Science of the Total Environment 712 (2020) 135889

Contents lists available at ScienceDirect

Science of the Total Environment

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

Long-range transport of legacy organic pollutants affects alpine fish eaten by ospreys in western Canada

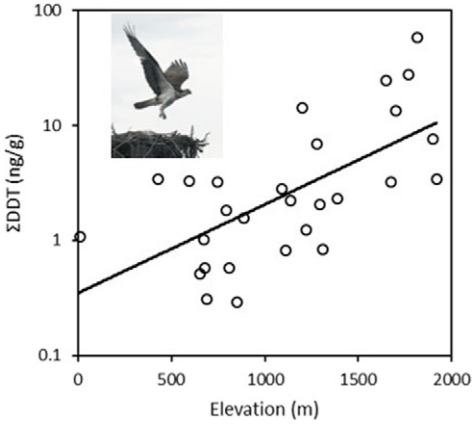
Philippe Grenier [a], John E. Elliott [b], Ken G. Drouillard [c], Mélanie F. Guigueno [d], Derek Muir [e], D. Patrick Shaw [f], Mark Wayland [g], Kyle H. Elliott [a,*]

[a] Department of Natural Resource Sciences, McGill University, Ste Anne-de-Bellevue, Canada
[b] Environment and Climate Change Canada, Delta, Canada
[c] Institute for Great Lakes Research, University of Windsor, Canada
[d] Department of Biology, McGill University, Montréal, Canada
[e] Environment and Climate Change Canada, Burlington, Canada
[f] Environment and Climate Change Canada, Vancouver, Canada
[g] Environment and Climate Change Canada, Saskatoon, Canada

HIGHLIGHTS

- Chemicals transported long distances condense in cold regions (i.e. alpine glaciers).
- We report levels of legacy POPs in osprey and fish from alpine Canada.
- Levels of DDT increased with elevation and were high in alpine lakes.
- Toxaphene congeners mostly implied long range transport.
- Food chain length (using baseline-corrected values) was also important.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 20 August 2019
Received in revised form 22 October 2019
Accepted 30 November 2019
Available online 6 December 2019

Editor: Yolanda Picó

Keywords:
POPs
Amino acid specific stable isotope analyses
Biomagnification

ABSTRACT

Persistent organic pollutants (POPs) contaminate pristine, alpine environments through long-range transport in the atmosphere and glacier trapping. To study variation in POPs levels in western Canada, we measured levels in the prey (fish) of osprey (*Pandion haliaetus*) during 1999–2004, and compared those to levels in eggs and chicks. Values in fish muscle (representing human consumption) correlated with whole carcasses (wildlife consumption) for all POPs, except toxaphene, allowing us to pool data. Biomagnification factors for osprey eggs were much higher than published values from Oregon, reflecting differences in local diet. We factored baseline-corrected food chain variation by using amino acid-specific analysis of osprey eggs, illustrating how top predators (ospreys) can indicate both ecosystem-wide baselines and contamination. Given that our biomagnification factors were so different from those for the same species from a nearby site, we argue that trophic magnification factors derived from baseline-corrected $\delta^{15}N$ are likely a more accurate method for estimating contamination. *Dichlorodiphenyltrichloroethane* (ΣDDT) concentrations were greatest in rainbow trout from a small lake at

* Corresponding author.
E-mail address: kyle.elliott@mcgill.ca (K.H. Elliott).

https://doi.org/10.1016/j.scitotenv.2019.135889

Osprey
Western Canada

1800 m, and those levels exceeded wildlife and human health guidelines. Indeed, once sites with known agricultural inputs were eliminated, elevation, percent lipids and baseline-corrected $\delta^{15}N$ (from amino acid specific isotope values) best predicted ΣDDT. Baseline-corrected, but not bulk, $\delta^{15}N$ was the main predictor of polychlorinated biphenyls (ΣPCB). Total toxaphene was consistently the major contaminant after ΣPCB and ΣDDT in osprey eggs, and was present in many fish samples. We concluded that toxaphene arrived from long range deposition due to high proportions of Parlar 40–50 congeners. The only exception was Paul Lake, where toxaphene was used as a piscicide, with a high concentrations of the Hex-Sed and Hep-Sed congeners at that site. We conclude that long-range transport and trophic position, not melting glaciers, were important determinants of some legacy POPs in fish and wildlife in alpine Canada.

1. Introduction

Persistent organic pollutants (POPs) can be transported long distances on air currents (Fernandez and Grimalt, 2003; Von Waldow et al., 2010; Steinlin et al., 2016). Combined with a resistance to environmental degradation, multiple volatilization-transport-deposition cycles can lead to concentration of POPs in cold regions (Blais et al., 2001a, 2001b; Semeena and Lammel, 2005; Luo et al., 2016). Thus, elevated levels of POPs can occur in remote Arctic and alpine habitats partially due to scavenging by snow crystals, possibly via adsorption or entrapment (Wania and Mackay, 1993; Blais et al., 1998; Franz and Eisenreich, 1998). Indeed, atmospheric transport of volatile contaminants has been recognized for some time (Atlas and Giam, 1981; Wania and Mackay, 1993; Jaffe et al., 2003; Elliott et al., 2012). Many of the transported POPs bioconcentrate and some can biomagnify in aquatic systems through trophic transfer (Kidd et al., 1995; Henny et al., 2003). Glacial runoff can interact with cold condensation further contaminating surrounding watersheds due to the minimal volatilization loss from low temperature runoff waters, limited catchment retention owing to low soil organic matter within such catchments and the rapid channeling of the runoff from the glacier surface (Blais et al., 2001a, 2001b; Milner et al., 2017). Alpine areas in western North America are one region where contaminants, such as POPs and mercury, from Asia or industrial regions of North America can be transported atmospherically and deposited (Blais et al., 2001a, 2001b; Jaffe et al., 2003), potentially contaminating wildlife and people that eat fish in alpine lakes (Elliott et al., 2012; Guigueno et al., 2012). The 1960–1980 global cold cycle trapped contaminants in glaciers, but these trapped contaminants would have largely melted out by 1980 (Elliott et al., 2012; Guigueno et al., 2012; Steinlin et al., 2016). Nonetheless, local processes coupled with ongoing POPs deposition in cold environments mean that significant levels of POPs remain in alpine environments worldwide (Elliott et al., 2012; Luo et al., 2016; Steinlin et al., 2016; Milner et al., 2017).

Studies on organochlorine residues in aquatic systems throughout Canada have shown elevated levels in fish tissues even in locations far removed from contaminant sources (Muir et al., 1990; Kidd et al., 1995; Donald et al., 1993, 1999). A broad geographic survey of contaminants in burbot liver (Muir et al., 1990) showed elevated levels of a wide range of organochlorine pesticides, generally increasing from south to north. Kidd et al. (1995) measured levels of the insecticide, toxaphene, in Yukon lakes at levels of health concern to human consumers. Likewise, high levels of chlorinated hydrocarbons, particularly PCBs and toxaphene, have been measured in lake trout from remote lakes in the Rocky Mountains with high glacial melt (Donald et al., 1999) and in burbot from large lakes in the Fraser River basin (Shaw and Gray, 2004). Point sources can also be important. Toxaphene was used as a piscicide in some lakes in western Canada prior to stocking (Stringer and McMynn, 1960), and remnant levels of toxaphene occur in those lakes (Miskimmin et al., 1995; Donald et al., 1999). Similarly, DDT was sprayed heavily in some of the intermontane valleys.

Whereas fish communities in lakes can be complex, apex piscivorous predators consolidate the information from the food chain to provide an overall index of contamination for lakes and other aquatic environments (Furness, 1993; Jaspers et al., 2011; Løseth et al., 2019). Apex predators are proven indicators of POPs contamination in aquatic ecosystems because they integrate signals across space and time, and are also, themselves, sensitive to impacts of such contaminants (Sergio et al., 2008; Elliott and Elliott, 2013). Thus using wildlife as a model for environmental pollution can provide data on both exposure and effect (Zhou et al., 2008; Cesh et al., 2010; Roos et al., 2012), a finding that dates to the earliest days of ecotoxicology (Risebrough et al., 1967; Spitzer et al., 1978), and a foundation that we build on here. To be useful in assessing risks to humans, species must occupy a high trophic status and bioaccumulate pollutants, making apex predators well suited for the role (Basu et al., 2007; Henny et al., 2010; Elliott and Elliott, 2013). For example, ospreys are excellent indicators because they: (1) have a widespread distribution allowing for direct comparisons from different populations across the globe; (2) are at the top of the aquatic avian food web and are obligate piscivores; (3) bioaccumulate hydrophobic contaminants such POPs; (4) have a restricted home range, typically foraging within 1–3 km of their nests; (5) have a well-known biology as they are common and easily studied in temperate regions, and their decline in the 1960s prompted many studies; (6) have well understood sensitivity to contaminants; and (7) have a booming population post-DDT era (Grove et al., 2009; Henny et al., 2010). Ospreys are also long-lived, adapt to human landscapes, tolerate short-term disturbances with little impact on nest success and are regularly spatially distributed, which make them a relatively easy species to study (Henny et al., 2010). Ospreys have been used as sentinels in western Canada to measure mercury and POPs contamination in alpine environments with variable precipitation, deposition patterns and glacial runoff (Elliott et al., 2012; Guigueno et al., 2012). Although 'legacy' POPs levels are declining globally, levels are often still higher than many 'emerging' contaminants and sometimes above thresholds for sublethal toxicity (Henny et al., 2009; Cesh et al., 2010; Roos et al., 2012; Ortiz-Santaliestra et al., 2015; Elliott et al., 2015; Brogan et al., 2017).

Diet can confound the interpretation of trends in top predators. Stable isotope ratios are biogeochemical tracers that are incorporated from diet into predator tissues, and therefore offer the ability to track chemical pollution through the food chain (Kidd et al., 1995; Elliott et al., 2012; Dolgova et al., 2018). The ratio of $^{13}C/^{12}C$ can be used to infer food source (e.g. benthic vs. pelagic) (Braune et al., 2002; Hobson, 1995). The ratio of $^{15}N/^{14}N$ increases systematically from prey to consumer, allowing estimation of the 'trophic position' of a consumer (Braune et al., 2002; Hobson, 1995; Kidd et al., 1995). Nonetheless, variation in nitrogen stable isotope ratios due to spatial variation in baseline levels may swamp trophic signals, and inferred variation due to 'biomagnification' may actually represent spatial variation in nitrogen inputs to the system (Pajuelo et al., 2010; Seminoff et al., 2012). Amino acid specific analyses can provide a more robust measure of trophic level by accounting for variation in background nitrogen stable isotope ratios (Lorrain et al., 2009; Pajuelo et al., 2010; Seminoff et al., 2012). The $\delta^{15}N$ values in 'source' amino acids such as phenylalanine ($\delta^{15}N_{phe}$) do not increase with trophic level and can therefore be used as a measure of baseline $\delta^{15}N$ (Lorrain et al., 2009; Seminoff et al.,

2012). In contrast, 'trophic' amino acids such as glutamate ($\delta^{15}N_{glu}$) increase systematically with trophic level (Lorrain et al., 2009; Seminoff et al., 2012). Thus, the difference between these two values, $\delta^{15}N_{phe}$-$\delta^{15}N_{glu}$, provides a baseline corrected estimate of trophic level (Lorrain et al., 2009; Seminoff et al., 2012; Elliott and Elliott, 2016; Dolgova et al., 2018). Once trophic level is accounted for, variation in POPs levels due to geography can be directly assessed.

We studied levels of legacy persistent organic pollutants (POPs) in osprey and their main prey, fish, to understand the processes and patterns of POP variation in alpine regions of western Canada. We were particularly interested in geographical and food web correlates of levels in fish, and the biomagnification from fish to ospreys, as geographical variation in osprey levels has already been reported (Morrissey et al., 2004; Guigueno et al., 2012; Elliott et al., 2012). In ospreys, percent lipids (primarily in eggs), food web factors (eggs: bulk $\delta^{13}C$; nestlings: bulk $\delta^{13}C + \delta^{15}N$) and geographical variables explained much of the variation in HCB, ΣDDT, ΣPCB, toxaphene and chlordane (Elliott et al., 2012). Typically, levels increased with elevation and proportion of glaciers, increased with watershed size and decreased with lake size (due to dilution), although typically only one or two variables were significant for any particular POP (Elliott et al., 2012). Furthermore, contaminant levels of osprey tracked from the Pacific Northwest were not associated with levels in fish at wintering sites in Latin America, implying that contaminants largely originate on the breeding grounds (Elliott et al., 2000; Elliott et al., 2007). In this study, we focus on ΣDDT, ΣPCB and toxaphene because they are the dominant legacy POPs in western Canada. Also, there were a large number of measurements of other compounds that were below the detection limit in fish compared with osprey. Because we had a mixture of estimates from muscle (representing human consumption) and whole carcasses (representing wildlife consumption), we first established relationships between those two. Next, we developed models for predicting POPs levels based on geographical variables (lake size, watershed size, percent glaciation, elevation) and diet, as inferred from nitrogen and carbon stable isotope ratios. Finally, we calculated biomagnification factors between fish and ospreys to understand how POPs in fish make their way into osprey.

2. Methods

2.1. Sampling and geographical variables

Between 1999 and 2004, we collected samples of osprey and fish populations in the western Canada (Table 1), targeting 15 different watersheds. Samples from 88 eggs, each from different nests, as well as plasma and feathers samples from 70 chicks, and 11 plasma samples from adults were taken. In addition, we caught 211 fish from sites in proximity to osprey nests or to include sites with expected high concentrations to determine contaminant range (e.g. Paul Lake, known site of toxaphene use as a piscicide). Fish were collected using angling, gill netting and electroshocking. Specific species and quantities were targeted when diet information was known. When unavailable, sports fish such as trout and char were favoured, as these potential osprey prey are also regularly consumed by humans. All contaminants data is uploaded as a Data Appendix.

Geographic variables were initially gathered using the Spatial Analyst toolbox and Hydrology toolset in ARCGIS 9.2. We determined the size (area) of 24 watershed drainage basins and of local osprey foraging lakes and rivers within the Canadian Rockies (ESRI, www.esri.com). Elevation (m) of osprey nest location and watershed area (km^2) were determined using the digital elevation model and glacier data archived at NRC Geomatics Canada (NRC, 2001a; NRC, 2001b) and Alberta Environment and Sustainable Resource Development (2015). Glacial areas (km^2) were calculated using glacier extents in 2001. All data were projected onto the same coordinate system UTM Zone 11N, NAD83. Postprocessing of data with the Hydrology toolset required sink-filling using the sink tool 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. Once watersheds were developed, areas of both watersheds and glaciers were calculated using the script within ARCGIS 9.2. (Elliott et al., 2012). We also recorded the proportion of agricultural land cover in each watershed ("agricultural input").

Fish from British Columbia were analyzed using both dorsal muscle samples (to assess human health implications) and combined composite samples (whole fish minus the dorsal muscle, to assess wildlife health implications), henceforth referred to as "carcass". The concentrations of major contaminants found in muscle tissue were compared to the concentrations found in carcass samples on both a wet-weight and a lipid-weight basis. Geometric means of concentrations found in fish of the same species for each sampled lake in British Columbia were calculated. Concentrations were adjusted for lipid weight by applying the ratio method described in Hebert and Keenleyside (1995). Comparing wet weight to lipid weight values as well as muscle and carcass concentrations enables determination if one measure could be used to approximate the other. For example, from equilibrium lipid partitioning principles, we expect wet weight concentrations to differ between paired samples of carcass and muscle, whereas lipid adjusted concentrations should be similar. Lakes in Revelstoke National Park were resampled in 2004 to compare with data from 1999.

To observe prey being delivered by osprey to their nestlings, we recorded prey at five nests at each of five lakes: Lillooet, Nakusp, Nicola, Oliver and Pitt Meadows (Morrissey et al., 2004). Study lakes were selected to represent a diversity of habitat characteristics in terms of elevation, hydrology, lake size and depth, in addition to biodiversity of fish species. All prey observations were conducted by a single observer assigned to each site to monitor five nests for the duration of the study period. Over a 12 week period, from May to early August 2001, observations were conducted at each nest to encompass the major phases of Osprey breeding (incubation, chick rearing and fledging). The study was designed to render a total of 1500 observation hours over the study period (60 h at each of the 25 nests). A schedule was devised to ensure that each nest would be observed for equal amounts of .time and at equal times of the day. If a nest failed during the observation period, another active nest in the same breeding stage was used as a replacement for the remainder of the study to ensure completeness of the dataset.

2.2. Lab analysis

For samples collected in B.C., fish were wrapped in solvent-cleaned aluminum foil and frozen on-site using dry ice. In the laboratory, fish were thawed, weighed and measured. Ageing structures (otoliths, fin rays or scales) were removed and forwarded to the Fish Ageing Laboratory at the Pacific Biological Station, Nanaimo for determination. Fish were sexed by internal examination. Skin-off fillets of dorsal muscle from 3 to 6 individuals of each species from each waterbody were dissected, wrapped in solvent-cleaned foil (pesticide-grade acetone/hexane) and re-frozen for individual analysis. The remaining carcasses (less dissected dorsal muscle) were combined into a single composite sample.

For DDT and other OC pesticides, and PCB congeners, samples were subsequently shipped to the National Wildlife Research Centre, Hull, Quebec for homogenization prior to chemical analysis. Homogenized samples were subsequently shipped to the analytical laboratory of the Great Lakes Institute for Environmental Research (GLIER) at the University of Windsor, which has been accredited by the Canadian Association for Environmental Analytical Laboratories (CAEAL), and followed procedures described in Guertin et al. (2010) and Elliott et al. (2015). Analysis of fish samples included determination of chlorobenzenes (ΣCBz = 1,2,4,5-tetrachlorobenzene, 1,2,3,4-tetrachlorobenzene, pentachlorobenzene, and exachlorobenzene), hexachlorocyclohexanes (ΣHCH = α-, β-, and γ-hexachlorocyclohexane), chlordane-related compounds (ΣCHLOR = oxychlordane, trans-chlordane, cis-chlordane, trans-nonachlor, cisnonachlor),dichlorodiphenyltrichloroethane (DDT) and

Table 1
Locations, species and biological data (means and range) for fish collected from waterbodies in western Canada.

Lake	Lat	Long	Species	*Formal name*	N	Length (mm)		Mass (g)		Age(y)	
Bridge river drainage											
	50° 51′	122° 30′	Dolly varden	*Salvelinus confluentus*	4	386	359–430	571	402–804	8	5–10
	50° 51′	122° 30′	Largescale sucker	*Catastomus macrocheilus*	5	289	280–297	279	244–318	6	6–7
Carpenter Lake	50° 51′	122° 30′	Mountain whitefish	*Prosopium williamsoni*	5	307	291–322	404	314–443	7	5–9
Downton Lake	50° 50′	122° 59′	Rainbow trout	*Onchorhynchus mykiss*	5	302	252–325	303	216–392	5	4–5
Gun Lake	50° 52′	122° 53′	Rainbow trout	*Onchorhynchus mykiss*	2	257	215–298	234	141–327	na	na
Columbia river drainage											
Columbia River nr Parson	51° 03′	116° 37′	Largescale sucker	*Catastomus macrocheilus*	5	333	277–389	392	239–644	na	na
	51° 03′	116° 37′	Pike minnow	*Ptychocheilus oregonensis*	5	292	271–311	264	237–321	na	na
Kinbasket; Bush Arm	51° 42′	117° 33′	Largescale sucker	*Catastomus macrocheilus*	5	308	263–370	348	216–549	na	na
	51° 42′	117° 33′	Pike minnow	*Ptychocheilus oregonensis*	5	289	223–335	326	162–453	na	na
Kinbasket Lake; Wood Arm	52° 07′	118° 24′	Bull trout	*Salvelinus confluentus*	5	466	349–534	1386	338–2164	8	5–12
	52° 07′	118° 24′	Largescale sucker	*Catastomus macrocheilus*	5	356	274–394	655	574–757	22	12–39
Revelstoke Lake, South	51° 09′	118° 12′	Bull trout	*Salvelinus malma*	4	348	303–396	401	278–544	6	3–7
	51° 09′	118° 12′	Whitefish	*Prosopium williamsoni*	5	304	295–315	367	294–456	6	4–9
Upper Arrow nr Nakusp	50° 12′	117° 47′	Bull trout	*Salvelinus malma*	5	402	332–438	684	358–908	6	5–9
	50° 27′	117° 53′	Kokanee	*Onchorhynchus nerka*	5	200	195–205	94	89–99	na	na
	50° 27′	117° 53′	Largescale sucker	*Catastomus macrocheilus*	5	395	383–409	785	683–927	na	na
	50° 12′	117° 47′	Rainbow trout	*Onchorhynchus mykiss*	5	330	287–359	394	271–524	4	2–6
	50° 27′	117° 53′	Mountain whitefish	*Prosopium williamsoni*	5	249	240–255	172	153–196	na	na
Nechako river drainage											
Ootsa Lake	53° 46′	126° 08′	Rainbow trout	*Onchorhynchus mykiss*	5	321	309–336	336	294–371	na	na
Okanagan river drainage											
Osoyoos Lake	49° 04′	119° 31′	Largescale sucker	*Catastomus macrocheilus*	5	432	406–460	1107	919–1492	na	na
	49° 04′	119° 31′	Lake whitefish	*Coregonus clupeaformis*	5	401	373–432	713	628–838	na	na
Mt. Revelstoke Park											
Eva Lake (1999)	51° 05′	118° 06′	Brook charr	*Salvelinus fontinalis*	2	175	171–188	62	51–77	6	3–10
Eva Lake (2004)	51° 05′	118° 06′	Brook charr	*Salvelinus fontinalis*	2	194	180–208	65	56–74	na	na
Lower Jade Lake	51° 04′	118° 03′	Rainbow trout	*Onchorhynchus mykiss*	14	209	167–270	94	51–171	na	na
Miller Lake	51° 04′	118° 06′	Brook charr	*Salvelinus fontinalis*	2	181	171–215	66	44–119	6	5–7
Upper Jade Lake (1999)	51° 04′	118° 04′	Rainbow trout	*Onchorhynchus mykiss*	2	236	206–280	158	103–225	8	6–10
Upper Jade Lake (2004)	51° 04′	118° 04′	Rainbow trout	*Onchorhynchus mykiss*	7	244	203–271	153	98–224	na	na
SW British Columbia											
Nicola Lake	50° 10′	120° 32′	Dolly varden	*Salvelinus confluentus*	3	522	510–538	1369	1344-1419	na	na
	50° 10′	120° 32′	Largescale sucker	*Catastomus macrocheilus*	5	329	265–395	455	215–855	8	5–10
	50° 11′	120° 32′	Peamouth chub	*Mylocheilus caurinus*	5	208	202–215	88	80–100	na	na
	50° 10′	120° 32′	Pike minnow	*Ptychocheilus oregonensis*	5	302	285–320	304	267–354	10	8–13
Paul Lake	50° 44′	120° 07′	Rainbow trout	*Onchorhynchus mykiss*	5	370	310–395	583	369–705	6	5–8
Pitt Lake (Main Lake)	49° 24′	122° 32′	Cutthroat trout	*Onchorhynhus clarki*	5	324	262–380	376	171–594	3	2–3
	49° 24′	122° 32′	Whitefish	*Prosopium williamsoni*	5	226	206–234	117	90–127	3	2–3
Pitt Lake (Katzie Slough)	49° 21′	122° 35′	Brown bullhead	*Ameiurus nebulosus*	5	237	229–250	172	159–194	na	na
N. BC/Yukon											
Atlin Lake	59° 34′	133° 43′	Lake trout	*Salvelinus namaycush*	3	394	380–415	636	201–1114	na	na
Williston Lk (Manson Arm)	55° 54′	123° 57′	Longnose sucker	*Catastomus catastomus*	5	354	339–370	517	456–572	na	na
	55° 54′	123° 57′	Pike minnow	*Ptychocheilus oregonensis*	5	277	246–331	237	155–385	na	na
	55° 54′	123° 57′	Mountain whitefish	*Prosopium williamsoni*	3	246	243–250	163	158–174	na	na
Alberta											
Athabasca River	52°52′	118°03′	Longnose sucker	*Catastomus catastomus*	5	418	398–455	932	760–1200	na	na
	52°52′	118°03′	Mountain whitefish	*Prosopium williamsoni*	5	273	254–285	271	210–300	na	na
Belly River	49°07′	113°42′	Longnose sucker	*Catastomus catastomus*	5	383	358–405	741	600–860	na	na
	51°00′	115°04′	Longnose sucker	*Catastomus catastomus*	5	437	424–470	1148	970–1400	na	na
Barrier Lake	51°00′	115°04′	Brown trout	*Salmo trutta*	2	436	434–437	649	600–697	na	na
	51°00′	115°04′	Mountain whitefish	*Prosopium williamsoni*	2	294	293–294	244	217–270	na	na
Bow Lake	51°39′	116°25′	Lake trout	*Salvelinus namaycush*	7	327	289–367	369	270–550	na	na
	51°39′	116°25′	Mountain whitefish	*Prosopium williamsoni*	1	285		290		na	na
Bow River (upstream)	51°15′	115°55′	Mountain whitefish	*Prosopium williamsoni*	3	218	198–229	97	70–115	na	na
	51°15′	115°55′	Longnose sucker	*Catastomus catastomus*	1	390		730		na	na
Bow River (downstream)	51°03′	115°10′	Mountain whitefish	*Prosopium williamsoni*	1	700		425		na	na
Cabin Lake	52°52′	118°07′	Rainbow trout	*Onchorhynchus mykiss*	8	267	192–314	198	70–285	na	na
Castle River	49° 33′	114° 02′	Longnose sucker	*Catastomus catastomus*	5	376	348–415	621	520–810	na	na
Chain Lakes	50°16′	114°13′	Longnose sucker	*Catastomus catastomus*	5	346	323–370	439	340–600	na	na
	50°16′	114°13′	Mountain whitefish	*Prosopium williamsoni*	1	285		255		na	na
	50°16′	114°13′	Rainbow trout	*Onchorhynchus mykiss*	1	280		115		na	na
Crowsnest River	49°33′	114°19′	Mountain whitefish	*Prosopium williamsoni*	4	359	318–400	570	450–750	na	na
	49°33′	114°19′	Rainbow trout	*Onchorhynchus mykiss*	2	382	377–387	565	540–590	na	na
Gap Lake	51°03′	115°14′	Longnose sucker	*Catastomus catastomus*	5	913	78–959	393	369–419	na	na
Goat Pond	51°01′	115°24′	Longnose sucker	*Catastomus catastomus*	5	392	373–407	722	590–855	na	na
	51°01′	115°24′	Cutthroat trout	*Catastomus catastomus*	1	277		220		na	na
Lower Kananaskis Lake	50°41′	115°10′	Longnose sucker	*Catastomus catastomus*	5	428	360–472	1026	620–1150	na	na
Miette River	52°52′	118°16′	Mountain whitefish	*Prosopium williamsoni*	5	217	190–255	119	60–185	na	na
Moose Lake	52°57′	118°55′	Longnose sucker	*Catastomus catastomus*	5	428	405–445	1093	935–1300	na	na

Table 1 (*continued*)

Lake	Lat	Long	Species	*Formal name*	N	Length (mm)		Mass (g)		Age(y)	
	52°57″	118°55″	Lake trout	*Salvelinus namaycush*	4	321	295–355	327	255–440	na	na
Pyramid Lake	52°55″	118°06″	Lake trout	*Salvelinus namaycush*	5	440	374–551	1012	590–1750	na	na

its metabolites [ΣDDT = p,p′-dichlorodiphenyltrichloroethane (p,p′-DDT),p,p′-dichlorodiphenyldichloroethylene (p,p′-DDE), and p,p′-dichlorodiphenyldichloroethane (p,p′-DDD)], mirex, PCBs (ΣPCBs = 39 congeners identified according to IUPAC numbers: 17/18, 28/31, 33, 44, 49, 52, 70, 74, 82, 87, 95, 99, 101, 105/132, 110, 118, 128, 138, 149, 153, 156/171, 158, 170, 177, 180, 183, 187, 191, 194, 195, 201, 205, 206, 208, and 209. Co-eluting congeners are reported together. Briefly, 2–4 g of liver homogenate was ground, mixed with anhydrous sodium sulfate (VWR) and added to a glass chromatography column containing hexane/dichloromethane (50% V/V, VWR, St. Catharines, Ontario, Canada). Each column was spiked with 200 ng of 1,3,5-tribromobenzene (TBB; AccuStandard, New Haven, Connecticut, USA) for use as a surrogate recovery standard. The column was eluted and further extracted with hexane/dichloromethane (50% V/V) and the eluant roto-evaporated and made up to a volume of 10 mL with hexane. A portion of the extracts (2 mL) were removed for lipid determination by gravimetric methods. Clean-up of the remaining extracts was performed by chromatography on Florisil. Fractions 1 and 2 were analyzed for OCPs and PCBs by gas chromatography–electron capture (GC–ECD). An Agilent gas chromatograph with a ^{63}Ni-electron capture detector, 7673 autosampler and DB-5 column (60 m × 0.250 mm × 0.1 μm DB-5; Chromatographic Specialties, Brockville, Ontario, Canada) was used for analysis. Samples were extracted and analyzed in batches of five or six. With each batch, a method blank (containing 5 g sodium sulfate) and an in-house reference sample [Detroit River carp (*Cyprinus carpio*) homogenate] were co-extracted and analyzed. For OCPs and PCBs, analytes were quantified using a single point external standard (Quebec Ministry of Environment PCB mixture and a custom ordered certified OCP mixture; AccuStandard; Chromatographic Specialties, Brockville,Ontario, Canada) injected at the start of each sample batch analysis with monthly audits of full standard curves being completed for each instrument. Quality assurance and control procedures were outlined in Guertin et al. (2010). Detection limits for individual analytes were approximately 0.02–0.05 ng/g wet weight.

Toxaphene measurements were conducted at the Canada Centre for Inland Waters, Burlington, Ontario from the fraction two extracts received from GLIER. Gas chromatography-mass spectrometry was carried out using an Agilent mass selective detector (HP5973 MSD) operated in negative ion mode. Ion source, quadruple and transfer liner were held at 150 °C, 100 °C and 250 °C, respectively. Methane of ultra high purity was used as ionization gas at a pressure of 1.9 × 10–4 Torr in the ion source. Separation was performed with a HP5 column (30 m × 0.25 mm id;film thickness, 0.25 μm) using a temperature program optimized earlier by Glassmeyer et al. (1999) and splitless injection. The following ions, corresponding to fragment ions (M-Cl)-, (M-Cl + 2)- and (M-Cl + 4)- for the hexa- (*m/z* 307, 309 and 311), hepta (*m/z* 341, 343 and 345), octa- (*m/z* 375, 377 and 379) and nona-CHBs (*m/z* 411 and *m/z* 413) were monitored. ^{13}C-mirex was added to each sample extract as an internal performance standard. Quantitative analysis of total toxaphene and homologs was carried out using an external standard of "Hercules" technical toxaphene (Ultra Scientific). In addition, 25 individual hexa- to nonachlorobornane congeners were determined using standards from Dr. Ehrenstorfer (Augsburg Germany) or Promochem (Wesel, Germany). Method detection limits (MDLs) for individual chlorobornane congeners were approximately 0.01 ng/g (wet wt) and 0.1 ng/g dw for total toxaphene and homologue groups. Fish from Alberta were analyzed for toxaphene at the GLIER using a GC-electron capture negative ion MS using a HP 5973MSD. Total toxaphene and homologs are determined by the method of Glassmeyer et al. (1999) with minor modifications.

Stable isotope analysis for δ^{15}N and δ^{13}C were conducted by the University of California Davis stable isotope facility. Carbon and nitrogen were simultaneously analyzed in 1 mg freeze-dried samples and analyzed using a continuous-flow isotope ratio mass spectrometer. Details are reported elsewhere (Guigueno et al., 2012; Elliott et al., 2012). Amino acid-specific analysis following the methods described by Yarnes et al. (2011). Amino acids were liberated via acid hydrolysis and derived by methyl chlorformate. Methoxycarbonyl amino acid methyl esters were then injected in splitless (^{15}N) mode and separated on an Agilent DB-23 column (30 m × 0.25 mm ID, 0.25 μm film thickness). Once separated, the esters were converted to N_2 in a combustion reactor at 1000 °C. Water was subsequently removed through a nafion dryer. During the final step of the analysis, N_2 entered the IRMS. Pure reference N_2 was used to calculate provisional δ- values of each sample peak. Next, isotopic values were adjusted to an internal standard (e.g. norleucine) of known isotopic composition. Final δ-values were obtained after adjusting the provisional values for changes in linearity and instrumental drift such that correct δ-values for laboratory standards were obtained. Laboratory standards were custom mixtures of commercially available amino acids that had been calibrated against IAEA-N1, IAEA-N2, IAEA-N3, USGS-40, and USGS-41. Baseline-corrected δ^{15}N was calculated as $\delta^{15}N_{bulk} - \delta^{15}N_{source}$, where $\delta^{15}N_{source}$ was the average of δ^{15}N for the source amino acids lysine, methionine and phenylalanine (Dolgova et al., 2018). Bulk δ^{13}C values were corrected for lipid content using the equation of Logan et al. (2008).

2.3. Statistical analysis

To compare muscle and carcass concentrations we used simple linear regression (Løseth et al., 2019). Both wet-weight and lipid-weight biomagnification factors (BMF) were calculated (following Henny et al., 2003) by taking the ratio of the contaminant concentrations found in Osprey eggs relative to locally collected fish as well as in osprey chicks compated to fish. Data on osprey eggs and chicks appeared in Elliott et al. (2012). To determine the prey basket relevant for ospreys, visual observations of osprey prey capture from five of the watersheds were used (Morrissey et al., 2004). To aid identification of fish from visual observations, remains in nests were measured for length, family and species, when possible. Like Henny et al. (2003) all prey for this study were categorized into major families (>10% of all observations), being here catostomid, salmonid and cyprinid. The ichtalurid family was disregarded as there were no fish from this family recorded in a watershed where egg or chick data were available. Given that ospreys are opportunistic foragers, fish occurrence in the diet was weighed equally for each recorded observation. A mean weight for each of the families was used to convert "percent occurrence in the diet" into "percent biomass". A geometric mean of wet weight concentrations for all available individuals in fish and osprey eggs and chicks was taken and then a weighted average diet OCP or PBDE concentration was used to calculate BMFs using the ratio method. The BMF for each of seven watersheds with both fish and osprey data was then averaged to come up with the actual BMF. Only carcass and wet weight concentrations were used for the fish values, as to match Henny's methods. Apart from BMF calculations, we included percent lipid as a covariate rather than using lipid weight measurements (Hebert and Keenleyside, 1995).

A separate analysis was conducted to explain variation in fish wet weight contaminants levels. Due to the number of potential explanatory variables (latitude, elevation, lake area, watershed area, glacier area, proportion of glacier area in the watershed, proportion of lake area in

Table 2
Linear model parameters for regression of muscle and carcass concentrations on a wet weight basis. The R^2 value for regressions on a lipid weight basis are also shown.

	% Lipids	ΣDDT	ΣChlordane	ΣPCB	HCB	Dieldrin	ΣToxaphene
Intercept (SE)	1.94 (0.48)	1.79 (1.50)	0.35 (0.23)	2.28 (0.85)	0.25 (0.07)	0.14 (0.06)	1.81 (0.59)
Slope (SE)	2.03 (0.22)	3.77 (0.07)	3.05 (0.20)	2.99 (0.13)	2.17 (0.25)	2.16 (0.30)	−0.07 (0.28)
R^2 (wet weight)	0.75	0.99	0.90	0.95	0.72	0.71	0.00
R^2 (lipid weight)		0.41	0.57	0.77	0.15	0.46	0.00
p-value	2.433e-10	2.2.e-16	8.553e-15	2.2e-16	2.06e-9	3.015e-9	0.81

the watershed, lipid %, $\delta^{15}N$ and $\delta^{13}C$ isotopes), exploratory statistics were used to highlight the most important variables that could explain contaminant concentration. Model selection and Pearson's correlation tests were done using R 3.5.1. Although stepwise selection has a tendency to overfit, stepwise selection was justifiable in this context because of the high number of variables considered. Competing linear models compared based on their variation explained (R^2) and AIC values. Models with the highest R2 and lowest AIC were selected as the best model. In principle, when the difference in AICc between two models exceeds a value of 2, the model with the lowest AICc is considered superior.

3. Results

3.1. Food web and geographic factors influencing POPs levels in fish

First, we established relationships between muscle and fish carcass so that we could report both levels relevant to humans (muscle) and wildlife (muscle + carcass). Using wet weight concentrations, muscle tissue values for major contaminants were significantly correlated using a linear model with carcass tissue concentrations for the same fish for ΣDDT, Σchlordane, PCB congeners, HCB and dieldrin. In all of the above, the linear model fits were highly significant ($p < .001$; Table 2). Carcass wet weight concentrations were always greater than muscle being between 2.2 and 3.8 fold higher. Muscle wet concentration was, however, not a good predictor of carcass concentrations for total toxaphene, possibly due to the large variability observed when toxaphene is analyzed with a technical standard rather than individual congeners (de Geus et al., 1999). The authors suggest that isomers should be treated individually during laboratory analysis to achieve results meaningful to environmental work. Using lipid adjusted concentrations to evaluate the relationship between muscle and carcass concentrations, muscle samples were generally less strongly linearly correlated to carcass samples (Table 2). Lipid-weight muscle concentrations were still significantly correlated to carcass concentrations for ΣDDT, Σchlordane, PCB congeners and dieldrin. Carcass values were

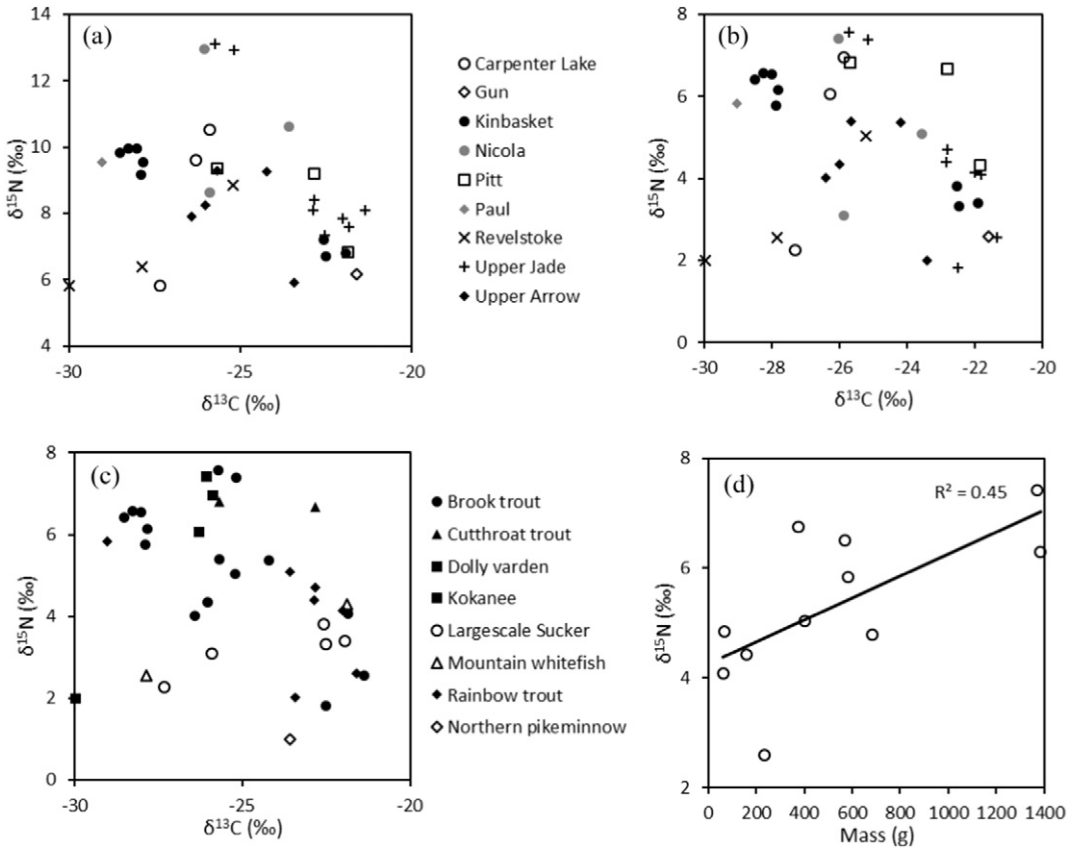


Fig. 1. Isotopic values ($\delta^{15}N$-$\delta^{13}C$) for freshwater fish sampled in alpine lakes in western Canada. (a) Uncorrected values organized by lake (black: lakes formed by dams on the Columbia River; grey: lakes in agricultural regions; open: lakes on the Pacific slope; cross: alpine lakes in Revelstoke Park); (b) Baseline-corrected values organized by lake; (c) Baseline-corrected values organized by species (black: salmonids; open: non-salmonids); and (d) Baseline-corrected (but not bulk) $\delta^{15}N$ correlated with salmonid weight.

always higher, ranging from ~2.0 times to ~56 times the muscle concentration. HCB was less strongly correlated and, again, no relationship was established from total toxaphene. Given the stronger relationships on a wet weight basis than a lipid weight basis, we used wet weight values to estimate carcass values in all subsequent analyses for samples where muscle but not carcass values were available.

Baseline-corrected $\delta^{15}N$ showed less variation among lakes than bulk $\delta^{15}N$, and, within a species group, the $\delta^{15}N$- $\delta^{13}C$ signal was quite consistent in each lake (Fig. 1a,b). In particular, lakes with extensive areas of agricultural land in the watersheds, such as Nicola Lake, had $\delta^{15}N$ values that were more similar to other lakes once baselines had been accounted for. Salmonids had higher $\delta^{15}N$ than non-salmonids (Fig. 1c). Kokanee salmon, which making spawning runs, were separate from the other salmonids in Fig. 1c. The spawning runs may mean that kokanee had substantial non-local isotopic signature leading to their separation from other salmon. When considering the salmonids (except kokanee), there was a positive relationship between mass and $\delta^{15}N$ ($R^2 = 0.45$, $t_{10} = 2.71$, $P = .02$). ΣPCB increased with baseline-corrected $\delta^{15}N$ ($R^2 = 0.52$, $t_{15} = 3.91$, $P = .002$), but not bulk $\delta^{15}N$ ($R^2 = 0.09$, $t_{15} = 1.18$, $P = .26$).

Next, we considered the combined effect of isotope levels and geographic variables. For ΣDDT, we excluded those sites with extensive agriculture upstream that may have had DDT use in the past (Nicola and Osoyoos Lakes; geometric mean = 6.7 ng/g compared with 2.3 for the remaining lakes). After excluding those sites, elevation was the best predictor of ΣDDT (Table 3, Fig. 2). For the subset of lakes with measured $\delta^{15}N$, baseline-corrected $\delta^{15}N$ and elevation were the best predictors. For example, the geometric mean values at the highest elevation lakes sampled (all nearby one another in Revelstoke National Park and sampled in 1999) were 206 ± 7 ng/g in the carcass (55 ± 10 ng/g in muscle) from rainbow trout collected at Upper Jade Lake (elevation: 1817 m), and 55 ± 25 ng/g (6.3 ± 1.6 ng/g in muscle) and 17.5 ± 0.5 ng/g (3.1 ± 0.3 ng/g in muscle) in brook trout carcasses collected at Miller (elevation: 1899 m) and Eva (elevation: 1920 m) Lakes, respectively. In 2004, Lower Jade Lake (elevation: 1767 m) had rainbow trout carcasses with 27 ± 42 ng/g (muscle: 28 ± 12 ng/g). In 1999, Revelstoke Lake had bull trout carcasses with 9.4 ± 6.0 ng/g. Thus, levels within these nearby lakes increased with elevation from mid-elevation (Revelstoke) to high elevation (Lower Jade) to highest elevation (Upper Jade), with lower levels on the wetter, more heavily treed west side of the park farther from glaciers (Eva and Miller). In 2004, rainbow trout carcasses from Upper Jade Lake had 97 ± 49 ng/g (muscle: 34 ± 34 ng/g) and brook trout muscle from Eva Lake reported had 3.65 ng/g (muscle: 3.5 ng/g). Thus, five years later, levels in carcasses were <25% (Eva) to 50% (Upper Jade) of what they were in 1999. Percent lipid was the best predictor for dieldrin and HCB, while percent lipid, elevation and $\delta^{13}C$ were the best predictors of toxaphene (Table 3). The best predictor of ΣPCB in fish was percent lipid, lake area, watershed area and the interaction of lake and watershed areas. However, once isotope data was included, then only percent lipid and bulk-corrected $\delta^{15}N$ were included in the best-fit models (Table 3, Fig. 3). There were not enough data on the ∑chlordane to generate meaningful models.

Examining toxaphene in more detail, there were four congeners that were never above detection limits (Parlar (P) 11–12, 25, 32, 56 and 69; note that Parlar 11–12, 15, 21, 25, 38, 51, 56 and Hex-sed were not analyzed for Alberta fish), four congeners that were recorded once (P31, 59 and 62 in fish from Garibaldi Lake, P32 in whitefish from Bow Lake) and three congeners that were recorded twice (P21 and 63 from Garibaldi Lake, and b8-1412 from mountain whitefish in the Miette River and lake trout in Pyramid Lake; b8-1412 was not analyzed for in British Columbia samples). In addition, three congeners were recorded thrice (P15, twice from rainbow trout at Paul Lake and once from a brook trout at Eva Lake; P58, all from Garibaldi Lake; P38, twice at Garibaldi and once from a rainbow trout at Downton), one that was recorded four times (P51, twice at Garibaldi, and once each in a Dolly Varden and mountain whitefish from Carpenter) and one that was recorded nine times (P39, once at Garibaldi; twice from rainbow trout at Upper Jade; once from rainbow trout at Downton; once each from longnose suckers at Lower Kananaskis, Goat Pond and Moose Lake; and once from whitefish at Bow Lake). Finally, the congener hex-sed (2-exo,3-endo,6-exo,8,9,10-hexachlorobornane, Stern et al., 1996) occurred in seven fish, all at Paul Lake, and the congener hep-sed (2-endo,3-exo,5-endo,6-exo,8,9,10- heptachlorobornane, Stern et al., 1996) occurred in ten fish: a rainbow trout from Paul Lake, longnose suckers from Belly River, Chain Lakes, Athabasca River, Lower Kananaskis and Moose Lake, whitefish from Athabasca River and Bow Lake, and lake trout from Pyramid Lake, Moose Lake and Bow Lake. A principal component analysis of the remaining five congeners that were recorded at least 20 times is shown in Fig. 4. Because several fish at Garibaldi Lake (a remote site where levels were likely due to glacial melt) had anomalously high values, we excluded those fish to retain

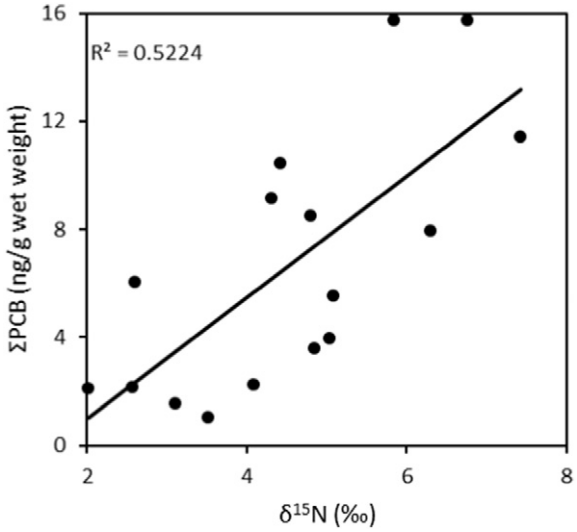


Fig. 2. Baseline-corrected (but not bulk) $\delta^{15}N$ increases with ΣPCB in fish from lakes in western Canada. Two fish from Carpenter Lake (near both a hydropower dam and the Bridge glacier) with very high ΣPCB were excluded.

Table 3
General linear models of explanatory variables for contamination in fish. Only includes models with ΔAICc < 2.0 shown.

Contaminant	Model variables	ΔAICc
ΣDDT	Lipid % + Elevation[1]	0.00
	Lipid %[2]	0.00
Dieldrin	Lipid % + Watershed Area	1.55
	Lipid % + Elevation	1.80
HCB	Lipid %[2]	0.00
	Lipid % + Elevation	1.87
ΣPCB	Lipid % + Lake Area + Watershed Area + Watershed * Lake[3]	0.00
	Lipid % + Watershed Area	1.98
Σtoxaphene	Lipid % + Elevation[4]	0.00

Only results of ΔAICc < 2.0 shown.

[1] Excludes sites with high agricultural use (Nicola and Osoyoos Lakes). The best-fit model for the subset of samples with isotope data was: lipid + elevation + bulk-corrected $\delta^{15}N$ (ΔAICc = 1.65 compared with model with $\delta^{15}N$ alone; ΔAICc = 1.77 compared with model with elevation alone; no other model had ΔAICc >2.0).

[2] The best-fit model for the subset of samples with isotope data was still percent lipid (models with $\delta^{15}N$: ΔAICc >2.0).

[3] Excludes Carpenter Lake, which is near a hydropower plant which likely caused elevated PCB levels. The best-fit model for the subset of samples with isotope data was: lipid + bulk-corrected $\delta^{15}N$ (ΔAICc = 1.65 compared with model with $\delta^{15}N$ alone; ΔAICc = 1.77 compared with model with elevation alone; no other model had ΔAICc > 2.0).

[4] The best-fit model for the subset of samples with isotope data was percent lipid + elevation + $\delta^{13}C$ (models with $\delta^{15}N$: ΔAICc > 2.0). Excludes Paul Lake where toxaphene was used as a piscicide.

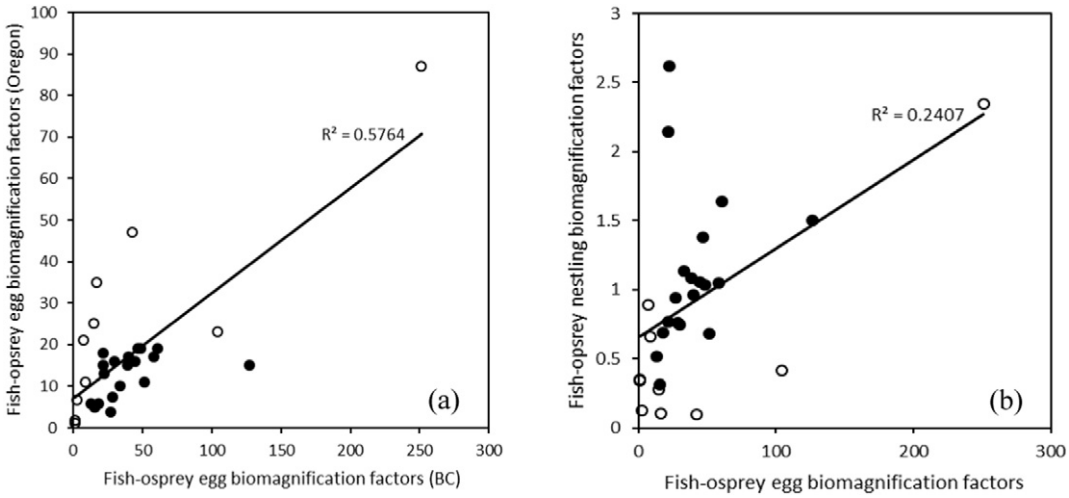


Fig. 3. Fish-Osprey biomagnification factors (a) for eggs in western Canada compared with Oregon and (b) for eggs compared with chicks. Values from Oregon are from Henny et al. (2003). Values for PCB congeners are shown in black; other organochlorines in open symbols. Biomagnification factors in BC were calculated assuming a diet of 39.7% salmonids, 42.7% catostomids and 17.6% cyprinids. We calculated the average contaminant value of the diet and of ospreys for each of seven watersheds, and averaged the biomagnification factor. If data were missing for a dietary item, then we assumed that the ratio of contaminants for that diet item was the same as the average across all other watersheds.

normality for the analysis. Congeners P40–41, 42 and 44 were heavily loaded together while congeners P26 and 50 were loaded together. Nicola tended to have relatively low levels of all congeners, with Paul (the only Nicola watershed point to the right of the origin) having higher levels; Paul Lake is the only lake within our dataset known to have had use of toxaphene as a piscicide. This is consistent with studies in other lakes that had toxaphene treatment (Miskimmin et al., 1995). Revelstoke fish had relatively high levels of most congeners, but relatively low levels of P26 and 60. Carpenter fish showed no clear pattern. For osprey eggs, Parlar 11, 15, 32, 39, 56, 58, 62 ad 69 were never reported, while for adult and nestling plasma, Parlar 11, 15, 56, 69 and 69 were never reported, with 31, 32, 58 and 62 only reported from a single nestling in the Athabasca River. For osprey eggs, hex-sed was reported for all eggs except Belly River and hep-sed was only recorded at Chaine Lakes and West Jasper, whereas for osprey nestlings both hex-sed and hep-sed were reported from one nest at each of Belly River and Athabasca River, while hep-sed alone was reported from a nest at each of Bow Lake and Pitt River. No other nestlings recorded either congener.For the remaining congeners, there was little separation between the Alberta and British Columbia eggs, although eggs from British Columbia tended to have lower 26 (unlike the fish), 50 (like the fish) and hex-sed (like the fish). The greater overlap in osprey eggs compared with fish may represent a signal from wintering grounds or other periods.

3.2. Biomagnification factors from fish to osprey

Salmonids made up almost one-third of prey delivered by osprey (Table 4), followed by suckers (castostomids) and cyprinids. Nonetheless, there was substantial variation among study locations. At Lillooet and Nicola Lakes, salmonids and suckers made up most of the prey while at Upper Arrow Lake salmonids alone made up most of the prey. At Oliver and Pitt Lakes, prey were more evenly spread among families with cyprinids also being significant. At Pitt, almost one third of prey deliveries were catfish. Thus, clearly diet varied across sites and we consequently averaged BMFs across sites.

Biomagnification from fish to osprey eggs was highest in PCB and DDT compounds. The highest wet-weight biomagnification factor was for CB-194 (127) and p,p′-DDE (251). Biomagnification from fish to osprey nestlings was significantly lower than between fish and eggs (e.g. 25 out of 34 studied contaminants had a BMF ≤ 1). The highest BMF was for PCB 171 (2.6), and p,p′-DDE (2.3). BMFs were higher in eggs for every compound except cis-chlordane (Table 5). Comparing our results for fish to egg biomagnification to Henny et al. (2003), all calculated PCB and DDT BMF were higher in the present study. For sixteen of the above contaminants, the eggs in our study site had BMF greater than twice those of the Willamette river study (Table 5, Fig. 5).

4. Discussion

Long-range transport leads to 'cold-trapping' of semi-volatile POPs at high elevations, with a 10–100 fold increase in POPs in snow at high elevations (3100 m) in western Canada (Blais et al., 1998). High levels of POPs also occur in soils at higher elevation in Mount Revelstoke. POPs, especially DDT, occur at these high elevation due to high precipitation rates, lower volatilization at colder temperatures, scavenging by the high surface area of falling snow, and accumulation in the snowpack by adsorption to organic matter (Fernandez and Grimalt, 2003; Burniston et al., 2007; Milner et al., 2017). Such long-range transport was evident in our study for DDT and toxaphene, both of which increased with elevation. As there was likely no historical use of DDT directly into the alpine lakes in Revelstoke National Park, DDT likely arrived via atmospheric transport from nearby valleys. One possible origin is the Okanagan Valley, where DDT was sprayed widely, which may have led to higher deposition on the southeasterly Jade Lakes compared with the northwesterly Miller and Eva Lakes. However, there was also spraying in valleys adjacent to the park, possibly including some within the park itself (e.g. https://search-bcarchives.royalbcmuseum.bc.ca/helicopter-spraying-ddt-at-invermere-3). On the other hand, the relatively high DDT:DDE ratio in our study suggests that it is linked to air masses arriving from Asia, as reported elsewhere for the Rocky Mountains, with potentially some contribute from ongoing use in Latin America. Similarly, Arctic air masses have higher DDT:DDE ratios when they originate in Asia. Similarly, the congener composition of toxaphene implied that toxaphene also arrived via long range transport, as was the case in ospreys.

Local sources of DDT were apparent in the Okanagan Valley and Nicola Lake area, where intensive agriculture occurs. Indeed, 30 years after DDT was restricted in Canada, levels in songbirds in the Okanagan Valley were so high (Elliott et al., 2005) that even 40 years later, peregrine falcons are unlikely to be able to successfully breed in the region. The highest levels of DDT in two species of birds of prey in British Columbia is also in the Okanagan region (Elliott et al., 2015).

Our trends were similar to those we reported previously in ospreys. However, in ospreys, no nests were found above 900 m in British Columbia, where long range transport from Asia may be stronger due to

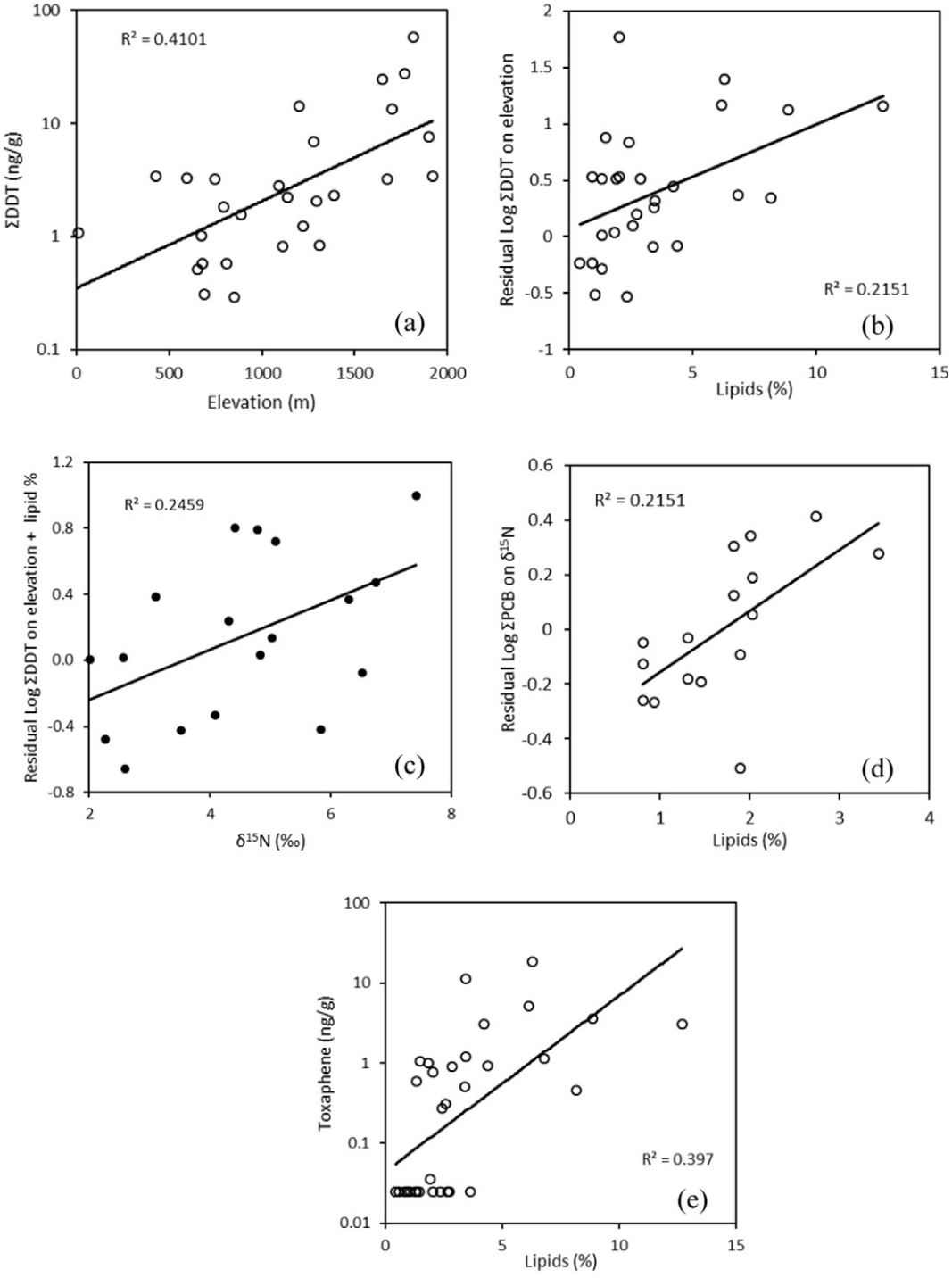


Fig. 4. ΣDDT increased with (a) elevation and (b) % lipids, and, (c) in the subset of fish with isotope data, with baseline-corrected $\delta^{15}N$. Nicola and Osoyoos Lake values were removed as they have known agricultural input. (d) ΣPCB increases with % lipids after correcting for baseline-corrected $\delta^{15}N$ (see Fig. 2). Carpenter Lake values were removed as the site has a hydropower plant nearby. (e) Toxaphene increased with percent lipids. Paul Lake value was removed due to known use of toxaphene as a piscicide.

proximity, and so elevation was less significant in some results (Elliott et al., 2012). Moreover, that analysis did not include baseline $\delta^{15}N$ values, and so levels of trophic position may have been skewed. Similar to the fish, elevation, bulk $\delta^{13}C$ and bulk $\delta^{15}N$ was the best model for explaining ΣDDT in osprey nestlings, along with percent lipid (Elliott et al., 2012). For eggs, which may integrate both breeding and non-breeding sources for highly recalcitrant compounds, bulk $\delta^{13}C$, glacier area, lake area and watershed area were all important (Elliott et al., 2012). The proportion of glacier in a given watershed was the only significant predictor of toxaphene in nestlings, whereas elevation, lake area, percent lipids and bulk $\delta^{13}C$ were significant for eggs. Lake area was the only significant predictor of ΣPCB in nestlings, whereas elevation, lake area, percent lipids and $\delta^{13}C$ were significant in eggs.

Apart from elevation and percent lipid (which is commonly correlated with contaminant levels for these lipophilic compounds), a major correlate of contaminant concentration in fish was trophic position, as assessed by $\delta^{15}N$. However, bulk $\delta^{15}N$ did not predict any contaminants. Rather, it was necessary to correct for baselines using levels of $\delta^{15}N$ in 'source' amino acids in osprey eggs. This interesting approach shows the potential for top predators, such as raptors, to act as

Table 4
Fish delivered (% in diet) to osprey nests at five study lakes in southern British Columbia in 2001. Five nests were recorded at each site for 60 h/site. Totals per family include unidentified species. Values in bold refer to family totals, including individuals known to family but not to species.

Species	Lillooet	Upper Arrow	Nicola	Oliver	Pitt
Cutthroat trout (*Oncorhynchus clarki*)					0.7
Kokanee (*Oncorhynchus nerka*)		37.0	31.9		
Mountain whitefish (*Prosopium williamsoni*)	2.2			4.3	7.9
Rainbow trout (*Oncorhynchus mykiss*)	37.0	27.0	5.6	5.3	
Salmonidae	**41.4**	**76.0**	**41.7**	**10.7**	**16.5**
Common carp (*Cyprinus carpio*)				12.8	2.9
Goldfish (*Carassius auratus*)					0.7
Northern pikeminnow (*Ptychocheilus oreronensis*)		7.8	1.4	3.2	10.0
Peamouth chub (*Mylocheilus caurinus*)			12.5		2.9
Tench (*Tinea tinea*)				3.2	
Cyprinidae		**7.8**	**13.9**	**19.2**	**24.4**
Bridgelip sucker (*Catostomus columbianus*)	47.8	3.9			
Longnose sucker (*Catostomus catostomus*)	2.2	3.9	12.5	1.1	5.7
Largescale sucker (*Catostomus macrocheilus*)			16.7	4.3	2.1
Catostomidae	**52.2**	**11.7**	**43.1**	**15.0**	**12.1**
Black crappie (*Pomoxis nirromaculatus*)				1.1	1.4
Smallmouth bass (*Microvterus dolomieu*)				5.3	0.7
Centrarchidae				**6.4**	**2.1**
Brown catfish (*Ameiurus nebulosus*)					30.1
Ictaluridae				**1.1**	**30.1**
Prickly sculpin (*Cottus asper*)					0.7
Yellow perch (*Perca flavescens*)				1.1	
Starry flounder (*Platichthys stellatus*)					1.4
Unidentified species	6.5	3.9	1.4	46.8	12.1

sentinels for the entire ecosystem, even providing baselines for other biota. Both PCB and DDT levels increased with baseline-corrected trophic level in fish.

Table 5
Biomagnification factors between fish and ospreys. Nestling values refer to plasma.

Contaminant	Prey-egg (BC)	Prey-egg (Oregon)	Prey-nestling (BC)
PCB 74	27	3.8	0.94
PCB 99	29	7.4	0.76
PCB 105	34	10	1.13
PCB 110	18	5.9	0.69
PCB 138	30	16	0.75
PCB 141	22	15	0.77
PCB 146	58	17	1.04
PCB 149	16	5	0.32
PCB 153	45	16	1.06
PCB 170 + 190	47	19	1.38
PCB 171	22	13	2.62
PCB 172	61	19	1.64
PCB 174	13	5.8	0.52
PCB 180	49	19	1.04
PCB 183	39	15	1.08
PCB 194	127	15	1.5
PCB 200	21	18	2.14
PCB 201	40	17	0.96
ΣPCB	51	11	0.68
HCB	1.3	1.2	0.35
p,p'-DDE	251	87	2.34
Mirex	17	35	0.11
Oxychloradane	7.4	21	0.89
Trans-nonachlor	1.1	1.8	0.34
Cis-nonachlor	8.5	11	0.66
p,p'-DDT	42	47	0.1
HE	15	25	0.28
Dieldrin	2.5	6.7	0.13
p,p'-DDD	104	23	0.42
ΣDDT	176		1.64
Toxaphene	190		

4.1. Biomagnification factors

The high BMF in osprey eggs for PCB and DDT was to be expected, as these are highly hydrophobic (lipophilic) substances ($LogK_{OW}$ from 4.5 to 8.3 for PCBs and from 6.02 to 6.91 for DDT) (Howard and Meylan, 1997; Zhou et al., 2005). Since both lipid-weight and wet-weight BMFs were <1 for most compounds in osprey chicks and since there was a large ratio between egg and chick BMF, suggesting that as osprey develop, their metabolism is capable of significantly reducing the concentration of toxic contaminants or contaminants are diluted during growth (Drouillard et al., 2003). Indeed, biomagnification in chicks was minimal.

The comparison between our study and Henny et al. (2003) hint that higher latitudes and elevation could possibly lead to higher contamination of PCB and DDT compounds, and less contamination of some chlordanes, mirex, HE and dieldrin. Both migratory osprey populations migrated to the same general areas of Latin America (Elliott et al., 2007) and were located in watersheds that had not been not subject to any important local contamination of PCB and DDT for decades. As such, the colder climate of our study site may be a reasonable hypothesis for explaining the large differences observed in BMF. Of course, latitude and elevation may not be the only factors at play here. A simple linear regression showed that there was a trend of an increase in ~1.2 times the concentration of contaminants in our study site (although the fit was not strong). A future analysis comparing the volatility of each contaminant as a measure of $\log(K_{OA})$ and the change in BMF from one study site to the other for each group of contaminants could help support or dismiss this hypothesis. Regardless, the variation in BMFs between two relatively similar studies demonstrates the limitations of the BMF approach and the need to use more robust trophic magnification factors (Borgå et al., 2012; Fremlin, 2018).

4.2. Melting glaciers and cold-trapping as a source of contaminants in alpine ospreys

We were unable to sample ospreys at high elevation in the west, as nests were sparse or adults only visited small alpine lakes post-breeding. However, our BMFs allow an estimation of levels expected in ospreys using these lakes. For example, combining our BMFs with measured POPs levels would predict that osprey eggs in nests around the alpine lakes at Revelstoke would have had 36 μg/g w.w. ΣDDT (six times the threshold for nest failure in ospreys; Wiemeyer et al., 1988) and around Garibaldi Lake would have had 9.0 μg/g w.w. toxaphene (well below any effect on fertility in chickens; Arscott et al., 1976). Thus, transport of chemicals, both locally within British Columbia and over longer ranges from Asia, followed by cold-trapping in the mountains of western Canada, caused negative impacts on wildlife health even thirty years after many of the chemicals were banned in Canada. Additionally, climate change may portend an increase in the elevation at which osprey and other species are able to successfully breed (Maggini et al., 2011).

Whereas climate change and chemical pollution are often considered separate 'planetary boundaries' within the Anthropocene, there is growing awareness that these 'boundaries' can interact (Jenssen, 2005; Noyes et al., 2009). Specifically, changing weather and other climatic patterns can alter deposition patterns of contaminants (Nadal et al., 2015; Foster et al., 2019). Climate change can also lead to changes in food web structure, leading to changes in bioaccumulation patterns in top predators (McKinney et al., 2015; Braune et al., 2015). Given the strong evidence for cold-trapping and trophic magnification for DDT, PCBs and toxaphene in alpine lakes, both pathways are clearly important in alpine Canada. As POPs accumulated in glaciers during the mid-century have long since melted out of glaciers, current long range transport is likely the main source of POPs in alpine Canada. Warming of alpine environments and disappearance of glaciers may reduce cold-trapping, therefore reducing contaminant levels in the alpine. In contrast, warmer temperatures in the future may facilitate fish growth,

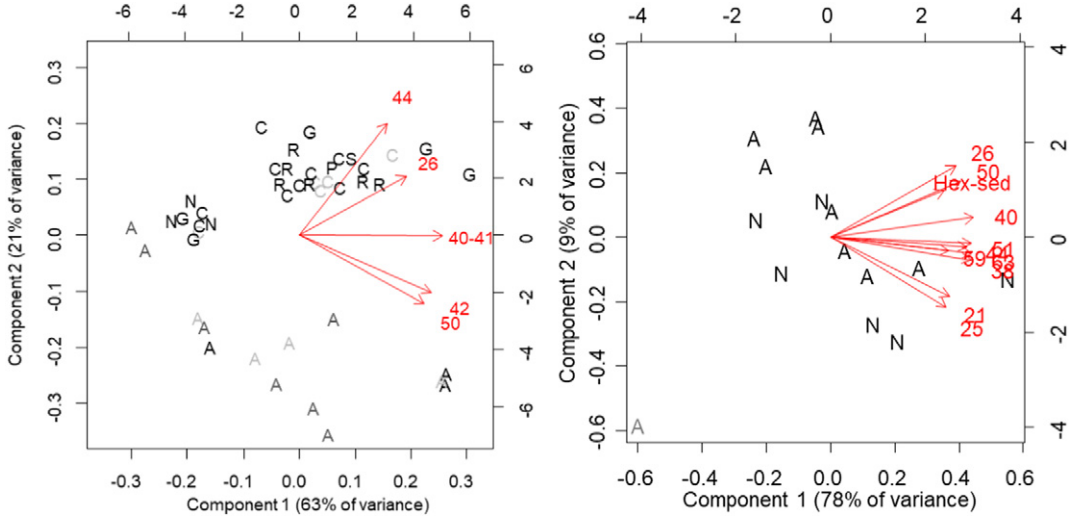


Fig. 5. (a) Principal component analysis of toxaphene congeners (log-transformed) in (a) fish and (b) osprey eggs from watersheds across western Canada (A = Alberta, C = Carpenter, N = Nicola, P = Paul, R = Revelstoke). All fish were salmonids except those in like grey (whitefish) and medium grey (suckers); the grey symbol for osprey eggs is an egg from the Belly River, outside of the montane region of Alberta. In total, 28 fish where no toxaphene was detected and 12 fish where only one toxaphene congener was detected are excluded from analyses to meet statistical assumptions.

potentially altering contaminant levels. The interaction of climate change and contaminants remains important to study.

Declaration of competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

M. Conner, A. Elliott, H. Gill, G. Kardosi, M. Machmer, C. Steeger, S. Weech, L. Wilson and, most of all, S. Lee 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. 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. This manuscript was based on PG's Honours thesis, and we thank the McGill School of Environment, and especially K. Roulet, for their help in developing the thesis.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.135889.

References

Alberta Environment and Sustainable Resource Development, 2015. Watersheds of Alberta. Retrieved from. https://maps.alberta.ca/genesis/rest/services/Watersheds_of_Alberta/Latest/MapServer.

Arscott, G.H., Michener, S., Bills, D.D., 1976. Effect of toxaphene on the performance of white leghorn layers and their progeny. Poultry Sci 55, 1130–1131.

Atlas, E., Giam, C.S., 1981. Global transport of organic pollutants: ambient concentrations in the remote marine atmosphere. Science 211, 163–165.

Basu, N., Scheuhammer, A.M., Bursian, S.J., Elliott, J., Rouvinen-Watt, K., Chan, H.M., 2007. Mink as a sentinel species in environmental health. Environ. Res. 103, 130–144.

Blais, J.M., Schindler, D.W., Muir, D.C.G., Kimpe, L.E., Donald, D.B., Rosenberg, B., 1998. Accumulation of persistent OC compounds in mountains of western Canada. Nature 395, 585–588.

Blais, J.M., Schindler, D.W., Muir, D.C.G., Sharp, M., Donald, D., Lafrenière, M., Strachan, W.M.J., 2001a. Melting glaciers: a major source of persistent organochlorines to Subalpine Bow Lake in Banff National Park, Canada. AMBIO 30, 410–415. https://doi.org/10.1579/0044-7447-30.7.410.

Blais, J.M., Schindler, D.W., Sharp, M., Braekevelt, E., Lafrenière, M., McDonald, K., Muir, D.C.G., Strachan, W.M.J., 2001b. Fluxes of semivolatile organochlorine compounds in Bow Lake, a high-altitude, glacier-fed, subalpine lake in the Canadian Rocky Mountains. Limnol. Oceanogr. 46, 2019–2031.

Borgå, K., Kidd, K.A., Muir, D.C., Berglund, O., Conder, J.M., Gobas, F.A., Kucklick, J., Malm, O., Powell, D.E., 2012. Trophic magnification factors: considerations of ecology, ecosystems, and study design. Integr. Environ. Assess. Manage. 8, 64–84.

Braune, B., Donaldson, G., Hobson, K., 2002. Contaminant residues in seabird eggs from the Canadian Arctic. II. Spatial trends and evidence from stable isotopes for intercolony differences. Environ. Poll. 117, 133–145. https://doi.org/10.1016/S0269-7491(01)00186-5.

Braune, B.M., Gaston, A.J., Hobson, K.A., Gilchrist, H.G., Mallory, M.L., 2015. Changes in trophic position affect rates of contaminant decline at two seabird colonies in the Canadian Arctic. Ecotoxicol. Environ. Safety 115, 7–13.

Brogan, J.M., Green, D.J., Maisonneuve, F., Elliott, J.E., 2017. An assessment of exposure and effects of persistent organic pollutants in an urban Cooper's hawk (Accipiter cooperii) population. Ecotoxicology 26, 32–45.

Burniston, D.A., Strachan, W.J.M., Hoff, J.T., Wania, F., 2007. Changes in surface area and concentrations of semivolatile organic contaminants in aging snow. Environ. Sci. Technol. 41, 4932–4937.

Cesh, L., Elliott, K.H., McKinney, M., Quade, S., Maisonneuve, F., Garcelon, D.K., Sandau, C.D., Letcher, R.J., Williams, T.D., Elliott, J.E., 2010. Polyhalogenated aromatic hydrocarbons and metabolites: relation to circulating thyroid hormone and retinol in nestling bald eagles (*Haliaeetus leucocephalus*). Environ. Toxicol. Chem. 29, 1301–1311.

de Geus, H.J., Besselink, H., Brouwer, A., Klungsøyr, J., McHugh, B., Nixon, E., de Boer, J., 1999. Environmental occurrence, analysis, and toxicology of toxaphene compounds. Environ. Health Perspectives 107S, 115–144. https://doi.org/10.1289/ehp.99107s1115.

Dolgova, S., Popp, B.N., Courtoreille, K., Espie, R.H., Maclean, B., McMaster, M., Straka, J.R., Tetreault, G.R., Wilkie, S., Hebert, C.E., 2018. Spatial trends in a biomagnifying contaminant: application of amino acid compound–specific stable nitrogen isotope analysis to the interpretation of bird mercury levels. Environ. Toxicol. Chem. 37, 1466–1475.

Donald, D., Bailey, R., Crosley, R., Muir, D., Shaw, P., Syrgiannis, J., 1993. Polychlorinated Biphenyls and Organochlorine Pesticides in the Aquatic Environment along the Continental Divide Region of Alberta and British Columbia. Environment Canada, Western and Northern Region, Water Quality Branch, Regina (Report).

Donald, D.B., Syrgiannis, J., Crosley, R.W., Holdsworth, G., Muir, D.C.G., Rosenberg, B., Sole, A., Schindler, D.W., 1999. Delayed deposition of organochlorine pesticides at a temperate glacier. Environ. Sci. Technol. 33, 1794–1798.

Drouillard, K.G., Norstrom, R.J., Fox, G.A., Gilman, A., Peakall, D.B., 2003. Development and validation of a herring gull embryo toxicokinetic model for PCBs. Ecotoxicology 12, 55–68.

Elliott, J.E., Elliott, K.H., 2013. Tracking marine pollution. Science 340, 556–558.
Elliott, K.H., Elliott, J.E., 2016. Origin of sulfur in diet drives spatial and temporal mercury trends in seabird eggs from Pacific Canada 1968–2015. Environ. Sci. Technol. 50, 13380–13386.
Elliott, J.E., Wilson, L.K., Henny, C.J., Machmer, M.M., 2000. Contaminants in ospreys from the Pacific Northwest: II. Patterns and trends in organochlorine pesticides, polychlorinated biphenyls and mercury in eggs and plasma. Arch. Environ. Contamin. Toxicol. 38, 93–106.
Elliott, J.E., Miller, M.J., Wilson, L.K., 2005. Assessing breeding potential of peregrine falcons based on chlorinated hydrocarbon concentrations in prey. Environ. Poll. 134, 353–361.
Elliott, J.E., Morrissey, C.A., Henny, C.J., Inzunza, E.R., Shaw, P., 2007. Satellite telemetry and prey sampling reveal contaminant sources to Pacific Northwest ospreys. Ecol. Appl. 17, 1223–1233.
Elliott, J.E., Levac, J., Guigueno, M.F., Shaw, D.P., Wayland, M., Morrissey, C.A., Elliott, K.H., 2012. Factors influencing legacy pollutant accumulation in alpine osprey: biology, topography, or melting glaciers? Environ. Sci. Technol. 46, 9681–9689. https://doi.org/10.1021/es301539b.
Elliott, J.E., Brogan, J., Lee, S.L., Drouillard, K.G., Elliott, K.H., 2015. PBDEs and other POPs in urban birds of prey partly explained by trophic level and carbon source. Sci. Tot. Environ. 524, 157–165.
Fernandez, P., Grimalt, J.O., 2003. On the global distribution of persistent organic pollutants. Chimia 57, 514–521.
Foster, K.L., Braune, B.M., Gaston, A.J., Mallory, M.L., 2019. Climate influence on legacy organochlorine pollutants in Arctic seabirds. Environ. Sci. Technol. 53, 2518–2528.
Franz, T.P., Eisenreich, S.J., 1998. Snow scavenging of polychlorinated biphenyls and polycyclic aromatic hydrocarbons in Minnesota. Environ. Sci. Technol. 32, 1771–1778.
Fremlin, K., 2018. Trophic Magnification of Legacy Persistent Organic Pollutants and Emergent Contaminants within a Terrestrial Food-Web of an Avian Apex Predator, the Cooper's Hawk (*Accipiter cooperii*). PhD dissertation. Simon Fraser University, Burnaby, BC, Canada.
Furness, R.W., 1993. Birds as monitors of pollution. In: Furness, R.W., Greenwood, J.J.D. (Eds.), Birds as Monitors of Environmental Change. Springer, Dordrecht, pp. 86–143.
Glassmeyer, S.T., Shanks, K.E., Hites, R.A., 1999. Automated toxaphene quantitation by GC/MS. Anal. Chem. 71 (7), 1448–1453.
Grove, R.A., Henny, C.J., Kaiser, J.L., 2009. Osprey: worldwide sentinel species for assessing and monitoring environmental contamination in rivers, lakes, reservoirs, and estuaries. J. Toxicol. Environ. Health 12, 25–44. https://doi.org/10.1080/10937400802545078.
Guertin, D.A., Ben-David, M., Harestad, A., Drouillard, K.G., Elliott, J.E., 2010. Non-invasive fecal sampling links individual river otters to chlorinated hydrocarbon contaminant exposure. Environ. Toxicol. Chem. 29, 275–284.
Guigueno, M.F., Elliott, K.H., Levac, J., Wayland, M., Elliott, J.E., 2012. Differential exposure of alpine ospreys to mercury: melting glaciers, hydrology or deposition patterns? Environ. Int. 40, 24–32. https://doi.org/10.1016/j.envint.2011.11.004.
Hebert, C.E., Keenleyside, K.A., 1995. To normalize or not to normalize? Fat is the question. Environ. Toxicol. Chem. 14, 801–807.
Henny, C.J., Kaiser, J.L., Grove, R.A., Bentley, V.R., Elliott, J.E., 2003. Biomagnification factors (fish to osprey eggs from Willamette River, Oregon, U.S.A.) for PCDDs, PCDFs, PCVs and OC pesticides. Environ. Monitoring Assess. 84, 275–315.
Henny, C.J., Kaiser, J.L., Grove, R.A., Johnson, B.L., Letcher, R.J., 2009. Polybrominated diphenyl ether flame retardants in eggs may reduce reproductive success of ospreys in Oregon and Washington, USA. Ecotoxicol 18, 802–813.
Henny, C.J., Grove, R.A., Kaiser, J.L., Johnson, B.L., 2010. North American osprey populations and contaminants: historic and contemporary perspectives. J. Toxicol. Environ. Health B 13, 579–603. https://doi.org/10.1080/10937404.2010.538658.
Hobson, K.A., 1995. Reconstructing avian diets using stable-carbon and nitrogen isotope analysis of egg components: patterns of isotopic fractionation and turnover. Condor 97, 752–762. https://doi.org/10.2307/1369183.
Howard, P., Meylan, W., 1997. Handbook of Physical Properties of Organic Chemicals. CRC Press, Lewis Publishers, Boca Raton, FL.
Jaffe, D., McKendry, I., Anderson, T., Price, H., 2003. Six 'new' eisodes of trans-Pacific transport of air pollutants. Atmosph. Environ. 37, 391–404.
Jaspers, V.L.B., Rodriguez, F.S., Boertmann, D., Sonne, C., Dietz, R., Rasmussen, L.M., Eens, M., Covaci, A., 2011. Body feathers as a potential new biomonitoring tool in raptors: a study on organohalogenated contaminants in different feather types and preen oil of West Greenland white-tailed eagles (*Haliaeetus albicilla*). Environ. Int. 37, 1349–1356.
Jenssen, B.M., 2005. Endocrine-disrupting chemicals and climate change: a worst-case combination for arctic marine mammals and seabirds? Environ. Health Perspectives 114, 76–80.
Kidd, K.A., Schindler, D.W., Muir, D.C., Lockhart, W.L., Hesslein, R.H., 1995. High concentrations of toxaphene in fishes from a subarctic lake. Science 269, 240–242.
Logan, J.M., Jardine, T.D., Miller, T.J., Bunn, S.E., Cunjak, R.A., Lutcavage, M.E., 2008. Lipid corrections in carbon and nitrogen stable isotope analyses: comparison of chemical extraction and modelling methods. J. Anim. Ecol. 77, 838–846.
Lorrain, A., Graham, B., Ménard, F., Popp, B., Bouillon, S., Van Breugel, P., Cherel, Y., 2009. Nitrogen and carbon isotope values of individual amino acids: a tool to study foraging ecology of penguins in the Southern Ocean. Mar. Ecol. Prog. Ser. 391, 293–306.
Løseth, M.E., Briels, N., Flo, J., Malarvannan, G., Poma, G., Covaci, A., Herzke, D., Nygård, T., Bustnes, J.O., Jenssen, B.M., Jaspers, V.L., 2019. White-tailed eagle (*Haliaeetus albicilla*) feathers from Norway are suitable for monitoring of legacy, but not emerging contaminants. Sci. Tot. Environ. 647, 525–533.
Luo, W., Gao, J., Bi, X., Xu, L., Guo, J., Zhang, Q., Romesh, K.Y., Giesy, J.P., Kang, S., 2016. Identification of sources of polycyclic aromatic hydrocarbons based on concentrations in soils from two sides of the Himalayas between China and Nepal. Environ. Poll. 212, 424–432.
Maggini, R., Lehmann, A., Kéry, M., Schmid, H., Beniston, M., Jenni, L., Zbinden, N., 2011. Are Swiss birds tracking climate change? Detecting elevational shifts using response curve shapes. Ecol. Model. 222, 21–32.
McKinney, M.A., Pedro, S., Dietz, R., Sonne, C., Fisk, A.T., Roy, D., Jenssen, B.M., Letcher, R.J., 2015. A review of ecological impacts of global climate change on persistent organic pollutant and mercury pathways and exposures in arctic marine ecosystems. Curr. Zool. 61 (4), 617–628.
Milner, K.R., Blais, J., Bogdal, C., Villa, S., Schwikowski, M., Pavlova, P., Steinlin, C., Gerbi, C., Kreutz, K.J., 2017. Legacy organochlorine pollutants in glacial watersheds: a review. Environ. Sci. Processes & Impacts 19 (12), 1474–1483.
Miskimmin, B.M., Muir, D.C.G., Schindler, D.W., Stern, G.A., Grift, N.P., 1995. Chlorobornanes in sediments and fish 30 years after toxaphene treatment of lakes. Environ. Sci. Technol. 29, 2490–2495.
Morrissey, C.A., Kardosi, G., Wilson, L.K., Elliott, J.E., 2004. Effect of food supply and contaminants on osprey productivity in southern British Columbia, Canada. Raptors Worldwide (Budapest, Hungary).
Muir, D.C., Ford, C.A., Grift, N.P., Metner, D.A., Lockhart, W.L., 1990. Geographic variation of chlorinated hydrocarbons in burbot (*Lota lota*) from remote lakes and rivers in Canada. Arch. Environ. Contamin. Toxicol. 19, 530–542.
Nadal, M., Marquès, M., Mari, M., Domingo, J.L., 2015. Climate change and environmental concentrations of POPs: a review. Environ. Res. 143, 177–185.
Natural Resources Canada, 2001a. Canada 3D:Product Standard: Sherbrooke, PQ.
Natural Resources Canada, 2001b. North American Atlas. Glaciers, Sherbrooke, PQ.
Noyes, P.D., McElwee, M.K., Miller, H.D., Clark, B.W., Van Tiem, L.A., Walcott, K.C., Erwin, K.N., Levin, E.D., 2009. The toxicology of climate change: environmental contaminants in a warming world. Environ. Int. 35, 971–986.
Ortiz-Santaliestra, M.E., Resano-Mayor, J., Hernández-Matías, A., Rodríguez-Estival, J., Camarero, P.R., Moleón, M., Real, J., Mateo, R., 2015. Pollutant accumulation patterns in nestlings of an avian top predator: biochemical and metabolic effects. Sci. Tot. Environ. 538, 692–702.
Pajuelo, M., Bjorndal, K.A., Alfaro-Shigueto, J., Seminoff, J.A., Mangel, J.C., Bolten, A.B., 2010. Stable isotope variation in loggerhead turtles reveals Pacific-Atlantic oceanographic differences. Mar. Ecol. Prog. Ser. 417, 277–285.
Risebrough, R.W., Menzel, D.B., Jun, D.J.M., Olcott, H.S., 1967. DDT residues in Pacific sea birds: a persistent insecticide in marine food chains. Nature 216, 589.
Roos, A.M., Bäcklin, B.M.V., Helander, B.O., Rigét, F.F., Eriksson, U.C., 2012. Improved reproductive success in otters (*Lutra lutra*), grey seals (*Halichoerus grypus*) and sea eagles (*Haliaeetus albicilla*) from Sweden in relation to concentrations of organochlorine contaminants. Environ. Poll. 170, 268–275.
Semeena, V.S., Lammel, G., 2005. The significance of the grasshopper effect on the atmospheric distribution of persistent organic substances. Geophys. Res. Lett. 32, L07804. https://doi.org/10.1029/2004GL022229.
Seminoff, J.A., Benson, S.R., Arthur, K.E., Eguchi, T., Dutton, P.H., Tapilatu, R.F., Popp, B.N., 2012. Stable isotope tracking of endangered sea turtles: validation with satellite telemetry and $\delta^{15}N$ analysis of amino acids. PLoS One 7, e37403.
Sergio, F., Caro, T., Brown, D., Clucas, B., Hunter, J., Ketchum, J., Hiraldo, F., 2008. Top predators as conservation tools: ecological rationale, assumptions, and efficacy. Ann. Rev. Ecol. Evol. System. 39, 1–19. https://doi.org/10.1146/annurev.ecolsys.39.110707.173545.
Shaw, P., Gray, C., 2004. Persistent organohalogen contaminants in fish from lakes and reservoirs in southwestern British Columbia, Canada. Proceedings of the 2003 Georgia Basin/Puget Sound Research Conference, Vancouver, BC. 31, pp. 1–5.
Spitzer, P.R., Risebrough, R.W., Walker, W., Hernandez, R., Poole, A., Puleston, D., Nisbet, I.C., 1978. Productivity of ospreys in Connecticut—Long Island increases as DDE residues decline. Science 202, 333–335.
Steinlin, C., Bogdal, C., Lüthi, M.P., Pavlova, P.A., Schwikowski, M., Zennegg, M., Schmid, P., Scheringer, M., Hungerbühler, K., 2016. A temperate alpine glacier as a reservoir of polychlorinated biphenyls: model results of incorporation, transport, and release. Environ. Sci. Technol. 50 (11), 5572–5579.
Stern, G.A., Loewen, M.D., Miskimmin, B.M., Muir, D.C.G., Westmore, J.B., 1996. Characterization of two major toxaphene components in treated lake sediment. Environ. Sci. Technol. 30, 2251–2258.
Stringer, O.E., McMynn, R.G., 1960. Three year's use of toxaphene as a fish toxicant in British Columbia. Can. Fish Culturist 28, 37–44.
Von Waldow, H., MacLeod, M., Jones, K., Scheringer, M., Hungerbuhler, K., 2010. Remoteness from emission sources explains the fractionation pattern of polychlorinated biphenyls in the Northern Hemisphere. Environ. Sci. Technol. 44, 6183–6188.
Wania, F., Mackay, D., 1993. Global fractionation and cold condensation of low volatility organochlorine compounds. Ambio 22, 10–18.
Wiemeyer, S.N., Bunck, C.M., Krynitsky, A.J., 1988. Organochlorine pesticides, polychlorinated biphenyls, and mercury in osprey eggs—1970–79—and their relationships to shell thinning and productivity. Arch. Environ. Contamin. Toxicol. 17, 767–787.
Yarnes, C., Santos, F., Singh, N., Abiven, S., Schmidt, M.W., Bird, J.A., 2011. Stable isotopic analysis of pyrogenic organic matter in soils by liquid chromatography–isotope-ratio mass spectrometry of benzene polycarboxylic acids. Rapid Commun. Mass Spectrom. 25 (24), 3723–3731.
Zhou, W., Zhai, Z., Wang, Z., Wang, L., 2005. Estimation of n-octanol/water partition coefficients (K_{ow}) of all PCB congeners by density functional theory. J. Mol. Structure: THEOCHEM 755, 137–145. https://doi.org/10.1016/J.THEOCHEM.2005.08.020.
Zhou, Q., Zhang, J., Fu, J., Shi, J., Jiang, G., 2008. Biomonitoring: an appealing tool for assessment of metal pollution in the aquatic ecosystem. Anal. Chim. Acta 606, 135–150. https://doi.org/10.1016/J.ACA.2007.11.018.