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Atmospherically Deposited PBDEs, Pesticides, PCBs, and PAHs in Western U.S. National Park Fish: Concentrations and Consumption Guidelines

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Concentrations of polybrominated diphenyl ethers (PBDEs), pesticides, polychlorinated biphenyls (PCBs), and polycyclic aromatic hydrocarbons were measured in 136 fish from 14 remote lakes in 8 western U.S. National Parks/Preserves between 2003 and 2005 and compared to human and wildlife contaminant health thresholds. A sensitive (median detection limit, −18 pg/g wet weight), efficient (61% recovery at 8 ng/g), reproducible (4.1% relative standard deviation (RSD)), and accurate (7% deviation from standard reference material (SRM)) analytical method was developed and validated for these analyses. Concentrations of PCBs, hexachlorobenzene, hexachlorocyclohexanes, DDTs, and chlordanes in western U.S. fish were comparable to or lower than mountain fish recently collected from Europe, Canada, and Asia. Dieldrin and PBDE concentrations were higher than recent measurements in mountain fish and Pacific Ocean salmon. Concentrations of most contaminants in western U.S. fish were 1–6 orders of magnitude below calculated recreational fishing contaminant health thresholds. However, lake average contaminant concentrations in fish exceeded subsistence fishing cancer thresholds in 8 of 14 lakes and wildlife contaminant health thresholds for piscivorous birds in 1of 14 lakes. These results indicate that atmospherically deposited organic contaminants can accumulate in high elevation fish, reaching concentrations relevant to human and wildlife health.

Introduction

Organic contaminant measurements in environmental compartments, such as snow (*1–3*), water (*3–5*), sediment (*6, 7*), vegetation (*8, 9*), and fish (*10–16*), have demonstrated that cold, high elevation ecosystems can selectively accumulate some semivolatile and persistent organic pollutants (*4*). Semivolatile organic compounds (SOCs) volatilize at warmer temperatures and condense or are scavenged to the earth's surface more readily at cold temperatures (*17, 18*). Furthermore, SOCs can be magnified across elevational, temperature, and precipitation gradients (*1, 18*) as well as up food chains. Additionally, pesticide deposition in the annual snowpack of western U.S. high-elevation ecosystems has been positively associated with proximity to past and current agricultural activity (*2*). Although enhanced accumulation of SOCs in high-elevation fish has been observed in European mountains (*10–12*) and the Canadian Rockies (*13*), there are very limited data on the accumulation of SOCs in high-elevation fish across the western U.S. (*15, 19, 20*), and what, if any, risks these contaminants may pose.

Chemical fate and transport studies in ecosystems benefit from the measurement of multiple SOCs with different sources and/or physical−chemical properties which aid in understanding mechanisms of chemical emission, distribution, transport, uptake, and degradation (*16, 21*). Because fish often contain the highest concentrations of organic contaminants in aquatic ecosystems (*22*), and because they are relevant to human as well as wildlife health, fish are an important environmental compartment for ecosystem fate and transport studies.

To date, most multicontaminant methods for fish have focused on similar classes of SOCs (*23, 24*) or are used as commercial food safety screening methods with high part-per-billion (ppb) detection limits (*25, 26*). These methods can fail to detect analytes in a large number of environmental samples (*27, 28*). Few methods have been shown to quantify more than two classes of SOCs at concentrations below 1 ppb (1 ng/g) in fish (*29*). Because pesticides and other SOCs are often estimated at less than 1 ppb in fish tissues, analytical methods are needed to quantify SOCs in fish at these concentrations (*10, 11, 30*).

Our objectives were to develop and validate an analytical method to measure polybrominated diphenyl ethers (PBDEs), pesticides, polychlorinated biphenyls (PCBs), and polycyclic aromatic hydrocarbons (PAHs) in fish, including concentrations below 1 ppb, and to assess if measured SOC concentrations in fish from remote western US. National Park/Preserve lakes were relevant to human and wildlife health.

Experimental Section

Sample Collection. A set of 136 fish from 14 lakes in 8 U.S. National Parks/Preserves (∼10/lake; Table S1, Figure S1) were selected for SOC analysis from 212 fish (∼16/lake) in order to achieve roughly equal age and sex distributions. Samples were collected as described previously (*31*) and in the Supporting Information. Brook or lake trout (*Salvelinus fontinalis* (*S. fontinalis*), *S. namaycush*) were primarily collected and, on occasion, cutthroat and rainbow trout (*Oncorhynchus clarki* (*O. clarki*), *O. mykiss*) were collected (Table S1). Burbot and whitefish (*Lota lota* (*L. lota*), *Prosopium cylindraceum* (*P. cylindraceum*) were collected from McLeod

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Lake in Denali National Park because trout were not prevalent in the lake. The average age of the fish collected ranged from 3 to 20 years old, while the average length ranged from 20 to 50 cm, the average mass ranged from 50 to 1400 g, the average growth rate ranged from 13 to 167 g/year, the average condition factor ranged from 0.62 to 1.19 cg/cm^3, and the average sex ratio (M:F) ranged from 0.7 to 4.0 (Table S1).

Field blanks were obtained by exposing foil, bags, and scalpels to air and water in the same manner and time as sample collection. The samples were stored in a −25 °C freezer until analysis. Fish Standard Reference Material (SRM) No. 1946 (Lake Superior lake trout, 1997) was purchased from the National Institutes of Standards and Technology (NIST, Gaithersburg, MD) for method validation. Chemical standards and reagents were purchased, prepared, and stored as described previously (*3*).

Sample Preparation and Analysis. Sample collection and preparation details can be found in the Supporting Information. Whole fish homogenates were analyzed to better assess piscivorous wildlife contaminant exposure. Whole fish and method blanks (150 g of baked sodium sulfate) were homogenized with liquid nitrogen in a food processor. Following the addition of 32 isotopically labeled recovery surrogates, samples (~20 g wet weight (ww), 1.8 g SRM) were extracted using pressurized liquid extraction. Interferants were removed from extracts using silica adsorption chromatography and gel permeation chromatography. Final extracts were reduced under nitrogen and spiked with four isotopically labeled internal standards prior to analysis by gas chromatographic mass spectrometry (GC/MS) with recovery correction from a deuterated or ^{13}C-substituted surrogate analogue (isotope dilution), or the closest eluting isotope-labeled surrogate of a similar chemical class.

Purified extracts were analyzed by GC/MS as described previously (*3, 32*). The SOCs measured, their characteristic ions, and SIM windows, as well as the GC column and oven temperature program were previously described for the electron impact ionization MS (EI-MS) analysis (*3*) and in Table S2 for the electron capture negative ionization-MS (ECNI-MS) analysis. Quality assurance and control procedures were followed, including frequent analysis of calibration checks (25% of analyses), blanks (10%), duplicate injections (10%), and SRM analysis (10%), and are described elsewhere (*3*). All reported sample concentrations had their method blanks subtracted, were corrected by their surrogates' sample specific recoveries, and were censored when below method detection limits or blanks greater than 33% of sample mass.

Method Validation. Estimated method detection limits (EDLs) were calculated in triplicate for three of the 136 fish samples, spanning species (lake, brook, and rainbow trout), age (*3–25*), and lipid ranges (1.8–8.6%) collected from remote lakes in Denali, Sequoia, and Rocky Mountain National Parks, respectively (Table S1). The procedure described in U.S. EPA Method 8280A was used to calculate EDLs from each analyte's signal-to-noise ratios in selected ion chromatograms of each fish extract and standards (*33*). This estimation method was conducted for these three fish samples, (spanning species, age, and location) to assess the consistency of the EDL determination method.

An additional lake trout collected from Wonder Lake in Denali National Park was selected for method recovery experiments due to its moderately high lipid content (10%), relatively low background concentrations of the target analytes, and sufficient sample mass to permit multiple analyses. Background SOC concentrations in this fish homogenate were measured in triplicate prior to triplicate fortification with target analytes (~8 ng/g ww). The three fortified aliquots were processed according to the analytical method except that recovery surrogates were not added to the sample prior to extraction to adjust for method recovery but instead were added to the extract just prior to analysis in order to calculate analyte loss over the entire analytical method. Analyte specific method recoveries were calculated from method blank and background subtracted concentrations in the final extracts of these fortified aliquots.

Method precision and accuracy were assessed using five aliquots of ~1.8 g of NIST SRM 1946, analyzed over a 5 month period. Less mass of the SRM was analyzed than in the U.S. National Park fish due to higher target analyte concentrations in the SRM. The average analyte concentrations measured in NIST SRM 1946 aliquots using this method were compared to certified values for 31 comparable analytes.

Contaminant Health Thresholds. Contaminant specific health thresholds were calculated for recreational and subsistence fishing consumption patterns using U.S. Environmental Protection Agency (EPA) risk assessment documents (*34*). The contaminant specific health thresholds discussed here were calculated fish contaminant concentrations that would likely lead to contaminant consumption exceeding U.S. EPA Integrated Risk Information System (IRIS) reference doses or acceptable cancer risk levels for individual contaminants only, assuming typical recreation or subsistence fish consumption patterns (*35*). Additionally, lake specific advisory fish consumption limits were calculated for recreational and subsistence fishing consumption using U.S. EPA guidance on additive cancer risks (*35*). U.S. EPA default recreational (17.5 g of fish/day) and subsistence fishing consumption rates (142 g of fish/day), adult body mass (70 kg individuals), and acceptable risk levels (lifetime excess cancer risk of 1:100000) were used to calculate the contaminant health thresholds and likely lake specific advisory fish consumption limits (*34*). Contaminant health thresholds for recreational fishing were adjusted to account for differences between the measured whole fish SOC concentrations and likely exposure in trimmed and cooked fish fillets by increasing the contaminant threshold by 32%, equivalent to an average 32% reduction in organochlorine exposure estimated to be achieved due to trimming and cooking (*34*). Subsistence fishing health thresholds were not adjusted for reductions from whole fish SOC concentrations since subsistence consumption patterns are reported to be highly variable and often include the whole fish (soups, stews, etc.) (*34, 36*).

Wildlife contaminant health thresholds were derived from wildlife fish contaminant consumption criteria in the literature (*37*) for nonlethal reproductive and developmental wildlife health end points identified by the U.S. EPA (*38*). For use in this work, the criteria were derived for piscivorous wildlife known to occur in the majority of National Parks and where nonlethal end points were used as indicators of a negative effect.

Results and Discussion

Chromotography and Blanks. This analytical method resulted in fish extract chromatograms with baseline separation of a large majority of the target analytes (Figure S2). In the few cases where complete baseline separation was not achieved, mass-spectral separation was achieved. Most analytes were not detected (nd) in method or field blanks, or measured concentrations were typically estimated below the quantitation limit (lowest point on the calibration curve). However, phenanthrene, PBDE 47, and PBDE 99 were measured in most method blanks with average mass corresponding to fish concentrations of 300, 15, and 25 pg/g, respectively. When analytes were quantified in method blanks, concentrations were less than 5% of sample concentrations for all method recovery and SRM experiments and averaged less than 5% of measured analyte values in western U.S. National Park fish.

Method Validation. Estimated method detection limits for the 91 SOC ranged from 0.2 to 920 pg/g ww, with a median of 18 pg/g ww (Table S3). The EDLs were fairly consistent across the three different fish species, with an average 11% relative standard deviation (RSD; $n = 3$). With the exception of nine PBDEs and endrin, all analyte EDLs were 100 pg/g ww or less. Analyte recovery over the entire method ranged from 31.4 to 98.3%, with an average recovery of 61.4% at environmentally relevant concentrations of ~8 ng/g ww. Analyte recoveries were reproducible, with an average standard deviation of 4.1% ($n = 3$). Individual target analyte recoveries and their surrogate's recoveries in the National Park fish differed by less than 12%, on average, suggesting recovery surrogates were generally representative of target analytes.

Concentrations for 19 of 31 comparable analytes with certified concentrations in NIST SRM 1946 were measured to within the NIST confidence intervals (Table S3). The concentrations of all 31 certified analytes averaged within 7% and ranged less than 30% from NIST certified values (Table S3). Additionally, eight SOCs were measured which were not previously reported and/or certified in SRM 1946 (b-HCH, heptachlor, endrin, dacthal, endosulfan I, endosulfan sulfate, PBDE 155, PBDE 183). The polarity range of compounds measured in the SRM demonstrates the utility of this analytical method for measuring a broad range of SOCs, at low concentrations in fish tissues.

SOCs in Western U.S. National Park Fish. The 10 most concentrated SOCs measured in >75% of fish from western U.S. National Parks were p,p-dichlorodiphenylethane (p,p-DDE), dieldrin, PBDE 47, 99, PCB 153, 138, dacthal, *trans*-nonachlor, hexachlorobenzene (HCB), and endosulfan sulfate, respectively. Dieldrin, *p,p′*-DDE, dacthal, and endosulfan sulfate concentrations were highest in Sequoia, Rocky Mountain, and Glacier National Park fish (Figure 1C,D). PBDE concentrations in fish across western U.S. National Parks varied less than most other SOCs, both within and between lakes. PBDE concentrations were highest in Mt. Rainier National Park fish and lowest in the Alaskan National Park fish. The fish concentrations of five major PCB congeners were comparable between the Alaskan and Pacific Coast National Park fish (Sequoia, Olympic, and Mt. Rainier National Parks) and lower in fish from the Rocky Mountains (Glacier and Rocky Mountain National Parks) (Figure 2A). In general, current use pesticide concentrations were highest in fish from Sequoia National Park, followed by Rocky Mountain and Glacier National Parks (Figure 1A,B) and lower in Pacific Northwestern (Olympic and Mt. Rainier National Parks) and Alaskan National Park fish (Denali and Noatak National Park/Preserve).

For most compounds, the variation in the fish SOC concentrations within lakes was almost as large as the variation in concentrations between lakes. Concentrations for only 28 of the most frequently detected SOCs are presented here. Concentrations of PAHs and other SOCs measured are not shown here because they were detected in less than 50% of samples, likely due to low ambient concentrations and rapid transformation and/or elimination from fish (*39*).

Comparisons to SOCs in Other Mountain Fish. When compared to salmonid fishes collected from similar high-elevation lakes throughout Europe (*10*), PBDE concentrations measured in western U.S. National Park fish were, on average, 3 times higher in concentration after adjusting for typical differences between muscle and whole tissue concentrations (*34*) (Table 1). Concentrations for most historic use SOCs (HCB, DDTs, and HCHs) were 2–9 times lower in western U.S. fish than European mountain fish (*11*). Because the European mountain fish and western U.S. National Park fish studied here were salmonids of similar age and condition (~6 years, ~1.0 $cg \cdot cm^{-3}$) collected from similarly cold, oligotrophic lakes within 3 years, it is unlikely that the observed differences in the SOC concentrations are due to fish accumulation differences or potentially changing PBDE emissions. PBDE accumulation in other European (*40*) and North American (*41*) aquatic ecosystems likely underwent a slowing, if not slight decline, during this time period. This indicates that PBDE emission trends do not account for the observed PBDE concentration differences. This may suggest that fish from western U.S. National Parks are exposed to higher PBDE concentrations than similar European mountain fish, which is consistent with PBDE concentrations measured in other North American and European environmental compartments (*42*), and more recent European fish samples (*16*).

When compared to fish collected from a transect of mountain lakes in Canada (*13*), HCHs and chlordanes were, on average, 4–5 times higher in concentration in the Canadian mountain fish (Table 1). The ΣDDTs and HCB concentrations in Canadian fish were comparable to western U.S. National Park fish, while dieldrin was 3 times higher in concentration in western U.S. National Park fish. The fish sampled in Canada and the western U.S.Parks were similar species (primarily brook, lake, and rainbow trout), sizes (~30 cm, ~425 g), and ages (~6 years) and had similar lipid levels (~3.7%). In addition, the samples were collected within a 4 year period from similar oligotrophic lakes located at similar elevations and had similar site temperatures. Because of these similarities, and because dieldrin and chlordanes are similarly persistent and bioaccumulative, the higher dieldrin and lower chlordane concentrations are unlikely to be due solely to fish accumulation differences. This suggests that fish in western U.S. Parks may be exposed to higher concentrations of dieldrin and/or lower concentrations of chlordanes and HCHs than similar fish from the southern Canadian Rocky Mountains. Elsewhere, fish collected from high-elevation lakes and rivers in the Asian Tibetan Plateau (*14*) had comparable DDT concentrations and 4–9 times higher HCB and HCH concentrations than these western U.S. fish (Table 1).

Although dacthal and endosulfan sulfate were among the most concentrated and frequently detected SOCs in this study, it is not known if their concentrations in western U.S. high elevation fish are comparable to or higher than fishes from other regions of the world due to limited reports in the literature. Together, these comparisons suggest that fish in western U.S. National Park lakes may be exposed to comparable or lower concentrations of most historic use pesticides than similar fish around the world, but higher concentrations of PBDEs and dieldrin.

Comparisons to SOCs in Pacific Salmon. The average concentrations of DDTs, dieldrin, and PBDEs were 2–5 times higher in these western U.S. National Park fish than ocean-caught Pacific salmon (*Oncorhynchus* spp), while HCB, HCH, and chlordane concentrations were 3–19 times lower (Table 1) (*43, 44*). Differences in the year of fish collection and analytical methodology likely do not account for these concentration differences because most samples were collected within a 3 year period and the analytical methods are similar, including the use of isotope-labeled surrogate recovery GC/MS (*44*). Because the accumulation of these organohalogens in fish is similarly affected by differences that affect fish lipid and age, it is not likely that lipid and age differences would explain both higher and lower SOC concentrations in the salmon. Differences in food webs, as well as ambient environmental concentrations, are likely responsible for the different SOC concentrations observed. This suggests that DDTs, dieldrin, and PBDEs are accumulated to higher concentrations in fish from high-elevation aquatic ecosystems of western U.S. National Park

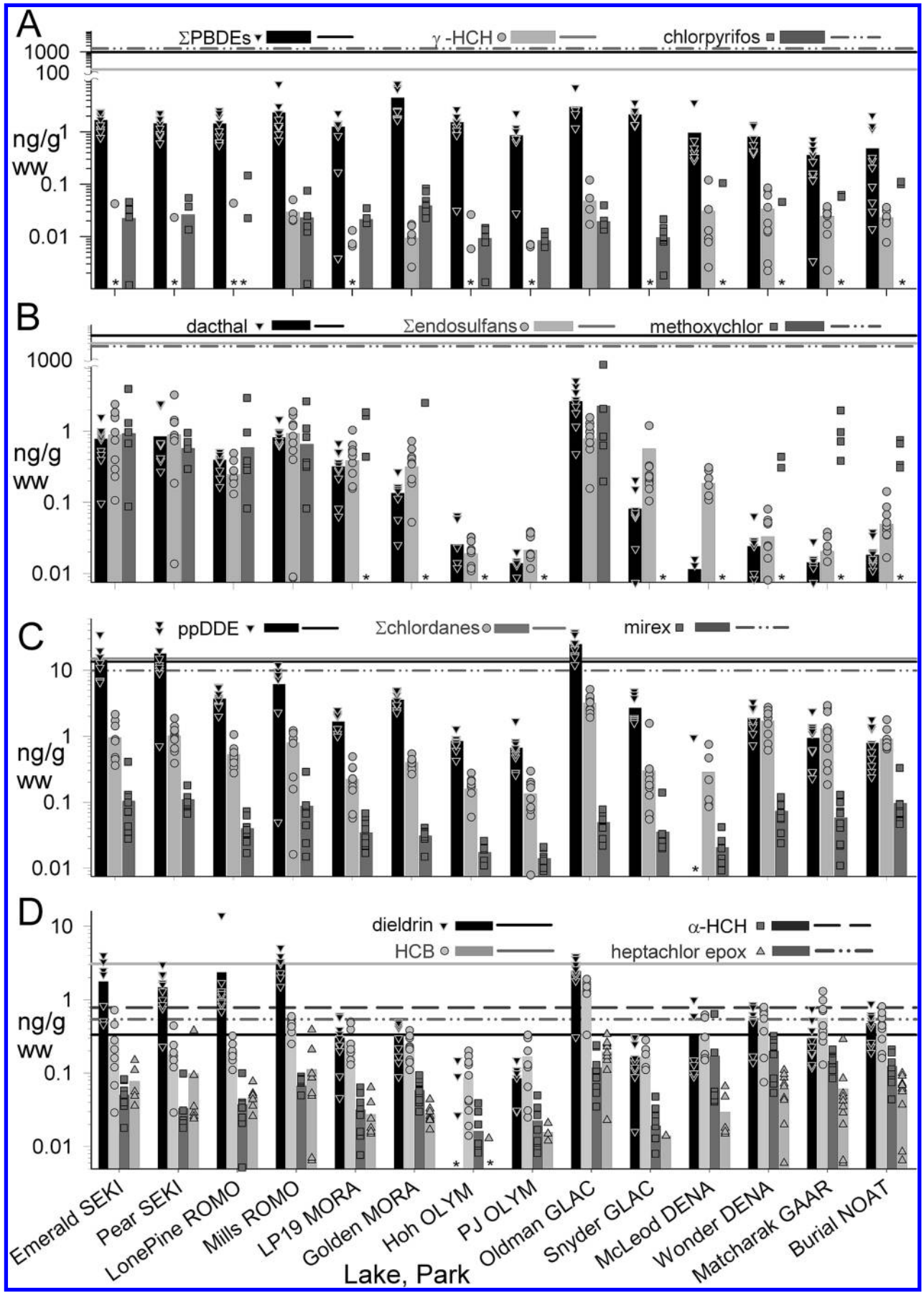

FIGURE 1. Concentrations of select current (A, B) and historic use SOCs (C, D) in individual fish (symbols) and lake average fish (bars) from western U.S. National Parks compared to estimated noncancer (A, B) and cancer (C, D) contaminant health thresholds for subsistence fishing (lines). ∗, nd > 50% of lake fish; SEKI, Sequoia; ROMO, Rocky Mountain; MORA, Mt. Rainier: OLYM, Olympic; GLAC, Glacier; DENA, Denali; GAAR, Gates of the Arctic National Parks; and NOAT, Noatak National Preserve.

TABLE 1. Comparison of Recent Contaminant Concentrations in Fish Muscle from Remote Areas (ng/(g ww))

location	concentration									
	ΣPBDE(av range)	dieldrin(av range)	HCB(av range)	ΣDDT(av range)	ΣHCH(av range)	Σchlordane(av range)	ΣPCB(av range)	*n*	year	ref.
Western U.S. Parks[a,b]	1.1(0.18–5.7)	0.68(0.01–9.5)	0.26(0.01–1.3)	4.3(0.16–34)	0.063(0.001–0.52)	0.058(0.005–3.5)	1.1(0.10–4.6)	136	2003–2005	this study
Tibetan Plateau	n/a[c]	n/a	1.0(0.31–3.2)	6.3(0.78–23)	0.91(0.13–2.6)	n/a	n/a	20	2005	*d*
European Mountains	0.38(0.07–1.1)	n/a	0.42(0.14–1.0)	19(0.25–65)	0.54(0.10–1.6)	n/a	6.3(0.68–17)	163	2000–2001	*e, f*
Canadian Mountains	n/a	0.23(nd[h]–1.0)	0.35(0.014–1.9)	5.6(0.17–52)	0.25(0.023–1.2)	0.23(0.03–1.0)	7.7(0.5–59)	91	1997, 2001, 2003	*g*
Pacific salmon	~0.2(~0.04 to ~5)	~0.3(~0.1 to ~1)	~0.8(~0.3 to ~3)	~2(~0.8 to ~10)	~0.75(~0.4 to ~11)	~1(~0.4 to ~2.75)	~2.5(~1.1 to ~10)	47	2002	*i, j*

[a] Muscle tissue concentrations estimated using 68% whole fish concentrations (32% reduction). [b] ΣPCB = PCB 74, 101, 118, 138, 153, 183, and 187. [c] n/a, not available. [d] Ref *14*. [e] Ref *11*. [f] Ref *10*. [g] Ref *13*. [h] nd, not determined. [i] Ref *43*. [j] Ref *44*.

lakes than salmon from the adjacent Northeast Pacific Ocean and its tributaries.

Human Fish Consumption Criteria. Dieldrin and/or *p,p′*-DDE concentrations in just over half (77 of 136) of the individual fish (in 11 of 14 lakes) exceeded calculated subsistence fishing human contaminant health thresholds. Lake average dieldrin and/or *p,p′*-DDE concentrations in fish exceeded the contaminant health thresholds for subsistence fishing for 8 of 14 lakes. These lakes are located in Sequoia, Rocky Mountain, Denali, and Glacier National Parks, as well as Noatak National Preserve (Figure 1C,D). The concentrations of 11 other SOCs (with IRIS toxicity data) ranged 1−6 orders of magnitude below contaminant health thresholds for subsistence fishing (Figure 1). Additive lifetime cancer risks from each lake's average fish SOC concentrations exceeded the acceptable risk level at the subsistence fishing consumption level in all but three lakes (Table S4) largely due to cancer risk contributions from dieldrin concentrations. Additive cancer risk of subsistence fishing consumption was calculated by multiplying the fish SOC concentrations by the assumed consumption rates and each SOCs' cancer slope factor, dividing it by the assumed body mass (70 kg), and summing these "risks" for all SOCs. Additive cancer risk exceeded acceptable thresholds when a lake's average fish SOC concentrations increased the lifetime risk of cancer by more than 1:100000 (0.001%).

Lake average SOC concentrations in western U.S. National Park fish were below recreational fishing contaminant health thresholds for all 13 SOCs with IRIS toxicity data (Figure S3). However, average dieldrin concentrations in fish from 4 of the 14 lakes (Rocky Mountain, Glacier, and Sequoia National Parks) were within a factor of 1.5 of the contaminant health thresholds, and the concentrations of dieldrin in 13 individual fish (Rocky Mountain, Glacier, and Sequoia National Parks) exceeded the lifetime cancer contaminant health threshold for recreational fishing (Figure S3D). No other fish SOC concentrations measured in western U.S. National Parks exceeded recreational fishing contaminant health thresholds and average concentrations in fish ranged 1−7 orders of magnitude below these health thresholds (Figure S3). Additive lifetime cancer risks from average fish SOC concentrations did not exceed the acceptable risk threshold for recreational fishing at any of the 14 western U.S. National Park/Preserve lakes (Table S4).

Calculated contaminant health thresholds for subsistence fishing were about 1 order of magnitude lower than recreational fishing due to higher estimated consumption rates and whole fish versus muscle consumption scenarios (*34, 36*). It should be noted that the likelihood of the above risks being realized at these sites is limited by the remote locations, which limits the number of people exposed and the extent of exposure. Also, the confidence in the cancer slope factor for dieldrin has been classified as "low" (*35*), and there is some debate about the accuracy in the use of upper-bound cancer slope factors and additive cancer risks employed in the U.S. EPA risk assessment methods (*45*). Together, this suggests that the human health risks from fish-SOC ingestion could be lower than those calculated here (*45*). Although this risk estimation methodology does not fully include potential SOC interactions beyond cancer risk additivity, it remains the sole health based consumption advice that uniformly treats lifetime health risk due to fish contaminant consumption for such a broad range of chemicals. The contaminant health thresholds were risk estimations and did not characterize nutritional or health benefits associated with salmonid consumption, such as cardiac health risk reduction from increased omega-3 fatty acid consumption or potential dietary reduction in unhealthy fats due to food substitutions.

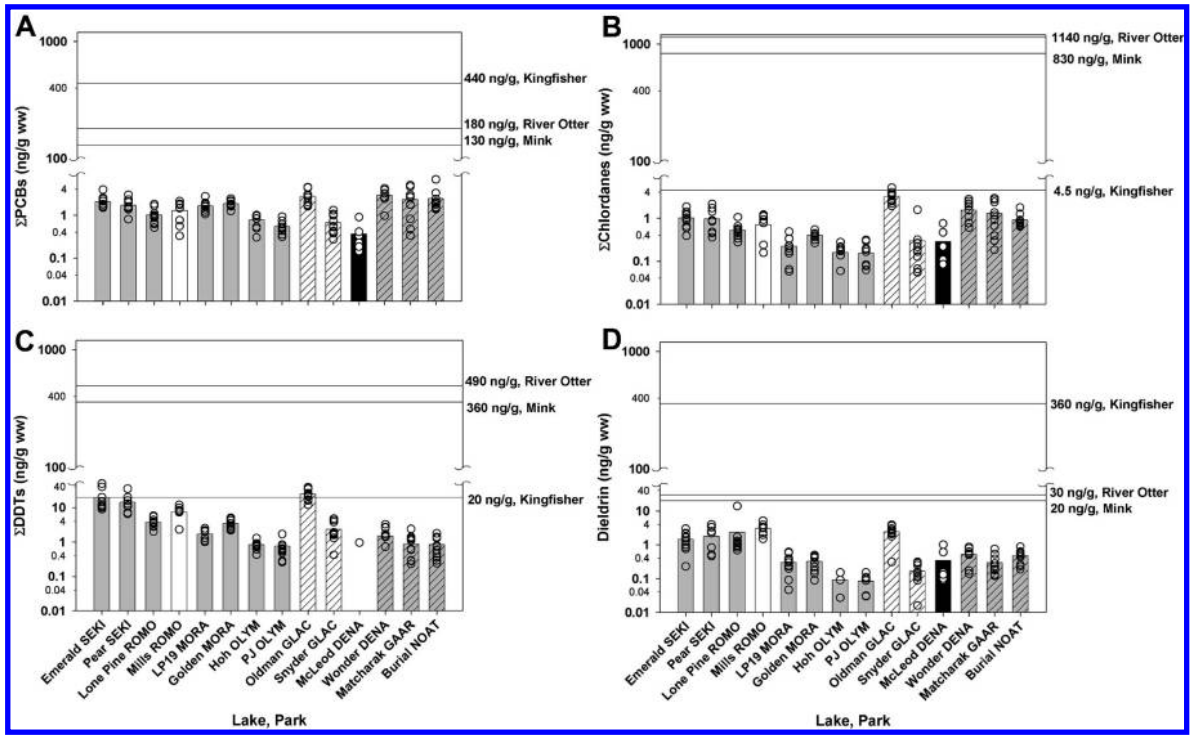

FIGURE 2. Concentrations of PCBs (A), chlordanes (B), DDTs (C), and dieldrin (D) in individual whole fish (symbols) and lake average fish (bars) from Western U.S. National Park lakes, compared to wildlife contaminant health thresholds (labeled lines). Gray bars are *Salvelinus* spp. brook trout (solid) and lake trout (hatched); white bars are *Oncorhynchus* spp. rainbow trout (solid) and cutthroat trout (hatched); the black bar is burbot and whitefish.

These benefits from salmonid consumption may help some fish consumers more than a reduction in contaminant exposure.

Wildlife Fish Consumption Criteria. Lake average ΣDDT concentrations in fish exceeded wildlife contaminant health thresholds for the belted Kingfisher (*Ceryle alcyon*) in Oldman Lake in Glacier National Park (Figure 2C). Chlordane concentrations in several individual fish from this lake also exceeded health thresholds for Kingfishers (Figure 2B), and ΣDDT concentrations in several individual fish from Emerald and Pear Lakes in Sequoia National Park exceeded thresholds for Kingfishers (Figure 2C). Contaminant health thresholds for American mink (*Mustela vison*) and river otters (*Lutra canadensis*) were not exceeded by fish contaminant concentrations in any of the national park lakes studied. Except for Glacier and Sequoia National Parks, contaminant concentrations in fish were sufficiently low to be protective of wildlife health (Figure 2).

Comparing concentrations of historic use pesticides in fish to wildlife health thresholds suggests that some SOC concentrations could negatively impact piscivorous wildlife in certain remote areas. However, it has been cautioned that the wildlife consequences of eating contaminated fish may vary with the health, sex, and reproductive status of the animal (*37*), as well as the amount of time the animals forage from the lake. The wildlife contaminant health thresholds (*37*) were developed using reproductive and developmental end points established by the U.S. EPA (*38*); therefore, observation of changes in population numbers and health would likely be delayed. The nutrition derived from fish consumption is necessary for the survival of largely piscivorous wildlife and a balance exists between nutrition and the detrimental effects of SOCs. Nonetheless, concentrations measured here suggest the potential for negative impact on some wildlife in some locations due to fish-SOC concentrations.

Using this multicontaminant method and U.S. EPA risk methodology, atmospherically deposited SOCs in fish from western U.S. National Parks were measured at concentrations relevant to human and wildlife health. The analytical method was validated to be sensitive, accurate, and reproducible and the concentrations of historic use pesticides were comparable to or lower than those measured in similar fish from Europe, Canada, and Asia. However, PBDE and dieldrin concentrations were uniformly higher in high-elevation fish collected from western U.S. National Parks/Preserves than similar high-elevation fish from elsewhere in the world and Pacific Ocean salmon.

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Supporting Information Available

Further information on methods, samples, sampling locations, and graphs of results described here. This information is available free of charge via the Internet at http://pubs.acs.org.

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