
- α - and β -endosulphane and endosulphane sulphate
- an expanded suite of PAHs.

The loadings have been determined in a manner consistent with previous IADN reports, although refinements to the model include an update to the database of physicochemical parameters used in the calculations and the use of measured rather than estimated wind speeds. As a result of these refinements, loadings presented here are not strictly comparable to those presented in previous reports. In order to develop a uniform picture of network loadings over time, results have been recalculated for the period 1992-1994 using the same model assumptions and parameters as used for 1995-96. Temporal trends discussed in this report are based on those recalculated values.

In examining the loadings of toxic substances to the Great Lakes, three fundamental issues are considered: the magnitude of the loadings, the manner in which each loading component contributes to the total, and the variation or trends in the loadings across the basin and over many years. These loadings are presented in terms of fluxes (mass/unit area/unit time; *viz.* ng/m²/d) in order to account for differences between lakes due to their areas.

Typical fluxes of banned organochlorine pesticides are on the order of 0.1 to 1 ng/m²/d and only regularly exceed 10 ng/m²/d for the gas exchange of α -HCH and dieldrin. Fluxes of individual PCB congeners tracked by IADN are typically between 0.1 and 1 ng/m²/d for each loading component, similar to many of the banned organochlorine pesticides, although seasonal volatilisation fluxes of Σ PCB can be higher than 50 ng/m²/d. HCB gas exchange fluxes are in the 1 to 10 ng/m²/d range and fluxes of current-use pesticides γ -HCH and the endosulphans are on the order of 1 to 5 ng/m²/d.

Inputs to the lakes of pesticides and PCBs are dominated by gas exchange and wet deposition. Dry particle concentration measurements ceased after 1995 due to low reported levels, but loading estimates presented here show that dry particle deposition of dieldrin, *p,p'*-DDD and PCBs may be significant when compared to the other deposition processes. Rates of dieldrin and Σ PCB volatilisation are greater than gas absorption so the lakes are acting as sources of these substances to the atmosphere.

Inputs of PAHs and metals are larger than those of pesticides and PCBs as expected by their continued emission to the environment. PAH fluxes range from 1 to 1000 ng/m²/d depending on species and loading process, and fluxes of trace metals reach values as high as 2000 ng/m²/d. Since metals are non-volatile, they are subject only to wet and dry deposition with the wet fluxes typically being the larger of the two. Available data indicate that PAH volatilisation from the lakes is small compared to the other flux terms, and gas absorption is substantial for phenanthrene and pyrene while the higher molecular weight PAHs are delivered mostly by wet and dry particle deposition.

As part of its quality assurance program, IADN has begun a new set of intercomparison studies between its participating agencies. Until results are available, comparisons of depositional behaviour between lakes and over time have been limited to those situations where data were generated by the same operating agency.

Wet deposition fluxes are generally decreasing in time for the banned OC pesticides while dry deposition and gas exchange fluxes have been variable. The temporal trend in gas exchange is generally toward air-water equilibrium. For PCBs, wet deposition is steady in time while dry deposition was increasing before measurements ceased in 1995. Gas exchange of PCBs is in the direction of volatilisation from all lakes but is generally approaching air-water equilibrium.

Wet and dry particle deposition of PAHs show no consistent trends in time though levels increase from west to east across the basin. Little net gas exchange flux information is presented for PAHs since water concentration data are sparse. Deposition of metals is limited to wet and dry deposition with wet deposition declining in time and dry deposition being variable.

Loading estimates produced by IADN have traditionally been based on the assumption that Master Stations located at remote sites on the lakes are characterizing the regional background deposition. However, strong inputs with more limited geographic influence

are also likely to exist near cities and industrial areas. Using the case of Chicago on Lake Michigan as an example, data from 1996 were used to assess the impact of air pollution from urban centres on the deposition to the lakes. The IADN calculation was modified to include a small lake sub-area influenced by the high concentrations measured at Chicago and, though results should be viewed as lower-bound estimates when compared to other studies, deposition from Chicago sources is still estimated to be substantial for certain pesticides and PCBs and for all PAHs. Further work is needed to correctly characterize the lake area affected by urban air pollution and deduce effective ways of incorporating significant urban centres in IADN loading estimates.

IADN Loadings (kg/yr) From 1992 to 1996 Lake Superior

		Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition		Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition	
α -HCH	1992	78	1.1	390	470	PCB101	1992	1.7	0.45	-42	-40
	1993	33	5.6	450	490		1993	1.6	0.74	-39	-37
	1994	38	12	710	760		1994	1.6	2.3	-25	-21
	1995	28	4.7	-230	-200		1995	3.1	1.4	-0.94	3.6
	1996	71	-	-240	-		1996	2.6	-	22	-
γ -HCH	1992	62	0.65	140	200	Σ PCB	1992	58	27	-1300	-1200
	1993	14	2.7	47	64		1993	110	25	-1200	-1100
	1994	19	2.4	95	120		1994	63	86	-1000	-850
	1995	9.5	1.9	65	76		1995	70	47	-300	-180
	1996	38	-	43	-		1996	90	-	-380	-
dieldrin	1992	21	7.4	-500	-470	phenanthrene	1992	260	100	-5500	-5100
	1993	62	6.3	-540	-470		1993	180	60	-6800	-6600
	1994	11	25	-500	-460		1994	130	310	-6800	-6400
	1995	34	15	-240	-190		1995	500	59	5700	6300
	1996	20	-	-200	-		1996	190	100	2200	2500
<i>p,p'</i> -DDD	1992	-	6.1	-	-	pyrene	1992	160	120	980	1300
	1993	17	0.1	-	-		1993	140	74	67	280
	1994	10	-	-	-		1994	210	220	-58	370
	1995	0.89	0.5	-9.3	-7.9		1995	460	54	2600	3100
	1996	0.8	-	4.3	-		1996	120	91	210	420
<i>p,p'</i> -DDE	1992	2.6	0.39	-	-	B(k)F	1992	120	52	140	310
	1993	3.8	1.2	-	-		1993	130	13	20	160
	1994	4	-	-	-		1994	92	58	70	220
	1995	4.6	0.96	-18	-12		1995	640	159	190	990
	1996	2.1	-	-14	-		1996	219	152	98	470
<i>p,p'</i> -DDT	1992	6.2	1.6	21	29	B(a)P	1992	140	58	22	220
	1993	59	1.9	12	73		1993	160	14	21	200
	1994	48	-	-	-		1994	92	39	35	170
	1995	1.7	2.4	2	6.1		1995	170	17	76	260
	1996	3.2	-	7.8	-		1996	49	35	34	120
HCB	1992	5.3	0.67	47	53	Pb	1992	-	-	-	-
	1993	25	19	15	59		1993	-	16000	-	-
	1994	1.2	0.37	16	18		1994	-	26000	-	-
	1995	1.5	0.42	24	26		1995	-	-	-	-
	1996	1.2	-	22	-		1996	-	-	-	-
PCB18	1992	0.92	0.26	-71	-70	As	1992	-	-	-	-
	1993	3.1	0.39	-74	-71		1993	-	5600	-	-
	1994	1.6	2.9	-71	-67		1994	-	2900	-	-
	1995	2	1.9	-14	-10		1995	-	-	-	-
	1996	3.4	-	14	-		1996	-	-	-	-
PCB44	1992	1.4	0.45	-19	-17	Se	1992	-	-	-	-
	1993	9.1	0.92	-14	-4		1993	-	1800	-	-
	1994	1.7	2.8	-7.3	-2.8		1994	-	3100	-	-
	1995	2.2	1.7	-8.3	-4.4		1995	-	-	-	-
	1996	1.9	-	51	-		1996	-	-	-	-
PCB52	1992	1.1	0.31	-13	-12	Cd	1992	-	-	-	-
	1993	1.8	0.56	-8	-5.6		1993	-	2100	-	-
	1994	2.1	3.1	5	10		1994	-	4400	-	-
	1995	2.6	2.4	-9.9	-4.9		1995	-	-	-	-
	1996	2.6	-	34	-		1996	-	-	-	-

IADN Loadings (kg/yr) From 1992 to 1996
Lake Michigan

		Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition		Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition	
α -HCH	1992	62	1.5	52	120	PCB101	1992	1.4	0.33	-26	-24
	1993	44	4	81	130		1993	0.81	0.66	-23	-22
	1994	98	6.3	120	220		1994	1.7	1.1	-29	-26
	1995	56	4.5	350	410		1995	2.1	1.6	-9.3	-5.6
	1996	15	-	300	-		1996	1.4	-	-9.1	-
γ -HCH	1992	65	1.1	870	940	Σ PCB	1992	52	16	-1300	-1200
	1993	120	2	250	370		1993	86	24	-1200	-1100
	1994	47	1.4	490	540		1994	71	39	-1400	-1300
	1995	26	2.2	190	220		1995	78	41	-330	-210
	1996	6.6	-	110	-		1996	48	-	-320	-
dieldrin	1992	58	8	-	-	phenan-threne	1992	350	110	-	-
	1993	55	7.2	-	-		1993	230	100	-	-
	1994	62	23	-	-		1994	160	160	-	-
	1995	47	20	-	-		1995	360	82	-	-
	1996	30	-	-	-		1996	220	100	-	-
<i>p,p'</i> -DDD	1992	-	3.8	-	-	pyrene	1992	220	140	-	-
	1993	6.4	0.98	-	-		1993	220	140	-	-
	1994	16	-	-	-		1994	130	170	-	-
	1995	1.6	1.9	-	-		1995	340	95	-	-
	1996	1.8	-	-	-		1996	140	110	-	-
<i>p,p'</i> -DDE	1992	3.8	0.48	-	-	B(k)F	1992	130	56	-	-
	1993	11	1.4	-	-		1993	110	43	-	-
	1994	3.5	-	-	-		1994	73	63	-	-
	1995	7.4	1.4	-	-		1995	480	198	-	-
	1996	3.9	-	-	-		1996	258	197	-	-
<i>p,p'</i> -DDT	1992	22	2.3	44	68	B(a)P	1992	170	77	-	-
	1993	56	6.2	35	97		1993	170	42	-	-
	1994	58	-	-	-		1994	77	63	-	-
	1995	9.7	0.67	-	-		1995	160	37	-	-
	1996	9	-	-	-		1996	84	41	-	-
HCB	1992	2.6	0.38	24	27	Pb	1992	-	-	-	-
	1993	13	13	1.2	27		1993	-	16000	-	-
	1994	1.1	0.19	-10	-8.7		1994	-	-	-	-
	1995	1.4	0.35	8.3	10		1995	-	-	-	-
	1996	0.91	-	8.3	-		1996	-	-	-	-
PCB18	1992	0.91	0.18	-69	-68	As	1992	-	-	-	-
	1993	1.3	0.46	-74	-72		1993	-	820	-	-
	1994	1.6	1.2	-75	-72		1994	-	1300	-	-
	1995	2.6	1.1	-24	-20		1995	-	-	-	-
	1996	1.7	-	-24	-		1996	-	-	-	-
PCB44	1992	1.4	0.32	-44	-42	Se	1992	-	-	-	-
	1993	1.2	0.67	-42	-40		1993	-	910	-	-
	1994	9.1	1.4	-52	-42		1994	-	-	-	-
	1995	1.8	1.1	-20	-17		1995	-	-	-	-
	1996	0.97	-	-22	-		1996	-	-	-	-
PCB52	1992	1.2	0.24	-56	-55	Cd	1992	-	-	-	-
	1993	1.1	0.55	-55	-53		1993	-	4500	-	-
	1994	2.3	1.4	-60	-56		1994	-	-	-	-
	1995	3	2	3	8		1995	-	-	-	-
	1996	2.2	-	2.2	-		1996	-	-	-	-

IADN Loadings (kg/yr) From 1992 to 1996 Lake Huron

		Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition		Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition	
α -HCH	1992	170	-	-	-	PCB101	1992	11	-	-	
	1993	140	-	-500	-		1993	6	-	0.77	
	1994	150	-	-490	-		1994	1.9	-	0.74	
	1995	220	-	-50	-		1995	-	-	-7.7	
	1996	160	-	-63	-		1996	-	-	-6.1	
γ -HCH	1992	-	-	-	-	Σ PCB	1992	180	-	-	
	1993	260	-	-34	-		1993	130	-	-490	
	1994	120	-	-19	-		1994	110	-	-460	
	1995	110	-	32	-		1995	-	-	-240	
	1996	93	-	29	-		1996	-	-	-230	
dieldrin	1992	-	-	-	-	phenanthrene	1992	-	-	-	
	1993	13	-	-760	-		1993	640	90	-	
	1994	15	-	-720	-		1994	320	71	-	
	1995	19	-	-	-		1995	250	63	-	
	1996	41	-	-	-		1996	390	110	-	
<i>p,p'</i> -DDD	1992	-	-	-	-	pyrene	1992	-	-	-	
	1993	-	-	-	-		1993	350	130	-	
	1994	1.8	-	-	-		1994	190	77	-	
	1995	3.8	-	-	-		1995	220	61	-	
	1996	6.7	-	-	-		1996	350	130	-	
<i>p,p'</i> -DDE	1992	10	-	-	-	B(k)F	1992	-	-	-	
	1993	-	-	-	-		1993	-	83	-	
	1994	3.4	-	-	-		1994	-	48	-	
	1995	9.6	-	-	-		1995	610	-	-	
	1996	7.8	-	-	-		1996	-	257	-	
<i>p,p'</i> -DDT	1992	22	-	-	-	B(a)P	1992	-	-	-	
	1993	-	-	2.5	-		1993	-	110	-	
	1994	4.1	-	2.5	-		1994	-	56	-	
	1995	10	-	-	-		1995	350	-	-	
	1996	18	-	-	-		1996	-	100	-	
HCB	1992	5.8	-	-35	-	Pb	1992	100000	11000	-	110000
	1993	11	-	-14	-		1993	64000	8000	-	72000
	1994	3.3	-	-18	-		1994	47000	11000	-	58000
	1995	3.6	-	-35	-		1995	15000	7600	-	23000
	1996	1.3	-	-28	-		1996	18000	13000	-	31000
PCB18	1992	17	-	-	-	As	1992	11000	2200	-	13000
	1993	4	-	-28	-		1993	7500	1700	-	9200
	1994	0.94	-	-29	-		1994	6500	1200	-	7700
	1995	-	-	-14	-		1995	2200	710	-	2900
	1996	-	-	-15	-		1996	2700	2900	-	5600
PCB44	1992	20	-	-	-	Se	1992	17000	2700	-	20000
	1993	5.2	-	-10	-		1993	12000	2400	-	14000
	1994	2.5	-	-10	-		1994	10000	2600	-	13000
	1995	-	-	-17	-		1995	2700	110	-	2800
	1996	-	-	-15	-		1996	3100	1100	-	4200
PCB52	1992	7.6	-	-	-	Cd	1992	6600	470	-	7100
	1993	11	-	-6.8	-		1993	2900	310	-	3200
	1994	2.6	-	-6.7	-		1994	2300	410	-	2700
	1995	-	-	-7.3	-		1995	1400	170	-	1600
	1996	-	-	-5.5	-		1996	2000	310	-	2300

IADN Loadings (kg/yr) From 1992 to 1996
Lake Erie

		Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition		Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition
α -HCH	1992	84	1.2	140	230	PCB101	0.48	0.28	-2.2	-1.4
	1993	35	2	290	330		0.81	0.37	1	2.2
	1994	19	3.4	300	320		1.1	0.73	-2.6	-0.77
	1995	29	0.9	120	150		1.4	1.4	-5.2	-2.4
	1996	6.3	-	44	-		0.51	-	-6.6	-
γ -HCH	1992	46	0.45	60	110	Σ PCB	21	16	-200	-160
	1993	23	1.3	83	110		26	14	-100	-60
	1994	22	0.83	68	91		41	29	-200	-130
	1995	13	0.6	45	59		58	32	-220	-130
	1996	1.7	-	47	-		18	-	-310	-
dieldrin	1992	28	5.6	-300	-270	phenanthrene	500	86	-	-
	1993	32	3.7	-120	-84		360	100	-	-
	1994	8.9	18	-110	-83		210	190	-	-
	1995	12	11	-110	-87		530	160	1600	2300
	1996	9.4	-	-110	-		91	190	-770	-490
<i>p,p'</i> -DDD	1992	1.9	2	-	-	pyrene	330	110	-	-
	1993	3.4	0.21	-	-		310	130	-	-
	1994	1.4	-	-	-		160	250	-	-
	1995	1.7	1.2	-	-		360	210	290	860
	1996	2	-	-	-		58	260	-80	240
<i>p,p'</i> -DDE	1992	4.6	0.53	-	-	B(k)F	150	60	-	-
	1993	4.6	0.65	-	-		140	84	-	-
	1994	3.6	-	-	-		81	100	-	-
	1995	7.8	1.2	-	-		560	580	52	1200
	1996	2.6	-	-	-		158	430	15	600
<i>p,p'</i> -DDT	1992	34	4.3	20	58	B(a)P	180	63	-	-
	1993	98	2.1	30	130		190	57	-	-
	1994	15	-	14	-		97	98	-	-
	1995	14	1.5	-	-		190	100	-0.75	290
	1996	4.9	-	-	-		50	120	-8	160
HCB	1992	0.88	0.2	-17	-16	Pb	-	-	-	-
	1993	5.4	6.4	0.76	13		-	13000	-	-
	1994	0.4	0.21	-7	-6.4		-	13000	-	-
	1995	0.73	0.22	-1	-0.05		-	-	-	-
	1996	0.34	-	-5.7	-		-	-	-	-
PCB18	1992	0.34	0.12	-17	-17	As	-	-	-	-
	1993	0.57	0.21	-15	-14		-	1500	-	-
	1994	0.56	0.74	-15	-14		-	1400	-	-
	1995	1.4	0.6	-22	-20		-	-	-	-
	1996	0.43	-	-26	-		-	-	-	-
PCB44	1992	0.55	0.23	-5.4	-4.6	Se	-	-	-	-
	1993	0.81	0.43	-1.1	0.14		-	2800	-	-
	1994	1.6	0.83	-5.2	-2.8		-	2400	-	-
	1995	0.96	1.2	-11	-8.8		-	-	-	-
	1996	0.32	-	-16	-		-	-	-	-
PCB52	1992	0.42	0.28	-5.8	-5.1	Cd	-	-	-	-
	1993	0.729	0.33	-2.2	-1.1		-	1100	-	-
	1994	1	0.85	-5.2	-3.4		-	1500	-	-
	1995	1.7	1.3	-8.6	-5.6		-	-	-	-
	1996	0.65	-	-13	-		-	-	-	-

**IADN Loadings (kg/yr) From 1992 to 1996
Lake Ontario**

		Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition		Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition	
α -HCH	1992	52	-	-80	-	PCB101	1992	1.7	-1.6	-	
	1993	32	-	-54	-		1993	2.3	-2.4	-	
	1994	33	-	-23	-		1994	0.62	-0.84	-	
	1995	21	-	-5.7	-		1995	1.8	-6.2	-	
	1996	31	-	-11	-		1996	1.3	-5.4	-	
γ -HCH	1992	50	-	-8.2	-	Σ PCB	1992	56	-450	-	
	1993	37	-	-1.5	-		1993	89	-570	-	
	1994	24	-	7.9	-		1994	15	-450	-	
	1995	13	-	12	-		1995	38	-230	-	
	1996	26	-	17	-		1996	26	-230	-	
dieldrin	1992	11	-	-330	-	phenanthrene	1992	70	-510	-400	
	1993	5.4	-	-200	-		1993	540	-	-	
	1994	3	-	-180	-		1994	380	-	-	
	1995	3.9	-	-230	-		1995	110	-	-	
	1996	4.5	-	-210	-		1996	250	-	-	
<i>p,p'</i> -DDD	1992	2.9	-	-	-	pyrene	1992	64	-46	110	
	1993	0.36	-	-	-		1993	470	-	-	
	1994	0.49	-	-	-		1994	220	-	-	
	1995	0.42	-	-	-		1995	130	-	-	
	1996	0.59	-	-	-		1996	260	-	-	
<i>p,p'</i> -DDE	1992	4.4	-	-96	-	B(k)F	1992	33	92	-9.7	120
	1993	2	-	-99	-		1993	-	79	-	-
	1994	0.61	-	-80	-		1994	-	29	-	-
	1995	5.3	-	-	-		1995	173	105	-	-
	1996	2	-	-	-		1996	311	249	-	-
<i>p,p'</i> -DDT	1992	3.3	-	4.4	-	B(a)P	1992	54	86	-	-
	1993	7.2	-	5.5	-		1993	-	79	-	-
	1994	1.1	-	8.2	-		1994	-	43	-	-
	1995	14	-	-	-		1995	63	29	-	-
	1996	4.1	-	-	-		1996	110	60	-	-
HCB	1992	6.1	-	-170	-	Pb	1992	40000	4500	-	45000
	1993	3	-	-190	-		1993	27000	5300	-	32000
	1994	0.62	-	-150	-		1994	15000	6100	-	21000
	1995	0.73	-	-32	-		1995	7600	3300	-	11000
	1996	0.87	-	-28	-		1996	5000	5100	-	10000
PCB18	1992	2.5	-	-19	-	As	1992	2900	570	-	3500
	1993	0.28	-	-22	-		1993	3100	720	-	3800
	1994	0.35	-	-19	-		1994	2100	630	-	2700
	1995	0.81	-	-18	-		1995	970	210	-	1200
	1996	0.71	-	-16	-		1996	580	610	-	1200
PCB44	1992	3.6	-	-19	-	Se	1992	5500	1000	-	6500
	1993	5.7	-	-21	-		1993	5000	1600	-	6600
	1994	0.66	-	-18	-		1994	3900	1800	-	5700
	1995	2	-	-20	-		1995	1300	150	-	1500
	1996	1.4	-	-19	-		1996	1100	340	-	1400
PCB52	1992	1.6	-	-18	-	Cd	1992	2600	96	-	2700
	1993	1.4	-	-22	-		1993	1300	99	-	1400
	1994	0.93	-	-18	-		1994	550	130	-	680
	1995	2.5	-	-11	-		1995	530	49	-	580
	1996	2.5	-	-9.5	-		1996	390	100	-	490