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CONTAMINANTS OF EMERGING CONCERN IN FISH FROM WESTERN U.S. AND ALASKAN NATIONAL PARKS — SPATIAL DISTRIBUTION AND HEALTH THRESHOLDS[1]

Colleen M. Flanagan Pritz, Jill E. Schrlau, Staci L. Massey Simonich, and Tamara F. Blett[2]

ABSTRACT: Remote national parks of the western U.S. and Alaska are not immune to contaminants of emerging concern. Semivolatile organic compounds (SOCs) such as pesticides and PCBs can selectively deposit from the atmosphere at higher rates in cold, high-elevation and high-latitude sites, potentially increasing risk to these ecosystems. In the environment, SOCs magnify up food chains and are known to increase health risks such as cancer and reproductive impairment. One hundred twenty-eight fish in 8 national parks in Alaska and the western U.S. were analyzed for contaminant concentrations, assessed by region, and compared to human and wildlife health thresholds. SOC concentrations from an additional 133 fish from a previous study were also included, for a total of 31 water bodies sampled. PCBs, endosulfan sulfate, and *p*,*p*′-DDE were among the most frequently detected contaminants. Concentrations of historic-use pesticides dieldrin, *p*,*p*′-DDE, and/or chlordanes in fish exceeded USEPA guidelines for human subsistence fish consumers and wildlife (kingfisher) health thresholds at 13 of 14 parks. Average concentrations in fish ranged from 0.6-280 ng/g lipid (0.02-7.3 μg/g ww). Contaminant loading was highest in fish from *Alaskan* and *Sierra Nevada* parks. Historic compounds were highest in *Alaskan* parks, while current-use pesticides were higher in the *Rockies* and *Sierra Nevada*. This study provides a rigorous analysis of CECs in fish from national parks and identifies regions at potential risk.

(KEY TERMS: semivolatile organic compounds; national parks; fish; consumption thresholds.)

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INTRODUCTION

Contaminants of emerging concern (CECs) include pollutants that have only been recently detected in the environment, as well as pollutants that have been measured at concentrations that have the potential to cause harmful effects in humans and wildlife (e.g., Battaglin *et al.*, 2007). Limited research on the presence and effects of contaminants in specific geographic regions also warrants classification of contaminants as CECs. CECs include a wide range of chemical classes such as pharmaceuticals, personal care products, and perfluorinated compounds. The focus of this research, semivolatile organic compounds (SOCs), comprise another suite of CECs and include historic- and current-use pesticides, industrial pollutants such as polychlorinated biphenyls

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(PCBs), and combustion by-products like polycyclic aromatic hydrocarbons (PAHs). SOCs have the potential to undergo regional and long-range transport followed by deposition to remote ecosystems.

Compounds such as pesticides, PCBs, and PAHs may not be emerging, but according to Landewe (2008), "the concern about the environmental impact of these compounds, some of which have been in existence for quite some time, is emerging" (in specific geographic regions). Daughton (2003) noted that the term "emerging" refers to old pollutants with new concerns and new pollutants with unknown issues. Although the presence of SOCs in fish has been observed in high-elevation and high-latitudinal areas across the Tibetan Plateau, European Alps, Pyrenees, Canadian Rockies, Sierra Nevada, Aleutian Islands, Arctic, and Yukon lakes (Allen-Gil *et al.*, 1997; Ohyama *et al.*, 2004; Vives *et al.*, 2004; Ryan *et al.*, 2005; Blais *et al.*, 2006; Gallego *et al.*, 2007; Ackerman *et al.*, 2008; Hardell *et al.*, 2010; Yang *et al.*, 2010), there are very limited data on the risks, if any, associated with the accumulation of SOCs in fish from remote regions.

Semivolatile organic compounds bioaccumulate and magnify within food chains, potentially threatening both human and wildlife health. Some SOCs are endocrine-disrupting compounds (EDCs), impairing reproductive function and the control of hormones that regulate growth and development (Colborn *et al.*, 1993; Longnecker *et al.*, 1997; Carpenter, 2006; Battaglin *et al.*, 2007). EDCs in fish are known to induce skewed sex ratios and intersex characteristics, degraded predator avoidance behavior, and cause reproductive failure and population collapse in sensitive fish species (Kidd *et al.*, 2007; Vajda *et al.*, 2008; McGee *et al.*, 2009; Painter *et al.*, 2009). Other endocrine effects in wildlife that have been measured include abnormal thyroid function, decreased fertility, reduced avian hatchling success, male feminization, and alteration of immune function (Colborn *et al.*, 1993; Jones and de Voogt, 1999; Iwanowicz and Ottinger, 2009). In addition, many SOCs are known or suspected carcinogens, or have been implicated in various chronic diseases, reduced IQ, altered behavior, impaired reproductive function, or an increased risk of developing cardiovascular and liver disease and diabetes (Colborn *et al.*, 1993; Longnecker *et al.*, 1997; Carpenter, 2006).

The Western Airborne Contaminants Assessment Project (WACAP) found SOCs in fish across high-elevation and high-latitude national parks in the western United States (U.S.) and Alaska, and assessed the potential ecological risks these contaminants may pose. The concentration of some contaminants — specifically dieldrin, *p,p′*-dichlorodiphenylethene (*p,p′*-DDE; a degradation product of DDT often found in fish), and mercury — in some fish exceeded thresholds established for human and wildlife health (Ackerman *et al.*, 2008; Schwindt *et al.*, 2008; Landers *et al.*, 2010). Given the remote nature of the study sites, the contaminant burden in fish was attributed to atmospheric sources. Pesticides and other SOCs have been documented in snowpack profiles from many of the same western U.S. and Alaskan national parks (Hageman *et al.*, 2006, 2010), supporting the case for atmospheric transport and deposition of SOCs to the study locations.

High-elevation and high-latitude areas are particularly susceptible to the atmospheric transport and deposition of SOCs due to the physical-chemical properties of SOCs, including vapor pressure, octanol-water partition coefficient (K_{OW}), and octanol-air partition coefficient (K_{OA}). These factors result in selective deposition of SOCs from the atmosphere at higher rates in cold, mountainous, and circumpolar regions (Wania and MacKay, 1993; Simonich and Hites, 1995; Wania *et al.*, 1998; Daly and Wania, 2005). These properties also influence what compounds are most prone to volatilization, transport, deposition, and accumulation in biological media (Schrlau *et al.*, 2011). Elevation (Blais *et al.*, 2003, 2006; Hageman *et al.*, 2006; Demers *et al.*, 2007; Landers *et al.*, 2010; Bradford *et al.*, 2013), proximity to source region (Hageman *et al.*, 2010), and input from bio-vectors like anadromous fish (Ewald *et al.*, 1998; Krummel *et al.*, 2005) are other factors known to affect contaminant accumulation in the ecosystem.

Fish in these remote areas are particularly at risk to contaminants not only because of the potentially harmful effects of bioaccumulation but also because these regions support fish with higher lipid storages and longer lives (Blais *et al.*, 2003). Along with the slower growth of aquatic species and a shorter growing season, these factors may exacerbate contaminant accumulation and heighten ecosystem sensitivity in high-elevation and high-latitude areas. Older fish are particularly at risk given the increased susceptibility for contaminant bioaccumulation over a long time period (Scheuhammer *et al.*, 1998; Rose *et al.*, 1999; Schwindt *et al.*, 2008). Trophic level of fish and length of food chain also influence variation among sites; for example, predatory fish are more likely to have elevated contaminant concentrations due to biomagnification within the food web (Scheuhammer *et al.*, 1998; Rose *et al.*, 1999; Blais *et al.*, 2003, 2006; Demers *et al.*, 2007). Water quality parameters such as organic carbon, sulfides, hardness, and pH can influence the bioavailability and toxicity of contaminants (e.g., Chapman *et al.*, 1998).

The U.S. National Park Service (NPS) safeguards over 400 special places across 84 million acres for the protection of unique natural resources and scenic beauty. National parks are often considered to be among the most pristine ecosystems in the U.S. The NPS is mandated to protect national park areas and associated resources, in an unimpaired condition for future generations (USDOJ, 1916). Therefore, understanding contaminant burdens and subsequent potential risks to humans and wildlife helps the NPS better manage its resources. The purpose of this study was to provide a rigorous spatial analysis (using legacy data and new data) of CECs in fish from national parks so to comprehend park or regional differences in contaminant burdens, and compare contaminant concentrations to human and wildlife health thresholds to better understand potential effects in these remote, relatively pristine areas.

MATERIALS AND METHODS

Study Area

Contaminant concentrations were analyzed in 128 fish from 18 water bodies in 8 U.S. National Park Service units: Great Sand Dunes National Park & Preserve (NP & Pres.) and Rocky Mountain National Park (NP) in Colorado; Lassen Volcanic NP, Sequoia & Kings Canyon NPs, and Yosemite NP in California; and Katmai NP & Pres., Lake Clark NP & Pres., and Wrangell-St. Elias NP & Pres. in Alaska (Table 1). Water bodies were remote, high-elevation and/or high-latitude, and were approximately one mile or more from anthropogenic influence (e.g., roads, latrines, developed areas) to isolate the contribution of contaminant loading by atmospheric deposition. Salmonids (male and female) were targeted to reduce variation associated with species differences and trophic status of fish, and to enable comparisons between sites. Brook or lake trout (*Salvelinus fontinalis*, *S. namaycush*) were primarily collected, as were cutthroat and rainbow trout (*Oncorhynchus clarki*, *O. mykiss*) (Table 1). Arctic char (*S. alpinus*) were collected from Idavain Lake in Katmai because trout were not prevalent in the lake.

The fish data presented herein are combined with legacy fish data from another set of 8 national parks in the western U.S. and Alaska previously analyzed for contaminants (Ackerman *et al.*, 2008; Landers *et al.*, 2010). The combined water bodies sampled (all lakes except Sand Creek at Great Sand Dunes NP & Pres.) ranged from high-latitude/low-elevation to mid-latitude/high-elevation subalpine and alpine lakes (see Table 1). Water quality data were not collected as part of this study.

Sampling Procedures

Fish were collected in 2008-2011 by NPS resource managers and in 2003-2006 by Landers *et al.* (2010) using gill nets, hook-and-line, or electroshocking techniques. With the exception of Katmai and Lake Clark NP & Pres. samples, fish were euthanized with a blow to the head before the gonads and spleen were removed for a separate study (see Schreck and Kent, 2013). The whole fish were then preserved on wet or dry ice in the field, frozen in the laboratory freezer (<4°C) at each park, shipped on dry ice to Oregon State University, and stored at −20°C until analyzed. See Ackerman *et al.* (2008) and Landers *et al.* (2010) for detailed collection procedures for the legacy fish samples (2003-2006).

Laboratory and Statistical Analyses

Whole fish homogenate was prepared, extracted, and analyzed for all fish samples (2003-2011) at Oregon State University's Simonich Laboratory following the method described in Ackerman *et al.* (2008) for 34 pesticides, 7 PCBs, and 18 PAHs (Table S1). SOCs were selected according to established laboratory methods, environmental relevancy, and consistency with previous studies. Fish extracts were analyzed for the same analytes as Landers *et al.* (2010), except that polybrominated diphenyl ethers were excluded from the current work. Quality assurance and control (QA/QC) procedures were followed according to Ackerman *et al.* (2008) and included calibration checks (25% frequency), method blanks (25% frequency), and standard reference material (NIST SRM 1946) extraction and analysis (10% frequency). A more detailed description of QA/QC guidelines is in Erway *et al.* (2004). Pesticides include current-use pesticides and historic-use pesticides (i.e., legacy contaminants now banned in North America). While the samples were analyzed for all 59 SOCs, only those detected in greater than 50% of the fish samples are discussed in depth. Sum PCBs, endosulfans, chlordanes, and PAHs are further defined in Table S1.

Pesticide concentrations were compared to U.S. Environmental Protection Agency (USEPA) contaminant health thresholds for recreation (17.5 g fish/day) and subsistence (142 g fish/day) fish consumption (Ackerman *et al.*, 2008). Calculations assume an adult body mass of 70 kg and an acceptable risk level (lifetime excess cancer risk of 1:100,000). Human health thresholds established for gamma-hexachlorohexane (γ-HCH), chlorpyrifos, dacthal, and endosulfans are defined for the non-cancer (i.e., chronic) end point; human health thresholds for *p,p′*-DDE, chlordanes, dieldrin, and hexachlorobenzene (HCB) are

TABLE 1. Fish from National Parks Analyzed for SOCs by Oregon State University, 2003-2011. Shaded cells indicate previously published data (Ackerman *et al.*, 2008; Landers *et al.*, 2010). The number of fish, n, for each park is listed. NA, not available; data on fish sex for certain parks is incomplete. Regional groups are *Alaskan* (AK), *Cascades* (CS), *Rockies* (RO), and *Sierra Nevada* (SN).

Park	Code	Region	Water Body	Elevation (m)	Species	Sex M	Sex F	Median Length (mm)	Year Collected
Great Sand Dunes NP & Pres. (n = 8)	GRSA	RO	Sand Creek	2,301	Cutthroat Trout	0	8	233	2009
Katmai NP & Pres. (n = 20)	KATM	AK	Idavain Lake	223	Arctic Char	NA		300	2011
			Lake Brooks	18	Lake Trout			536	
Lake Clark NP & Pres. (n = 20)	LACL	AK	Kijik Lake	106	Lake Trout	NA		473	2011
			Lake Kontrashibuna	140				456	
Lassen Volcanic NP (n = 8)	LAVO	SN	Summit Lake	2,072	Brook Trout	4	4	282	2009
Rocky Mountain NP (n = 15)	ROMO	RO	Dream Lake	3,048	Cutthroat Trout	2	0	282	2006
			Haynach Lake	3,413		2	1	319	
			Lake Haiyaha	3,115		2	0	281	
			Lone Pine Lake	3,024	Brook Trout	2	0	262	
			Spirit Lake	3,208		2	0	260	
			Nanita Lake	3,285	Cutthroat Trout	2	0	230	2008
			Ypsilon Lake	3,304		2	0	238	2009
Sequoia & Kings Canyon NPs (n = 35)	SEKI	SN	Bench Lake	3,249	Brook Trout	10	6	148	2009
			Kern Point Lake	3,000	Rainbow Trout	9	8	195	
Wrangell-St. Elias NP & Pres. (n = 12)	WRST	AK	Copper Lake	885	Lake Trout	0	6	500	2008
			Tanada Lake	879		0	6	512	2009
Yosemite NP (n = 10)	YOSE	SN	Mildred Lake	2,987	Brook Trout	6	4	267	2009
Denali NP & Pres. (n = 13)	DENA	AK	McLeod Lake	609	Burbot/ Whitefish	3	0	178	2004-05
			Wonder Lake	610	Lake Trout	6	4	460	
Gates of the Arctic NP & Pres. (n = 10)	GAAR	AK	Matcharak Lake	488	Lake Trout	5	5	515	2004
Glacier NP (n = 20)	GLAC	RO	Oldman Lake	2,026	Cutthroat Trout	6	4	387	2005
			Snyder Lake	1,600		5	5	172	
Mount Rainier NP (n = 20)	MORA	CS	Golden Lake	1,372	Brook Trout	7	3	229	2005
			Lake LP19	1,372		5	5	238	
Noatak NPres. (n = 10)	NOAT	AK	Burial Lake	427	Lake Trout	5	5	423	2004
Olympic NP (n = 20)	OLYM	CS	Hoh Lake	1,384	Brook Trout	5	5	205	2005
			PJ Lake	1,433		5	5	71	2003, 2005
Rocky Mountain NP (n = 20)	ROMO	RO	Mills Lake	3,030	Rainbow Trout	4	6	225	2003
			Lone Pine Lake	3,024		4	6	243	
Sequoia & Kings Canyon NPs (n = 20)	SEKI	SN	Emerald Lake	2,800	Brook Trout	6	4	199	2003
			Pear Lake	2,904		8	2	207	

defined for the cancer end point. Contaminant health thresholds for piscivorous wildlife (kingfisher, mink, and river otter) were derived using USEPA nonlethal reproductive and developmental wildlife health end points as indicators of a negative effect (Ackerman *et al.*, 2008). Concentrations of PCBs in fish are not compared to human health thresholds because different PCB congeners were analyzed than are used in the USEPA health threshold. Also, the frequency of PAH detection was too low (i.e., 11% average detection) to perform appropriate human and wildlife health risk estimations. SOC concentrations in fish were expressed as ng/g lipid except when compared to health thresholds, which are defined in ng/g wet weight (ww). Estimated detection limits were included in the graphs and statistical analyses only when they occurred at a frequency of less than 50% in the dataset (consistent with Ackerman *et al.*, 2008).

The Tukey-Kramer honestly significant test (SAS 9.3) was used to compare SOC concentrations (ng/g lipid) in 128 fish by park and site for the 8 national parks in this study (2006-2011). SOC concentrations from an additional 133 legacy fish samples collected in 2003-2005 (Ackerman *et al.*, 2008; Landers *et al.*, 2010) were incorporated into the statistical analyses for a more robust dataset. The Tukey-Kramer test was used on the total dataset ($n = 261$) to meta-analyze SOC concentrations from 31 water bodies in 14 national parks. All statistically significant differences had a p-value < 0.05. The variability in contaminant concentrations in fish was tested by park, region, water body, elevation, species, fish length, and sex. Fish characteristics (species, length, sex) increase understanding of vulnerability to SOC accumulation; geographic characteristics (water body, elevation) help understand how site factors affect SOC distribution in remote areas; and spatial characteristics help the NPS understand if SOC concentrations in fish from some parks or regions are higher than others.

All fish species were categorized into seven taxa: lake trout, burbot, whitefish, Arctic char, cutthroat trout, brook trout, and rainbow trout (Table 1). The rainbow trout hybrid collected at Sequoia & Kings Canyon (Kern Point) and the cutthroat trout hybrid collected at Great Sand Dunes (Sand Creek) were categorized as rainbow trout and cutthroat trout, accordingly. Elevation was determined as (m): low (<1,000), mid (1,000-2,500), and high (>2,500). Fish length, a surrogate for fish age, was categorized as (mm): small (50-190), small-medium (190-225), medium (225-280), medium-large (280-455), and large (455-620). Median fish length ranged from 71 mm at Olympic (PJ) to 536 mm at Katmai (Brooks) (Table 1).

RESULTS

SOCs in Fish from Western U.S. and Alaskan National Parks

The 10 most concentrated SOCs measured in >95% of the fish (ng/g lipid) collected in 2006-2011 from 8 western U.S. and Alaskan national parks as part of this study were *p,p′*-DDE, HCB, chlordanes (*trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor), endosulfan sulfate, and PCB 138, 153, 183, 187 (Figure 1, Figure S1). Concentrations of HCB, chlordanes, and PCBs were significantly higher at Lake Clark compared to the other parks ($p < 0.05$), while *p,p′*-DDE concentrations were significantly higher at Lake Clark and Yosemite than the other parks ($p < 0.05$). Endosulfan sulfate was significantly higher at Rocky Mountain and Yosemite ($p < 0.05$) (Figure 1). Dacthal and chlorpyrifos were detected in 82% and 67% of the fish samples, respectively, and concentrations of both were significantly higher at Yosemite than the other parks ($p < 0.05$). Dieldrin and methoxychlor were detected in 73% and 72% of the fish samples, respectively. The highest average dieldrin concentration was measured at Lake Clark (11 ng/g lipid), whereas the highest average methoxychlor concentration was measured at Wrangell-St. Elias (76 ng/g lipid). Mirex concentrations, detected in 56% of the fish samples, were significantly higher at Lake Clark compared to the other parks ($p < 0.05$). The lowest detected SOC (measured in 2% of the fish samples), γ-HCH, was detected at Wrangell-St. Elias (Copper) and Rocky Mountain (Spirit).

Legacy data representing SOC concentrations (ng/g lipid) in fish collected in national parks from 2003-2005 (Ackerman *et al.*, 2008; Landers *et al.*, 2010) were combined with SOC concentrations (ng/g lipid) in fish collected in 2006-2011 for the meta-analysis. When considering all 261 fish from 14 national parks, the 10 most concentrated SOCs were as follows: *p,p′*-DDE, HCB, chlordanes (*trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor), and PCBs (PCB 101, 138, 153, 183, 187) (Figures S2F-S2H, S2M). Park-by-park comparisons showed significantly higher concentrations of mirex, chlordanes, and PCBs at Lake Clark as compared to the other 13 parks ($p < 0.05$). HCB concentrations in fish were significantly higher at Lake Clark and Wrangell-St. Elias, whereas *p,p′*-DDE concentrations were significantly higher at Yosemite and Lake Clark ($p < 0.05$). Site-by-site comparisons showed that concentrations of HCB, mirex, *p,p′*-DDE, chlordanes, and PCBs were significantly higher in fish from Kijik Lake at Lake Clark than the other 30 water bodies sampled ($p < 0.05$). While based on a small sample size ($n = 2$), endosulfan sulfate in fish was significantly higher from Dream Lake at Rocky Mountain than all other water bodies ($p < 0.05$).

SOC Concentrations by Geospatial Characteristics

The 14 total parks were grouped into geographic areas centered around mountain ranges to determine which regions, if any, were susceptible to specific SOCs. The four groups of parks were defined as *Alaskan* (Denali, Gates of the Arctic, Katmai, Lake Clark, Noatak, and Wrangell-St. Elias), *Cascades* (Mount Rainier and Olympic), *Sierra Nevada* (Lassen Volcanic, Sequoia & Kings Canyon, and Yosemite), and *Rockies* (Glacier, Great Sand Dunes, and Rocky

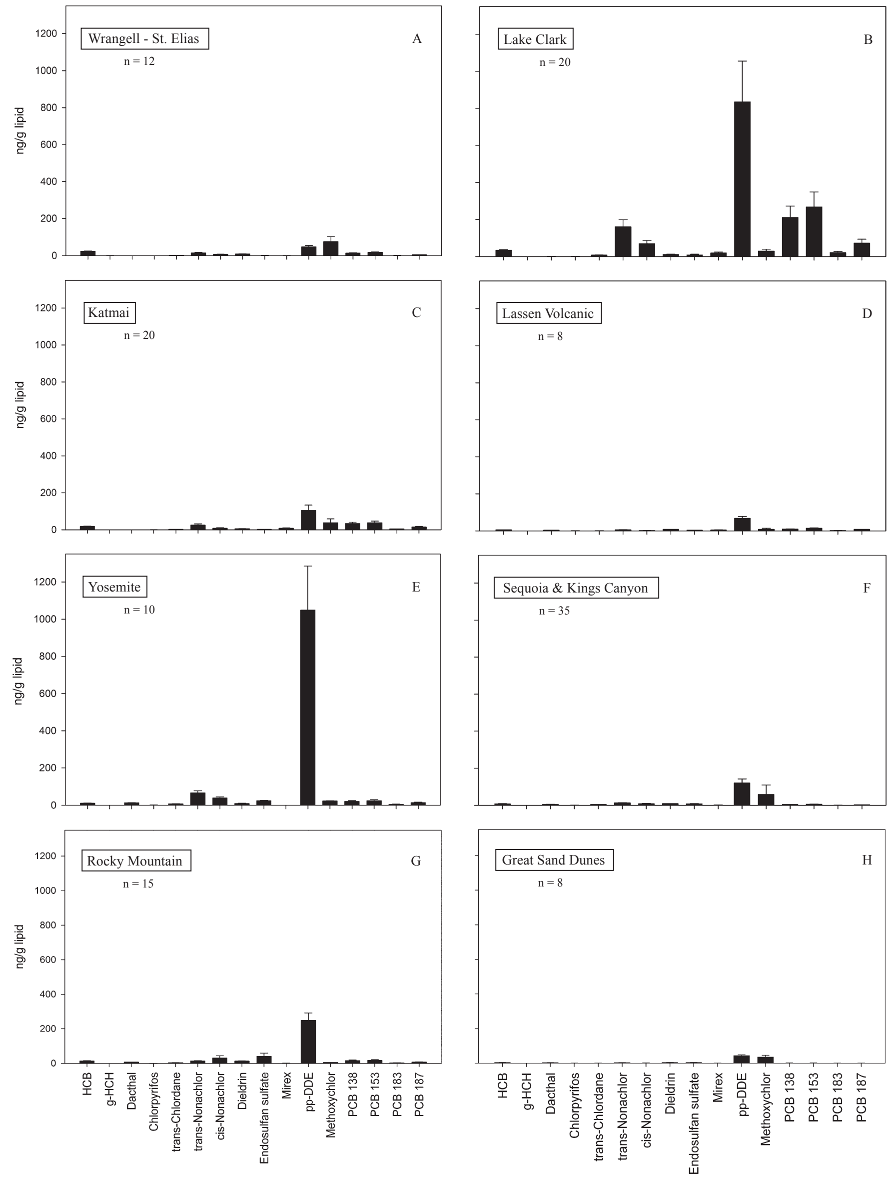

FIGURE 1. Mean SOC Concentrations in Fish (ng/g lipid), by National Park, in the Current Study. Only the most frequently detected and environmentally relevant SOCs are included here. Bars represent standard error. The number of fish, *n*, for each park is listed on the graph. ND, no detect; *, ND > 50% of lake fish.

Mountain). Fish from the *Alaskan* region had significantly higher concentrations of mirex, HCB, chlordanes, and PCBs than fish from the other regions (*Cascades*, *Rockies*, *Sierra Nevada*) ($p < 0.05$). Along with fish from the *Alaskan* region, fish from the *Sierra Nevada* had significantly higher concentrations of *p*,*p*′-DDE; whereas fish from both the *Sierra Nevada* and *Rockies* had significantly higher concentrations of dacthal and endosulfans ($p < 0.05$). Figure 2 illustrates the relative concentration of selected SOCs in fish between the regions. Fish from low-elevation sites had significantly higher concentrations of HCB, chlordanes, PCBs, and mirex ($p < 0.05$), whereas fish from high-elevation sites had significantly higher concentrations of dieldrin and endosulfan sulfate ($p < 0.05$).

SOC Concentrations by Fish Characteristics

Tukey-Kramer test results on all fish species indicate that HCB and chlordane concentrations (ng/g lipid) were significantly higher in lake trout, whitefish, and burbot than in Arctic char and brook, cutthroat, and rainbow trout; while mirex and PCB concentrations were significantly higher in lake trout, whitefish, burbot, and Arctic char as compared to brook, cutthroat, and rainbow trout ($p < 0.05$). The

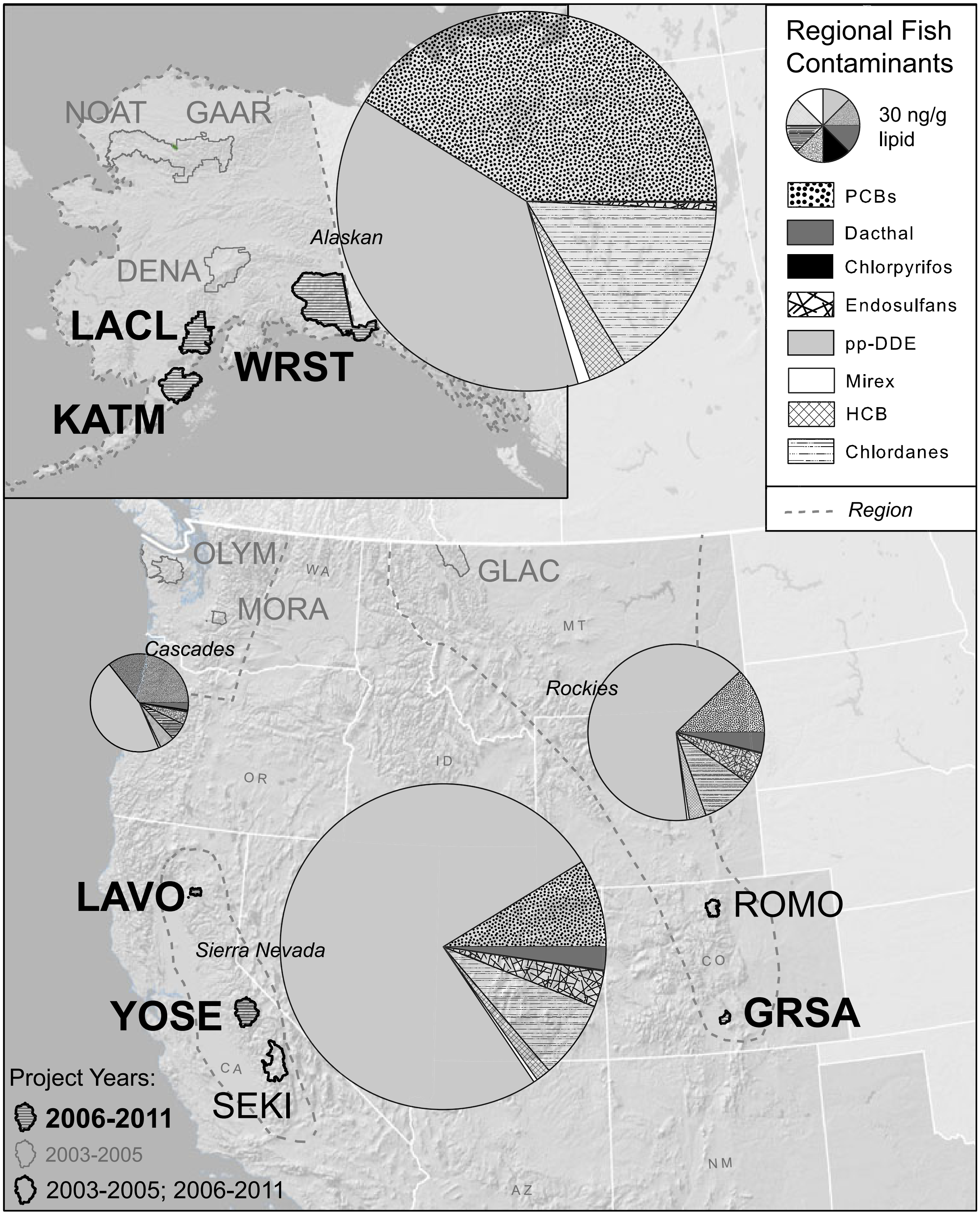

FIGURE 2. Contaminant Concentrations (ng/g lipid) in Fish by Region (*Alaskan, Cascades, Rockies, Sierra Nevada*) Overlaid on a Map of National Parks with Fish SOC Data from the Simonich Laboratory, 2003-2005 and 2006-2011. Fish from SEKI and ROMO were collected for both efforts. Circle area is proportional to total SOC concentration. Park codes are identified in Table 1.

remaining SOCs were similar for all species. Medium-large and large fish from all sites had significantly higher concentrations of HCB, chlordanes, PCBs, and mirex ($p < 0.05$). Small-medium and medium-large fish had significantly higher concentrations of dacthal ($p < 0.05$). Male fish had significantly

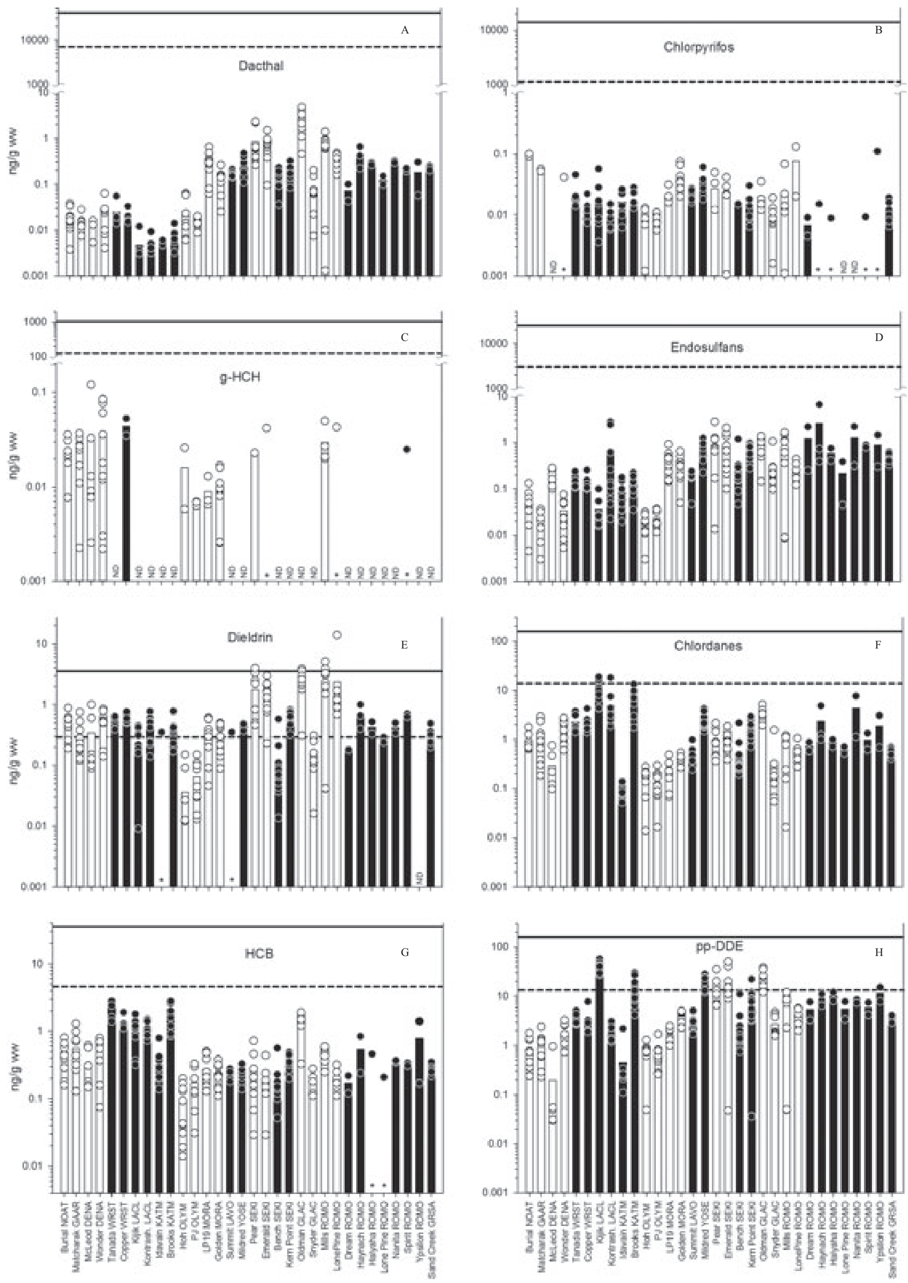

FIGURE 3. Concentrations of SOCs in Fish (ng/g ww) Compared to Human Health Fish Consumption Thresholds. Dashed lines represent the subsistence threshold; solid lines represent the recreational threshold. Top of bar indicates the mean concentration and the circles indicate concentrations of individual fish. Black bars depict data from the current study; white bars depict previously published data (Ackerman *et al.*, 2008; Landers *et al.*, 2010). ND, no detect; *, ND > 50% of lake fish. Parks ordered by region on the *x*-axis (L to R: *Alaskan, Cascades, Sierra Nevada, Rockies*); park codes identified in Table 1. Data are plotted on a $\log_{10}$ scale.

higher concentrations of PCBs, *p,p′*-DDE, and endosulfan sulfate, whereas female fish had significantly higher concentrations of methoxychlor ($p < 0.05$).

Comparing SOC Concentrations in Fish to Contaminant Health Thresholds

Contaminant health thresholds for humans were adopted from the USEPA to evaluate non-cancer (i.e., chronic) and cancer contaminant health thresholds for human consumption of fish at recreational and subsistence levels, similar to Ackerman *et al.* (2008). These data are shown in Figure 3, Figure S3, and Table S2. The legacy data (shown in white bars in Figures 3, S3) are not discussed in detail in this report (see Ackerman *et al.*, 2008; Landers *et al.*, 2010), but were included in the meta-analysis to present the largest possible sample size of all fish collected in national parks from 2003-2011 and ana-

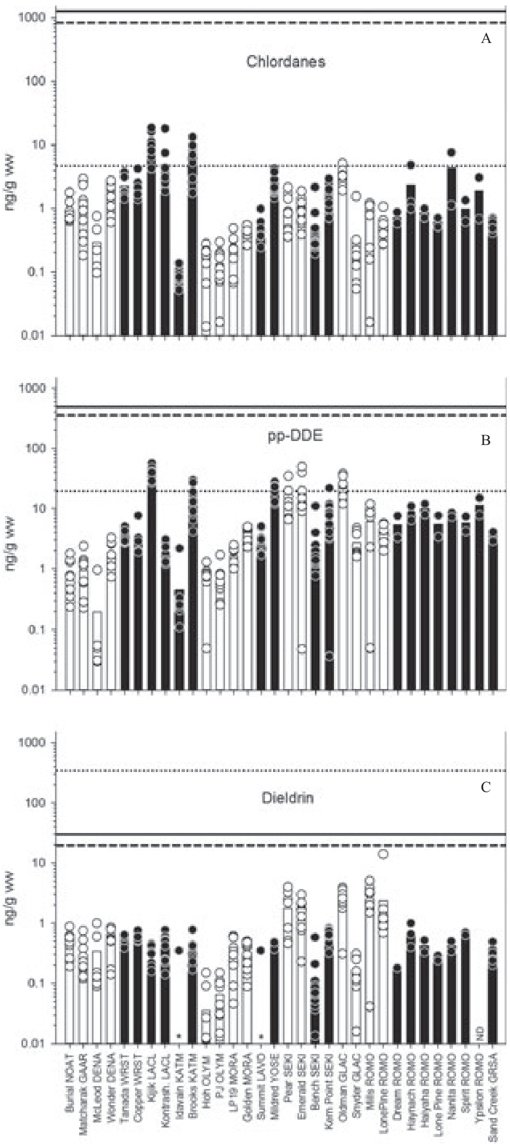

FIGURE 4. Concentrations of Historic-Use Pesticides Chlordanes, *p,p′*-DDE, and Dieldrin in Fish (ng/g ww) Compared to Piscivorous Wildlife Health Thresholds for Kingfisher (dotted line), Mink (dashed line), and River Otter (solid line). Top of bar indicates the mean concentration and the circles represent the concentrations of individual fish. Black bars depict data from the current study; white bars depict previously published data (Ackerman *et al.*, 2008; Landers *et al.*, 2010). ND, no detect; *, ND > 50% of lake fish. Parks ordered by region on the *x*-axis (L to R: *Alaskan, Cascades, Sierra Nevada, Rockies*); park codes identified in Table 1. Data are plotted on a $\log_{10}$ scale.

lyzed with the same protocols. Fish SOC concentrations at the 8 parks collected in this study (shown in black bars in Figures 3, S3) did not exceed any contaminant health thresholds for fish consumption by recreational users. However, concentrations of three historic-use pesticides (dieldrin, *p,p′*-DDE, and chlordanes) exceeded the guideline for fish consumption by subsistence users (Table S2). Dieldrin concentrations in fish at 15 of 18 sites exceeded the subsistence health threshold. The lake-average dieldrin concentration in fish exceeded the guideline for subsistence fish consumption at nine sites: Wrangell-St. Elias (Tanada, Copper), Lake Clark (Kontrashibuna), Katmai (Brooks), Sequoia & Kings Canyon (Kern Point), and Rocky Mountain (Haynach, Haiyaha, Nanita, Spirit) (Figure 3E). Dieldrin concentrations in individual fish exceeded the subsistence threshold at Lake Clark (Kijik), Katmai (Idavain), Lassen Volcanic (Summit), Yosemite (Mildred), Sequoia & Kings Canyon (Bench), and Great Sand Dunes (Sand Creek). The lake-average *p,p′*-DDE concentration in fish exceeded the guideline for subsistence fish consumption at Lake Clark (Kijik), Katmai (Brooks), and Yosemite (Mildred) (Figure 3H). The *p,p′*-DDE concentrations in individual fish at Sequoia & Kings Canyon (Kern Point) and Rocky Mountain (Ypsilon) also exceeded the subsistence threshold. Chlordane concentrations in three individual fish at 2 of 18 sites, Lake Clark (Kijik and Kontrashibuna), exceeded the guideline for subsistence fish consumption (Figure 3F). Concentrations of the historic-use pesticide, HCB (Figure 3G), recently banned pesticides γ-HCH and endosulfans (Figures 3C-3D), and current-use pesticides dacthal and chlorpyrifos (Figures 3A-3B) did not exceed any health thresholds. Historic-use pesticides mirex, alpha-hexachlorohexane (α-HCH), heptachlor epoxide, and methoxychlor also did not exceed human health thresholds (Figure S3).

Concentrations of chlordanes, *p,p′*-DDE, and dieldrin in fish were compared to contaminant health thresholds established for fish-eating wildlife (kingfishers, mink, river otter) (Figure 4, Table 2, Table S3). Concentrations of chlordanes and *p,p′*-DDE in fish exceeded kingfisher health thresholds, but concentrations in fish did not exceed mink and river otter health thresholds. The lake-average chlordane concentration in fish from Lake Clark (Kijik, Kontrashibuna) and Katmai (Brooks) exceeded the kingfisher health threshold. Individual fish exceeded the chlordane threshold for kingfisher health at Rocky Mountain (Haynach, Nanita) (Figure 4A). The lake-average concentration of *p,p′*-DDE in fish exceeded the threshold for kingfisher health at Lake Clark (Kijik), whereas *p,p′*-DDE concentrations in individual fish exceeded the kingfisher threshold at Katmai (Brooks), Yosemite (Mildred), and Sequoia & Kings Canyon (Kern Point) (Figure 4B). All individual fish were below dieldrin wildlife thresholds (Figure 4C; Table S3).

Of the total fish dataset in the meta-analysis, dieldrin, *p,p′*-DDE, and/or chlordane concentrations in over half (153 of 261) of the individual fish (in 28 of 31 water bodies) exceeded human health thresholds for subsistence fish consumption. Lake-average

TABLE 2. Selected Chemical Residue Concentrations (μg/g ww) in 261 Whole-Body Fish Samples from 31 Water Bodies in National Parks and Comparison to Established Fish Toxicity and Wildlife Health Thresholds (μg/g ww).

	Concentrations			Thresholds						
Pesticide	**Mean**	**Median**	**Maximum**	**Kingfisher[1]**	**Mink[1]**	**Otter[1]**	**Bald Eagle[2]**	**Fish**	**No. of Fish Exceeding Thresholds**	**No. of Water Bodies Exceeding Thresholds**
HCB	0.52	0.31	2.8	NA	NA	NA	2,250	330[3]	0	0
γ-HCH	0.024	0.018	0.12	NA	NA	NA	20,000	100[3]	0	0
Dieldrin	0.71	0.42	14	360	20	30	770	120-5,650[3]	0	0
Chlordanes	1.7	0.77	19	4.5	830	1,140	2,140[5]	300[3,5]	20	6
Endosulfans	0.41	0.20	6.7	NA	NA	NA	100,000	NA[3]	0	0
p,p'-DDE	7.3	3.0	57	20	360	490	NA	600[4]	27	7
DDT[6]	7.7	3.2	68	NA	NA	NA	270	150-3,000[3]	0	0
				No. of Fish Exceeding Thresholds						
				38	0	0	0	0		
				No. of Water Bodies Exceeding Thresholds						
				10	0	0	0	0		

NA, data not available
[1]Ackerman *et al.* (2008); Lazorchak *et al.* (2003).
[2]Hinck *et al.* (2009), No Effects Hazard Concentration (NEHC).
[3]Hinck *et al.* (2009), toxicity thresholds for fish and wildlife.
[4]Beckvar *et al.* (2005).
[5]Chlordanes = sum of heptachlor epoxide, chlordanes (*cis, trans, oxy*), and nonachlor (*cis, trans*).
[6]DDT = sum of *o,p'*- and *p,p'*- DDT, DDD, and DDE.

dieldrin and/or *p,p'*-DDE concentrations in fish exceeded the human health thresholds for subsistence fish consumption in 20 of 31 water bodies. These water bodies are located in Noatak, Denali, Wrangell-St. Elias, Lake Clark, Katmai, Mt. Rainier, Yosemite, Sequoia & Kings Canyon, Glacier, and Rocky Mountain. The concentrations of five other SOCs ranged from 1-6 orders of magnitude below contaminant health thresholds for subsistence fish consumption (Figures 3A-3D, 3G). Chlordane and/or *p,p'*-DDE concentrations in less than one-sixth (38 of 261) of the individual fish (in 10 of 31 water bodies) exceeded piscivorous wildlife (i.e., kingfisher) health thresholds. Lake-average chlordane and/or *p,p'*-DDE concentrations in fish exceeded the kingfisher health threshold for safe fish consumption in 4 of 31 water bodies. These water bodies are located in Lake Clark, Katmai, and Glacier.

DISCUSSION

This research analyzed contaminant concentrations in 128 fish from 18 water bodies in 8 western U.S. and Alaskan national parks, and compared concentrations to human and wildlife health thresholds established for fish consumption. This dataset was also analyzed alongside legacy data from another 133 fish at 14 water bodies in 8 western U.S. and Alaskan national parks collected and analyzed using similar methods (Ackerman *et al.*, 2008; Landers *et al.*, 2010). This meta-analysis totaled 261 fish from 31 distinct water bodies in 14 unique national parks and assessed SOC distribution in fish given certain geographic and fish parameters. Findings indicate that historic-use pesticides (e.g., HCBs, chlordanes) and banned industrial compounds like PCBs are especially high in Alaska, whereas current-use pesticides (chlorpyrifos, dacthal, and endosulfans [recently banned]) are particularly high in the *Sierra Nevada* and *Rockies* (Figure 2). The findings in fish are consistent with the relative concentrations of pesticides detected in snowpack and vegetation profiles from these parks/regions. Sequoia & Kings Canyon and Rocky Mountain snowpack and vegetation pesticide profiles include dacthal, chlorpyrifos, endosulfans; whereas the snowpack and vegetation pesticide profile in parks from Alaska includes historic-use pesticides like HCB and chlordanes at a larger percentage of the total concentration than is found in the *Rockies*, *Cascades*, and *Sierra Nevada* (Figure 2) (Hageman *et al.*, 2010; Landers *et al.*, 2010; Schrlau *et al.*, 2011).

Historic-use pesticides dieldrin and *p,p'*-DDE are the general exception in that they are not regionally concentrated and they are ubiquitously present throughout the parks. Dieldrin was banned in the U.S. in 1987 (ATSDR, 2002a) but has a half-life in soil of 730 days (Mackay *et al.*, 2000) and a high Log K_{OA} (9.04) (EPA, 2012). DDT was banned in 1972 and is readily degraded to *p,p′*-DDE through photolysis and microorganisms (ATSDR, 2002b). The half-life of *p,p′*-DDE in soil is 2-15 years depending on the soil conditions (ATSDR, 2002b), and also has a high Log K_{OA} (8.78) (EPA, 2012). Both dieldrin and *p,p′*-DDE are persistent and bioaccumulative, and undergo slow volatilization from soil because of their relatively high Log K_{OA} (Table S4).

The relative concentrations of pesticides — particularly current-use pesticides dacthal, chlorpyrifos, and endosulfans — in fish from national parks in the western U.S. and Alaska appear to be highest in parks located in close proximity to agricultural areas, a probable source of both current-use and historic-use pesticides (Hageman *et al.*, 2006, 2010). This finding is also consistent with Landers *et al.* (2010). Elevated concentrations of the banned compounds *p,p′*-DDE, mirex, chlordanes, PCBs, and HCB in fish at Lake Clark and Wrangell-St. Elias, and the *Alaskan* parks in general, which are far removed from agricultural areas, are the likely result of global transport and colder air temperatures that enhance SOC deposition in circumpolar regions (Wania and MacKay, 1993; Simonich and Hites, 1995; Wania *et al.*, 1998; Daly and Wania, 2005).

Lake-average dieldrin and *p,p′*-DDE concentrations in fish exceeded human health thresholds in nearly two-thirds of the total water bodies sampled at 10 of 14 national parks in all four regions. Exceedances imply that a lifetime consumption of contaminated fish may increase the risk of developing cancer by more than 1 in 100,000. Fish consumption advice is not provided here because national parks are encouraged to work with the respective state agency (e.g., departments of health, natural resources) and the NPS Office of Public Health to issue related warnings and communicate guidance regarding contaminants in fish (DOI, 2012). Criteria for providing lake-specific fish consumption warnings and advice to the public considers fish species, length, sample size, waterbody location, and the cultural and health benefits associated with salmonid consumption, such as improved cardiac health from increased omega-3 fatty acid consumption or potential reduced intake of unhealthy fats due to food substitutions.

Lake-average chlordane and *p,p′*-DDE concentrations in fish exceeded kingfisher health thresholds in one-third of the total water bodies sampled at 3 of 14 national parks in the *Alaskan* and *Rockies* regions (Lake Clark, Katmai, and Glacier). Lake-average concentrations in fish did not exceed kingfisher health thresholds at national parks in the *Sierra Nevada*, nor those previously studied in the *Cascades* (Ackerman *et al.*, 2008; Landers *et al.*, 2010). Individual fish exceeded the kingfisher threshold for *p,p′*-DDE and/or chlordanes at Yosemite, Sequoia & Kings Canyon, and Rocky Mountain. The risk to kingfisher health is best understood when compared to prey fish, such as cutthroat, rainbow, and smaller brook trout from parks in the conterminous U.S. (e.g., Glacier) rather than large, predatory fish like lake trout from Lake Clark and Katmai that are unattainable for consumption by kingfisher. The nonlethal reproductive and developmental wildlife health end points pertinent to kingfisher health include eggshell thinning, reduced egg production, embryo mortality, behavioral modifications, and stunted growth of hatchlings (EPA-OWS, 1995).

Concentrations of HCB, γ-HCH, dieldrin, chlordanes, DDT, and endosulfan in fish were compared to no effects hazard concentrations (NEHCs) established for bald eagles (Hinck *et al.*, 2009) — a more likely predator than kingfishers of large fish such as the lake trout from Lake Clark and Katmai. Zero of 261 fish exceeded the bald eagle thresholds (Table 2). However, it is worth noting that the thresholds may be inaccurate estimates of sensitivity, given that the NEHC was a screening level assessment based on dietary or tissue studies, some of which were derived from studies conducted several decades ago (Hinck *et al.*, 2009).

Given the above comparisons to human and wildlife health thresholds, we conclude that fish concentrations of the pesticides dieldrin, *p,p′*-DDE, and chlordanes, in some national parks, may cause potential health effects for kingfishers and human subsistence fish consumers. However, little is known about contaminant concentrations in fish (i.e., critical body residues) that may cause adverse effects to the fish itself, as compared to the fish consumer. Concentrations of HCB, γ-HCH, dieldrin, chlordanes, and DDT in all fish from the meta-analysis were compared to fish toxicity thresholds established for and compiled by Hinck *et al.* (2009). Zero of 261 fish exceeded those thresholds (Table 2). Fish were also compared to a *p,p′*-DDE fish toxicity threshold reported by Beckvar *et al.* (2005) and similarly, 0 of 261 fish exceed that protective threshold (Table 2). However, as above, the authors note that the threshold may not be fully protective because the value was derived from older studies that emphasize lethality rather than the potentially more sensitive nonlethal end points. Schwindt *et al.* (2009) documented intersex, another nonlethal biological end point, in fish samples from national parks (2003-2006), and identified suspected SOCs that might be associated with reproductive biomarkers, but did not suggest a cause-and-effect relationship.

The selection of appropriate endpoints in the analysis is important. Mink, otter, and kingfisher are considered to be ubiquitous among these sites of varying elevation and latitude. Mink and sometimes otter consume large, adult fish and the belted kingfisher consumes smaller fish that are presumed to have lower contaminant concentrations, so the risk may be overestimated for kingfisher dependent upon dietary preferences. The diet of bald eagles also includes large, adult fish such as those collected and analyzed in this study. Because it is unlikely that the diet of the piscivorous wildlife consists entirely of fish from one site, threshold exceedances applied herein may overestimate impacts to wildlife. However, nestling birds or cubs fed locally procured prey items may better estimate risk. That said, as reported by Hinck *et al.* (2009), "the risk of accumulative contaminants to nestlings/cubs and juveniles may be greater because of their small body size and correspondingly higher food ingestion rates compared to adult wildlife." One last consideration, similar to the Hinck *et al.* (2009) study: cumulative effects of the varying contaminants are not represented here. While many of the contaminants co-occur at the sites, this assessment assumes that the contaminants act independently.

Comparisons of contaminant concentrations between fish species showed that lake trout, whitefish, burbot, and Arctic char had higher concentrations of HCB, chlordane, mirex, and PCB concentrations than brook, cutthroat, and rainbow trout. All lake trout, whitefish, burbot, and Arctic char samples were collected from national parks located in Alaska, where concentrations of these historic-use contaminants were highest (Figure 2). Similarly, fish from the large length category had higher concentration of the same four historic-use compounds. An assessment of fish length (age) suggests that the average *Alaskan* fish was older than the fish collected in the conterminous U.S. The larger fish are susceptible to high contaminant concentrations because of longer life spans, lower growth rates, and lower metabolism (Scheuhammer *et al.*, 1998; Rose *et al.*, 1999; Schwindt *et al.*, 2008). Furthermore, lake trout are predatory and thus more prone to elevated contaminant concentrations. Small-medium to medium-large fish (i.e., Glacier, Lassen Volcanic, Rocky Mountain, Sequoia & Kings Canyon, Yosemite) had high concentrations of dacthal, also consistent with the relative proportion of this current-use pesticide on a regional level (Figure 2). Fish from high-elevation sites (i.e., *Rockies*, *Sierra Nevada*) had higher concentrations of endosulfan sulfate, and fish from the low-elevation sites (i.e., *Alaskan*) had higher concentrations of HCB, chlordane, mirex, and PCBs — also consistent with the regional analysis (Figure 2).

Due largely to this study's sampling design and specifically what fish were available and where, a gradient of fish species, length, and elevation was not defined within parks and therefore cannot be accurately tested herein. The differences in both fish and geospatial characteristics are evident only between parks. For instance, the large fish (i.e., lake trout) and low-elevation sites were concentrated in *Alaskan* parks, whereas smaller fish and the high-elevation parks were concentrated in the *Rockies* and *Sierra Nevada*. We conclude that species, elevation, and length differences are an artifact of the sampling design and species distribution. Statistical analyses were not performed to determine drivers in the variability, but region and proximity to sources appeared to influence the distribution of SOCs and contaminant loading/accumulation in fish. While no size-adjusted analyses were conducted, fish length (age) also likely influenced the resultant contaminant accumulation in fish.

Figure 2 illustrates the elevated SOC accumulation in both *Alaskan* and *Sierra Nevada* fish (155% and 133% of average total loading, respectively), likely attributable to fish age in *Alaskan* parks and source proximity in *Sierra Nevada* parks. SOC concentrations in *Sierra Nevada* fish are unique in that total contaminant loading is very high, yet fish are younger in age (mid-length in size). Contaminant loading in fish from the *Cascades* was lowest (40% of average total loading), followed by fish from the *Rockies* (72% of average total loading). *Alaskan* and *Cascades* fish had similar proportions of PCBs and *p,p′*-DDE, whereas fish from the *Rockies* and *Sierra Nevada* were dominated by *p,p′*-DDE. However, the current-use pesticide dacthal and recently-banned endosulfans were present in greater relative proportions in fish from parks in the conterminous U.S. than fish from *Alaskan* parks.

In addition to atmospheric sources, another vector of SOCs and source specific to anadromous Alaskan lakes — mainly Kijik Lake at Lake Clark, Brooks Lake at Katmai, and Tanada Lake at Wrangell-St. Elias (Shearer, J. and E. Veach, National Park Service, personal communication) — may be migrating sockeye salmon (*Oncorhynchus nerka*). Studies show that these marine-derived fish deliver SOCs including PCBs and DDT into lake sediments, and hence the food chain, upon returning to the freshwaters to spawn and die (Ewald *et al.*, 1998; Krummel *et al.*, 2005; Baker *et al.*, 2009). While the anadromous sockeye salmon were not sampled for this research, fish from the above anadromous lakes had concentrations of dieldrin, *p,p′*-DDE, and chlordanes seven times higher than the nonanadromous lakes sampled at those three national parks. However, it is unknown how factors including bioaccumulation rates, length/age, elevation, water chemistry, etc. contribute to the variation.

CONCLUSION

Results of this meta-analysis provide further evidence for the atmospheric transport and deposition of airborne contaminants, and accumulation in fish, at national parks in high-elevation and high-latitude areas of the western U.S. and Alaska. Concentrations of the historic-use pesticides dieldrin, *p,p'*-DDE, and/or chlordanes in fish exceeded human and/or wildlife health thresholds in 13 of 14 national parks (all parks studied except Olympic), bolstering previous findings on potential contaminant impacts throughout the study area. This is a particular concern because the national parks are provided for the public to enjoy, and the Organic Act that established the national parks mandates that resources remain unimpaired for future generations (USDOJ, 1916). Contaminants in fish differ by region, with the highest loadings reported in *Alaskan* and *Sierra Nevada* parks, while SOC distribution seemingly varies according to source proximity. The relatively high concentrations of current-use pesticides (e.g., dacthal) in fish from parks near agriculturally dense areas suggest that these areas are a probable source of contaminants, a finding consistent with other studies. In contrast, the concentrations of historic-use compounds are relatively high in national parks in Alaska, far from agriculture, a finding that supports the global distribution of SOCs and at some sites, the potential influence of marine-derived contamination. A similar SOC distribution in *Cascades* parks implies a global atmospheric signal, rather than local. SOC profiles in fish are consistent with profiles previously defined for snowpack at many of the same locations. Contributions from this research improve understanding of contaminant sources and resultant accumulation in fish. This work illustrates that even remote, mountainous, relatively pristine locations are not immune to contaminants of emerging concern. Given the findings, further work is warranted in such contaminant hot spots as Alaska and the Sierra Nevada.

Future work in this arena can reduce uncertainties associated with the findings presented herein. The thresholds applied in this study were commonly used and relevant to consider; however, they are not universally applicable and hence result in unlikely scenarios. Thus, regionally specific effects thresholds would increase relevancy and allow a better estimation of risk. The sampling of small, adult prey fish would more appropriately assess potential wildlife exposures to contaminants. Updated thresholds for various ecosystem end points would also allow a more accurate risk assessment of biological and toxicological effects in fish and other ecosystem components. Additional research in the areas of source attribution would shed light on how best to manage the risk, and the role anadromous fish play in defining that risk. Furthermore, multivariate analyses could be employed to tease out the variables and better quantify what is driving contaminant distribution. In closing, with few exceptions, research on other CECs such as pharmaceuticals and personal care products is largely lacking from high-elevation and high-latitude areas. Given the popularity of national parks among the public, with humans as one of many vectors of such CECs, investigations in parks may be warranted.

SUPPORTING INFORMATION

Additional Supporting Information may be found in the online version of this article:

Table S1. SOCs measured in fish by two modes of ionization.

Table S2. Number of fish exceeding subsistence fish consumption threshold established for human health. Numerator indicates number of fish exceeding consumption limit and denominator indicates number of fish analyzed. Shaded cells depict an exceedance. No fish exceeded the recreational fish consumption threshold.

Table S3. Number of fish exceeding kingfisher health thresholds. Numerator indicates number of fish exceeding consumption limit and denominator indicates number of fish analyzed. Shaded cells depict an exceedance. No fish exceeded the mink or river otter health thresholds. Park codes identified in Table 1.

Table S4. Subcooled Vapor Pressure (Pa), Log K_{OW}, and Log K_{OA} at 25°C (EPA, 2012) for 12 pesticides frequently measured in fish.

Figure S1. Mean SOC concentrations in fish (ng/g lipid), by national park, in the current research (2006-2011). Those SOCs excluded from Figure 1 are displayed here. Bars represent standard error. The number of fish, *n*, for each park is listed on the graph. ND, no detect; *, ND > 50% of lake fish.

Figure S2. Concentrations of SOCs in fish (ng/g lipid) from current research (2006-2011) and previous study (2003-2005). Top of bar indicates the mean concentration and the circles indicate concentrations of individual fish. Black bars depict data from the current, post-WACAP study; white bars depict previously published data from WACAP (Ackerman *et al.*, 2008; Landers *et al.*, 2010). ND, no detect; *, ND > 50% of lake fish. Parks ordered by region on the *x*-axis (L to R: *Alaskan, Cascades, Sierra Nevada, Rockies*); park codes identified in Table 1. Data are plotted on a $\log_{10}$ scale.

Figure S3. Concentrations of SOCs in fish (ng/g ww) compared to human health fish consumption thresholds. Dashed lines represent the subsistence threshold; solid lines represent the recreational threshold. Top of bar indicates the mean concentration and the circles indicate concentrations of individual fish. Black bars depict data from the current study (2006-2011); white bars depict previously published data (2003-2005) (Ackerman *et al.*, 2008; Landers *et al.*, 2010). ND, no detect; *, ND > 50% of lake fish. Parks ordered by region on the *x*-axis (L to R: *Alaskan*, *Cascades, Sierra Nevada*, *Rockies*); park codes identified in Table 1. Data are plotted on a $\log_{10}$ scale.

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LITERATURE CITED

Ackerman, L., A. Schwindt, D. Koch, G. Wilson, and S.L. Simonich, 2008. Atmospherically Deposited PBDEs, Pesticides, and PAHs in Western U.S. National Park Fish: Concentrations and Consumption Guidelines. Environmental Science and Technology 42(7):2334-2341.

Allen-Gil, S.M., C.P. Gubala, R. Wilson, D.H. Landers, T.L. Wade, J.L. Sericano, and L.R. Curtis, 1997. Organochlorine Pesticides and Polychlorinated Biphenyls (PCBs) in Sediments and Biota from Four US Arctic Lakes. Archives of Environmental Contamination and Toxicology 33(4):378-387.

ATSDR (Agency for Toxic Substances and Disease Registration), 2002a. Frequently Asked Questions about Aldrin/Dieldrin Fact Sheet. http://www.atsdr.cdc.gov/toxfaqs/tf.asp?id=316&tid=56, *accessed* February 2013.

ATSDR (Agency for Toxic Substances and Disease Registration), 2002b. Frequently Asked Questions about DDT & DDE Fact Sheet. http://www.atsdr.cdc.gov/toxfaqs/tf.asp?id=80&tid=20, *accessed* February 2013.

Baker, M.R., D.E. Schindler, G.W. Holtgrieve, and V.L. St. Louis, 2009. Bioaccumulation and Transport of Contaminants: Migrating Sockeye Salmon as Vectors of Mercury. Environmental Science and Technology 43:8840-8846.

Battaglin, W., J. Drewes, B. Bruce, and M. McHugh, 2007. Introduction: Contaminants of Emerging Concern in the Environment. Water Resources Impact 9(3):3-4.

Beckvar, N., T.M. Dillion, and L.B. Read, 2005. Approaches for Linking Whole-Body Fish Tissue Residues of Mercury and DDT to Biological Effects Thresholds. Environmental Toxicology and Chemistry 24(8):2094-2105.

Blais, J.M., S. Charpentie, F. Pick, L.E. Kimpe, A.S. Amand, and C. Regnault-Roger, 2006. Mercury, Polybrominated Diphenyl Ether, Organochlorine Pesticide, and Polychlorinated Biphenyl Concentrations in Fish From Lakes along an Elevation Transect in the French Pyrenees. Ecotoxicology and Environmental Safety 63(1):91-99.

Blais, J.M., F. Wilhelm, K.A. Kidd, D.C. Muir, D.B. Donald, and D.W. Schindler, 2003. Concentrations of Organochlorine Pesticides and Polychlorinated Biphenyls in Amphipods (*Gammarus Lacustris*) Along an Elevation Gradient in Mountain Lakes of Western Canada. Environmental Toxicology and Chemistry 22(11):2605-2613.

Bradford, D.F., K.A. Stanley, N.G. Tallent, D.W. Sparling, M.S. Nash, R.A. Knapp, L.L. McConnell, and S.L. Massey Simonich, 2013. Temporal and Spatial Variation of Atmospherically Deposited Organic Contaminants at High Elevation in Yosemite National Park, California. USA Environmental Toxicology and Chemistry 32(3):517-525.

Carpenter, D.O., 2006. Polychlorinated Biphenyls (PCBs): Routes of Exposure and Effects on Human Health. Reviews on Environmental Health 21(1):1-24.

Chapman, P.M., F. Wang, C. Janssen, J. Persoonne, and H.E. Allen, 1998. Ecotoxicology of Metals in Aquatic Sediments: Binding and Release, Bioavailability, Risk Assessment, and Remediation. Canadian Journal of Fisheries and Aquatic Sciences 55:2221-2243.

Colborn, T., F.S. vom Saal, and A.M. Soto. 1993. Developmental Effects of Endocrine-Disrupting Chemicals in Wildlife and Humans. Environmental Health Perspectives 101(5):378.

Daly, G.L. and F. Wania, 2005. Organic Contaminants in Mountains. Environmental Science and Technology 39:385-398.

Daughton, C.G., 2003. Non-Regulated Contaminants: Emerging Issues. Presentation. Environmental Health Sciences, Research, and Medicine (EHSRT), Workshop #5: From Source Water to Drinking Water: Emerging Challenges for Public Health. National Academies, Institute of Medicine, Washington, D.C., October 16, 2003. http://www.epa.gov/esd/chemistry/ppcp/images/iom-2003.pdf, *accessed* February 2013.

Demers, M.J., E.N. Kelly, J.M. Blais, F.R. Pick, V.L. St. Louis, and D.W. Schindler, 2007. Organochlorine Compounds in Trout from Lakes over a 1600 Meter Elevation Gradient in the Canadian Rocky Mountains. Environmental Science and Technology 41: 2723-2729.

DOI (U.S. Department of Interior), 2012. Chapter 5: Communication of Fish and Shellfish Consumption Advisories. Departmental Manual, Series: Environmental Quality Programs, Part 515: Environmental Management.

EPA, 2012. Estimation Programs Interface Suite™ for Microsoft® Windows, v 4.11, United States Environmental Protection Agency, Washington, D.C.

EPA-OWS, 1995. Great Lakes Water Quality Initiative Criteria Documents for the Protection of Wildlife: DDT, Mercury, 2,3,7,8-TCDD, PCBs. EPA-820-B-95-008. USEPA, Washington, D.C.

Erway, M.M., S. Simonich, D.H. Campbell, A. Schwindt, L. Geiser, K. Hageman, D. Schmedding, G. Wilson, S. Usenko, L. Ackerman, S. Echols, and D. Landers, 2004. Western Airborne Contaminants Assessment Project | Quality Assurance Project Plan. http://www.epa.gov/wed/pages/publications/authored/WACAP%20QA%20Project%20Plan%20May04.pdf.

Ewald, G., P. Larsson, H. Linge, L. Okla, and N. Szarzi, 1998. Biotransport of Organic Pollutants to an Inland Alaska Lake by Migrating Sockeye Salmon (*Oncorhynchus Nerka*). Arctic 51(1): 40-47.

Gallego, E., J.O. Grimalt, M. Bartrons, J.F. Lopez, L. Camarero, J. Catalan, E. Stuchlik, and R. Battarbee, 2007. Altitudinal Gradients of PBDEs and PCBs in Fish from European High Mountain Lakes. Environmental Science and Technology 41:2196-2202.

Hageman, K.J., W.D. Hafner, D.H. Campbell, D.A. Jaffe, D.H. Landers, and S.L. Massey Simonich, 2010. Variability in Pesticide Deposition and Source Contributions to Snowpack in Western U.S. National Parks. Environmental Science & Technology 44:4452-4458.

Hageman, K.J., S.L. Simonich, D.H. Campbell, G.R. Wilson, and D.H. Landers, 2006. Atmospheric Deposition of Current-Use and Historic-Use Pesticides in Snow at National Parks in the Western United States. Environmental Science and Technology 40(10):3174-3180.

Hardell, S., H. Tilander, G. Welfinger-Smith, J. Burger, and D.O. Carpenter, 2010. Levels of Polychlorinated Biphenyls (PCBs) and Three Organochlorine Pesticides in Fish from the Aleutian Islands of Alaska. PLoS ONE 5(8):e12396.

Hinck, J.E., C.J. Schmitt, K.A. Chojnacki, and D.E. Tillitt, 2009. Environmental Contaminants in Freshwater Fish and Their Risk to Piscivorous Wildlife Based on a National Monitoring Program. Environmental Monitoring and Assessment 152(1-4):469-494.

Iwanowicz, L.R. and C.A. Ottinger, 2009. Estrogens, Estrogen Receptors and Their Role as Immunoregulators in Fish. *In*: Fish Defenses, Volume 1: Immunology, G. Zaccone, J. Meseguer, A. Garcia-Ayala, and B.G. Kapoor (Editors). Science Publishers, Enfield, New Hampshire, pp. 277-322.

Jones, K.C. and P. de Voogt, 1999. Persistent Organic Pollutants (POPs): State of the Science. Environmental Pollution 100(1): 209-221.

Kidd, K.A., P.J. Blanchfield, K.H. Mills, V.P. Palance, R.E. Evans, J.M. Lazorchak, and R.W. Flick, 2007. Collapse of a Fish Population after Exposure to a Synthetic Estrogen. Proceedings of the National Academy of Sciences 104:8897-8901.

Krummel, E.M., I. Gregory-Eaves, R.W. MacDonald, L.E. Kimpe, M.J. Demers, J.P. Smol, B. Finney, and J.M. Blais, 2005. Concentrations and Fluxes of Salmon-Derived Polychlorinated Biphenyls (PCBs) in Lake Sediments. Environmental Science and Technology 39:7020-7026.

Landers, D.H., S. Massey Simonich, D. Jaffe, L. Geiser, D.H. Campbell, A. Schwindt, C. Schreck, M. Kent, W. Hafner, H.E. Taylor, K. Hageman, S. Usenko, L. Ackerman, J. Schrlau, N. Rose, T. Blett, and M. Morrison Erway, 2010. The Western Airborne Contaminant Assessment Project (WACAP): An Interdisciplinary Evaluation of the Impacts of Airborne Contaminants. Environmental Science and Technology 44(3):855-859.

Landewe, R.A., 2008. Scope of Contaminants of Emerging Concern in National Parks. Natural Resource Report NPS/NRPC/NRR–2008/032. National Park Service, Fort Collins, Colorado.

Lazorchak, J.M., F.H. McCormick, T.R. Henry, and A.T. Herlihy, 2003. Contamination of Fish in Streams of the Mid-Atlantic Region: An Approach to Regional Indicator Selection and Wildlife Assessment. Environmental Toxicology and Chemistry 22(3):545-553.

Longnecker, M.P., W.J. Rogan, and G. Lucier, 1997. The Human Health Effects of DDT (Dichlorodiphenyltrichloroethane) and PCBs (Polychlorinated Biphenyls) and an Overview of Organochlorines in Public Health. Annual Review of Public Health 8:211-244.

Mackay, D., W.Y. Shiu, and K.C. Ma, 2000. Physical-Chemical Properties and Environmental Fate Handbook. CRC Press, Lewix Publishers, Boca Raton, Florida.

McGee, M.R., M.L. Julius, A.M. Vajda, D.O. Norris, L.B. Barber, and H.L. Schoenfuss, 2009. Predator Avoidance Performance of Larval Fathead Minnows (*Pimephales Promelas*) Following Short-Term Exposure to Estrogen Mixtures. Aquatic Toxicology 91:355-361.

Ohyama, K.J., J. Angermann, D.Y. Dunlap, and F. Matsumura, 2004. Distribution of Polychlorinated Biphenyls and Chlorinated Pesticide Residues in Trout in the Sierra Nevada. Journal of Environmental Quality 33:1752-1764.

Painter, M.M., M.A. Buerkley, M.L. Julius, A.M. Vajda, D.O. Norris, L.B. Barber, E.T. Furlong, M.M. Schultz, and H.L. Schoenfuss, 2009. Antidepressants at Environmentally Relevant Concentrations Affect Predator Avoidance Behavior of Larval Fathead Minnows (*Pimephales Promelas*). Environmental Toxicology and Chemistry 28: 2677-2684.

Rose, J., M.S. Hutcheson, C.R. West, O. Pancorbo, K. Hulme, A. Cooperman, G. Decesare, R Isaac, and A. Screpetis, 1999. Fish Mercury Distribution in Massachusetts, USA Lakes. Environmental Toxicology and Chemistry 18:1370-1379.

Ryan, M.J., G.A. Stern, M. Diamond, M.V. Croft, P. Roach, and K. Kidd, 2005. Temporal Trends of Organochlorine Contaminants in Burbot and Lake Trout from Three Selected Yukon Lakes. Science of the Total Environment 351:501-522.

Scheuhammer, A.M., C.M. Atchison, A.H.K. Wong, and D.C. Evers, 1998. Mercury Exposure in Breeding Common Loons (Gavia Immer) in Central Ontario, Canada. Environmental Toxicology and Chemistry 17:191-196.

Schreck, C. and M. Kent, 2013. Extent of Endocrine Disruption in Fish of Western and Alaskan National Parks. National Park Service-Oregon State University Task Agreement J8W07080024. NPS report available at https://irma.nps.gov/App/Reference/Profile/2195337.

Schrlau, J.E., L. Geiser, K.J. Hageman, D.H. Landers, and S.M. Simonich, 2011. Comparison of Lichen, Conifer Needles, Passive Air Sampling Devices, and Snowpack as Passive Sampling Media to Measure Semi-Volatile Organic Compounds in Remote Atmospheres. Environmental Science and Technology 45(24): 10354-10361.

Schwindt, A.R., J.W. Fournie, D.H. Landers, C.B. Schreck, and M.L. Kent, 2008. Mercury Concentrations in Salmonids from Western U.S. National Parks and Relationships with Age and Macrophage Aggregates. Environmental Science and Technology 42(4):1365-1370.

Schwindt, A.R., M.L. Kent, L.K. Ackerman, S.L. Massey Simonich, D.H. Landers, T. Blett, and C.B. Schreck, 2009. Reproductive Abnormalities in Trout from Western U.S. National Parks. Transactions of the American Fisheries Society 138:522-531.

Simonich, S.L. and R.A. Hites, 1995. Global Distribution of Persistent Organochlorine Compounds Science 269:1851-1854.

USDOJ (U.S. Department of Justice), 1916. National Park Service Organic Act of 1916. 16 U.S.C. §§ 1-18f, 39 Stat. 535.

Vajda, A.M., L.B. Barber, J.L. Gray, E.M. Lopez, J.D. Woodling, and D.O. Norris, 2008. Reproductive Disruption in Fish Downstream from and Estrogenic Wastewater Effluent. Environmental Science and Technology 42:3407-3414.

Vives, I., J.O. Grimalt, J. Catalan, B.O. Rosseland, and R.W. Battarbee, 2004. Influence of Altitude and Age in the Accumulation of Organochlorine Compounds in Fish from High Mountain Lakes. Environmental Science and Technology 38:690-698.

Wania, F., J.E. Haugen, Y.D. Lei, and D. Mackay, 1998. Temperature Dependence of Atmospheric Concentrations of Semi-Volatile Organic Compounds. Environmental Science and Technology 32:1013-1021.

Wania, F. and D. MacKay, 1993. Global Fractionation and Cold Condensation of Low Volatility Organo-Chlorine Compounds in Polar Regions. Ambio 22:10-18.

Yang, R., Y. Wang, A. Li, Q. Zhang, C. Jing, T. Wang, and G. Jiang, 2010. Organochlorine Pesticides and PCBs in Fish from Lakes of the Tibetan Plateau and the Implications. Environmental Pollution 158(6):2310-2316.