Environmental Pollution 157 (2009) 755–762

Contents lists available at ScienceDirect

Environmental Pollution

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

Monitoring organic contaminants in eggs of glaucous and glaucous-winged gulls (*Larus hyperboreus* and *Larus glaucescens*) from Alaska

Stacy S. Vander Pol [a,*], Paul R. Becker [a], Michael B. Ellisor [a], Amanda J. Moors [a], Rebecca S. Pugh [a], David G. Roseneau [b]

[a] National Institute of Standards and Technology, Hollings Marine Laboratory, 331 Fort Johnson Road, Charleston, SC 29412, USA
[b] U.S. Fish and Wildlife Service, Alaska Maritime National Wildlife Refuge, 95 Sterling Highway, Suite 1, Homer, AK 99603, USA

Organic contaminant concentrations in Alaskan gull eggs could possibly be affecting chick growth and survival rates, but the eggs should be safe for humans to eat in small quantities.

ARTICLE INFO

Article history:
Received 23 September 2008
Received in revised form
14 November 2008
Accepted 19 November 2008

Keywords:
Organic contaminant
Glaucous gull
Glaucous-winged gull
Seabird egg
Subsistence food

ABSTRACT

Gull eggs have been used to monitor contaminants in many parts of the world. The Seabird Tissue Archival and Monitoring Project (STAMP) is a long-term program designed to track trends in pollutants in northern marine environments using seabird eggs. Glaucous and glaucous-winged gull (*Larus hyperboreus* and *Larus glaucescens*) eggs collected in 2005 from seven Alaskan colonies were analyzed for organic contaminants. Concentrations ranged from below detection limits to 322 ng g^{-1} wet mass in one egg for 4,4′-DDE and differed among the samples collected in the Gulf of Alaska and Bering and Chukchi Seas. Chick growth and survival rates may be affected by the contaminant levels found in the eggs, but the eggs should be safe for human consumption if they are eaten in small quantities. STAMP plans to continue collecting and banking gull eggs for future real-time and retrospective analyses.

Published by Elsevier Ltd.

1. Introduction

Gulls have been used to monitor contaminants in many parts of the world. The Canadian Great Lakes herring gull monitoring program is probably the most extensive (see Hebert et al., 1999), but the German, and Norwegian efforts are also noteworthy (see Marth et al., 2000; Borgå et al., 2001; Bustnes et al., 2003; Mallory et al., 2006; Verreault et al., 2006). In Alaska, only two studies have collected information on contaminant levels in gull eggs (Ohlendorf et al., 1982; Jack and Martinez, 2003). During 1973–1976, Ohlendorf et al. (1982) collected and analyzed glaucous-winged gull eggs from 12 locations in the Gulf of Alaska, Aleutian Islands, and southeastern Bering Sea (Copper River delta; Amalik Bay; Barren, Semidi, Shumagin Hinchinbrook, Middleton, Kodiak, Ugaiushak, Buldir, Bogoslof, and Shaiak Islands), and Jack and Martinez (2003) collected and analyzed glaucous gull eggs from the Kotzebue and Nunivak Island areas, and glaucous-winged gull eggs from the Togiak, Dutch Harbor, and Sitka vicinities in 2000.

In Alaska, glaucous gulls (*Larus hyperboreus*) nest coastally from the Beaufort Sea southward to St. Lawrence and Nunivak Islands and the Cape Peirce vicinity in the Bering Sea (ASIS, 2006a; Gilchrist, 2001, see Supplemental Fig. 1). Most of the Alaskan birds winter south of the pack ice in the Aleutian Islands and southern Bering Sea, but some individuals stay in open leads in ice-covered areas of the Chukchi and Bering Seas while others venture into the coastal waters of Russia as far south as the Kamchatka Peninsula (Troy Ecological Research Associates, 2004). Some birds also migrate along the North American coast as far south as Oregon and California. Glaucous-winged gulls (*Larus glaucescens*) breed from Cape Romanzof in western Alaska southward to Bristol Bay and the Alaska Peninsula (Verbeek, 1993). They also nest on the Pribilof and St. Matthew – Hall islands in the Bering Sea and throughout the Aleutian Islands and Gulf of Alaska. Some birds also breed on the Commander Islands in the Russian Far East and along the North American coast as far south as northwestern Oregon. Most of the Alaskan birds winter southward from the Bering Sea ice-front into the North Pacific and Gulf of Alaska, but some birds venture as far west as northern Japan and the Kurile Islands in the Russian Far East and as far south as California and the Baja Peninsula in North America (ASIS, 2006b; Verbeek, 1993; Supplemental Fig. 1).

Both gull species are surface feeders that tend to utilize nearshore environments. They are opportunistic predators and scavengers, feeding on a variety of fish and invertebrates and bird and

* Corresponding author. Tel.: +1 843 762 8994; fax: +1 843 762 8742.
E-mail address: stacy.vanderpol@nist.gov (S.S. Vander Pol).

0269-7491/$ – see front matter Published by Elsevier Ltd.
doi:10.1016/j.envpol.2008.11.026

mammal carcasses in both marine and terrestrial habitats. They also take bird eggs and chicks, and feed on human refuse and occasionally even seaweed and berries. Both species lay 2–3 eggs per clutch that may be replaced if lost early in the breeding season (Gilchrist, 2001; Verbeek, 1993). Seabird eggs have been used to monitor contaminants, and they are representative of the females at the time of laying (Verreault et al., 2006). Gulls and their eggs are still important in subsistence diets in many parts of rural Alaska.

The Seabird Tissue Archival and Monitoring Project (STAMP) was developed in 1999 as a long-term co-operative program among the U.S. Fish and Wildlife Service (USFWS), the U.S. Geological Survey (USGS), the Bureau of Indian Affairs (BIA), and the National Institute of Standards and Technology (NIST) to collect, cryogenically store (on the decadal scale), and analyze seabird tissues (primarily eggs) for chemical contaminants (e.g., polychlorinated biphenyls [PCBs], organochlorine pesticides [e.g., DDTs and chlordanes], polybrominated diphenyl ethers [BDEs], and metals and organometals [e.g., mercury, methylmercury, and butyltins]). In addition to collecting glaucous and glaucous-winged gull eggs, STAMP has also collected eggs from three other seabird species based on their feeding behavior and prey species: common and thick-billed murres (*Uria aalge* and *Uria lomvia*) and black-legged kittiwakes (*Rissa tridactyla*).

This paper reports organic contaminant levels in gull eggs collected at seven Alaskan colonies from the initial collections to serve as baseline data for STAMP and compares them with literature values. Geographic differences are also discussed, along with information related to human and environmental safety.

2. Materials and methods

2.1. Sample collection and processing

Glaucous and glaucous-winged gull clutches (1–3 eggs) were collected at seven colonies in the Bering and Chukchi seas and Gulf of Alaska in 2005 (Fig. 1) and processed at the USGS Alaska Science Center in Anchorage, Alaska using STAMP protocols (see Roseneau et al., 2008) between 16 Jun and 21 Oct 2005. Briefly, the eggs were cleaned with Type 1 water and measured (length, width, whole egg mass) before they were cut in half under a positive pressure laminar flow hood with a custom-made titanium knife. Eggs from the same clutch were pooled (Table 1). Egg shells were rinsed with Type 1 water, dried, weighed, placed in labeled plastic bags, and shipped to the University of Alaska Museum of the North in Fairbanks, Alaska for long-term storage.

The contents from each clutch were combined in a clean glass beaker and homogenized with a stainless steel kitchen hand blender (Oster 2614, Rye, New York). The blender blades and beaker were washed with soap and rinsed with Type 1 water, methanol, acetone, and hexane before they were used to process another clutch. Aliquots of the homogenized contents were put into Teflon PFA jars (Savillex, Minnetonka, Minnesota) and cryogenic polypropylene vials (Nunc International, Rochester, New York) and frozen before being shipped to NIST in liquid nitrogen vapor dry shippers. The samples were stored at −150 °C in liquid nitrogen vapor freezers at the Marine Environmental Specimen Bank (MESB) in the Hollings Marine Laboratory in Charleston, South Carolina for future analyses.

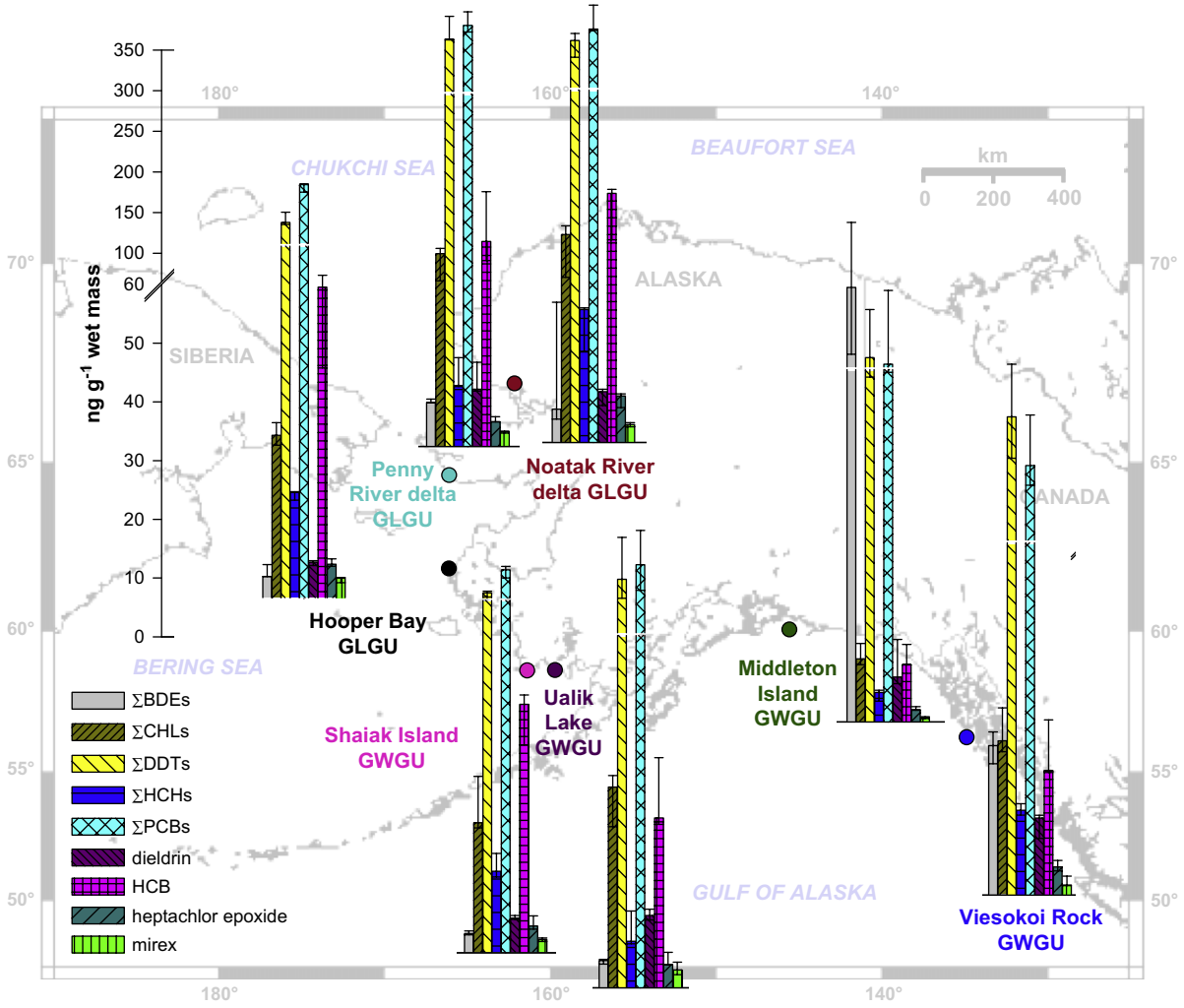

Fig. 1. Locations and organic contaminant concentrations (medians with standard errors in ng g^{-1} wet mass) in glaucous gull (GLGU) and glaucous-winged gull (GWGU) eggs collected at Alaskan colonies in 2005 ($n = 3$ clutches for each colony).

Table 1
Alaskan glaucous gull and glaucous-winged gull egg information.

Egg #	Storage ID	Field ID	Collection location	Collection date	Species	Sample notes
576	ST07E576C	TOGI02GWGU2005	Shaiak I. Bristol Bay, Bering Sea	11-May-05	*Larus glaucescens*	1 of ? eggs
578	ST07E578C	TOGI04GWGU2005	Shaiak I. Bristol Bay, Bering Sea	11-May-05	*L. glaucescens*	1 of ? eggs
579	ST07E579C	TOGI05GWGU2005	Shaiak I. Bristol Bay, Bering Sea	11-May-05	*L. glaucescens*	1 of ? eggs
611	ST07E611C	NOAT01GLGU2005	Noatak River delta, Chukchi Sea	1-Jun-05	*Larus hyperboreus*	2 of 2 eggs, both with small embryos
614	ST07E614C	NOAT04GLGU2005	Noatak River delta, Chukchi Sea	1-Jun-05	*L. hyperboreus*	2 of 2 eggs, possibly re-lays, both had some mold
616	ST07E616C	NOAT06GLGU2005	Noatak River delta, Chukchi Sea	1-Jun-05	*L. hyperboreus*	2 of 2 eggs, possibly re-lays, both had soft shells
617	ST07E617C	HOOP01GLGU2005	Hooper Bay, Bering Sea	29-May-05	*L. hyperboreus*	1 of ? eggs
619	ST07E619C	HOOP03GLGU2005	Hooper Bay, Bering Sea	29-May-05	*L. hyperboreus*	1 of ? eggs
622	ST07E622C	HOOP07GLGU2005	Hooper Bay, Bering Sea	29-May-05	*L. hyperboreus*	1 of ? eggs
641	ST07E641C	SITK01GWGU2005	Viesekoi Rocks, Gulf of Alaska	5-Jun-05	*L. glaucescens*	2 of 3 eggs, had some mold
642	ST07E642C	SITK02GWGU2005	Viesekoi Rocks, Gulf of Alaska	5-Jun-05	*L. glaucescens*	1 of 3 eggs, had some mold
644	ST07E644C	SITK04GWGU2005	Viesekoi Rocks, Gulf of Alaska	5-Jun-05	*L. glaucescens*	3 of 3 eggs, had some mold, may not be well homogenized
655	ST07E655C	UALK02GWGU2005	Ualik Lake, Dillingham, Bering Sea	12-Jun-05	*L. glaucescens*	1 of 3 eggs, had some mold
658	ST07E658C	UALK05GWGU2005	Ualik Lake, Dillingham, Bering Sea	12-Jun-05	*L. glaucescens*	1 of 3 eggs, had some mold
661	ST07E661C	UALK09GWGU2005	Ualik Lake, Dillingham, Bering Sea	12-Jun-05	*L. glaucescens*	2 of 2 eggs, had some mold
664	ST07E664C	MIDD07GWGU2005	Middleton I., Gulf of Alaska	31-May-05	*L. glaucescens*	1 of 3 eggs, had some mold
666	ST07E666C	MIDD09GWGU2005	Middleton I., Gulf of Alaska	31-May-05	*L. glaucescens*	1 of 3 eggs, had some mold
669	ST07E669C	MIDD12GWGU2005	Middleton I., Gulf of Alaska	31-May-05	*L. glaucescens*	2 of 3 eggs, had some mold
670	ST07E670C	NOME01GLGU2005	Penny River delta, Nome, Bering Sea	5-Jun-05	*L. hyperboreus*	2 of 2 eggs, had some mold
673	ST07E673C	NOME04GLGU2005	Penny River delta, Nome, Bering Sea	5-Jun-05	*L. hyperboreus*	1 of 2 eggs, had some mold
674	ST07E674C	NOME07GLGU2005	Penny River delta, Nome, Bering Sea	5-Jun-05	*L. hyperboreus*	1 of 2 eggs, had some mold

2.2. Sample preparation

Three clutch samples stored in Teflon jars in the MESB were randomly chosen from each colony (see Table 1). Approximately 3 g of material was removed from each sample, weighed on a three-place analytical balance and mixed with 8 g of diatomaceous earth that had been combusted at 650 °C for 12 h and cooled in a desiccator prior to use. The mixture was transferred to a 33 mL pressurized fluid extraction (PFE) cell (ASE Dionex, Salt Lake City, Utah) and extracted as previously described by Schantz et al. (1997). One half (0.5) mL of a mixed internal standard solution was added to the PFE cell using a gas-tight syringe that was weighed on a five-place analytical balance before and after dispensing the liquid into the cell. The internal standard solution contained ^{13}C labeled PCB congeners 28, 52, 118, 153, 180, 194, and 206; ^{13}C labeled BDE congeners 28, 47, 99, 100, 153, 154, and 209, ^{13}C labeled 4,4-DDE, 4,4′-DDT, HCB, dieldrin, oxychlordane, *trans*-chlordane, *trans*-nonachlor, deuterated 4,4′-DDD (d_8); and F-labeled BDE congener 208. From 80 ng to 250 ng of each compound was added to the samples. Aliquots of Standard Reference Material (SRM) 1946 and murre egg homogenate control material (CM; see Vander Pol et al., 2007) were prepared using the same techniques along with six external calibration solutions ranging from approximately 0.3 ng g^{-1} to 250 ng g^{-1} (BDEs ranged from 0.12 ng g^{-1} to 28 ng g^{-1}) and a method blank. The calibration solutions contained SRMs 2261 Chlorinated Pesticides in Hexane, 2262 Chlorinated Biphenyl Congeners in Isooctane, 2274 PCB Congener Solution-II in Isooctane, 2275 Chlorinated Pesticide Solution-II in Hexane, PCB Congener Solution-III (containing 15 PCB congeners), and PCB Congener Solution-IV (containing 31 PCB congeners and pentachlorobenzene), BDE Congener Solution (containing 26 BDE congeners), and octachlorostyrene (AccuStandard, New Haven, CT).

Following extraction, samples were reduced in volume, an aliquot was removed for lipid analysis, high molecular mass compounds were removed by size exclusion chromatography (SEC) and further clean-up was conducted using solid phase extraction (SPE) as described previously (Litz et al., 2007).

2.3. Sample analysis

Samples were analyzed using an electron impact (EI) gas chromatography/mass spectrometry (GC/MS) instrument (Agilent 6890N/5973, Palo Alto, California) operated in the selected ion monitoring mode (SIM) for most of the chlorinated pesticides and all of the PCB and BDE congeners. The instrument was equipped with a 60 m × 0.25 mm × 0.25 μm i.d. DB-5MS column (J&W Scientific, Folsom, California) with a 5 m × 0.25 mm retention gap added to the beginning of the column. A PTV injector (Agilent 6850) was used to introduce the sample. Liquid nitrogen vapor at 84.0 mL/min was used to cool the inlet to 10 °C for 1.6 min during the injection of 20 μL (4 × 5 μL) of the sample onto the column. The inlet was then heated at 720° C/min to the final transfer temperature of 250 °C with no hold time. The vent flow was 65.0 mL/min at 0 kPa until 1.5 min, purged at 100.0 mL/min at 2.30 min, and then conditions were 1.2 mL/min at 167 kPa. The GC oven was held at 100 °C for 1.5 min, ramped to 150 °C at 25 °C/min, ramped to 200 °C at 0.75 °C/min, and then ramped to 300 °C at 3 °C/min and held isothermally for 27 min (135.5 min total run time). Helium was the carrier gas set a constant flow of 1.2 mL/min. Selected organochlorine pesticides were analyzed by GC/MS in the negative ion (NCI) mode using SIM equipped with a 30 m × 0.18 mm × 0.18 μm i.d. DB-5MS column (J&W Scientific). Methane was used as the reaction gas. The samples were injected using the PTV as detailed above. The GC oven was held at 80 °C for 1.5 min, ramped to 170 °C at 25 °C/min, ramped to 250 °C at 2 °C/min, and then ramped to 325 °C at 25 °C/min and held isothermally for 10 min (58.1 min total run time). Data were quantified by using at least three calibration points and allowing the intercept to float.

2.4. Statistics

Multivariate Analysis of Variance (MANOVA) was run on the normally distributed lipid-adjusted compound classes by region and species because degrees of freedom were constrained. Compounds included ΣPCBs, ΣBDEs, ΣDDTs, ΣHCHs, Σchlordanes, heptachlor epoxide, HCB, dieldrin, and mirex. If differences were significant ($P < 0.05$), individual ANOVAs and Tukey–Kramer post-hoc tests were used to identify the compounds and species that were different. To help visualize the results, a principal components analysis using a correlation matrix was also run on a lipid mass basis for the percentages of total compounds with no samples that fell below detection limits (36 PCB congeners, 14 organochlorine compounds, and 5 BDE congeners). Statistical tests were conducted using commercially available software (SAS Institute, JMP 3.26, Cary, North Carolina). Because of human consumption concerns, acceptable/tolerable daily intake (ADI/TDI) values (number of eggs day^{-1}) for a 70 kg person were calculated using the following formula: ADI/TDI for the contaminant (μg kg^{-1} body weight day^{-1}) from Van Oostdam et al. (1999) × 70 kg × contaminant concentration in the egg^{-1} (μg g^{-1}) × egg mass^{-1} (g; mass was averaged for eggs belonging to multiple egg clutches).

2.5. Literature comparisons

The literature was searched for persistent pollutant data on glaucous and glaucous-winged gulls. If necessary, values were converted to ng g^{-1} wet mass by using stated percent lipid. Data were then organized by contaminant, region, and year of collection. Information from similar regions and years were sometimes combined by taking the central tendency (means or medians as reported) multiplied by the number of samples and dividing by the total number of samples for an arithmetic mean. If not given, ranges were calculated by multiplying the standard deviation by 3 which covers 99% of the values based on a Gaussian distribution. If data were presented with 95% confidence intervals or standard errors, the values were converted to standard deviations before calculating ranges. These conversions were only made to help provide more accurate visual comparisons in large sets of data.

3. Results and discussion

3.1. Contaminant concentrations

Contaminant concentrations in the eggs varied from below detection limits (0.1 ng g^{-1} wet mass) to 322 ng g^{-1} wet mass for 4,4′-DDE in one egg from Viesokoi Rock near Sitka in the southeastern Gulf of Alaska (the mean relative standard deviation within a colony was 45.6%; see Fig. 1 and Supplemental Tables 1–3). All

Table 2
Contaminant concentrations (medians in ng g^{-1} lipid mass with ranges shown in parentheses) in glaucous gull (GLGU) and glaucous-winged gull (GWGU) eggs from Alaska ($n = 3$ clutches for each colony). ANOVA F ratios and probabilities are shown following significant MANOVAs (Wilks' $\lambda = 0.0326$, approximate $F_{27,26.9} = 2.25$, $P = 0.0212$). Groups with different letters were significantly different based on Tukey–Kramer HSD post-hoc tests (e.g., for ΣHCHs, Chukchi Sea GLGU eggs (A) were significantly different than Gulf of Alaska GWGU eggs (B), but Bering Sea eggs (AB) were not different from either colony). Percent lipid values and statistics are shown for reference only.

Compound	Chuckchi Sea	Bering Sea		Gulf of Alaska	F Ratio
	GLGU	GLGU	GWGU	GWGU	Probability
Percent	7.96	8.02	7.13	7.70	0.640
Lipid	(7.24–8.34)	(5.90–10.2)	(5.70–8.23)	(7.00–8.84)	0.60
ΣBDEs	76.8 (59.5–470)	92.8 (51.1–142)	61.4 (47.2–99.2)	348 (243–4130)	2.49 0.096
ΣCHLs	330 (279–408)	287 (253–452)	308 (225–530)	180 (96.2–325)	2.62 0.084
ΣDDTs	1470 (1280–1910)	1590 (1040–2020)	1060 (893–3780)	1960 (594–3700)	0.357 0.79
ΣHCHs	278^{A} (194–313)	215^{AB} (98.3–274)	184^{AB} (85.6–243)	116^{B} (33.0–221)	3.40 0.0418*
ΣPCBs	1720 (1620–2500)	1690 (1580–2280)	1400 (1130–3870)	1710 (700–2920)	0.0345 0.99
Dieldrin	104 (78.8–130)	115 (60.2–164)	118 (78.4–247)	134 (78.0–221)	0.443 0.73
HCB	524^{A} (433–585)	588^{A} (342–758)	547^{A} (326–622)	216^{B} (133–388)	8.49 0.0011*
Heptachlor Epoxide	94.8^{A} (74.4–119)	76.3^{AB} (40.8–88.2)	67.0^{AB} (46.3–98.2)	42.3^{B} (25.1–74.8)	4.48 0.0172*
Mirex	36.2 (33.3–49.6)	38.4 (25.7–43.5)	33.3 (19.0–92.0)	14.8 (5.96–45.3)	1.95 0.16

values fell within the range reported for murre eggs from the same regions (Roseneau et al., 2008; Vander Pol et al., 2004), and lipids ranged from 5.7% to 10.2% and did not differ among colonies (Table 2 and Supplemental Table 1). To meet normality assumptions, all tests were run on a lipid mass basis. The values generated on the reference materials fell within previously reported ranges indicating that the analyses were in control.

3.2. Geographical and literature comparisons

The MANOVA comparing regions and species was significantly different (Wilks' $\lambda = 0.0326$, approximate $F_{27,26.9} = 2.25$, $P = 0.0212$). Eggs from glaucous-winged gull colonies in the Gulf of Alaska contained lower levels of ΣHCHs, HCB, and heptachlor epoxide (Table 2) than eggs from the Bering Sea, which was

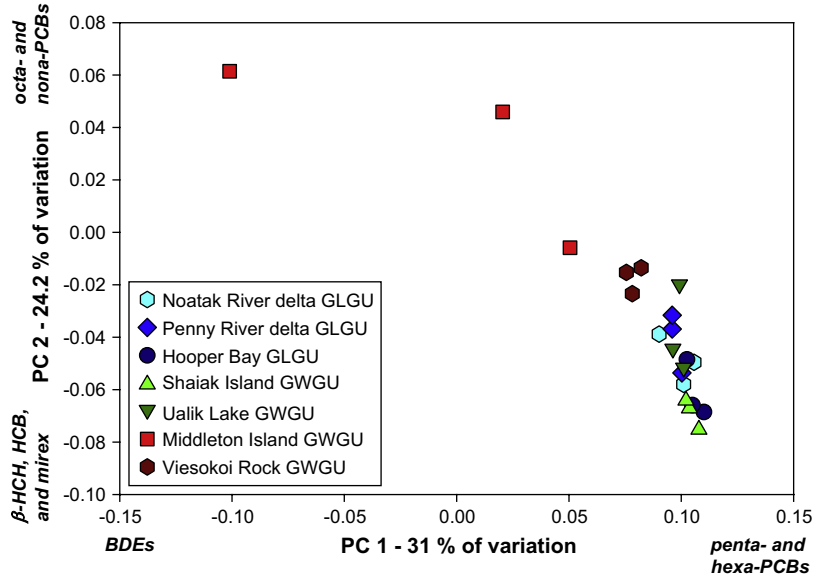

Fig. 2. Principal components analysis of Alaskan glaucous gull (GLGU) and glaucous-winged gull (GWGU) eggs collected at Alaskan colonies in 2005. Compounds contributing to the loadings are shown along the axes.

consistent with what is known about global organic contaminant transport (Shen and Wania, 2005; Wania and Dugani, 2003; Wania, 2006). The other compounds were not significantly different (Table 2). Murre eggs collected in the same regions only followed this pattern for HCB. The concentration levels of DDE and ΣPCBs in the murre eggs were higher in the Gulf of Alaska than in the Bering Sea (see Table 2 and Vander Pol et al., 2004).

The principal components analysis did not reveal any differences in contaminant patterns based on species (Fig. 2). However, the Middleton Island glaucous-winged gull eggs were distinct from the other colonies because of their higher proportions of BDEs and higher chlorinated PCBs. Also, the eggs from Viesokoi Rock near Sitka showed a slight amount of separation (see Fig. 2), something that was also noted in murre eggs from this same region (Vander Pol et al., 2004; Roseneau et al., 2008). This suggests a possible difference in regional contaminant levels. In comparison, a Norwegian gull study concluded that geographical contaminant patterns were more dependent on biomagnification than on global distillation/fractionation theory (Steffen et al., 2006). A stable isotope study of the Alaska gull eggs is underway that will help determine if there are trophic differences among individuals and colonies.

More data are needed to explain the variations in BDE levels found in the Middleton Island gull eggs (Fig. 1 and Supplemental Table 3). The congener patterns of BDEs within the eggs were also variable (Fig. 3 and Supplemental Table 3), with BDE 47 standing out as the predominant congener. The exceptions were glaucous-winged gull eggs from Shaiak Island near the entrance to Bristol Bay in the Bering Sea where BDE 154 was the dominate substance, and egg 619 from Hooper Bay in western Alaska where BDE 100 comprised 57.7% of the total BDEs. BDE 47 was also the major flame retardant (44.7% of the total) found in Norwegian glaucous gull eggs (Verreault et al., 2007). However the order of the other BDEs reported in the Norwegian study was congeners 100, 154 (with co-elution of polybrominated biphenyl [BB] congener 153), 99, and 153 (5–10% each) compared to the importance of the congeners found in this study (congeners 99 [5–40%], 154 [2–27%], 100 [7–58%], and 49 [0.2–21%]; see Fig. 3 and Verreault et al., 2007). The large range in the percentages of the major BDEs found during this study demonstrated that the pattern in congeners was highly variable among both the colonies and individuals with a mean relative standard deviation (RSD) of 52.1% for the compounds found at the colonies (glaucous gull eggs from the Penny River delta had consistent BDE congener patterns with a mean RSD of

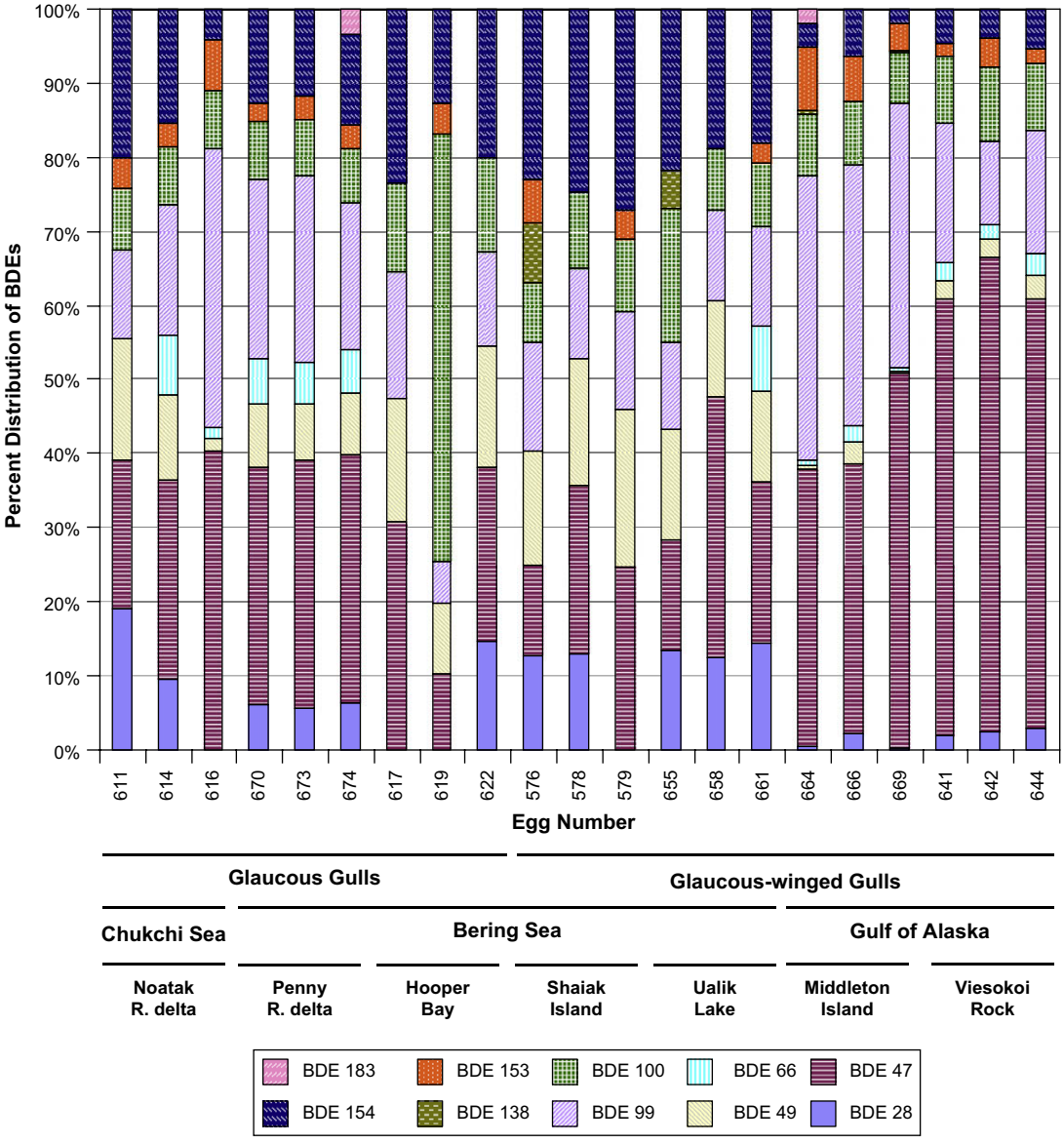

Fig. 3. Percentage distribution of brominated diphenyl ether congeners in glaucous and glaucous-winged gull eggs collected at Alaskan colonies in 2005.

7.2%; see Fig. 3). Because gulls are opportunistic predators and scavengers, their diet includes a broad range of items that vary from berries and fish to invertebrates, marine mammal carcasses and human refuse in landfills, which markedly increases their exposure to BDEs. Feeding on refuse in dumps may help explain the high variation in BDE concentrations and congener patterns. ΣBDE levels at Middleton Island were 162, 21, and 302 ng g^{-1} wet mass in eggs 664, 666, 669, respectively, compared to 3.3–34 ng g^{-1} wet mass in all of the other eggs analyzed during this study. Middleton Island supported an active Civil Aeronautics Authority (CAA) and Federal

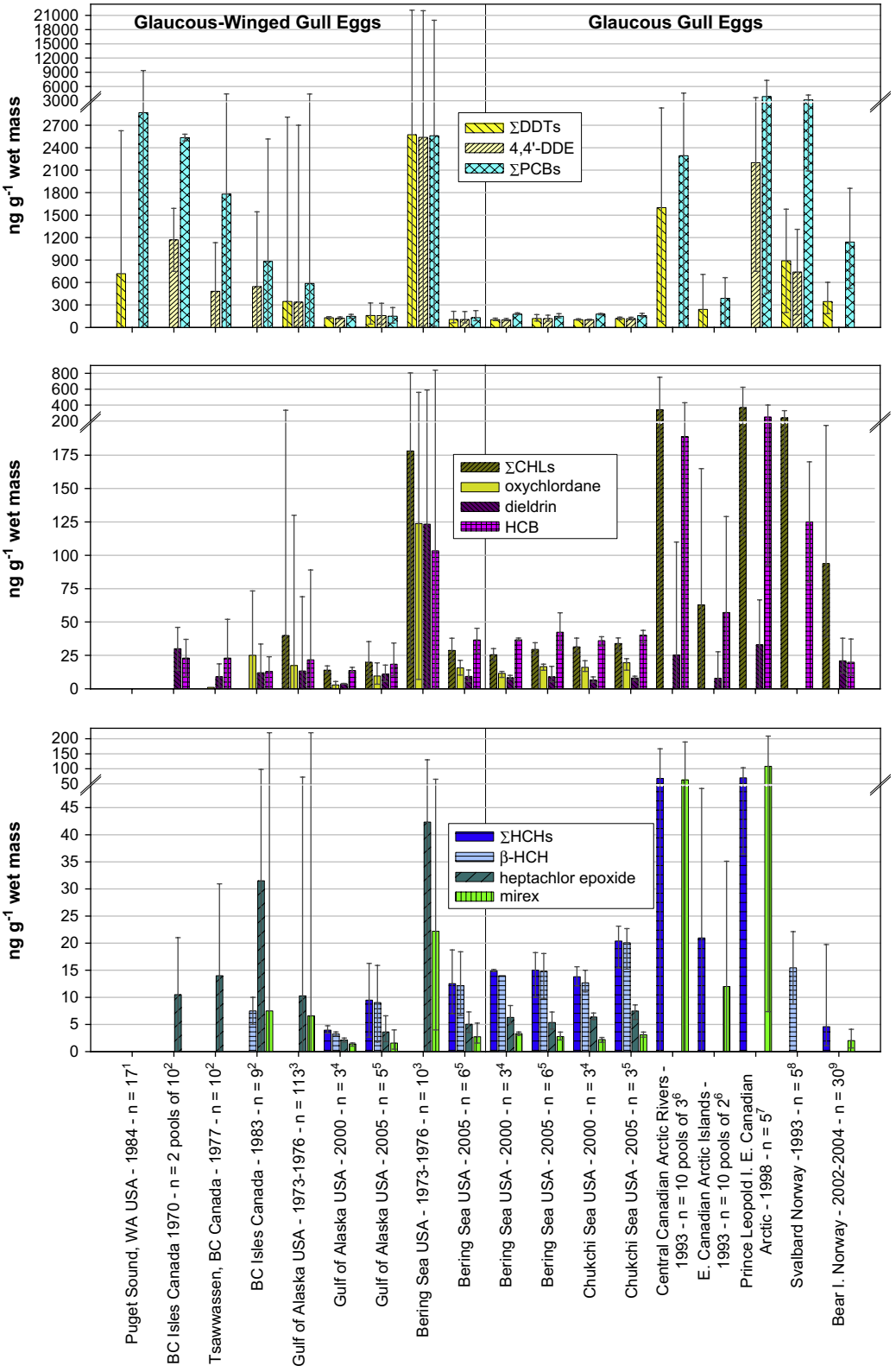

Fig. 4. Contaminant concentrations (means with ranges in ng g^{-1} wet mass) for glaucous gull and glaucous-winged gull eggs from the literature compared to the results of this study ([1]Speich et al., 1992, [2]Elliott et al., 1989, [3]Ohlendorf et al., 1982, [4]Jack and Martinez, 2003, [5]this study, [6]Braune et al., 2002, [7]1999; [8]Barrett et al., 1996; [9]Verreault et al., 2005).

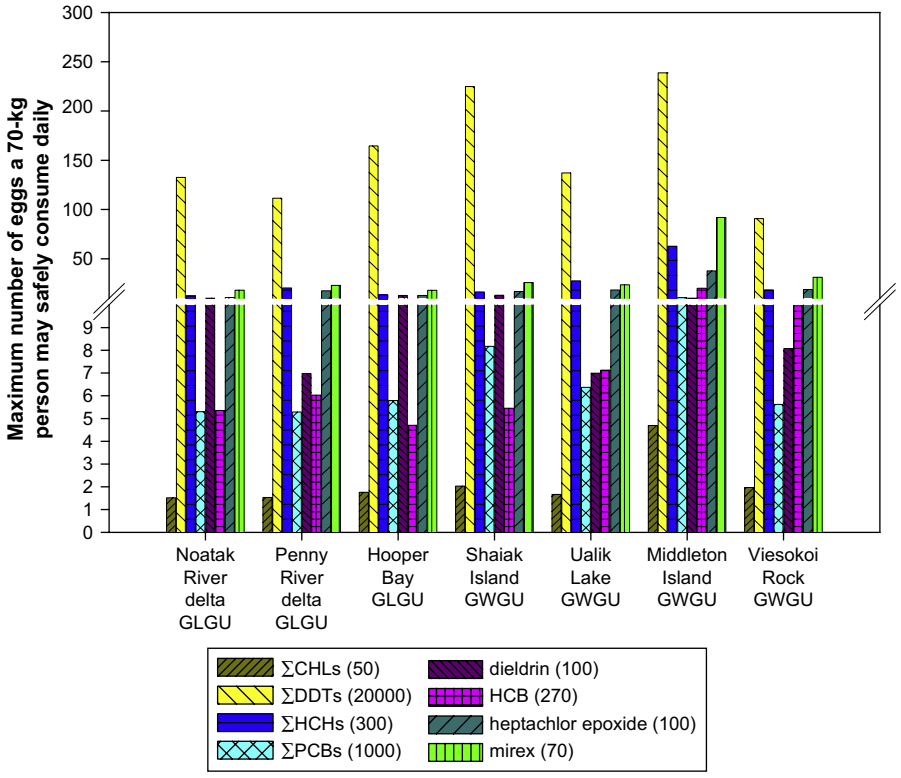

Fig. 5. Maximum number of glaucous gull (GLGU) and glaucous-winged gull (GWGU) eggs from Alaskan colonies that may be safely eaten by a 50-kg person based on the Canadian Acceptable/Tolerable Daily Intakes (ADI/TDI). Numbers in parentheses are ADI/TDIs for each contaminant in ng kg^{-1} d^{-1} (see Van Oostdam et al., 1999).

Aviation Agency (FAA) Flight Service Station during the 1940s–1970s (the CAA was renamed the FAA in 1958). It also supported an active U.S. Air Force Aerospace Control and Warning Site (ACWS) from 1958 to 1963 and an active White Alice Communications Site (WACS) from 1956 to 1985. Since the mid-1980s, an automated National Weather Service weather radar and FAA flight service facility has been located on the island that is intermittently maintained by personnel that commute to the site from Anchorage. A small team of USGS scientists has also visited the island almost every summer since the late 1970s to study seabirds. Although current anthropogenic influences are minimal, some of the historical operations may still play roles in exposure to some types of contaminants (e.g., BDEs and PCBs leaching out of old dump sites) and a recent study has shown the impacts of research stations and BDE contamination of the local ecosystem in Antarctica (Hale et al., 2008).

Also, contaminant patterns may have varied among first-, second-, and third-laid eggs (see Verreault et al., 2006), and this factor may help explain some of the Middleton Island variation because two of the samples were from single eggs belonging to three-egg clutches (see Figs. 2 and 3 and Table 1). However, if this factor played a role, greater variation should have occurred at several of the other colonies where only single eggs from multi-egg clutches were analyzed (the other eggs belonging to these clutches were broken in transit).

PCB, DDT, and mirex levels in the eggs were generally lower than those found in the literature (Fig. 4). HCB was generally higher than the literature values with the exception of the Canadian gull eggs. Other contaminant concentrations were similar to reported levels, including those found during the most recent Alaskan gull study (Jack and Martinez, 2003).

3.3. Human consumption and environmental safety

In general, the gull eggs are safe for daily consumption based on the levels of organic contaminants observed in this study (Fig. 5). Σchlordanes are of the most concern because in most cases a 70 kg person can only eat one to two eggs per day (range 1.5–4.7) before they exceed the recommended Canadian Acceptable/Tolerable Daily Intake level (see Van Oostdam et al., 1999). Based on these same standards, a 70 kg person can consume at least 4 eggs per day before they exceed the recommended levels for the other contaminant groups. In comparison to other potential marine subsistence foods in Alaska, gull eggs are at least an order of magnitude greater in concentration than salmon (*Oncorhynchus* spp.; ADEC, 2008), similar to two times greater than walleye pollock (*Theragra chalcogramma*; de Brito et al., 2002), and similar to an order of magnitude lower than walrus (*Odobenus rosmarus*) and ringed seal (*Phoca hispida*) blubber (Kucklick et al., 2006).

Estimating environmentally safe consumption for individuals, populations, and predators is always difficult for contaminants. Few toxicological data exist for most species and contaminants are usually tested alone or in combination with only a few others in spite of the possible synergistic effects of large mixtures. Bustnes et al. (2003) conducted a three-year glaucous gull study that correlated male and female blood contaminant levels with survival and fitness endpoints. Using this data, egg burdens were estimated by multiplying the ratio of the egg concentrations to the male or female plasma concentrations (after converting them to wet mass based on the percent lipids) reported from the same colonies by Verreault et al. (2005) and these values were compared with the current results (see Supplemental Table 4). Given that laying dates appear to be negatively correlated with HCB, β-HCH, oxychlordane, DDE, and persistent PCBs (see Bustnes et al., 2003), the body mass and survival of the chicks, if they had been allowed to live, might have been lower than normal.

4. Conclusions

Contaminant levels in the gull eggs analyzed during this study were similar to murre egg values reported from these same regions,

and based on this information, they appear to be safe for humans to eat in small quantities, and the local public health authorities may be contacted for more personalized assessments. It is possible that chick growth and survival rates may have been affected by the contaminant concentrations. Some geographical separation was evident between the Chukchi/Bering sea and Gulf of Alaska colonies. More information is needed to help understand the large variations in contaminant levels found in the gull eggs from Middleton Island. STAMP plans to continue collecting and banking eggs from both gull species to monitor long-term trends and conduct retrospective analyses.

5. Disclaimer

Any mention of commercial products is for information only; it does not imply recommendation or endorsement by NIST.

Acknowledgements

We would like to thank Austin Ahmasuk, D. Doderer, Peter Lockuk Sr., Jack Lorrigan, Rob MacDonald, Enoch Shiedt, Albert Simon, and A. Ramey for collecting the eggs, and Kristin Simac and Lesley Chappel for helping to process them. We would also like to thank Glenn Chen of the Bureau of Indian Affairs and the North Pacific Research Board for providing supplemental funding and support that allowed STAMP to conduct some of this work (NPRB Project 0534). This paper is NPRB publication No. 192.

Appendix. Supplementary material

Supplementary data associated with this article can be found in the online version, at doi:10.1016/j.envpol.2008.11.026.

References

ADEC (Alaska Department of Environmental Conservation), 2008. Alaska Department of Environmental Conservation Fish Monitoring Program: analysis of organic contaminants, p. 31. http://www.dec.state.ak.us/eh/docs/vet/FMP%20Organic%20data%206-08.pdf (accessed 8 Nov 2008).

ASIS (Alaska Seabird Information Series), 2006a. Glaucous gull. U.S. Fish and Wildlife Service Migratory Bird Management Report. U.S. Fish and Wildlife Service, Anchorage, Alaska.

ASIS (Alaska Seabird Information Series), 2006b. Glaucous-winged gull. U.S. Fish and Wildlife Service Migratory Bird Management Report. U.S. Fish and Wildlife Service, Anchorage, Alaska.

Barrett, R.T., Skaare, J.U., Gabrielsen, G.W., 1996. Recent changes in levels of persistent organochlorines and mercury in eggs of seabirds from the Barents Sea. Environmental Pollution 92 (1), 13–18.

Borgå, K., Gabrielsen, G.W., Skaare, J.U., 2001. Biomagnification of organochlorines along a Barents Sea food chain. Environmental Pollution 113 (2), 187–198.

Braune, B.M., Wakeford, B., Gaston, A., 1999. Contaminants in Arctic seabird eggs. Synopsis of Research Conducted Under the Northern Contaminants Program 1998–1999. Department of Indian Affairs and Northern Development, Ottawa, Canada. pp. 77–80.

Braune, B.M., Donaldson, G.M., Hobson, K.A., 2002. Contaminant residues in seabird eggs from the Canadian Arctic. II. Spatial trends and evidence from stable isotopes for intercolony differences. Environmental Pollution 117 (1), 133–145.

de Brito, A.P.X., Ueno, D., Takahasi, S., Tanabe, S., 2002. Organochlorine and butyltin residues in walleye pollock (*Theragra chalcogramma*) from Bering Sea, Gulf of Alaska and Japan Sea. Chemosphere 46, 401–411.

Bustnes, J.O., Erikstad, K.E., Skaare, J.U., Bakken, V., Mehlum, F., 2003. Ecological effects of organochlorine pollutants in the arctic: a study of the glaucous gull. Ecological Applications 13 (2), 504–514.

Elliott, J.E., Noble, D.G., Norstrom, R.J., Whitehead, P.E., 1989. Organochlorine contaminants in seabird eggs from the pacific coast of Canada, 1971-1986. Environmental Monitoring and Assessment 12 (1), 67–82.

Gilchrist, H.G., 2001. Glaucous gull (*Larus hyperboreus*). In: Poole, A., Gill, F. (Eds.), The Birds of North America. The Birds of North America, Inc., Philadelphia, PA No. 573.

Hale, R.C., Kim, S.L., Harvey, E., La Guardia, M.J., Mainor, T.M., Bush, E.O., Jacobs, E.M., 2008. Antarctic research bases: local sources of polybrominated diphenyl ether (PBDE) flame retardants. Environmental Science and Technology 42 (5), 1452–1457.

Hebert, C.E., Norstrom, R.J., Weseloh, D.V.C., 1999. A quarter century of environmental surveillance: the Canadian Wildlife Service's Great Lakes herring gull monitoring program. Environmental Reviews 7 (4), 147–166.

Jack, L., Martinez, D., 2003. Design & field test a prototype for a PBT screening tool for a selected Alaska tribal subsistence food. The Alaska Sea Otter and Sea Lion Commission, Anchorage, AK, TASSC Report 03-01.

Kucklick, J.R., Krahn, M.M., Becker, P.R., Porter, B.J., Schantz, M.M., York, G.S., O'Hara, T.M., Wise, S.A., 2006. Persistent organic pollutants in Alaskan ringed seal (*Phoca hispida*) and walrus (*Odobenus rosmarus*) blubber. Journal of Environmental Monitoring 8, 848–854.

Litz, J.A., Garrison, L.P., Fieber, L.A., Martinez, A., Contillo, J.P., Kucklick, J.R., 2007. Fine-scale spatial variation of persistent organic pollutants in bottlenose dolphins (*Tursiops truncatus*) in Biscayne Bay, Florida. Environmental Science and Technology 41 (21), 7222–7228.

Mallory, M.L., Gilchrist, H.G., Braune, B.M., Gaston, A.G., 2006. Marine birds as indicators of Arctic marine ecosystem health: linking the northern ecosystem initiative to long-term studies. Environmental Monitoring and Assessment 113 (1–3), 31–48.

Marth, P., Martens, D., Schramm, K.-W., Schmitzer, J., Oxynos, K., Kettrup, A., 2000. Environmental specimen banking. Herring gull eggs and breams as bioindicators for monitoring long-term and spatial trends of chlorinated hydrocarbons. Pure and Applied Chemistry 72 (6), 1027–1034.

Ohlendorf, H.M., Bartonek, J.C., Divoky, G.J., Klaas, E.E., Krynitsky, A.J., 1982. Organochlorine residues in eggs of Alaskan seabirds. United States Department of the Interior, Fish and Wildlife Service, Washington, D.C. Special Scientific Report – Wildlife No. 245.

Roseneau, D.G., Becker, P.R., York, G.S., Vander Pol, S.S., Day, R.D., Point, D., Simac, K.S., Moors, A.J., Ellisor, M.B., Pugh, R.S., 2008. Expanding the Seabird Tissue Archival and Monitoring Project (STAMP) in the North Pacific: geographic patterns in contaminant residues in seabird eggs used in rural subsistence diets. North Pacific Research Board Final Report Project 0534.

Schantz, M.M., Nichols, J.J., Wise, S.A., 1997. Evaluation of pressurized fluid extraction for the extraction of environmental matrix reference materials. Analytical Chemistry 69 (20), 4210–4219.

Shen, L., Wania, F., 2005. Compilation, evaluation, and selection of physical-chemical property data for organochlorine pesticides. Journal of Chemical and Engineering Data 50 (3), 742–768.

Speich, S.M., Calambokidas, J., Shea, D.W., Peard, J., Witter, M., Fry, M., 1992. Eggshell thinning and organochlorine contaminants in western Washington waterbirds. Colonial Waterbirds 15 (1), 103–112.

Steffen, C., Borgå, K., Skaare, J., Bustness, J.O., 2006. The occurrence of organochlorines in marine avian top predators along a latitudinal gradient. Environmental Science and Technology 40 (16), 5139–5146.

Troy Ecological Research Associates, 2004. Movements of glaucous gull trapped at the Barrow landfill. Results from a 2003 Pilot Study. Troy Ecological Research Associates, Anchorage, Alaska.

Van Oostdam, J., Gilman, A., Dewailly, E., Usher, P., Wheatley, B., Kuhnlein, H., Neve, S., Walker, J., Tracy, B., Feeley, M., Jerome, V., Kwavnick, B., 1999. Human health implications of environmental contaminants in Arctic Canada: a review. Science of the Total Environment 230 (1–3), 1–82.

Vander Pol, S.S., Becker, P.R., Kucklick, J.R., Pugh, R.S., Roseneau, D.G., Simac, K.S., 2004. Persistent organic pollutants in Alaskan murre (*Uria* spp.) eggs: Geographical, species, and temporal comparisons. Environmental Science and Technology 38 (5), 1305–1312.

Vander Pol, S.S., Ellisor, M.B., Pugh, R.S., Becker, P.R., Poster, D.L., Schantz, M.M., Leigh, S.D., Wakeford, B.J., Roseneau, D.G., Simac, K.S., 2007. Development of a murre (*Uria* spp.) egg control material. Analytical & Bioanalytical Chemistry 387 (7), 2357–2363.

Verbeek, N.A.M., 1993. Glaucous-winged gull (*Larus glaucescens*). No. 59. In: Poole, A., Gill, F. (Eds.), The Birds of North America. The Academy of Natural Sciences, Philadelphia The American Ornithologists Union, Washington, D.C.

Verreault, J., Letcher, R.J., Muir, D.C.G., Cuh, S., Gebbink, W.A., Gabrielsen, G.W., 2005. New organochlorine contaminants and metabolites in plasma and eggs of glaucous gulls (*Larus hyperboreus*) from the Norwegian Arctic. Environmental Toxicology and Chemistry 24 (10), 2486–2499.

Verreault, J., Villa, R.A., Gabrielsen, G.W., Skaare, J.U., Letcher, R.J., 2006. Maternal transfer of organohalogen contaminants and metabolites to eggs of Arctic-breeding glaucous gulls. Environmental Pollution 144 (3), 1053–1060.

Verreault, J., Gebbink, W.A., Gauthier, L.T., Gabrielsen, G.W., Letcher, R.J., 2007. Brominated flame retardants in glaucous gulls from the Norwegian arctic: more than just an issue of polybrominated diphenyl ethers. Environmental Science and Technology 41 (14), 4925–4931.

Wania, F., 2006. Potential of degradable organic chemicals for absolute and relative enrichment in the Arctic. Environmental Science and Technology 40 (2), 569–577.

Wania, F., Dugani, C.B., 2003. Assessing the long-range transport potential of polybrominated diphenyl ethers: a comparison of four multimedia models. Environmental Toxicology and Chemistry 22 (6), 1252–1261.