Environment International 40 (2012) 24–32

Contents lists available at SciVerse ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Differential exposure of alpine ospreys to mercury: Melting glaciers, hydrology or deposition patterns?

Mélanie F. Guigueno [a], Kyle H. Elliott [a], Joshua Levac [a], Mark Wayland [b], John E. Elliott [c,*]

[a] Department of Biological Sciences, University of Manitoba, Winnipeg, MB, Canada R3T 2N2
[b] Canadian Wildlife Service, Environment Canada, Saskatoon, SK, Canada, S7N 0X4
[c] Science & Technology Branch, Environment Canada, Delta, BC, Canada V4K 3N2

ARTICLE INFO

Article history:
Received 13 August 2011
Accepted 15 November 2011
Available online 27 December 2011

Keywords:
Atmospheric deposition
Canadian Cordillera
Mercury (Hg)
Osprey
Pandion haliaetus
Stable isotopes

ABSTRACT

Mercury (Hg) is a global contaminant impacting even remote environments. In alpine watersheds, glacial meltwater is a source of Hg, which accumulated in glaciers during the 1960–1980 cooling cycle. The considerable variation observed for Hg exposure of alpine animals in proximal watersheds could result from differences among those watersheds in Hg loading from glacial meltwater. Alternatively, variation may be the result of hydrology, atmospheric Hg deposition patterns, or food web characteristics. To examine those possibilities, we measured Hg in ospreys (*Pandion haliaetus*), apex predators in 15 watersheds in western Canada. Mercury levels in feathers of nestlings increased with increasing modeled atmospheric deposition rates and decreased with lake size. In eggs mercury decreased with $\delta^{13}C$, an indicator of food web structure, and with pH and elevation. Thus, Hg levels in chicks were strongly associated with local patterns relevant when the chicks were growing (e.g. the period post-snow melt: Hg deposition, lake size) while Hg levels in eggs were weakly associated with local patterns relevant during the snow melt (elevation, $\delta^{13}C$), with the remainder of the Hg variation in eggs determined by other factors such as possible Hg accumulation by the adult elsewhere. Modeled atmospheric deposition from prevailing upwind locations including Asia, followed by runoff into small lakes, were related to Hg patterns in osprey, with little apparent role for recent melting of glaciers. Our study highlights the importance of physical patterns to the environmental chemistry of top predators.

1. Introduction

Mercury (Hg) is a global contaminant, which appears to be increasing recently even in remote environments (Braune et al., 2006; Wang et al., 2004). Whereas Hg was once associated with industrial or agricultural point sources, Hg is now primarily distributed around the globe by low level atmospheric release from combustion sources and subsequent transport and deposition elsewhere (Scheuhammer et al., 2007; Schuster et al., 2002; Wang et al., 2004). Thus, while early human health issues focused on acute toxicity from point sources, human and environmental health concerns now include chronic exposure through the food chain from widespread, relatively low levels of Hg (Braune, 2007; Braune et al., 2006; Scheuhammer et al., 2007).

Geographically distant from sources of Hg, arctic and alpine regions nonetheless can have elevated levels of Hg due to long range transport (Schuster et al., 2002). Ice cores of alpine glaciers in western North America showed high sustained levels in the second part of the 20th century (Fig. 1). As industrial point sources in North America have been increasingly regulated, variation in Hg levels within lakes in western North America is likely associated with long-range transport from upwind sources primarily from combustion of fossil fuels in Asia (Jaffe et al., 2005). Long-range transport has been shown to affect geographical variation in Hg levels of a piscivorous bird in eastern North America, the common loon (*Gavia immer*), in accordance with predictions from transport and deposition models of Hg release from coal fired plants and other point sources mainly in the American Midwest (Evers et al., 1998).

The rapid efflux of trapped Hg from melting glaciers may also be a source of Hg in biota inhabiting glaciated watersheds. Glacial runoff is routed rapidly to surface waters after being channelized on the glacial surface (Blais et al., 2001). The runoff can be enriched with Hg because glacial meltwater has minimal contact with soils and sediments, creating little opportunity for Hg to dissipate through evaporation and binding to organic-poor glacial sediments (Bettinetti et al., 2008; Blais et al., 1998, 2001; Grimalt et al., 2001). The global melting of continental glaciers and icefields (Haeberli et al., 1999; Fig. 1) could lead to a rapid increase in Hg levels of alpine wildlife as Hg that has accumulated in glaciers over the past two centuries is released into aquatic ecosystems. Similarly, melting glaciers are believed to have resulted in increased levels of persistent organic pollutants (POPs) in alpine lakes (Donald

* Corresponding author.
E-mail address: John.elliott@ec.gc.ca (J.E. Elliott).

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doi:10.1016/j.envint.2011.11.004

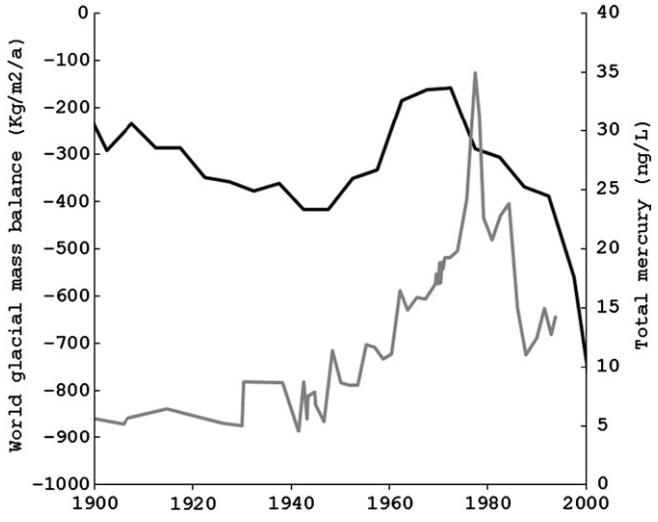

Fig. 1. World cumulative glacier volume relative to 1850 (black line, includes all glaciers excluding the Antarctic and Greenland ice sheets and recreated from data appendix in Cogley, 2009 available at http://people.trentu.ca/~gcogley/glaciology/glglmbal.htm) and Hg deposition at the Fremont Glacier, Wyoming (gray line, recreated from data appendix in Schuster et al., 2002) during 1900–2000. The peak in Hg deposition in 1980 represents the Mount St. Helens volcanic eruption.

et al., 1998; Grimalt et al., 2001; Kidd et al., 1995). Specifically, high industrial Hg deposition rates during 1960–1980 coincided with a cool period of glacial expansion, and Hg deposited during that period may be currently melting out (Fig. 1).

Ospreys (*Pandion haliaetus*) are apex predators in these ecosystems, feeding on fish in alpine lakes (Elliott et al., 2007; Grove et al., 2009; Henny et al., 2008, 2009a). They are at a similar trophic position to human fishers in these lakes and may serve as a sentinel species for human exposure to Hg through consumption of fish caught in alpine lakes (Elliott et al., 2001; Grove et al., 2009; Henny et al., 2008, 2009a,b). Because Hg biomagnifies through the food chain, ospreys are among the most highly exposed wildlife species to Hg in those systems (DesGranges et al., 1998; Henny et al., 2009a; Rattner et al., 2008; Toschik et al., 2009). Furthermore, because ospreys integrate information from the entire food chain, they are effective sentinels of environmental contamination (Elliott et al., 1998; Henny et al., 2008, 2009a,b; Toschik et al., 2009).

As many contaminants biomagnify within food chains and disperse more uniformly in aquatic compared with terrestrial ecosystems, trophic position ($\delta^{15}N$) and aquatic input ($\delta^{13}C$) often explain much of the variation in Hg levels in waterbirds (Braune et al., 2002; Elliott, 2005). We used stable isotopes to determine $\delta^{15}N$, an indicator of trophic position, and the amount of terrestrial versus aquatic input to lake ecosystems ($\delta^{13}C$) because little is known about the ecology of ospreys in this environment and osprey contaminant levels are often highly variable among nearby nests (Elliott et al., 1998, 2001). The ratio of ^{15}N to ^{14}N ($\delta^{15}N$) increases by about 2–5‰ per trophic position while the ratio of ^{13}C to ^{12}C ($\delta^{13}C$) varies depending on the carbon source for the local community, with terrestrial communities usually being depleted in ^{13}C compared to aquatic communities (Ethier et al., 2008; Hoekstra et al., 2003; Kiriluk et al., 1995; Ruus et al., 2002).

To examine the effect of melting glaciers, Hg deposition from upwind sources and food web considerations on Hg in ospreys from alpine regions of Western Canada, we measured Hg in osprey eggs and chick and adult feathers from a range of sites during 1999–2003. Western Canada is an ideal location to examine the effect of long-range deposition because extreme variation in precipitation means that wet deposition rates are highly variable (the region includes the entire range of deposition rates measured throughout North America), and because there are relatively few local sources of Hg (Bullock and Brehme, 2002; Jaffe et al., 2005). Sites include rivers, human-created lakes (hydroelectric reservoirs) and natural lakes. Here we used an information theoretic approach to examine the effects of variables that describe Hg deposition rates, watershed characteristics and trophic factors on Hg levels in ospreys. We predicted that contamination would be higher for birds feeding at higher trophic positions, and in water basins with extensive glaciation and high Hg deposition rates from the atmosphere.

2. Materials and methods

From 1999 to 2003, we sampled at 15 watersheds of the Canadian Cordillera in Alberta, British Columbia and the Yukon (Table 1, Fig. 2). We collected 83 osprey eggs, usually one per nest (data for multiple eggs from the same nest were combined using the geometric mean), 63 feathers (one per bird) plucked from the breast area, and plasma samples taken by syringe from the brachial vein of 51 nestlings and 12 adults. Nests were accessed with the assistance of electrical utility workers and the use of bucket truck when they were on utility poles or similar structures, by a professional climber when they were in trees or dead snags on land, and by leaning from a boat or helicopter or via a small net on a telescoping pole when they were in flooded snags in reservoir lakes. We captured adults using a domed noose carpet (bal-chatri) placed over the active nest or modified mist nets and attached a satellite transmitter to their back (see Elliott et al., 2007). We weighed, measured and banded each bird with a U.S. Fish and Wildlife Service band on one leg and a red alphanumeric band on the other leg. Further details on egg and blood sampling and handling are available (Elliott et al., 2000).

Adult ospreys molt body feathers year-round, but especially when they arrive at the wintering grounds (Prevost, 1983). Because Hg is sequestered in the feathers, Hg levels in adult feathers are believed to represent the integrated level within the body (Bond, 2010; Burger, 1993) and therefore reflect Hg obtained throughout the annual cycle (Bearhop et al., 2000; Monteiro and Furness, 2001). Ospreys begin producing eggs about two weeks prior to laying while female ospreys are at their breeding site about four weeks prior to laying and about two weeks after arriving at the breeding site after migration from the wintering grounds (Elliott et al., 2007). Thus, eggs likely represent exogenous stores local to the breeding grounds, as is the case in most birds (Jacobs et al., 2009; Oppel et al., 2010).

2.1. Hg analysis

All samples were shipped frozen to the Environment Canada laboratory at the National Wildlife Research Centre (NWRC) in Ottawa. We used feathers to measure Hg exposure as most of the sampled blood volume was needed for analysis of a range of organic contaminants. Feather samples were washed by shaking them sequentially in containers of acetone, dilute Triton-X and deionized water. After drying in a clean air hood overnight, the feathers were freeze-dried for 24 h.

Samples collected until 2000 were analyzed for total mercury by continuous-flow cold vapor atomic absorption spectrophotometry (CVT-AAS, Perkin-Elmer 3030B with VGA-76), as described by Scheuhammer and Bond (1991). For eggs, approximately 0.5 g of homogenate was transferred into a pre-weighed acid-washed test tube and freeze dried for at least 24 h until constant weight was obtained. Dry weights of samples were recorded and moisture contents were calculated so that concentrations could be expressed on either a dry or wet weight basis, as appropriate. Dried samples of eggs and feather were measured out into plastic, acid washed test tubes, then 0.25 mL of deionized water and 0.5 mL of HNO_3 (70%) were added to each dry sample. The samples sat overnight at room temperature. The next day they were heated, loosely capped, at 100 °C in dry heating blocks for 4 h. The cool samples were diluted to 2.0 mL in deionized water and analyzed by CVT-AAS.

Table 1
Summary information on the watersheds included in the study. If samples were taken on more than one lake within a watershed, geometric means were calculated. Hg values are in μg/g. Stable isotope values ($\delta^{15}N/\delta^{13}C$) are averages for chick plasma. For more information, please refer to Appendix A.

Water body	Watershed size (km^2)	Lake size (km^2)	Percent glaciated	Elevation (m)	pH	Sample size[a]	Egg Hg	Chick Hg	Adult Hg	$\delta^{15}N$	$\delta^{13}C$
Athabasca	2505	6	9	1210	8.4	6 (5, 5, 0)	0.21	3.08	–	9.3	−26.6
Atlin	8820	586	9	675	7.8	6 (6, 5, 3)	0.46	3.34	4.66	9.6	−24.3
Bow	2596	8	1	1546	8.0	6 (6, 4, 1)	0.17	2.81	1.05	11.4	−27.3
Columbia	16,330	24	9	808	8.0	8 (7, 8, 2)	0.39	7.36	6.60	8.8	−26.6
Downton–Carpenter	2014	45	0	676	7.3	5 (6, 3, 0)	0.54	6.01	NS	9.7	−27.0
Kinbasket	14,773	402	11	745	8.0	6 (6, 0, 0)	0.40	NS[b]	NS	NS	NS
Nicola	4296	24	0	645	8.2	8 (10, 0, 0)	0.43	NS	NS	NS	NS
Oldman	8808	6	0	c	8.3	7 (4, 8, 1)	0.25	3.59	2.11	13.1	−28.2
Ootsa	13,225	365	1	852	6.7	11 (7, 10, 1)	0.77	2.84	5.54	10.6	−29.2
Osoyoos	8203	369	0	305	8.3	5 (4, 5, 1)	0.37	2.33	4.71	13.9	−26.3
Pitt	1170	70	12	10	6.9	6 (5, 2, 2)	0.52	10.48	17.32	10.0	−25.7
Revelstoke	5253	66	11	594	7.7	4 (4, 0, 0)	0.54	NS	NS	NS	NS
Snafu	1508	9	0	793	7.7	1 (1, 1, 0)	0.12	20.50	NS	10.4	−26.9
Upper Arrow	7643	228	4	431	7.9	5 (5, 0, 1)	0.26	NS	12.31	8.1	−27.3
Williston	66,623	1660	0	690	7.8	7 (7, 0, 0)	0.36	NS	NS	NS	NS

[a] Number of nests (number of eggs, number of chicks, number of adults).
[b] NS — not sampled.

For samples from 2001 onward, concentrations of total Hg in freeze-dried egg and feather samples were determined directly using an automated mercury analyzer (AMA-254; Canalytical, Burlington, ON, Canada). The AMA-254 uses a combustion/catalyst tube to break down the sample in an oxygen-rich environment thus removing interfering elements. All Hg is then trapped by a gold amalgamator from the evolved gasses and a dual-path length cuvette/spectrophotometer determines Hg content. The practical detection limit of the instrument is 0.12 ng Hg, which corresponds to 0.006 μg g^{-1} in the average 0.020 g dry mass sample.

Quality control included repeated concurrent analysis of procedural blanks, and certified and in-house standard reference materials (SRMs). Analytical variability was determined by including duplicate samples in the analyses. Certified standard reference materials used for total Hg analysis included: National Research Council of Canada (NRCC) DOLT-2 (dogfish liver) and NRCC DORM-2 (dogfish muscle), ERM®-CE278 Mussel Tissue and BCR®-463 Tuna Fish. The accuracy of the method was confirmed by analyzing the concentration of certified reference materials DOLT-3 and TORT-2 from NRRC and Oyster Tissue 1566b from NIST. National Wildlife Research Centre in-house standard reference materials consisting of pooled egg samples from herring gulls that have been repeatedly analyzed over several years were also used for total Hg to insure comparability of results following method changes. All results are reported in μg g^{-1} dry weight.

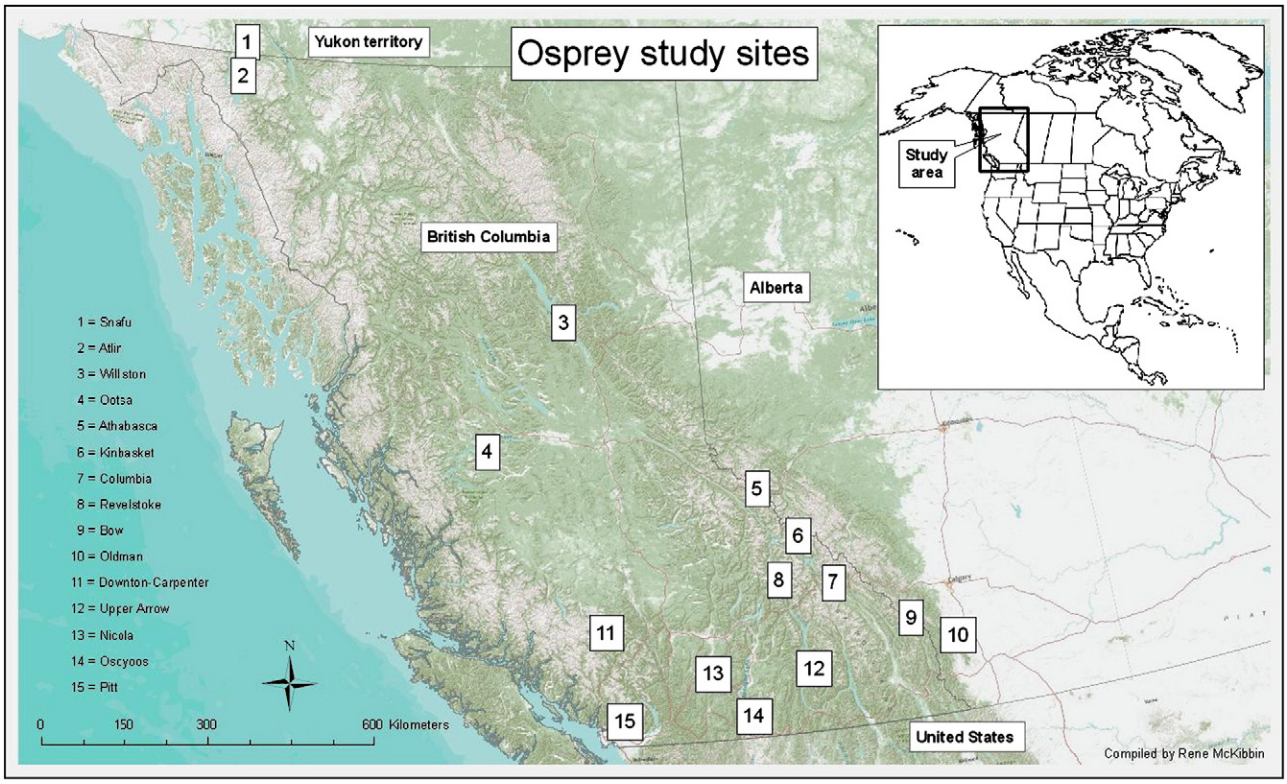

Fig. 2. Sampling location for osprey eggs and feathers in western Canada.

2.2. Se analysis

Selenium was measured in a subset of 30 chick feathers. Following the same sample preparation as for mercury analyses, selenium concentrations were determined by the method described in Neugebauer et al. (2000), except that Perkin-Elmer Analyst 800 Graphite Furnace Atomic Absorption Spectrometer (Waltham MA) with Zeeman background correction was used. The method detection limit in the digest under these conditions was 20 ng/mL (0.15 μg/g in 0.03–0.05 g dry mass) for selenium. The recoveries of the reference material samples were within the confidence interval. NRCC Dorm-2 and Dolt-2 certified reference materials were analyzed to confirm accuracy. Reagent blanks were included with each sample set. One feather sample was analyzed in triplicate.

2.3. Stable isotope analysis

We did not analyze feathers for SI, because feathers integrate Hg exposure, nitrogen isotopes and carbon isotopes over different time scales (Bond, 2010). Rather, we analyzed SI in plasma which provides an indication of the trophic position at which the chick is being fed. We also analyzed SI in the egg, which provides an indication of the trophic position of nutrients deposited by the female at the time the egg is made (~two–three weeks prior to the lay date up to the lay date, all of which time the female would already be at the breeding location; Elliott et al., 2007). Stable-carbon and nitrogen isotope assays of freeze-dried plasma and eggs were performed on 1 mg subsamples of powdered material at the stable isotope facility of the University of California Davis. Samples were first loaded into tin cups and combusted in a Robo-Prep elemental analyzer at 1200 °C. The resultant CO_2 and N_2 gasses were separated and analyzed using an interfaced Europa 20:20 continuous-flow isotope ratio mass spectrometer. Results were reported in delta notation in parts per thousand (‰) relative to Air ($\delta^{15}N$) and Vienna Pee Dee Belemnite ($\delta^{13}C$) (see Becker et al., 2007; Bond, 2010; Elliott et al., 2007).

We did not extract lipids as variance in carbon isotopes due to lipid fractionation (~0.4–0.5‰ greater variance for piscivorous birds; reviewed in Becker et al., 2007) is much lower than spatial variation in carbon isotopes within our study population (chick plasma SD = 2.04‰; egg SD = 1.96‰). We only consider correlations between Hg and $\delta^{13}C$, not the absolute value of $\delta^{13}C$, so consistent depletion of ^{13}C in yolk due to lipid fractionation would make no difference in the strength of correlations; also, $\delta^{13}C$ was unrelated to percent lipids (KHE, unpubl. data). Similarly, we did not measure baseline levels of $\delta^{15}N$ at each water body. Variation in baseline levels measured from two snail species within 23 mountain lakes in western Canada (SD = 0.77‰ and 0.80‰; range = 1.64‰ and 1.88‰; S. Lord, pers. comm.) were about one-third the variation within our osprey population (SD = 1.83 (chick plasma), 1.61 (egg); range = 6.8 (chick plasma), 6.6 (egg)), suggesting that variation in baseline $\delta^{15}N$ is less important than other factors affecting osprey $\delta^{15}N$. The two sites that had elevated $\delta^{15}N$ values (Oldman and Osoyoos) drain regions with high anthropogenic input, and subtracting 4.2 units to obtain an average $\delta^{15}N$ value for both basins similar to other sites did not change the ranking of any of the models. Furthermore, provided the $\delta^{15}N$ baselines were not correlated with Hg levels, variation due to baselines would insert random noise that would increase variance without changing the direction of the relationship.

2.4. Calculation of geographic features

We used the Spatial Analyst toolbox and Hydrology toolset in ARCGIS 9.2 to measure the size of watershed basins of osprey foraging on lakes and rivers within the Canadian Cordillera (ESRI, www.esri.com). We calculated the elevation of osprey nest location and the area of the watershed (km^2) using the digital elevation model and glacier data archived at Natural Resources Canada (2001). Glacial areas (km^2) were calculated using current glacier extents. All data were re-projected into the same coordinate system UTM Zone 11N, NAD83 to ensure aerial conformity. Post-processing of data with the Hydrology toolset required sink-filling using the sink tool (see Jenson and Domingue, 1998 for the algorithm) and the calculation of slope, aspect, flow direction and accumulation of water, and stream networks using flow direction and accumulation, watershed and stream network algorithms (Strahler, 1957; Mark, 1988; Tarboton et al., 1991). Once data for watersheds were developed, areas of both watersheds and glaciers were calculated using the script within ARCGIS 9.2.

2.5. Hg deposition

We estimated the Hg wet deposition rate from the CMAQv4.4 model based on 2001 Hg deposition monitoring (www.cmascenter.org). The CMAQ modeling system is "a comprehensive air quality model designed to operate on a range of domain sizes from urban to continental" (Bullock et al., 2008). Using emissions estimates, meteorological data and pollutant concentrations along the boundaries of the modeling domain, the model simulates atmospheric processes, including non-linear photochemistry and reaction products associated with atmospheric chemicals, within a three-dimensional array of pre-defined finite volume elements and can model complex interactions between all of the pollutants in air and cloud water that might exist within each element (Bullock et al., 2008). The model predicts wet mercury deposition measured at ground stations across North America (Bullock and Brehme, 2002; Bullock et al., 2008) and has the lowest bias and mean error of any of the models considered in the North American Mercury Model Intercomparison Study (Bullock et al., 2008, 2009). The main factor influencing the accuracy of the model is precipitation (Bullock et al., 2008, 2009). Indeed, the model outputs from all of the mercury models considered in the above study track precipitation levels across British Columbia closely, and therefore agree with one another on relative geographic variation (Bullock et al., 2008, 2009).

2.6. Statistical analysis

We used the Akaike Information Criterion (AIC) to examine general linear models that included pH, predicted Hg deposition rate, isotopic ($\delta^{15}N$, $\delta^{13}C$) abundance, year, nestling age (estimated from wing length; for nestlings only) and all geographic variables (elevation, watershed area, lake area, water body type and percentage of a watershed that is covered by glaciers) as explanatory variables and dry-weight Hg in osprey chick feathers and eggs as a response variable. We completed separate analyses for eggs, nestling feathers and adult feathers, except for an initial model that examined gross differences between the three groups. We log-transformed all total Hg concentrations in feathers and eggs so that they were normally distributed. We included the null model in all model selection procedures. As information theoretic approaches (AIC) are fundamentally different than frequentist (P-value) approaches, we largely avoid the presentation of P-values. Nonetheless, all of our major results from the general linear models remain statistically significant under a frequentist (P-value) approach. The only P-values presented were for the basic analyses in the preliminary portion of the Results section.

3. Results

All raw data can be found in the Appendix A. When all data were pooled, sample type (egg, chick feather or adult feather) was the most important variable in a general linear model of Hg including all explanatory variables ($F_{2,71} = 144.5$, $P < 0.0001$). Hg levels were lower in eggs than in chick feathers (mean = 1.06 ± 0.15 ppm greater in chicks) and lower in chick feathers than adult feathers (mean = 0.30 ± 0.26 ppm

Table 2
ΔAIC values for general linear models explaining variation in Hg in BC and Alberta osprey chick feathers and eggs (only models with ΔAIC < 1.5 are shown). Dependent variables include pH, Hg deposition rate (Hg deposition rate), elevation, lake area (Lake area), glacier area, watershed area, the proportion of glaciers in watersheds, the relative size of lakes to watersheds, δN, and δC (Aquatic input). The sign (+ or −) indicates whether the relationship was positive or negative.

	Egg			Chick		
	ΔAICc	AICc weights	Log-likelihood	ΔAICc	AICc weights	Log likelihood
Null	10.90	0.00	46.71	24.75	0.00	47.42
−Elevation−Aquatic input−pH	0.00	0.31	55.16			
−Elevation−pH	0.45	0.20	53.94			
Hg deposition rate — Lake area				0.00	0.37	61.80
Hg deposition rate — Glacier area−Lake area				0.61	0.20	62.50

greater in adults; Scheffe's Test: $F_{0.05} = 3.13$). Hg levels were highly correlated between chick and adult feathers from the same nest ($R^2 = 0.64$, $t_6 = 2.89$, $P = 0.03$; regression equation: Adult = 1.06 ± 0.68 * Chick + 0.08 ± 0.56) and within two chicks from the same nest ($R^2 = 0.51$, $t_8 = 2.54$, $P = 0.04$; regression equation: Chick One = 0.89 ± 0.52 * Chick Two + 0.13 ± 0.73; where chick one and chick two are two chicks from the same nest, not necessarily hatched in that order), with no difference from the expected 1:1 relationship.

Trophic position, as indicated by $\delta^{15}N$, was correlated between chicks and adults ($R^2 = 0.96$, $t_4 = 3.33$, $P = 0.03$), chicks and eggs ($R^2 = 0.70$, $t_{29} = 7.90$, $P < 0.0001$) and between two chicks from the same nest ($R^2 = 0.96$, $t_6 = 11.49$, $P < 0.0001$). Terrestrial input, as indicated by $\delta^{13}C$ was correlated between chicks and adults ($R^2 = 0.89$, $t_4 = 2.95$, $P = 0.04$), chicks and eggs ($R^2 = 0.19$, $t_{29} = 2.38$, $P = 0.02$) and within two chicks from the same nest ($R^2 = 0.98$, $t_6 = 13.37$, $P < 0.0001$). Furthermore, $\delta^{15}N$ correlated weakly with increased $\delta^{13}C$ (feathers: $R^2 = 0.05$, $t_{45} = -2.13$, $P = 0.04$; egg: $R^2 = 0.03$, $t46 = -1.27$, $P = 0.21$).

3.1. Feathers

Adult feather Hg averaged 5.6 μg/g and varied between 1.1 μg/g (Exshaw, Alberta, along the Bow River) to 30.4 μg/g (along the Pitt River just below Pitt Lake, British Columbia). Chick feather Hg averaged 3.9 μg/g and varied between 1.8 (at the highest elevation nest sampled, Goat Pond, Bow Valley, Alberta) and 20.5 μg/g (Snafu Lake, Yukon). When all dependent variables were included, modeled Hg deposition rate (from the CMAQv4.4 model) and lake area were in all of the most parsimonious models that predicted Hg levels in feathers of chicks (Table 2). Hg levels increased with modeled Hg deposition rate and decreased with lake area (Fig. 3).

3.2. Eggs

Mercury averaged 0.38 μg/g. The lowest concentration (0.04 μg/g) was at the highest elevation nest sampled (Goat Pond, Bow Valley, Alberta) and the highest concentration (2.02 μg/g) was at a middle elevation nest (Ootsa Lake, British Columbia). When all response variables were included, elevation and water body pH were consistently in the most parsimonious models that predicted levels of Hg in osprey eggs (Table 2). Aquatic input ($\delta^{13}C$) was also included in the most parsimonious model (Table 2). Mercury levels in eggs decreased with elevation, pH and aquatic input (Fig. 4), but relationships were generally not as strong as for feathers.

4. Discussion

Despite living in pristine environments, far from point sources of contamination, apex predators in Canadian alpine lakes had highly variable levels of Hg. Non-biological variables—Hg deposition rates and pH—explained the large variation in osprey contaminant levels (e.g., chlorinated hydrocarbons) in western Canada commented on by previous authors (Elliott et al., 1998, 2001, 2007). Hg levels of piscivorous birds across North America correlate with Hg deposition rates, with higher levels in eastern North America due to proximity to industrial sources and higher levels in the Arctic due to long-range transport and deposition (Evers et al., 1998; Scheuhammer et al., 2008). Across our smaller study area, the pattern was reversed; Hg levels were highest in the west, probably indicating the role of upwind sources of Hg from industrial activities and coal-fired power generation in Asia (Jaffe et al., 2005). Similarly, persistent organic pollutants are known to bioaccumulate in alpine lakes in North America and Europe through long-range transport (Campbell et al., 2000; Carrera et al., 2001, 2002; Villa et al., 2003). After evaporating in warmer regions, POPs condense in colder regions and pulses of contaminants can be detected after snowfalls in the alpine region (Villa et al., 2003).

Feather Hg was about an order of magnitude higher than egg Hg, as is the case in most birds because birds exude Hg in growing feathers (Bond and Diamond, 2009; Burger et al., 2008; Lewis et al., 1993). In some adult ospreys the feather Hg concentration was

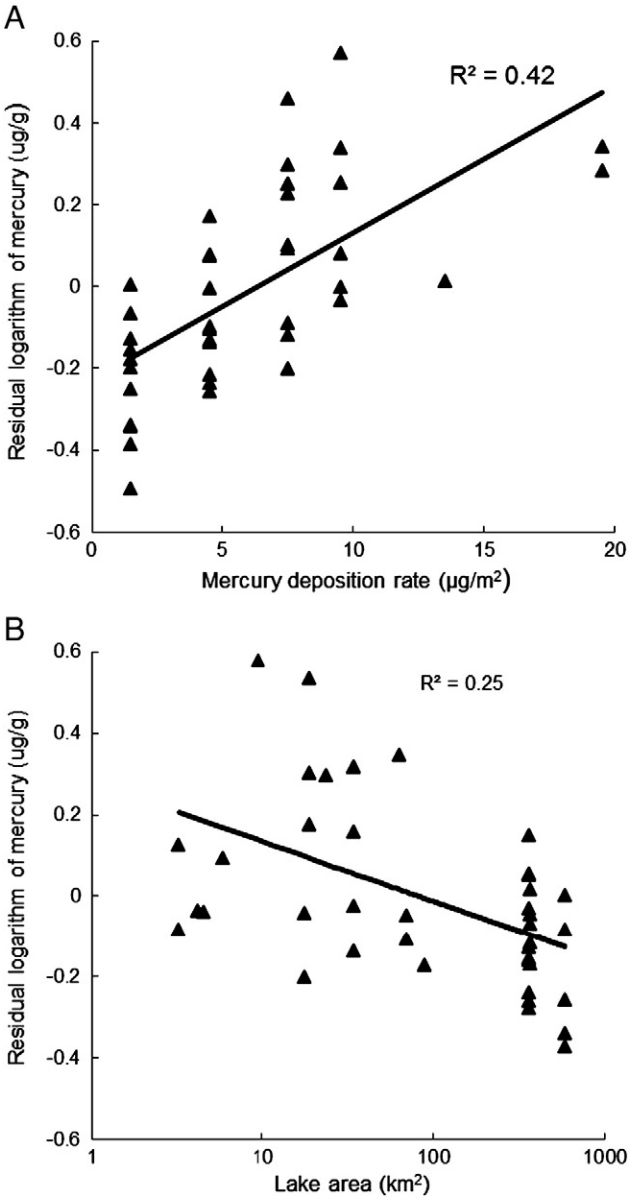

Fig. 3. Natural logarithm of Hg (ppm) (A) increased with Hg deposition rate (from the CMAQv4.4 model) and (B) decreased with lake area for Hg in chick osprey feathers from Western Canada. To account for the effect of both variables, the value for lake area is the residual of the logarithm of Hg on Hg deposition rate and the value for Hg deposition rate is the residual of the logarithm of Hg on lake area.

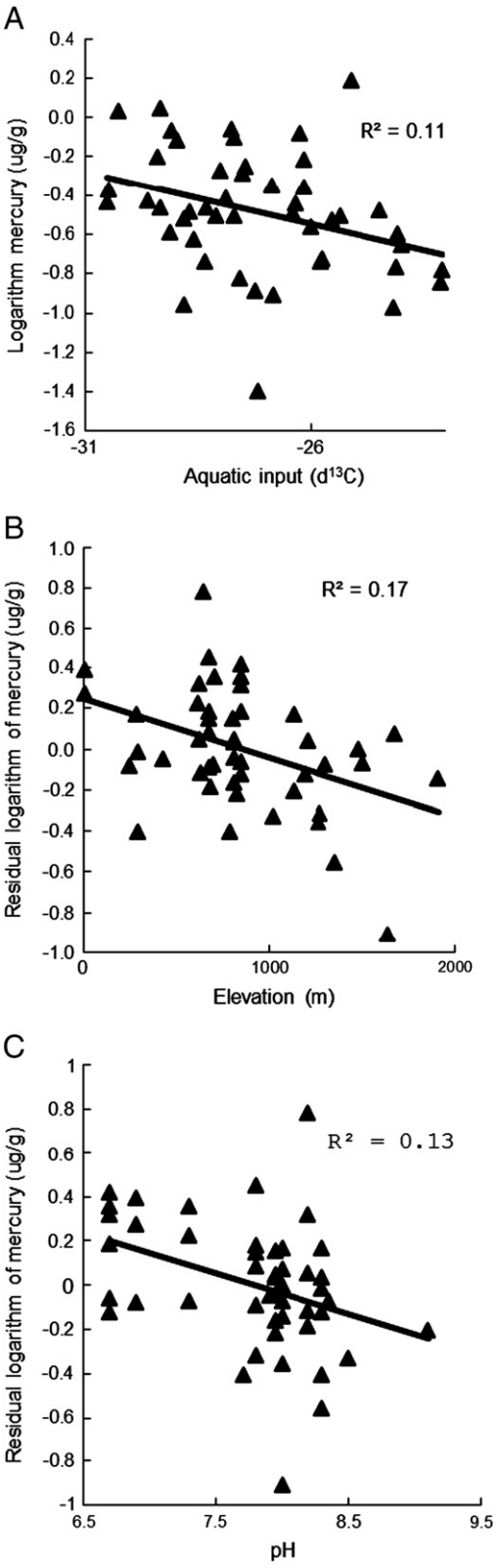

Fig. 4. Natural logarithm of Hg (ppm) decreased with (A) aquatic input, (B) elevation and (C) pH for osprey eggs from Western Canada. To account for the effect of all three variables, the value for elevation is the residual of the logarithm of Hg on both aquatic input and pH and the value for pH is the residual of the logarithm of Hg on both aquatic input and elevation.

greater than 20 μg/g, suggested by some authors as potentially associated with deleterious effects, such as impaired reproduction (Ackerman et al., 2008). As expected, Hg levels were highly correlated between feathers from two chicks at the same nest, as well as between chicks and adults at the same nest. Similar correlations occur in eagles nesting on lakes within our study area (Weech et al., 2006; but see Bond and Diamond, 2009).

Variation in mercury levels in chick feathers was largely explained by modeled atmospheric deposition rates (Fig. 3). The CMAQv4.4 model predicted that Hg deposition rates would be highest where rainfall was greatest and closest to Asian sources. Specifically, the CMAQ model predicted that Hg deposition would be highest in Coast Mountains and along the western edge of the Rockies and lowest in the dry southern Interior valleys (e.g. Okanagan) and Alberta foothills. The modeled deposition rates closely mimicked variation in osprey feathers. Lake area also played a role, with Hg levels lowest in larger lakes, perhaps because lake volume is larger and Hg therefore more diluted, or because a larger proportion of sediments are deposited in deeper water and fish feeding on benthic food chains were less accessible to osprey. The finding that Hg was negatively associated with $\delta^{13}C$ indicates that more terrestrial food sources were less contaminated with mercury. Osprey at many of the study sites consumed a significant dietary component of benthic feeding catostomid sucker and cyprinid carp species (Morrissey et al., 2004), which presumably would be at a lower trophic level than the more pelagic feeding salmonids which dominated the diet at other sites. We would have expected pelagic food chains in larger lakes to have longer food webs; however, there was no relationship between osprey Hg loads and trophic level as $\delta^{15}N$. Other factors being equal, Hg levels would presumably be highest in chicks fed from small lakes on the windward side of northern Vancouver Island or the northern mainland coast because of the high precipitation and relative proximity to Asian Hg sources, but we did not sample those locations.

Osprey eggs had lowest levels of Hg where water body pH, elevation and nutrient input from aquatic sources were highest (Fig. 4). Thus, ospreys feeding in the alkaline, high elevation lakes of the Canadian Rockies on food chains more enriched in ^{13}C, had lowest Hg levels. A study of organochlorines in our highest elevation site, Bow Lake, found significant negative relations between, for example, toxaphene and $\delta^{13}C$ in biota (Campbell et al., 2000). More efficient transfer of organochlorines to pelagic food webs than to benthic food webs was believed to have been due to higher lipid content of pelagic organisms, particularly some zooplankton species. Similarly, efficient transfer of methyl-mercury to lipid-enriched pelagic food chains may partially explain the patterns with ^{13}C observed in our study. Other factors that may be in effect at higher elevation sites include: low sediment fluxes of Hg probably related to low concentrations of organic particles, low net production of methyl-mercury related to low temperature and low concentration of dissolved and sediment organic carbon, higher rates of demethylation related to high transparency and low temperature, and short food chains as suggested in a study of fish at alpine lakes in Europe (Rognerud et al., 2002; but see Blais et al., 2006 where Hg levels increased with elevation in fish within a small geographical area).

Models were poorer (lower R^2 values) for eggs than for feathers (Figs. 3 and 4), perhaps reflecting some variation due to Hg accumulation in the female during migration or while on wintering grounds. Ospreys winter in Latin America and contaminant levels in eggs may reflect some Hg exposure on the wintering grounds, although egg levels of organochlorines were not related to wintering ground exposure for ospreys satellite-tracked from the Canadian Rockies (Elliott et al., 2007). Nonetheless, egg Hg levels were higher in acidic lakes, similar to patterns found in the Upper Great Lakes (Evers et al., 1998; Scheuhammer et al., 2007; Wang et al., 2004). Indeed, whereas chick feathers strongly ($R^2 > 0.5$) represented local patterns relevant when the chicks were growing (e.g. the period post-snow melt: Hg deposition, lake size), Hg levels in eggs were weakly ($R^2 < 0.2$) associated with local patterns relevant during the snow melt (elevation, $\delta^{13}C$). Elevation influences snow pack levels and timing of the snow

melt relative to breeding while the terrestrial signal ($\delta^{13}C$) would be associated with runoff brought into the system during the snow melt. Other factors we were unable to quantify, such as inter-annual variation in snow pack accumulation and snowpack runoff patterns, may also have influenced the variability of egg Hg concentrations.

Whereas most studies have focused on local processes affecting Hg at relatively smaller scales, especially at locations where Hg levels are particularly high due to local sources, our study focused on Hg levels over a large area with few sources of local Hg. Consequently, exposure of ospreys to Hg was relatively low in our study area (Table 3). Indeed, our values for eggs were lower than virtually all other studies except at some reference locations while our values for chick feathers were typical of most other studies (Table 3) Adult feathers, however, were lower than most other values (Table 3). Because local sources were unimportant in contributing to Hg loading in our study area, it is not surprising that we were able to detect variation in deposition rates across this large scale. Presumably, our study reflects the trends in exposure of most wildlife, which are not directly impacted by local sources. The studies by Evers et al. (1998) and Scheuhammer et al. (2008) of the common loon showed an increasing trend of Hg progressing from east to west across North America largely determined by the increasing deposition from industrial sources in the mid-western U.S. and eastern Canada. However, ours is the first study of Hg trends in a fish-eating bird across western Canada with its rather unique topography characterized by three major north–south mountain ranges reaching to 4000 m elevation, the Coast, the Columbia and Rocky Mountains interspersed with small ranges to 3000 m, and thus having an important affect on east–west air flow and deposition patterns of atmospherically transported contaminants.

Our patterns do not support the idea that melting glaciers were a major source of Hg contamination for wildlife in the Canadian Rockies. If melting glaciers were an important source, we might expect those watersheds with large amounts of glacial coverage and at higher elevations to have the highest levels of contamination (Bettinetti et al., 2008; Bizzotto et al., 2009; Blais et al., 1998, 2001). We observed the opposite patterns. Glacial extent during our study (1999–2003) suggests that the material melting out of a typical glacier would date from more than a century previous, well prior to industrial Hg deposition (Fig. 1), although apparently 10% of the glacial melt during 1998 in one of our study lakes, Bow Lake, originated from ice deposited during the 1950–1970 period (Blais et al., 2001). Rather than representing historical deposition, we suggest that Hg melts out during the spring snow pack melt and accumulates in lakes (Villa et al., 2003). Given that glaciers have, on average, been receding for over a century, Hg accumulated in a particular winter would presumably melt out in the subsequent few summers; the glacial ice currently melting in late summer often pre-dates the Industrial Revolution and likely dilutes Hg concentrations. Ospreys in small lakes at low elevations had the highest Hg levels, possibly because Hg was less able to disperse in small lakes and because flow rates are lowest at low altitudes, where the incline is less. Additionally, atmospheric Hg deposition from Asia is now much higher than in the 1960–1980s, and is it likely over-riding any effects of glacial melting.

We also found little support for biomagnification of Hg within lake food chains affecting osprey Hg loads. Similarly, a recent study examining Hg in alpine fish and osprey found that Hg did not biomagnify from fish to osprey, and the authors suggested that biomagnification occurs at lower levels of lake food webs (Grove et al., 2009). Hg levels in ospreys were better predicted by nutrient source than by trophic level, a finding similar to that of a study of persistent organic pollutants in eagles from western Canada (Elliott et al., 2009) Collectively, these results suggest that limnology may play a more important role than trophic position in affecting Hg levels in piscivorous wildlife in western North America. Trophic position is generally a weaker predictor of Hg levels than of organochlorines for piscivorous birds (Elliott, 2005; Weech et al., 2006).

Thus, whereas ecological factors play a preeminent role in determining Hg levels in some other regions (Braune et al., 2006; Mallory et al., 2004), modeled Hg deposition rates and hydrology better predicted Hg levels in alpine ospreys of western Canada than glacial melt or trophic position, likely reflecting the substantial variation in Hg deposition rates across our study area. Differential Hg levels in ospreys were influenced principally by a combination of differences in Hg deposition rates from upwind areas, likely in Asia, as well as hydrology (lake size, elevation), water chemistry (pH) and relative terrestrial and aquatic resource inputs to the watershed.

Table 3
Hg levels (μg/g DM) in ospreys from different regions. Values shown are average ± SD, with sample size in parentheses. If SD was not given, then range is shown. Feathers are body contour feathers, unless otherwise indicated. Values in bold are in wet weight.

Location	Adult feather	Chick feather	Egg	Source
Clear Lake, California, USA	15.5 ± 4.0 (39)[a]	5.25 ± 2.22 (12)		Anderson et al., 2008
Eagle Lake, California, USA	3.7 ± 1.5 (not reported)[a]			Buck and Kaiser, 2011
Coeur d'Alene, Idaho, USA		2.17 ± 1.00 (18)		Cahill et al., 1998
St Joe, Idaho, USA		2.25 (mdl-3.5) (18)		Cahill et al., 1998
Bahia de Los Angeles, Baja California, Mexico		Not detected (5)		Cahill et al., 1998
Portland Harbor reach and vicinity, Oregon, USA			<**0.5** (15)	Buck and Kaiser, 2011
Northern Quebec (La Grande Reservoir), Canada (built-up environments)	58.1 ± 51.3 (31)[b]	37.4 ± 20.1 (78)[c]	**0.22 ± 0.10** (18)	DesGranges et al., 1998
LaGrange Reservoir (natural environments)	16.5 ± 12.8 (29)[b]	6.96 ± 4.32 (63)	**0.18 ± 0.11** (33)	DesGranges et al., 1998
Finland and Lapland		3.4 to 24.7 (137)	0.73 ± 0.47 (16)[d]	Häkkinen and Häsänen, 1980
Atlantic coast, New Jersey		3.35 ± 0.94 (7)		
Delaware Bay, New Jersey		2.14 ± 0.53 (14)	0.42 ± 0.21 (5)	Hughes et al., 1997
St. Mary's River, Ontario/Michigan	28.8 ± 16.2 (2)	7.40 ± 1.36 (12)	0.64 ± 0.21 (8)	Hughes et al., 1997
Georgian Bay, Ontario	21.1 ± 15.8 (5)	4.56 ± 1.60 (13)	0.78 ± 0.55 (17)	Hughes et al., 1997
Kawartha Lakes, Ontario	6.70 ± 1.00 (4)	3.26 ± 1.29 (19)	0.44 ± 0.26 (22)	Hughes et al., 1997
Ogoki Reservoir, Ontario		10.98 ± 3.98 (5)	1.40 ± 0.46 (8)	Hughes et al., 1997
Florida Bay, Florida	16.4 ± 1.51 (17)	13.7 ± 5.76 (15)		Lounsbury-Billie et al. (2008)
Lake Åsnen, Sweden		6.7 ± 0.18 (83)[e]		Odsjö et al., 2004
British Columbia and Alberta	6.79 ± 5.43 (12)	6.23 ± 5.64 (51)[f]	0.39 ± 0.17 (93)	Present study

[a] Remiges, rectrices, other large feathers.
[b] Various feather types, collected beneath the nest.
[c] Fourth and fifth primary and secondary flight feathers and two covert feathers from each wing, two rectrices, and eight body feathers.
[d] Addled eggs.
[e] Tail feathers.
[f] Arithmetic average across basins presented in Table 1.

Acknowledgments

M. Conner, A. Elliott, H. Gill, G. Kardosi, M. Machmer, C. Steeger, S. Weech, L. Wilson and, most of all, S. Lee-Ortiz assisted with trapping and handling of ospreys in British Columbia and P. Amann, J. Keating, M. Miller and P. Vidalin assisted with field work in Alberta. Staff of BC Hydro, West Kootenay Power and Trans-Alta Utilities—D. Williams (Merritt), K. Brown, D. Hamp, B. Morris, V. Bidnyk (Golden), G. Peterson, V. Gerlib, K. Curly (Nakusp), R. Royer, J. Pickard, E. Marback, P. Quaedvlieg, R. Sherbinin (Oliver)—assisted in accessing nests, as did J. Oseychuk, A. Palmer, T. Tal, R. Hobbis, M. Martin, and S. Oleman. R. McNeil and L. Hemsen coordinated preparation, storage and shipping of samples in Ottawa. L. Wilson and C. Morrissey assisted with organization and management of the data. S. Lee-Ortiz and R. Mckibbin created the map. Thanks to C. Alpers, B. Branfireun and B. Braune for excellent comments on a draft of this manuscript. L. Hrenchuk allowed us access to a scale to weigh out samples for encapsulation. Funding was provided by Environment Canada, in part, through its Toxic Substances Research Initiative. Samples were obtained in accordance with permits provided by Alberta Sustainable Resource Development, BC Ministry of the Environment, Parks Canada and Environment Canada following protocols approved by Animal Care Committees acting on behalf of the Canadian Council of Animal Care.

Appendix A. Supplementary data

Supplementary data to this article can be found online at doi:10.1016/j.envint.2011.11.004.

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