Forty-Year Decline of Organic Contaminants in Eggs of Herring Gulls (Larus argentatus) from the Great Lakes, 1974 to 2013

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Forty-year Decline of Organic Contaminants in Eggs of Herring Gulls (*Larus argentatus*) from the Great Lakes, 1974 to 2013

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Abstract.—Following the discovery of widespread adverse reproductive effects in fish-eating colonial waterbirds nesting in the Canadian Great Lakes, Environment Canada started monitoring contaminants in Herring Gull (*Larus argentatus*) eggs in 1974. Current and historical concentrations and rates of decline of legacy contaminants (Polychlorinated Biphenyls [PCBs], 2,3,7,8-tetrachlorodibenzo-*p*-dioxin [TCDD] and organochlorine pesticides) in Herring Gull eggs from 15 Great Lakes colonies over 40 years are reported here. Large declines in contaminant concentrations were found in all colonies from the first year of reporting to 2013, with mean percent declines ranging from 72.7% for Σ chlordane to 95.2% for Mirex, indicating reduced availability of contaminants to wildlife. First-order exponential decay regressions indicated that rates of decline in eggs varied among compounds. Herring Gulls from Strachan Island (St. Lawrence River), for example, had the highest rates of decline for Dieldrin and Hexachlor Epoxide, whereas those from Middle Island (Lake Erie) had the lowest rates of decline for TCDD and PCBs, and those from Gull Island (Lake Michigan) had the lowest rates of decline for HCB and Mirex. Exponential rates of decline in Herring Gulls mirrored those previously reported for Great Lakes fish, thus demonstrating that Herring Gulls are a good fish-eating indicator species. *Received 2 June 2014, accepted 15 July 2015.*

Key words.—Great Lakes, Herring Gull, *Larus argentatus*, organochlorine pesticide, PCBs, TCDD.

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In the late 1960s, eggs of Herring Gulls (*Larus argentatus*) in Lake Michigan were discovered to contain elevated concentrations of organic contaminants, and the species was suffering from high embryonic mortality, eggshell thinning and poor reproductive success (Keith 1966). In the early 1970s, some of these same adverse reproductive effects were found to be prevalent in Herring Gulls and other fish-eating colonial waterbirds nesting in Lake Ontario and other areas of the Canadian Great Lakes, and were severely limiting populations of colonial waterbirds (Gilbertson and Reynolds 1972; Gilbertson 1974; Gilbertson and Hale 1974; Gilbertson and Fox 1977). Those studies soon led to the adoption of the Herring Gull and its eggs by the Canadian Wildlife Service-Environment Canada as an indicator species of health and contaminant exposure for fish-eating wildlife in the Great Lakes (Gilman *et al.* 1977; Fox *et al.* 1978); therefore, the Great Lakes Herring Gull Monitoring Program (GLHGMP) was born and continues to this day.

The Herring Gull is an excellent indicator species for organic contaminants on the Great Lakes. It breeds on all five of the individual lakes, including most connecting channels, and its eggs are relatively high in lipid content where many contaminants accumulate. Herring Gulls are primarily fish-eaters and therefore have a relatively high exposure to lipophilic contaminants (Weseloh 2012). They are relatively resistant to effects of contaminants and, as such, it takes very elevated concentrations of contaminants to cause adverse health effects in the species (Weseloh 2012). Adult Herring Gulls are non-migratory on the Great Lakes and therefore only reflect contaminants from the Great Lakes (Weseloh 2012). Lastly, they are well-studied and well-known to the public.

In its current form, the GLHGMP has three different components: 1) the tracking of temporal and spatial trends of various contaminants in eggs (and other tissues) of Herring Gulls (Norstrom *et al.* 2002; Gauthier *et al.* 2008; Weseloh *et al.* 2011); 2) contaminant-related effects on the health, physiology and genetics of Herring Gulls (Rutkiewicz *et al.* 2010); and 3) the search for new emerging contaminants in the Great Lakes ecosystem (Gauthier *et al.* 2007; Letcher *et al.* 2015; Su *et al.* 2015). The entire program was reviewed (Hebert *et al.* 1999), which assessed how successful the program has been in achieving its main goals. Lastly, although not an integral part of the program, the GLHGMP has proved valuable in contributing to our understanding of food web dynamics and the importance of understanding diet in interpreting contaminant and related data (e.g., Hebert *et al.* 1999, 2008; Hebert and Weseloh 2006).

A very practical application of all three of the program's components is to assess environmental problems that have impaired wildlife, such as in Great Lakes Areas of Concern (AOCs), as well as sites that are not AOCs (Weseloh *et al.* 1995). The 1987 Protocol to the 1978 Great Lakes Water Quality Agreement has committed both Canada and the United States to the "virtual elimination of persistent toxic substances" (International Joint Commission 1988). Forty-three areas in the Great Lakes-St. Lawrence basin were designated as AOCs based on impairment of beneficial uses and the inability of the regions to support healthy aquatic life (International Joint Commission 1988). Delisting of AOCs is dependent upon the remediation of the causes of beneficial use impairments. Several Herring Gull colonies are monitored within or near the boundaries of AOCs that include Cornwall, Toronto, Hamilton, and Detroit River; the data collected on health and contaminant burdens are used to evaluate beneficial use impairments that pertain to wildlife.

Previous publications have dealt with similar data for earlier time periods on a lake by lake basis or for a specific AOC (e.g., Weseloh *et al.* 1990, 1994, 2005), as well as on a Great Lakes-wide basis (e.g., Hebert *et al.* 1994; Pekarik and Weseloh 1998; Weseloh *et al.* 2006). Many past temporal analyses of contaminants in gull (Laridae) eggs or other biota used log transformed linear regressions, or similar permutations of linear regression, or used "equilibrium lipid partitioning concentrations" to express concentrations of PCBs in air, water, sediments, fish, and Larid eggs on a common equilibrium partitioning basis. Webster *et al.* (1999) used first order exponential decay models for determining trends for Lake Ontario and Lake Superior. Furthermore, because most of the mass of PCBs and most other legacy persistent organic pollutants (POPs) in the environment are contained in sediments and soils, and because temporal trends in these substrates after POP production ceased generally follow first order decay models (Van Metre *et al.* 1998; Ayris and Harrad 1999; Sinkkonen and Paasivirta 2000), one would expect that trends in biota would similarly follow exponential decay models. Therefore, our objective, which deals with the first component of the GLHGMP, is to report on current and historical concentrations as well as changes in rates of decline, or stabilization, of legacy contaminants (Polychlorinated Biphenyls [PCBs], 2,3,7,8-tetrachlorodibenzo-*p*-dioxin [TCDD], and organochlorine pesticides) in Herring Gull eggs following 40 years of monitoring from the early 1970s to 2013.

METHODS

Egg Collection

Herring Gull eggs have been collected annually as part of the GLHGMP at 15 Herring Gull colonies throughout the Great Lakes and connecting channels since as early as 1974 (Weseloh *et al.* 2011). Colony names and locations are: Granite Island (48° 43' 12.39" N, 88° 27' 32.04" W) and Agawa Rocks (47° 21' 32.04" N, 84° 41' 35.88" W) on Lake Superior; Gull Island (45° 42' 36.41" N, 85° 50' 17.51" W) and Big Sister Island (45° 13' 26.65" N, 87° 8' 48.22" W) on Lake Michigan; Chantry Island (44° 29' 19.67" N, 81° 24' 9.57" W), Double Island (46° 10' 24.54" N, 82° 51' 45.93" W), and Channel-Shelter Island (43° 40' 12.24" N, 83° 49' 23.04" W) on Lake Huron; Fighting Island (42° 11' 27.11" N, 83° 6' 52.43" W) on the Detroit River; Middle Island (41° 40' 54.01" N, 82° 40' 54.12" W) and the Port Colborne Breakwater (42° 52' 5.99" N, 79° 15' 29.88" W) on Lake Erie; Weseloh Rocks (43° 4' 31.55" N, 79° 4' 12.65" W) on the Niagara River; Neare and North Islands and Eastport Pier 27 in Hamilton Harbour (~43° 18' 9" N, 79° 48' 9" W), Leslie Street Spit in Toronto Harbour (43° 37' 19.25" N, 79° 20' 25.95" W) and Snake Island (44° 11' 26.81" N, 76° 32' 35.61" W) on Lake Ontario; and Strachan Island (45° 1' 15.45" N, 74° 48' 43.36" W) just above the dam at Cornwall, Ontario, on the St. Lawrence River (Fig. 1). Eggs were collected annually from

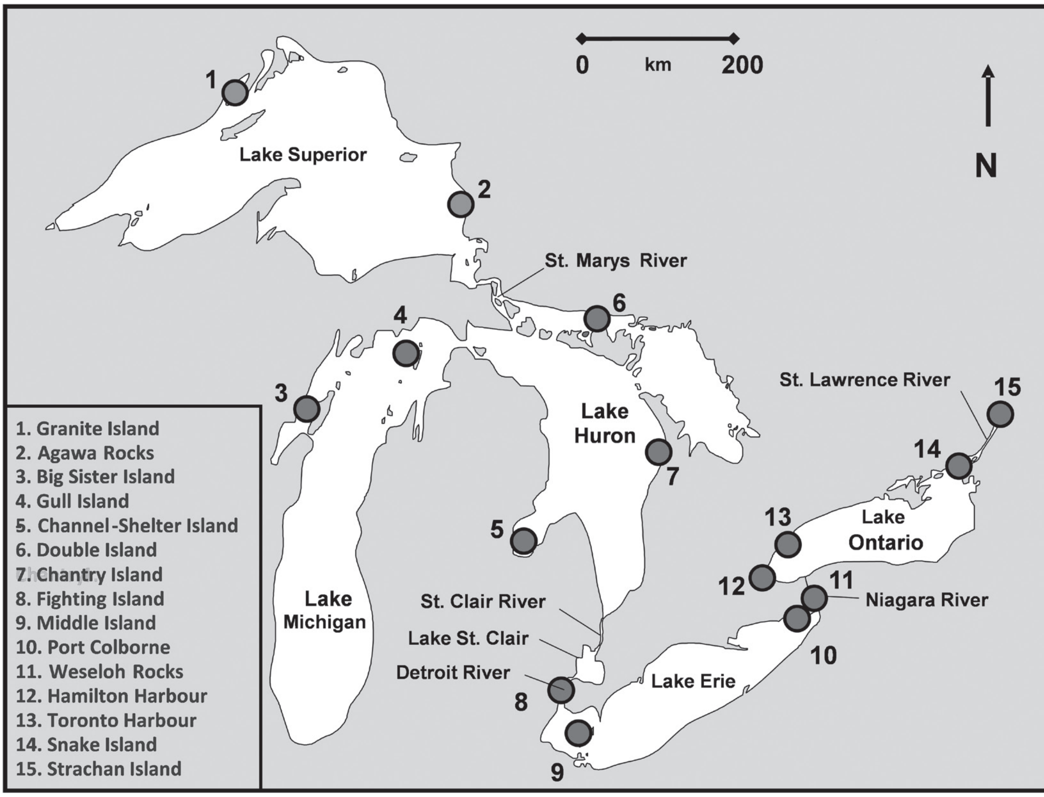

Figure 1. Herring Gull annual monitoring colonies in the Great Lakes and connecting channels, 1974-2013.

each colony generally from 1974 to 2013, usually during late April-early May; dates of first egg collection are summarized in Table 1. With some exceptions, for each year, a single egg was collected from one of 9-13 freshly-incubated Herring Gull clutches and then either analyzed as individual eggs (1974-1985) for which annual means were calculated or pooled for a single chemical analysis (1986 to present). Eggs were collected soon after the last egg was laid and clutches with two or fewer eggs were generally not targeted for egg collection. From each clutch, an egg was haphazardly collected; therefore, eggs were not biased toward eggs of a certain size or order of laying. After collection, egg contents were placed in acetone- and hexane-rinsed amber jars and frozen prior to chemical analysis. Furthermore, eggs were not collected from Fighting Island post-2010 because no breeding colonies were found on the Canadian side of the Detroit River.

Chemical Analyses

From the early 1970s to 1985, individual eggs from collections were analyzed for PCBs and other organochlorine pesticides at the Ontario Research Foundation - currently Process Research ORTECH Inc. - in Mississauga, Ontario, using high resolution gas chromatography-electron capture detection (GC-ECD; Reynolds and Cooper 1975; Norstrom *et al.* 1980). Concentrations of PCBs were expressed as a mixture of Aroclor 1254:1260 (1:1) and estimated by a single peak corresponding to PCB 138 (Turle *et al.* 1991). Starting in 1986, egg samples were analyzed as pooled samples at the National Wildlife Research Centre in Ottawa. PCBs and other organochlorines were quantified by GC-ECD for egg samples collected from 1986-1996 and then by gas chromatography with mass selective detection for egg samples collected from 1997-2013. For further details of chemical methods, see de Solla *et al.* (2010). To allow comparisons with earlier data, PCBs were expressed as Aroclor 1254:1260 1:1 equivalents. Due to space limitation, we report on concentrations of the six most abundant organochlorine pesticides or metabolites found in eggs. These include the contaminants that contributed the most to the ranking of Great Lakes colonies based on their concentrations relative to exceedances of fish flesh criteria for the protection of piscivorous wildlife (Weseloh *et al.* 2006): 1,1-bis-(4-chlorophenyl)-2,2-dichloroethene (*p,p'*-DDE), Dieldrin, Mirex, Σ chlordane (as the sum concentration of *cis*, *trans*, *oxy*-chlordane, *cis*- and *trans*-nonachlor), Hexachlorobenzene (HCB), and Heptachlor Epoxide (HE). Minimum detection limits (MDLs) for organochlorine and chemical analyses ranged between 0.0001 μg/g to 0.005 μg/g. Although changes in analytical methods used to quantify compounds during the study period could influence reported trends (de Solla *et al.* 2010), we were unable to create adjustment methods for the earlier data from 1974 to 1985, so we used unadjusted data. Gas chromatography/mass spectrometry was used to measure TCDD in Herring Gull eggs beginning in 1981 (Norstrom and Simon 1991; Simon and Wakeford 2000). All samples were above MDLs for TCDD, which ranged from 0.1-2 pg/g. Spikes, duplicates, blanks, and certified reference material were analyzed concurrently with the Herring Gull eggs and met quality assurance standards.

Statistical Analyses

Past analyses of temporal trends of contaminants in Herring Gull eggs used either log transformed linear regressions or change point regressions, which are also linear regressions (e.g., Norstrom *et al.* 2002; Hebert and Weseloh 2006; Weseloh *et al.* 2006; de Solla *et al.* 2010). In this study, we examined exponential decay models and calculated both decay constants and half-lives for compounds at all colonies to assess temporal trends. Regressions were performed using arithmetic means, where individual analyses of eggs were conducted, or as single values for pooled eggs. Earlier studies have determined that single values of pooled samples are equivalent to the mean concentrations calculated from the same eggs analyzed individually (Turle and Collins 1992). Regressions were used to assess temporal trends of contaminants in Herring Gull eggs; using both linear regressions of ln transformed contaminant concentrations, and by using a first-order exponential decay model:

$$C_t = C_0 \times e^{-\lambda t}$$

where C_t is the concentration at time t, C_0 is the concentration at the first year that data were reported, λ is the decay constant (where the concentration decreases at a rate proportional to its current value), and e is Euler's constant. The first year that egg collections were performed and reported at a Great Lakes colony was considered as $t = 1$. To compare decay constants among colonies or contaminants, 84% confidence intervals were calculated. When comparing decay constants among colonies, statistical significance was assessed as a non-overlap of confidence intervals of the decay constants among treatments. A confidence interval of 84% was used because this confidence interval is approximately equivalent to the nominal $\alpha = 0.05$ when using frequentist tests, assuming that the ratio of the two confidence intervals are one (Payton *et al.* 2003). The farther the ratio is from one, the smaller the discrepancy between the nominal value of $\alpha = 0.05$ and the actual α; as the ratio approaches ∞, the discrepancy between the nominal and actual α approaches zero. Contaminant half-lives were calculated as:

$$t_{1/2} = \ln(2)/\lambda$$

Analyses were completed using STATISTICA software (StatSoft, Inc. 2004) with a significance level of $\alpha = 0.05$.

RESULTS

Burdens of POPs in eggs of Herring Gulls have declined considerably at all colonies from the 1970s or early 1980s to 2013 (Table 1). Concentrations of PCBs (Aroclor 1254:1260 1:1 equivalents), for example, ranged between 50.1 and 165.6 µg/g among the 10 colonies monitored in the early 1970s, whereas by 2013 the highest value was 14.8 µg/g. Similar trends were observed for the organochlorine pesticides. Mirex declined the most overall, with concentrations in 2013 that were on average only 4.9% of the concentrations in the year of first reporting (Table 2). For all contaminants, the regression slopes were significantly less than 1 (all $P \leq 0.05$) for all colonies, and the mean percent declines from the first year of reporting to 2013 ranged from 72.7% for Σ chlordane to 95.1% for Mirex (Table 2). The colony that had the largest overall percent decline for all contaminants was Big Sister Island (Lake Michigan), with an average decline of 96.4%, whereas Leslie St. Spit (Toronto Harbour) had the lowest overall decline of 82.1%. For Aroclor 1254:1260 1:1 Middle Island had the lowest rate of decline; it had a significantly greater decay constant (i.e., lower rate of decline) compared to all other colonies, with the exception of Channel-Shelter Island (Lake Michigan).

When using linear regressions of ln transformed contaminant data, it was clear that, even for transformed data, rates of decline were not constant across years for all contaminants or colonies. By visually examining the data, in many cases it appeared that the rates of decline diminished in later years. We calculated change point regressions, with 2003 as a change point, for all comparisons, and compared the slopes before and after 2003. For 40% of the comparisons (9 contaminants × 15 colonies), the slopes from 2003 onward were significantly greater (less negative) than the slopes from the first year of reporting to 2002. In general, the temporal trends in contaminants better fit an exponential decay model, rather than a combination of single or break point linear regressions. As an example, the temporal

Table 1. Concentrations of Polychlorinated Biphenyls (PCBs) and organochlorine pesticides (µg/g, wet weight) and 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) (pg/g, wet weight) in Herring Gull eggs from the Great Lakes in the first and last years of reporting. Data shown represent either an arithmetic mean concentration for individual eggs or a single concentration for a pooled sample of eggs. DDE = 1,1-bis-(4-chlorophenyl)-2,2-dichloroethene; HE = Heptachlor Epoxide; HCB = Hexachlorobenzene.

Colony	Year	PCB 1260	PCB 1:1	Σ PCBs[1]	*p,p'*-DDE	HE	Σ Chlordane	HCB	Mirex	Dieldrin	TCDD[2]
Strachan Island	1986	19.82	35.79	18.81	7.44	0.06	0.22	0.07	0.94	0.16	57.0
	2013	2.68	5.33	3.03	0.65	0.01	0.03	0.01	0.12	0.02	4.8
Snake Island	1974	106.41	140.51	12.32	21.37	0.17	0.25	0.56	6.59	0.47	185.0
	2013	3.21	7.01	3.48	0.79	0.01	0.06	0.03	0.15	0.03	9.7
Leslie St. Spit	1974	125.80	165.56	9.51	23.32	0.14	0.17	0.60	7.44	0.46	60.0
	2013	4.45	8.78	4.56	1.48	0.02	0.13	0.03	0.20	0.08	11.2
Hamilton Harbour	1981	71.46	79.33	15.51	11.10	0.12	0.72	0.23	1.94	0.26	50.0
	2013	3.93	7.05	3.76	0.83	0.01	0.06	0.02	0.10	0.03	6.6
Port Colborne	1974	55.10	72.56	7.96	8.71	0.16	0.16	0.21	0.84	0.37	32.0
	2013	3.81	5.82	3.30	0.42	0.01	0.05	0.02	0.02	0.04	2.9
Weseloh Rocks	1979	44.14	50.47	5.56	4.01	0.09	0.24	0.17	0.49	0.20	87.0
	2013	3.57	5.90	3.39	0.52	0.01	0.05	0.03	0.04	0.04	4.3
Middle Island	1974	55.04	72.36	16.95	5.55	0.16	0.24	0.38	0.44	0.34	25.0
	2013	9.85	14.79	8.29	0.69	0.01	0.06	0.02	0.01	0.04	6.7
Fighting Island	1972	137.00	115.09	52.47	48.10	0.08	0.20	0.31	0.13	0.27	49.0
	2010	12.58	15.52	9.80	0.73	0.01	0.04	0.01	0.01	0.02	2.9
Double Island	1974	42.10	56.34	4.58	13.83	0.16	0.40	0.30	0.52	0.53	28.0
	2013	2.34	4.14	2.20	0.47	0.04	0.17	0.02	0.03	0.02	4.2
Gull Island	1977	96.48	111.60	10.16	27.76	0.26	0.89	0.12	0.21	0.72	58.0
	2013	3.43	7.49	3.75	1.00	0.02	0.13	0.03	0.02	0.06	3.2
Agawa Rocks	1974	37.32	50.07	5.47	14.19	0.13	0.38	0.29	0.76	0.42	79.0
	2013	1.70	3.38	1.68	0.34	0.02	0.07	0.02	0.02	0.03	3.0
Granite Island	1973	50.73	75.43	7.19	25.25	0.06	0.08	0.21	1.35	0.35	14.0
	2013	1.42	3.15	1.76	0.40	0.01	0.06	0.02	0.02	0.03	3.0

[1]Σ PCBs were not measured until 1986.
[2]TCDD was not measured until 1981 at the earliest.

Table 1. (Continued) Concentrations of Polychlorinated Biphenyls (PCBs) and organochlorine pesticides (µg/g, wet weight) and 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) (pg/g, wet weight) in Herring Gull eggs from the Great Lakes in the first and last years of reporting. Data shown represent either an arithmetic mean concentration for individual eggs or a single concentration for a pooled sample of eggs. DDE = 1,1-bis-(4-chlorophenyl)-2,2-dichloroethene; HE = Heptachlor Epoxide; HCB = Hexachlorobenzene.

Colony	Year	PCB 1260	PCB 1:1	Σ PCBs[1]	*p,p'*-DDE	HE	Σ Chlordane	HCB	Mirex	Dieldrin	TCDD[2]
Channel-Shelter Island	1980	68.85	69.55	22.03	8.90	0.13	0.29	0.19	0.20	0.18	155.0
	2013	5.05	10.64	6.59	1.04	0.01	0.04	0.08	0.02	0.02	25.0
Chantry Island	1974	64.54	85.67	3.97	20.97	0.16	0.36	0.47	2.16	0.47	45.0
	2013	1.48	2.84	1.49	0.42	0.01	0.05	0.02	0.04	0.02	4.4
Big Sister Island	1971	134.09	141.67	22.91	60.98	0.39	0.62	0.42	0.68	0.83	45.0
	2013	1.85	4.15	2.67	0.69	0.01	0.05	0.02	0.01	0.04	1.5

[1]Σ PCBs were not measured until 1986.
[2]TCDD was not measured until 1981 at the earliest.

trend for Aroclor 1254:1260 1:1 in Herring Gull eggs from Port Colborne shows different rates of decline from 1974 to 2002 compared to 2003 and onward, which could be better explained using two linear regressions (Fig. 2A). However, the first order exponential decay exhibited a better fit overall (Fig. 2B). It was not always the case; HE did not fit any distribution particularly well for Big Sister Island, including exponential decay.

Decay constants varied among colonies for each contaminant (Table 2). Strachan Island, for example, had the largest decay constants (i.e., had the highest rate of decline) for Dieldrin and HE, whereas Middle Island had the smallest decay constants for TCDD and PCBs, and Gull Island had the smallest for HCB and Mirex. For all contaminants, the colonies that had the fastest and slowest rates of decline had decay constants that were significantly different from each other (Table 2). In some cases, such as for Aroclor 1254:1260 1:1 at Big Sister Island and Middle Island, Big Sister Island initially had higher concentrations in eggs compared to those from Middle Island, but by the late 2000s the situation had reversed (Table 1). Mean half-lives for all colonies combined ranged from 5.5 to 13.7 years for PCBs, TCDD and the six organochlorines. For Σ PCBs, the half-lives ranged from 9.9 to 24.3 years among colonies, with Middle Island having the longest half-life. Overall, Middle, Granite and Gull islands had the longest half-lives for POPs.

DISCUSSION

At all colonies of Herring Gulls monitored in the Great Lakes, concentrations of PCBs and organochlorine pesticides have fallen dramatically since the 1970s. We found that, in general, trends in contaminant burdens followed an exponential decline from the 1970s to 2013, i.e., the rate of decline was proportional to the concentration at that time. Many other studies, primarily on fish, have found similar results for the Great Lakes region. Gewurtz *et al.* (2011) found that PCBs and total 1,1'-(2,2,2-trichloroethane-1,1-diyl)bis(4-chlorobenzene) (DDT)

Table 2. Mean percent declines (SD), mean decay constants (SD) and mean half-lives (SD) for Polychlorinated Biphenyls (PCBs), organochlorine pesticides and 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) in Herring Gull eggs from 15 Great Lakes colonies from the first year of reporting to 2013 (with the exception of Fighting Island where the last year of reporting was 2010). Minimum and maximum values and associated colonies are also shown. Note that colonies identified with the smallest (minimum) decay constant also have the longest (maximum) half-life and vice versa. DDE = 1,1-bis-(4-chlorophenyl)-2,2-dichloroethene; HE = Heptachlor Epoxide; HCB = Hexachlorobenzene; I = Island.

Parameter	PCB 1260	PCB 1:1	Σ PCBs	*p,p'*-DDE	HE	Σ Chlordane	HCB	Mirex	Dieldrin	TCDD
Mean	-93.66%	-91.05%	-65.17%	-94.42%	-89.25%	-72.74%	-88.61%	-95.15%	-90.15%	-88.84%
SD	0.04	0.05	0.13	0.04	0.06	0.21	0.11	0.04	0.05	0.07
Minimum	-82.10%	-79.55%	-39.08%	-87.14%	-77.17%	-23.63%	-57.16%	-87.23%	-80.40%	-73.28%
Maximum	-98.62%	-97.07%	-88.33%	-98.87%	-96.78%	-91.94%	-96.92%	-98.90%	-96.12%	-96.69%
Minimum Colony	Middle I	Middle I	Weseloh Rocks	Weseloh Rocks	Double I	Leslie Street Spit	Channel-Shelter I	Strachan I	Weseloh Rocks	Middle I
Maximum Colony	Big Sister I	Big Sister I	Big Sister I	Big Sister I	Big Sister I	Big Sister I	Fighting I	Big Sister I	Double I	Big Sister I
Decay Constant (λ)										
Mean	-0.116	-0.091	-0.054	-0.123	-0.063	-0.056	-0.140	-0.236	-0.075	-0.097
SD	0.029	0.025	0.010	0.056	0.019	0.020	0.045	0.343	0.017	0.028
Minimum λ	-0.056	-0.042	-0.029	-0.044	-0.039	-0.034	-0.072	-0.065	-0.055	-0.041
Maximum λ	-0.161	-0.128	-0.070	-0.244	-0.103	-0.110	-0.234	-1.334	-0.119	-0.146
Minimum Colony	Middle I	Middle I	Middle I	Channel-Shelter I	Granite I	Granite I	Gull I	Gull I	Granite I	Middle I
Maximum Colony	Gull I	Chantry I	Strachan I	Fighting I	Strachan I	Hamilton Harbour	Chantry I	Chantry I	Strachan I	Weseloh Rocks
Half-life (years)										
Mean	6.42	8.43	13.43	6.86	11.82	13.67	5.46	6.03	9.58	7.91
SD	2.06	3.24	3.43	3.28	3.16	3.72	1.77	3.10	1.87	3.07
Minimum	4.29	5.42	9.92	2.84	6.72	6.33	2.96	0.52	5.80	4.73
Maximum	12.41	16.35	24.27	15.63	17.77	20.24	9.57	10.67	12.63	16.95

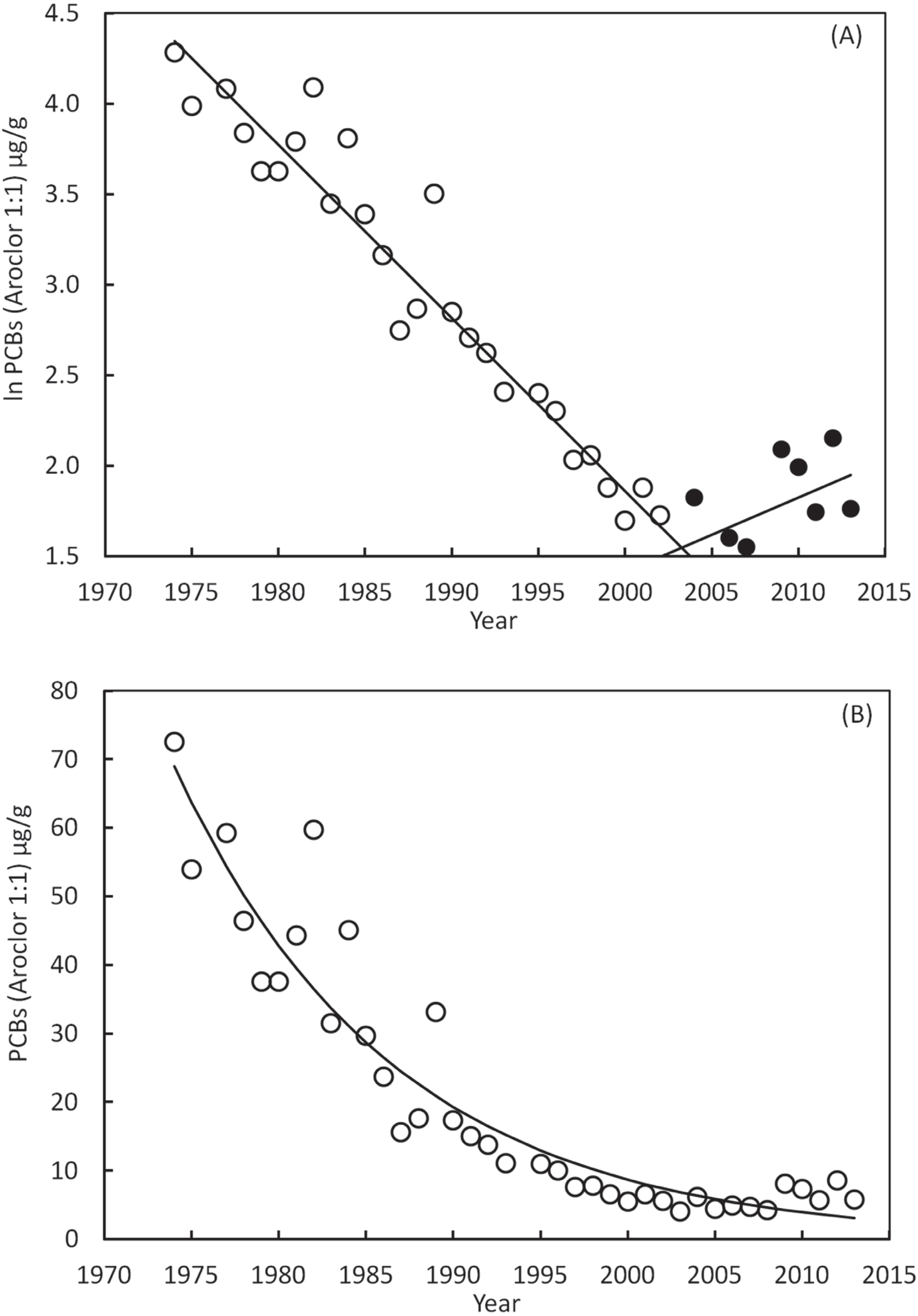

Figure 2. Temporal changes in concentrations of PCBs (Aroclor 1254:1260 1:1 equivalents; µg/g ww) in Herring Gull eggs from Port Colborne, 1974-2013. A breakpoint regression (A) ln transformed and exponential decay (B) regression was used to elucidate the nature of the decline in concentrations.

concentrations in fillets of lake trout (*Salvelinus namaycush*) and lake whitefish (*Coregonus clupeaformis*) from Lake Simcoe, Ontario, declined exponentially, but stabilized in the 1990s. Hickey *et al.* (2006) also found, using both single and double exponential models, that concentrations of PCBs and organochlorine pesticides initially declined in Great Lakes lake trout and walleye (*Sander vitreus*), but that there was a stable contaminant source resulting in more constant trends in later years. Prior to 1980, the half-life of PCBs was 2 years, but after 1980 the half-life was 13.6 years (Hickey *et al.* 2006). Sadraddini *et al.* (2011) also used a variety of exponential decay models to estimate contaminant trends in fish and found that PCB concentrations in walleye increased after the mid-1990s from Lake Erie. Similar trends have been observed outside of the Great Lakes. Because fish in the Great Lakes are showing such patterns of decline in POPs, it is not surprising that the contaminant body burdens of a primarily fish-eating bird – the Herring Gull – also exhibited a similar pattern of decline in the Great Lakes.

The rates of decline in POPs in Herring Gull eggs are generally lower in later years, and for many colonies, concentrations have stabilized in the last few years (Fig. 3). The colony that had the lowest rate of decline for PCBs and TCDD was Middle Island, which is downstream of the Detroit River. In another contaminants study, mean concentrations of PCBs in caged eastern elliptio mussels (*Elliptio complanata*) in the Detroit River from 1996 to 2010 revealed that although PCBs had declined between 2003 and 2010 at one monitoring site, burdens in mussels showed no trends at two sites and even had increasing trends at one site (Drouillard *et al.* 2013). Reasons for the stabilization or even slight increase in PCB burdens in Herring Gull eggs from the Detroit River or western Lake Erie are unknown, but may be related to resuspension of PCBs from contaminated sediment along the Detroit River. Similar long term declines in PCBs and other organochlorines in suspended sediment and fish from the Great Lakes have been reported since the 1970s/1980s (Gewurtz *et al.* 2011; Drouillard *et al.* 2013). Mercury in Herring Gull eggs from the Great Lakes have shown similar temporal trends to POPs; Weseloh *et al.* (2011) found that although overall mercury concentrations declined in Herring Gull eggs from 1974 to 2009, there were few changes in concentrations in the last 15 years of their study. Similarly, concentrations of mercury in walleye and northern pike (*Esox lucius*) generally increased (0.01-0.27 μg/g per decade) between 1995 and 2012 in Ontario (Gandhi *et al.* 2014).

Although we found dramatic declines in contaminant burdens, not all of these changes are due solely to the elimination of the contaminants in the environment. Changes in food web components may affect exposure to contaminants and body burdens in wildlife. If Herring Gulls switch to feeding lower in the food web, their rate of contaminant intake, and thus the contaminant load in their eggs, would decline. Bunnell *et al.* (2014) suggested that in many of the Great Lakes, there has been an increase in water clarity concurrent with declines in phytoplankton, invertebrates (excluding invasive species), and prey fish. Declines in prey fish were most obvious in the upper Great Lakes. By using stable isotopes and essential fatty acids, Hebert and Weseloh (2006) found that not only did Herring Gull diets and trophic level change at many Great Lakes colonies between 1974 and 2003, but when the effect of changing trophic level was removed, rates of contaminant declines were reduced. Unless researchers conducting monitoring programs are aware of a change in food web structure, they will, in this situation, overestimate the long term temporal declines in PCBs and organochlorine pesticides in fish-eating birds (Hebert and Weseloh 2006; Hebert *et al.* 2008). In actual fact, the Herring Gulls have only switched to eating a cleaner food source; therefore, the actual change in contaminants in the food web is likely lower than what we found, due to the confounding effect of the change in diet.

Not only has the fish community changed in the Great Lakes, but the food quality may have changed as well. Neff *et al.* (2012) found that lipid content has declined, in

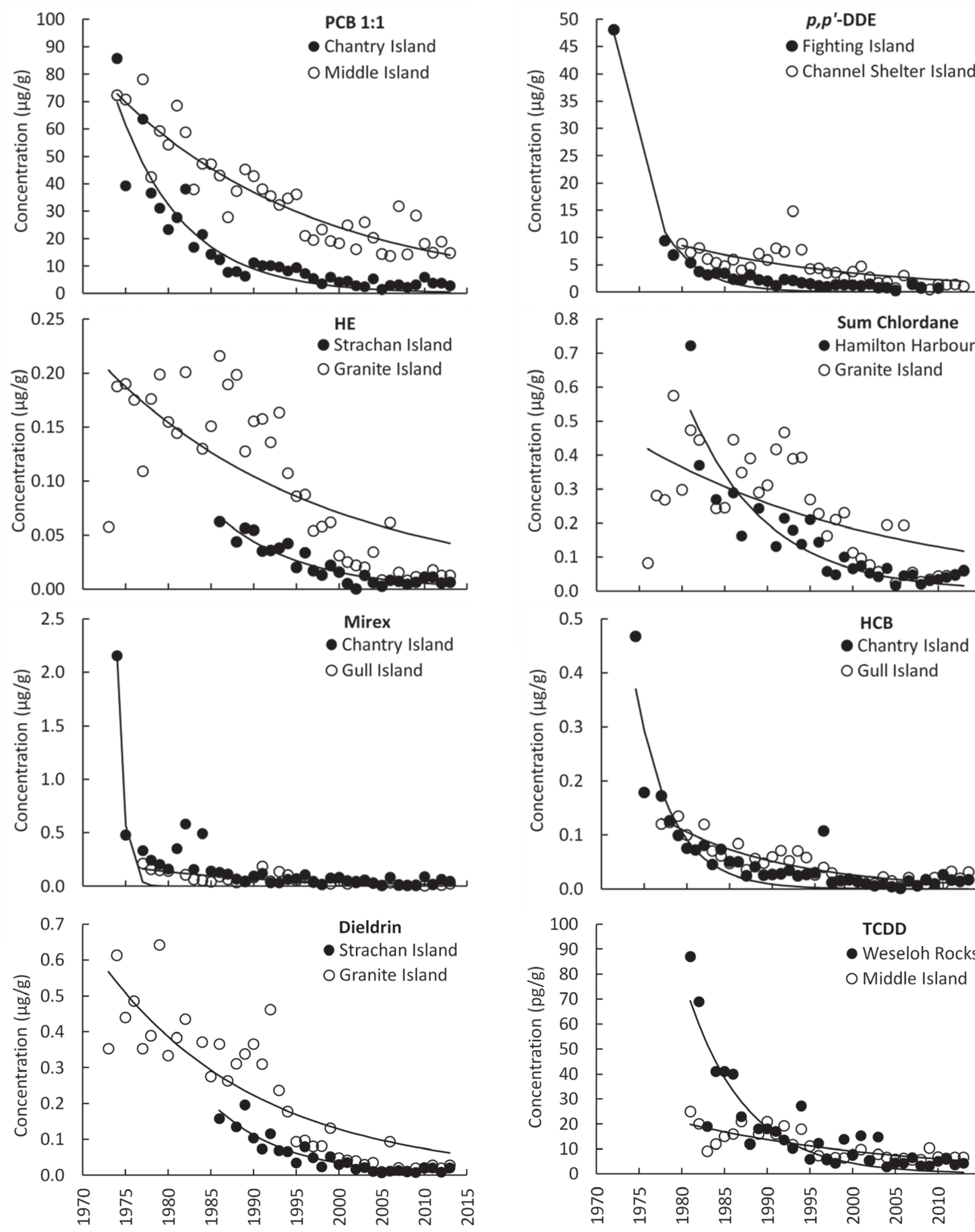

Figure 3. Temporal changes (exponential models) in concentrations of PCBs (Aroclor 1254:1260 1:1 equivalents) and six organochlorine pesticides (μg/g, wet weight) and 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) (pg/g, wet weight) in Herring Gull eggs from the Great Lakes, 1974-2013. For each compound, data were reported for the two colonies that had the highest (●) and lowest (○) decay constants.

general, in the muscle of commercial fish over a 35-year period. Therefore, not only may the energetic (and presumably nutritional) composition of fish have changed in the Great Lakes, but because POPs sequester primarily in the lipids of animals, changes in lipid composition in fish may have altered the partitioning of POPs in the food web.

Measurements of body burdens in wildlife integrate the net effect of factors such as bioavailability, temperature, growth rates, food chain dynamics, and chemical partitioning behavior, among others.

The mechanisms behind the manner in how these legacy POPs are declining are not established. The degree that factors such as degradation, metabolism, loss via atmospheric or aqueous transport, sequestering in specific environmental compartments, or changes in food web dynamics are driving the changes is not known. Nonetheless, exponential models are often assumed when estimating rates of decline of legacy POPs in abiotic compartments (e.g., Mackay *et al.* 1994). Van Metre *et al.* (1998) found that trends in concentrations of PCBs and total DDT decreased exponentially from the 1960s to 1990s in sediment cores taken from riverine systems, and suggested a first-order decay rate process. The rates of elimination of body burdens for POPs are much faster than the rates of environmental degradation; hence changes in body burdens reflect changes in the bioavailability of POPs. Degradation half-lives of tri- to hepta-chlorinated PCBs in sediment were estimated to range between 3 and 38 years, with the congeners frequently found in Herring Gull eggs having half-lives of 10 to 19 years in sediment (Sinkkonen and Paasivirta 2000). Conversely, the half-life of *p,p'*-DDE in Herring Gulls was estimated to be 264 days (Norstrom *et al.* 1986), with half-lives for PCBs likely to be similar. Hence, Herring Gulls respond faster to inputs of POPs through their diet than the degradation rate of POPs in the general environment. Abiotic factors, such as global emissions or atmospheric transport, climate change, and local geochemistry may also affect bioavailability of POPs to Larids, and thus their response time and magnitude of changes in burdens (e.g., Gandhi *et al.* 2014).

Regardless, the main determinant in the declines we observed in Herring Gull eggs is likely the availability of POPs. Although PCBs and organochlorine pesticides are very persistent, they may become increasingly unavailable to fish-eating birds, either through degradation, or by sequestering into environmental compartments that are no longer available for exposure to the food web, such as being buried in sediment. Ultimately, declines in the body burdens of these compounds can be attributed to bans in the production and severe restrictions on the use of these chemicals, improved industrial practices and the effectiveness of remedial activities such as dredging and disposal of contaminated substrates, thereby reducing the chemical inputs into the environment. Despite bans in production, the persistent nature of these compounds means that they continue to be available and accumulate in biota. For example, although production of PCBs in North America was banned in 1977, loadings of PCBs were higher than those of Polybrominated Diphenyl Ethers (PBDEs) from the city of Toronto to Lake Ontario in 2007 and 2008 (Melymuk *et al.* 2014). Although PCB production was restricted in 1974 onward to use in closed systems only, prior use in open use products such as caulking and sealants means that PCBs are still being released into the environment, thus maintaining their persistence. Assuming that the rate of degradation of POPs or loss in non-available compartments (e.g., buried in sediment, or transported outside of the Great Lakes through atmospheric transport) is exponential, not only is the temporal change in Herring Gulls likely to be exponential, but if the rates of decline of POPs slows in the Great Lakes, such changes are likely to be reflected within a similar timeframe in Herring Gull eggs.

There is little recent evidence that population numbers of colonial waterbirds in the Great Lakes are currently being limited by the direct effects of contaminants (Weseloh 2012), and productivity of Herring Gulls from colonies that once were reduced due to contaminants has returned to normal at many colonies in the Great Lakes. Nonetheless, contaminants may be having sublethal effects on gene expression or physiological responses of colonial waterbirds. Airborne polycyclic aromatic hydrocarbon exposure has been linked to the induction of mutations in Double-crested Cormorants (*Phala-*

crocorax auritus; King *et al.* 2014) in Hamilton Harbour, and PBDEs have been associated with the suppression of transthyretin in neuronal cell lines of Herring Gulls (Crump *et al.* 2008). Recent evidence suggests that reduced prey availability may be contributing to population declines reported for colonial waterbirds such as Herring Gulls and Great Black-backed Gulls (*L. marinus*) nesting on the Great Lakes (Morris *et al.* 2003; Pekarik *et al.* 2016). By using ecological tracers, Hebert *et al.* (2008) found a temporal relationship between diet in Herring Gulls and a long-term decline in pelagic prey fish abundance. Concurrent with these changes, Hebert *et al.* (2009) found that egg volume of Herring Gulls also declined, putatively due to the reduction in fish prey availability. During periods of ecological stress, the effects of POPs may be exacerbated by worsened body conditions in Larids that are induced by limited food availability (Bustnes *et al.* 2008). Hence, the relationship among ecological changes, such as changes in food web structure or disease, may have complex interactions with contaminants in Great Lakes Herring Gulls.

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