

Levels of Polychlorinated Biphenyls (PCBs) and Three Organochlorine Pesticides in Fish from the Aleutian Islands of Alaska

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Abstract

Background: Persistent organic pollutants (POPs), including polychlorinated biphenyls (PCBs) and chlorinated pesticides, have been shown to have many adverse human health effects. These contaminants therefore may pose a risk to Alaska Natives that follow a traditional diet high in marine mammals and fish, in which POPs bioaccumulate.

Methods and Findings: This study examined the levels of PCBs and three pesticides [*p*, *p'*-DDE, mirex, and hexachlorobenzene (HCB)] in muscle tissue from nine fish species from several locations around the Aleutian Islands of Alaska. The highest median PCB level was found in rock sole (*Lepidopsetta bilineata*, 285 ppb, wet weight), while the lowest level was found in rock greenling (*Hexagrammos lagocephalus*, 104 ppb, wet weight). Lipid adjusted PCB values were also calculated and significant interspecies differences were found. Again, rock sole had the highest level (68,536 ppb, lipid weight). Concerning the PCB congener patterns, the more highly chlorinated congeners were most common as would be expected due to their greater persistence. Among the pesticides, *p*, *p'*-DDE generally dominated, and the highest level was found in sockeye salmon (*Oncorhynchus nerka*, 6.9 ppb, wet weight). The methodology developed by U.S. Environmental Protection Agency (USEPA) was used to calculate risk-based consumption limits for the analyzed fish species. For cancer health endpoints for PCBs, all species would trigger strict advisories of between two and six meals per year, depending upon species. For noncancer effects by PCBs, advisories of between seven and twenty-two meals per year were triggered. None of the pesticides triggered consumption limits.

Conclusion: The fish analyzed, mainly from Adak, contain significant concentrations of POPs, in particular PCBs, which raises the question whether these fish are safe to eat, particularly for sensitive populations. However when assessing any risk of the traditional diet, one must also consider the many health and cultural benefits from eating fish.

Citation: Hardell S, Tilander H, Welfinger-Smith G, Burger J, Carpenter DO (2010) Levels of Polychlorinated Biphenyls (PCBs) and Three Organochlorine Pesticides in Fish from the Aleutian Islands of Alaska. PLoS ONE 5(8): e12396. doi:10.1371/journal.pone.0012396

Editor: Matthew Baylis, University of Liverpool, United Kingdom

Received: September 11, 2009; **Accepted:** March 23, 2010; **Published:** August 25, 2010

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Funding: This work was supported by the Institute for Health and Environment of the University at Albany. Travel support to SH and HT was received from Linköping University. The funders had no role in study design, data collection and analysis, decision to publish or preparation of the manuscript.

Competing Interests: The authors have declared that no competing interests exist.

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Introduction

Polychlorinated biphenyls (PCBs) and organochlorine (OC) pesticides are members of the group of persistent organic pollutants (POPs), an important class of environmental contaminants. POPs are semi-volatile, lipophilic, toxic and persistent in the environment [1–3]. The global spread of POPs and their effects on humans and the environment have been extensively studied during the last decades and measures have been taken to reduce future environmental burdens (e.g. the Aarhus Protocol and the Stockholm Convention) [1]. The Arctic regions are particularly at risk since POPs tend to bioaccumulate in these regions. As a consequence the traditional food, including fish, on which the indigenous people depend, is also contaminated [4].

Atmospheric transport of evaporated POPs is a major pathway for global spread and distribution [5–7]. POPs volatilize from lower latitudes, where they were predominantly used, and are

transported to the Arctic where they are then deposited via precipitation and through other mechanisms [3,8]. This process is called “global distillation” or “cold condensation” and as a consequence POPs tend to concentrate in the polar regions [2,9–11]. POPs can also be transported to the Arctic via the hydrosphere, by rivers and ocean currents [4]. Former military installations in the region are local sources of contamination. An additional factor affecting the levels of POPs in the Arctic is global warming. Permafrost has acted as a sink for POPs, and as it melts the contaminants are released [12]. Eventually, some of the POPs will end up in the sea where they are taken up by biota including phytoplankton, algae, microorganisms, and plants. These species are in turn consumed by fish, resulting in their biomagnification within food chains. Since POPs have a high affinity for lipids and a low elimination rate, they tend to bioaccumulate in fatty tissue [13]. For fish POPs can also be taken up via gill respiration, depending on how lipophilic the contaminants are [4]. Humans

are on top of the food chains and are therefore at risk of being exposed to high levels of contaminants [14–16], with an increase exposure risk to those consuming the Arctic fish that eat higher on the food chain.

PCBs are synthetic organochlorine chemicals that were produced by various companies around the world after 1929. The total amount produced worldwide has been estimated to be as high as 1.326 million tons [17]. PCBs are chemically stable, heat resistant, and have electrical insulating properties and were used in a variety of areas. These include as plasticizers, in carbonless copy paper, lubricating oils, inks, paints, ceiling/floor tiles, window caulking, in nominally closed systems as hydraulic and heat transfer fluids, and as insulators in transformers and electrical capacitors [1,2,18]. The use of PCBs was phased out in northern Europe and North America in the mid-1970s, but they were still produced in Russia until 1993 [1,2,4]. Manufacture and use of PCBs was prohibited by the United Nations Stockholm Convention on Persistent Organic Pollutants [1]. However, despite virtual elimination of production and use, PCB levels in the environment are only slowly declining. One reason is because of the high persistence of PCBs in biological systems. PCBs are also still leaking out from old industrial plants, waste disposal sites and buildings and are migrating from contaminated bodies of water [2,4,18].

There are 209 congeners of PCB, each with different physical activity and biological properties, depending on the number and location of the chlorine atoms [4,18]. Some congeners assume a so-called co-planar configuration, which give them dioxin-like properties [2,18]. The degree of persistence of a congener is influenced by the number of chlorine atoms on the biphenyl rings. In general, highly chlorinated PCB congeners are more stable in the environment than are the lower (1–3 chlorines) chlorinated ones [2,19]. Most PCBs are bound to soil, from which they get into the food supply and bioconcentrate in animal lipids. However, POPs, including PCBs, will also evaporate and can therefore be found in the atmosphere [2,3].

PCBs have many different health effects. They are carcinogenic, cause immune suppression, neurobehavioral effects, hypothyroidism, infertility and reproductive system disorders, cardiovascular disease and elevated serum lipids, hypertension, diabetes, liver disease, asthma, arthritis, and low birth weight [4,18]. Maternal exposure is of special concern since PCBs cross the placenta freely because of their lipophilicity, and the developing fetus is therefore exposed to the same contaminant levels as the mother [20,21]. PCBs are also found in breast milk [22]. Prenatal PCB exposure can reduce the ratio of male to female births and also cause IQ deficits in the baby [17,23]. Exposure has also been shown to increase the risk of testicular cancer in sons of women with higher PCB levels [24].

DDT (dichlorodiphenyltrichloroethane), mirex, and hexachlorobenzene (HCB) are organochlorine pesticides [25,26]. Their use is currently restricted in many countries, even though DDT is still used in mosquito control programs in some malaria endemic tropical areas. Like PCBs, these agents are persistent in the environment. The DDT metabolite, DDE (dichlorophenyldichloroethylene), is the most persistent [4]. The health effects of the organochlorine pesticides are similar to those of PCBs. All have been shown to increase the risk of cancers, have reproductive and developmental effects, cause immunosuppression, decrease IQ, and cause endocrine dysfunction [4,26,27].

The Aleutian Islands extend west from mainland Alaska in the Northern Pacific Ocean, and most became a National Wildlife Refuge in 1913 [28]. Subsistence food is very important for the indigenous people in the Aleutian Islands, accounting for 30–90%

of the diet. Traditionally fish and marine mammals are a principal component of this diet and contaminants in these species are therefore of special concern for the people [29,30]. Commercial fishing is the major source of income for the Aleuts; Dutch Harbor in the Aleutian region had the highest tonnage of fish landings in the world in 2002 [4]. Therefore the contaminants in the fish potentially affect many populations in addition to the Aleuts.

In recent years, the diet of many indigenous people in the Arctic has changed. This is partly due to an increased availability of imported foods and partly to fear of contaminants in subsistence foods [30–32]. However, the Alaska Division of Public Health has expressed concern that changing diet and lifestyle could impose a greater risk than the contaminants themselves [31]. Today, unrestricted fish consumption is recommended for everyone except women who are or can be pregnant, nursing mothers, and children aged 12 years and under. For these groups there are some recommended restrictions regarding certain species, even though unrestricted consumption is still recommended for the species known to be low in mercury [33]. However, in order to provide up to date consumption advice, the Alaska Division of Public Health states that “there is a pressing need to obtain additional information regarding PCB concentrations in Alaskan subsistence foods and sport fish” [34].

The metal levels in subsistence food from the Aleutians have been extensively studied [28,29,35–39]. There are also a number of previous studies of organochlorine contaminants in Alaskan communities [10,40]. However, there has been a lack of studies concerning POPs in the Aleutians in particular. In this paper, we examine the levels of PCB and the OC pesticides *p*, *p'*-DDE, HCB, and mirex in muscle tissue of nine different fish species from the Aleutian Islands in Alaska. The aim of the study was to determine whether the levels of POPs in Aleutian fish pose risks to the people that consumed the fish on the basis of fish consumption advisories developed by the U.S. Environmental Protection Agency (USEPA), and whether there were interspecies differences in the levels of contaminants.

Materials and Methods

Study sites

Fish were collected from the waters surrounding Adak ($n = 56$), Amchitka ($n = 4$), Atka ($n = 8$) and Kiska ($n = 1$) Islands in the Aleutian Chain of Alaska. Adak is located 1,850 km west-southwest of Anchorage, Alaska, and has been a site of U.S. military activity during and since World War II. A large Naval Air facility was located on the island from 1942 until 1997. Today about 200 people live in the island and there is an airport and a harbor [29,37,38]. Amchitka Island has also been subjected to military activities; it was a military base during World War II and it is one of the sites that comprise the Department of Energy’s “Nuclear Weapons Complex”. The USA detonated underground tests three times here between 1965 and 1971 [39]. Amchitka is uninhabited today, but the Aleutian people still consider it part of their homeland and fishermen sometimes go fishing near the island [36,39]. Atka, on the other hand, has a population of about 100 people and there is a harbor on the island. Kiska is an uninhabited island but is also traditional Aleut homeland. It was occupied by Japanese military during the World War II and has been abandoned since [35].

Sample collection

Fish were collected from the four islands in 2004 by the Consortium for Risk Evaluation with Stakeholder Participation (CRESP) primarily to examine whether radionuclides remaining

from the underground nuclear tests posed a risk to seafood safety [41]. The fish were caught either from land or boat with rod and reel by Aleuts and by scientists, and also by underwater spearing by scientists. A previous study has shown that there were few differences in the types and sizes of fish collected by Aleuts and scientists [42]. The number of samples chosen for analysis was a result of selecting among the fish collected that reflect the species Aleuts consume regularly. Fish were immediately measured, weighted and dissected, and samples of muscle were frozen for later analysis. A total of 69 samples from 9 different species were analyzed individually (Table 1). The samples used in this study have previously been analyzed for metals by the Environmental and Occupational Health Sciences Institute of Rutgers University [28,29,36].

Chemicals and Glassware

The solvents used for extraction and sample clean-up were hexane and acetone, both HPLC grade, and both were purchased by lot and analyzed prior to receipt to verify purity. The purity check consists of reducing 500 ml of solvent to 0.5 ml in an evaporator bottle with a 1.5 ml graduated stem (Labconco, Inc., Kansas City, MO) on an N₂ evaporator unit (Rapid Vap, Labconco, Inc.), followed by gas chromatography (GC) with electron capture detection (ECD) analysis. Fully activated Florisil (magnesium silicate) was purchased by lot and deactivated to a final concentration of 4%. The deactivation was executed via the dropwise addition of distilled/deionized water with constant shaking, followed with rolling over night. The Florisil was then calibrated with an extraction column and the addition of 1 ml of 20-ng/ml mixed Aroclor calibration standard to 10 g of Florisil, topped with ~2 g of anhydrous sodium sulfate, and then eluted with hexane. A 30 ml fraction was first collected followed by three 10 ml fractions, which were then individually analyzed by GC-ECD to determine the necessary volume to elute the congeners. Anhydrous sodium sulfate was baked in a muffle oven at 450°C for 4 hours and stored in a hexane-washed glass container with a Polytetrafluoroethylene (PTFE) lid. Reusable glassware including flasks, beakers, concentration tubes, and chromatography columns were soap and water washed, dried, and baked at 450°C for 4 hours to minimize residual contamination.

Instrumentation and Analysis

The details of the analytical procedure used for analysis of PCBs and three pesticides have been published [43]. This reference lists

the 101 specific congeners, their detection limits and relative retention times, the QA/QC procedures applied, and shows representative chromatograms. High resolution, ultratrace, congener-specific analysis was performed using a parallel dual-column (splitless injection) gas chromatography (GC) with electron capture detection (ECD). This method quantitates eighty-three individual PCB congeners and eighteen congeners as pairs or triplets. In addition three OC pesticides are detected, *p*, *p'*-DDE, mirex, and HCB, *p*, *p'*-DDE coelutes with PCB congener 85 under the present GC conditions. A Hewlett-Packard Ultra II 5% phenylmethyloctadecylsilyl-bonded (DB-5) fused silica (25 m, 0.33 μm film thickness, 0.25 mm inner diameter) capillary column and a fused silica Apiezon-L (30m, 0.25 μm film thickness, 0.25 mm inner diameter) capillary column were employed. The instrumentation used was a Hewlett-Packard 6890 GC equipped with electron capture detectors and auto sampler. Helium was used as the carrier gas at ~2 ml/min with a linear flow rate of 30 cm/s. All analysis batches contained a sequence of a hexane blank, QC Aroclor check standard, a congener check standard, the method blank, six unknown samples, and either a method control sample or unknown sample duplicate.

Tissue Extraction

Every extraction batch consisted of a method blank, six unknown samples and either a method control or unknown duplicate sample. Approximately 5 grams of muscle tissue was weighed into a 50 ml Erlenmeyer flask. The method blank was an empty Erlenmeyer flask which was carried through the extraction process, and the method controls were 1 ml of a 200 ppb mixed Aroclor standard in a 50 ml Erlenmeyer flask. 10 ml of 1:1 hexane:acetone, approximately 5 g of anhydrous sodium sulfate, and 5 μl of 1 ng/μl solution of two surrogate standards (IUPAC 125 and 192) were then added to each sample. Samples were then homogenized until the sample and Na₂SO₄ formed a slurry, ~2 minutes, using an Omni International TH tissue homogenizer. The extract was then transferred to a 50 ml volumetric flask using a Pasteur pipette. The hexane/acetone extraction was repeated twice and the extract transferred to the appropriate volumetric flask. The volume of the collected extracts was brought to a final volume of 50 ml with hexane. 5 ml of each of the combined extracts was placed in tarred weighing tins for the gravimetric determination of the total fat in each sample. The remainder of each extract was transferred to 600 ml evaporator bottles with

Table 1. Trophic level and lipid content of the analyzed fish species.

Common name	Scientific name	Trophic level	Number	Lipid content muscle, mean (%)	Lipid content muscle, range (%)
Sockeye salmon	<i>Oncorhynchus nerka</i>	low	13	5.8	1.3–18.8
Pacific cod	<i>Gadus macrocephalus</i>	high	7	0.38	0.090–0.54
Rock sole	<i>Lepidopsetta bilineata</i>	medium	8	0.47	0.29–0.65
Flathead sole	<i>Hippoglossoides elassodon</i>	medium	12	0.36	0.19–0.54
Dolly varden	<i>Salvelinus malma</i>	low	8	2.5	1.1–5.4
Great sculpin	<i>Myoxocephalus polyacanthocephalus</i>	high	12	0.60	0.29–1.1
Rock greenling	<i>Hexagrammos lagocephalus</i>	medium	6	0.47	0.37–0.58
Black rockfish	<i>Sebastes melanops</i>	medium	2	0.58	0.58–0.59
Pacific halibut	<i>Hippoglossus stenolepis</i>	Medium	1	4.7	NA ¹

¹Not available.

doi:10.1371/journal.pone.0012396.t001

1.5 ml graduated stems (Labconco, Inc., Kansas City, MO) and were then placed on an N₂ evaporator unit (RapidVap, Labconco, Inc.) and concentrated to a volume less than 2 ml.

Sample Cleanup

Extract interferences, including polar lipids, were removed from the sample extracts through Florisil adsorption. Once sample extracts were concentrated to less than 2 ml they were transferred to 1×15 cm glass columns packed with 10 g of calibrated, 4% deactivated Florisil, and were overlaid with ~2 g of anhydrous sodium sulfate. The column was then eluted with hexane until the predetermined volume of eluate was collected. This volume was based on the prior calibration of the Florisil. Sample eluates were transferred to clean evaporator bottles and concentrated to a volume of less than 1.0 ml. Samples were brought to a final volume of 1 ml with hexane and transferred to glass GC vials where 1 µl of 1ng/µl IUPAC 104 solution in hexane was added. Each sample was then split into a second GC vial and capped with a PTFE-lined silicon rubber GC vial cap prior to analysis.

Statistical Analysis

The non-parametric Kruskal-Wallis test was used to analyze the overall difference between the species for the agents. If a statistically significant result was found, subsequent analyses using Wilcoxon rank-sum test with Bonferroni correction were performed for pairwise comparisons between the species. The significance level was set at 0.05. All analyses were done using StataSE 10.1 (Stata/SE 10.1 for Windows; StataCorp., College Station TX).

Calculation of risk-based consumption limits

To derive advisory consumption recommendations for the fish species analyzed, the methodology developed by USEPA [44] was applied. The equation used for cancer health endpoints is based on the cancer slope factors for each compound (Table 2), and calculated to prevent 1 excess cancer in 100,000 over a 70-year exposure:

$$CR_{lim} = \frac{ARL \times BW}{CSF \times C_m}, \quad (1)$$

where CR_{lim} is the maximum allowable fish consumption rate (kg/d), ARL the maximum acceptable individual lifetime risk level (10⁻⁵, unit-less), BW is the consumer body weight (70 kg), C_m the measured concentration of a chemical contaminant *m* in a given species of fish (mg/kg), and CSF is the cancer slope factor [(mg/kg-d)⁻¹].

For non-cancer endpoints, based on the reference dose for each compound (Table 2), the following equation was used:

$$CR_{lim} = \frac{RfD \times BW}{C_m}, \quad (2)$$

where CR_{lim} is the maximum allowable fish consumption rate (kg/d), RfD the reference dose (mg/kg-d), BW the consumer body weight (70 kg), and C_m is the measured concentration of chemical contaminant *m* in a given species of fish (mg/kg).

To calculate the allowable number of fish meals of a specified meal size that may be consumed over a given time period, equation (3) was used:

$$CR_{mm} = \frac{CR_{lim} \times T_{ap}}{MS}, \quad (3)$$

where CR_{mm} is the maximum allowable fish consumption rate (meals per year), CR_{lim} is the maximum allowable fish consumption rate (kg/d), T_{ap} the time averaging period (365.25 days per year), and MS is the meal size (0.227 kg fish/meal).

Equation (4) was used to calculate CR_{lim} for carcinogenic health endpoints for the mixture of all the analyzed contaminants in each species.

$$CR_{lim} = \frac{(ARL)(BW)}{\sum_{m=1}^X C_m(CSF_m)}, \quad (4)$$

This equation assumes that the carcinogenic effects of the contaminants are additive. Meal consumption limits for mixtures of carcinogens were then calculated using equation (3).

Toxic equivalency factors (TEFs) were calculated using the 2006 TEF values [45] and based on the concentrations of the 5 dioxin-like PCB congeners that were analyzed in this study. These are 0.0001 for PCB 77 and 0.00003 for congeners 105, 114, 118, and 156. In total, 12 congeners have been recognized by WHO [45] for determining the toxic equivalencies (TEQs) in relation to 2,3,7,8-tetrachlorodibenzo-p-dioxin, but the others, including some of the most potent congeners, were not measured in our method. Equations (1) and (3) were then used to calculate consumption limits concerning TEQ.

Results

PCB levels ranged from 5.8 ng/g (ppb), wet weight, in a sockeye salmon sample to 776 ng/g (ppb), wet weight, in a Pacific cod sample. The highest median PCB levels were found in rock sole (285 ppb, wet weight) and the lowest levels were found in rock

Table 2. USEPA risk values [43], used in risk-based consumption limits.

Contaminant	RfD ¹ (mg/kg-d)	CSF ² (mg/kg-d) ⁻¹
Total PCBs	2×10 ⁻⁵	2.0
Mirex	2×10 ⁻⁴	NA ³
Hexachlorobenzene	8×10 ⁻⁴	1.6
Total DDT (sum of 4,4'- and 2,4'- isomers of DDT, DDE, and DDD)	5×10 ⁻⁴	0.34
Dioxins/furans	NA ³	1.56×10 ⁵

¹Reference Dose.

²Cancer Slope Factor.

³Not available.

doi:10.1371/journal.pone.0012396.t002

greenling (104 ppb, wet weight) (Figure 1). No significant interspecies differences were found regarding total PCB levels, however, the differences were statistically significant for the lipid adjusted PCB levels (Table 3). The median lipid adjusted PCB level was highest in rock sole (68,536 ppb, lipid weight) and lowest in sockeye salmon (3,246 ppb, lipid weight) (Figure 2). Salmon and dolly varden, the two species lowest on the food chain, had the lowest lipid adjusted PCB levels.

Black rockfish and Pacific halibut were excluded from the statistical analysis, as well as from the calculations of median values, since there were too few samples of these species. The mean PCB level for the two black rockfish samples was 15 ppb, wet weight, and the level in the single Pacific halibut sample was 364 ppb, wet weight.

The more highly chlorinated congeners dominated in all species except for black rockfish which had a relatively high proportion of lower chlorinated congeners. In general, the most prevalent congeners expressed as percent of total PCBs were IUPAC congeners 164+163+138, 153, 180, 90+101, 187, 118, 132, and 170 (Figure 3).

Among the pesticides, *p, p'*-DDE (measured as the sum of *p, p'*-DDE and PCB congener 85) dominated with median values ranging from 1.1 ppb, wet weight, in dolly varden to 6.9 ppb, wet weight, in sockeye salmon. The median values for HCB ranged from 0.19 ppb, wet weight, in flathead sole to 2.1 ppb, wet weight, in salmon. The levels of mirex were generally lowest, with median values ranging from 0.14 ppb, wet weight, in rock greenling to 0.68 ppb, wet weight, in rock sole. Significant interspecies differences were found for HCB and mirex (Table 3). Again, black rockfish and Pacific halibut were excluded from the analysis.

Figure 4 shows a calculation of consumption limits based on the risk for noncancer health endpoints considering PCB. The calculations were based on equations (2) and (3) as recommended by USEPA [44]. Black rockfish and Pacific halibut were excluded from all calculations of consumption limits, because of the limited

sample size. The most restrictive advisory was triggered by rock sole, for which 7–8 meals can be consumed each year. Rock greenling triggered the least restrictive advisory of 21–22 meals per year.

Risk-based consumption limits for noncancer health endpoints concerning the pesticides were also calculated using equations (2) and (3). These results allowed unrestricted consumption.

Figure 5 shows the result for carcinogenic health endpoints for PCBs alone and for the mixture of PCBs, HCB, and *p, p'*-DDE+85, respectively. The calculations were based on equations (1) and (3). As can be seen in the figure, the consumption advisory changed only little when the pesticides were included in the analysis. Compared to the advisories for non-cancer health endpoints, the advisories for carcinogenic health endpoints were more restrictive and ranged from 2 meals per year for rock sole to 5–6 meals per year for rock greenling. For both of these two species the calculated rates were the same for only PCB and for the mixture of PCB and the pesticides.

The calculated total TEQ values ranged from 0.15 pg/g (ppt), wet weight, in rock greenling to 0.58 pg/g (ppt), wet weight, in Pacific cod. The calculated USEPA consumption limits for cancer health endpoints concerning dioxin-like compounds ranged from 12–13 meals per year for Pacific cod to 49–50 meals per year for rock greenling (Figure 6).

Discussion

Many of the fish species studied are important parts of the traditional diet for the Aleutian people [46]. Fish is a staple food item in the Aleutians and a study in one of the islands concluded that at least 19 different species of fish were consumed by the people [36,46]. Salmon, halibut, dolly varden, and cod are among the more popular species, with fish estimated to account for 59% of the subsistence harvests [30,31,46]. The mean daily intake of fish and shellfish of Alaska Natives has been calculated to be 109 g,

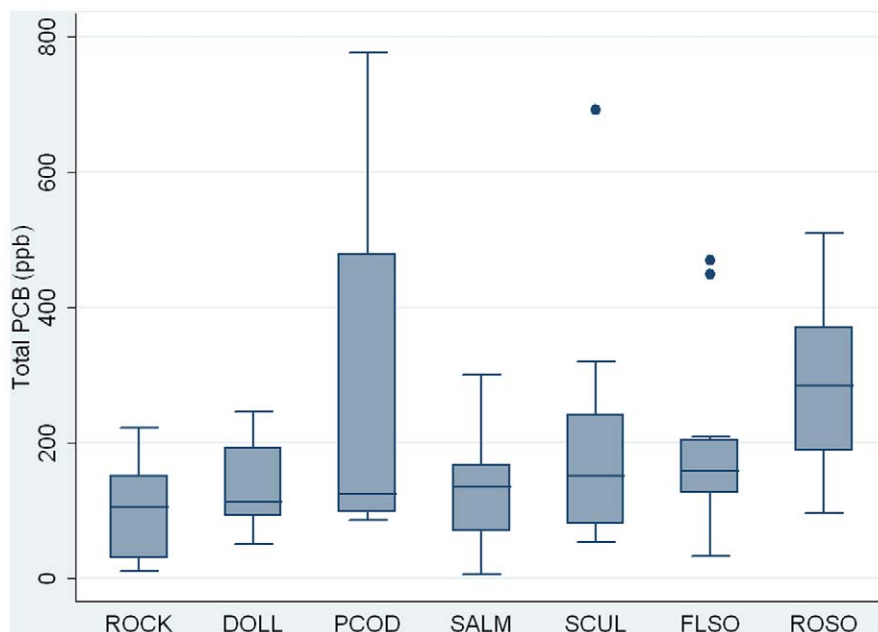


Figure 1. Box plot showing total PCB concentrations. Concentrations are in ng/g (ppb), wet weight. The box represents 50 percent of the values. The median, as well as the sample maximum (excluding outliers) and minimum are shown. Outliers are represented by dots. The species are arranged after median values in ascending order. Black rockfish and Pacific halibut were excluded. ROCK, rock greenling. DOLL, dolly varden. PCOD, Pacific cod. SALM, sockeye salmon. SCUL, great sculpin. FLSO, flathead sole. ROSO, rock sole. doi:10.1371/journal.pone.0012396.g001

Table 3. Contaminant levels and p values of the fish species included in the statistical analysis.

Contaminant	Sockeye Salmon, median (range)	Pacific Cod, median (range)	Rock Sole, median (range)	Flathead Sole, median (range)	Dolly Varden, median (range)	Great Sculpin, median (range)	Rock Greenling, median (range)	P ¹
Total PCBs	135 (5.8–301)	124 (85–776)	285 (96–510)	158 (32–470)	113 (50–245)	151 (54–691)	104 (10–222)	0.15
Total PCBs, lipid adjusted	3246 (234–8131)	40201 (21122–168081)	68536 (16611–96759)	42268 (14514–211555)	5765 (933–16581)	31134 (6833–235078)	22534 (1933–59812)	0.0001 ^a
HCB	2.1 (0.21–11)	0.72 (0.28–1.2)	0.38 (0.21–0.66)	0.19 (0.07–0.44)	0.77 (0.49–1.9)	0.92 (0.27–1.7)	0.59 (0.22–0.86)	0.0001 ^a
DDE+85	6.9 (0–56)	2.6 (1.2–7.3)	2.7 (1.6–6.8)	1.4 (0.26–6.1)	1.1 (0.74–8.0)	3.1 (0–6.3)	1.8 (0–3.3)	0.13
Mirex	0.34 (0.02–1.9)	0.25 (0.08–0.53)	0.68 (0.35–1.0)	0.21 (0–0.55)	0.19 (0.05–0.25)	0.24 (0–0.69)	0.14 (0–0.25)	0.002 ^a

Concentrations are in ng/g (ppb), wet weight.

^aSignificant differences² were found between the following species for each of the contaminants below.

Total PCBs, lipid adjusted.

Rock sole and sockeye salmon (p = 0.004).

Flathead sole and sockeye salmon (p < 0.002).

Great sculpin and sockeye salmon (p < 0.002).

Pacific cod and sockeye salmon (p = 0.01).

Rock sole and dolly varden (p = 0.02).

Flathead sole and dolly varden (p = 0.01).

Pacific cod and dolly varden (p = 0.03).

HCB.

Sockeye salmon and flathead sole (p = 0.002).

Dolly varden and flathead sole (p = 0.004).

Great sculpin and flathead sole (p = 0.004).

Pacific cod and flathead sole (p = 0.03).

Mirex.

Rock sole and flathead sole (p = 0.03).

Rock sole and dolly varden (p = 0.02).

Rock sole and great sculpin (p = 0.04).

Rock sole and rock greenling (p = 0.04).

¹Kruskal-Wallis test.

²Wilcoxon rank-sum test.

doi:10.1371/journal.pone.0012396.t003

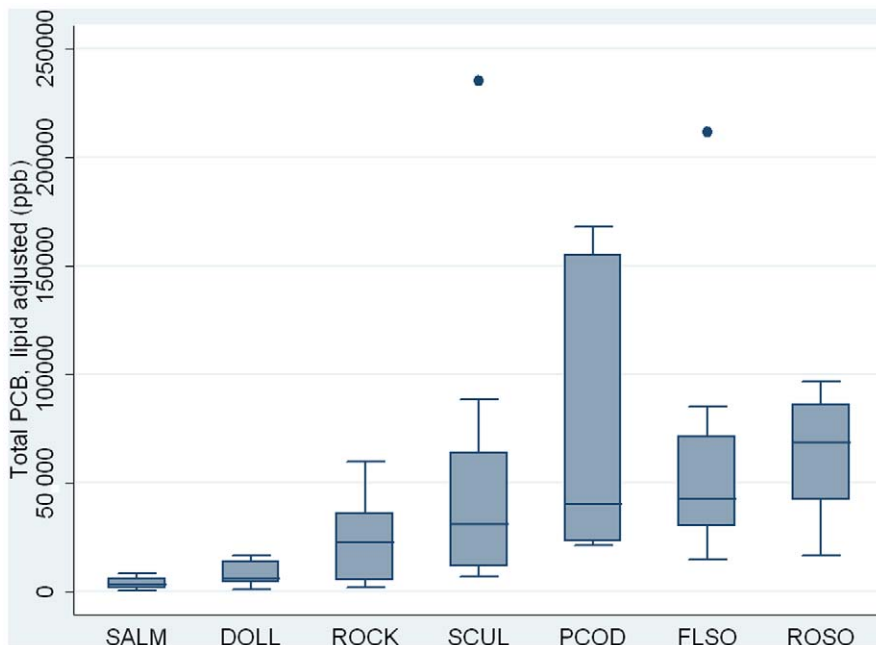


Figure 2. Box plot showing lipid adjusted total PCB levels. Concentrations are in ng/g (ppb), lipid weight. The box represents 50 percent of the values. The median, as well as the sample maximum (excluding outliers) and minimum are shown. Outliers are represented by dots. The species are arranged after median values in ascending order. Black rockfish and Pacific halibut were excluded. SALM, sockeye salmon. DOLL, dolly varden. ROCK, rock greenling. SCUL, great sculpin. PCOD, Pacific cod. FLSO, flathead sole. ROSO, rock sole.

doi:10.1371/journal.pone.0012396.g002

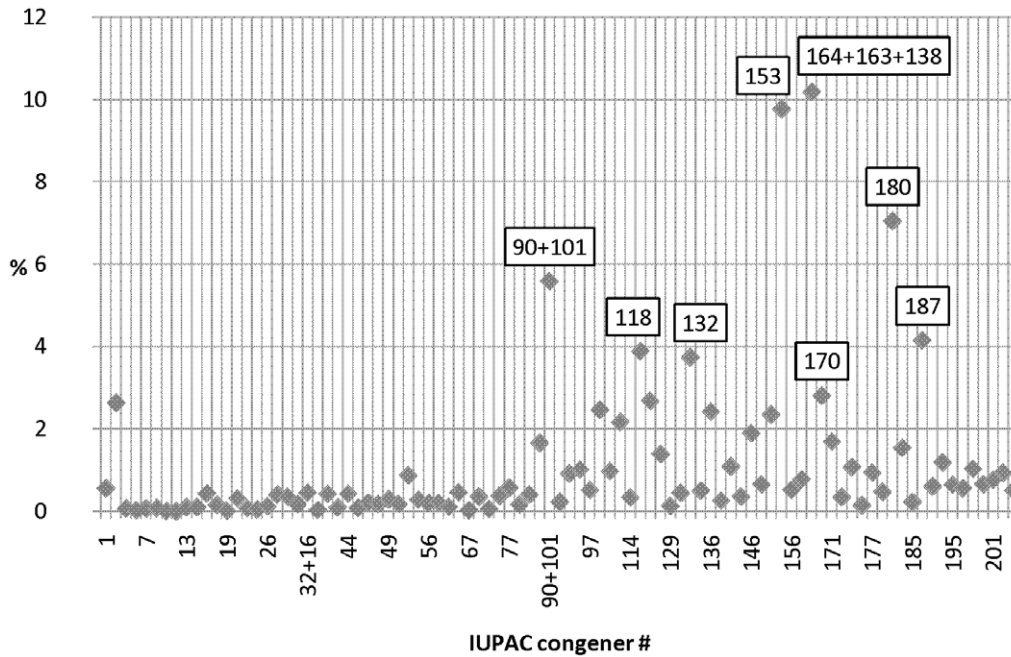


Figure 3. Average PCB congener pattern. The values are expressed as percent of total PCBs. Black rockfish and Pacific halibut were excluded from the calculations.
doi:10.1371/journal.pone.0012396.g003

which is approximately six times greater than the American average [47]. Much higher intakes have been estimated for other native groups [48]. The high fish consumption results in the Aleutian people having a greater exposure to the contaminants in fish than other populations.

It is not surprising that the higher chlorinated PCB congeners dominated in the analyzed fish samples. In general, the congener pattern changes in the food chain, with the higher chlorinated congeners dominating in species on the top trophic levels. This is attributable to the slower metabolism and higher persistence of these congeners [2,16]. Even though not all of the species studied

are on the highest trophic level, they all are predators and can therefore be expected to bioaccumulate higher chlorinated congeners. The congener pattern for black rockfish needs further study, as our material just contained two samples of this species.

In order to determine to which extent PCB exposure from fish contributes to total PCB body burden and the associated adverse health effects, it is important that there be further study of the correlation between the congener patterns in diet with the congener patterns found in human serum. It is also important to recognize that different PCB congeners exert their effects via different mechanisms and can therefore cause different health

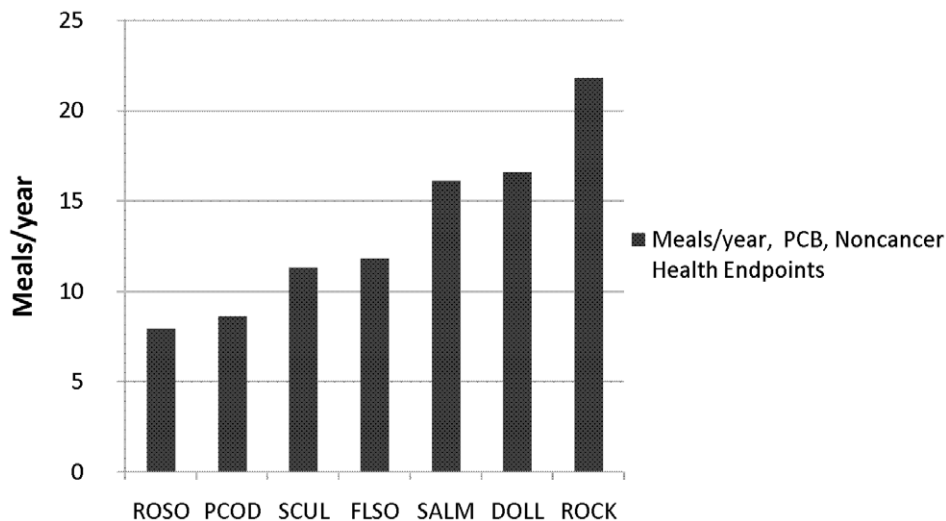


Figure 4. Consumption advisories for noncancer health effects based on PCB concentrations. The advisories are in meals/year and based on USEPA reference dose (RfD). The species are arranged after recommended meals per year in ascending order. ROSO, rock sole. PCOD, Pacific cod. SCUL, great sculpin. FLSO, flathead sole. SALM, sockeye salmon. DOLL, dolly varden. ROCK, rock greenling.
doi:10.1371/journal.pone.0012396.g004

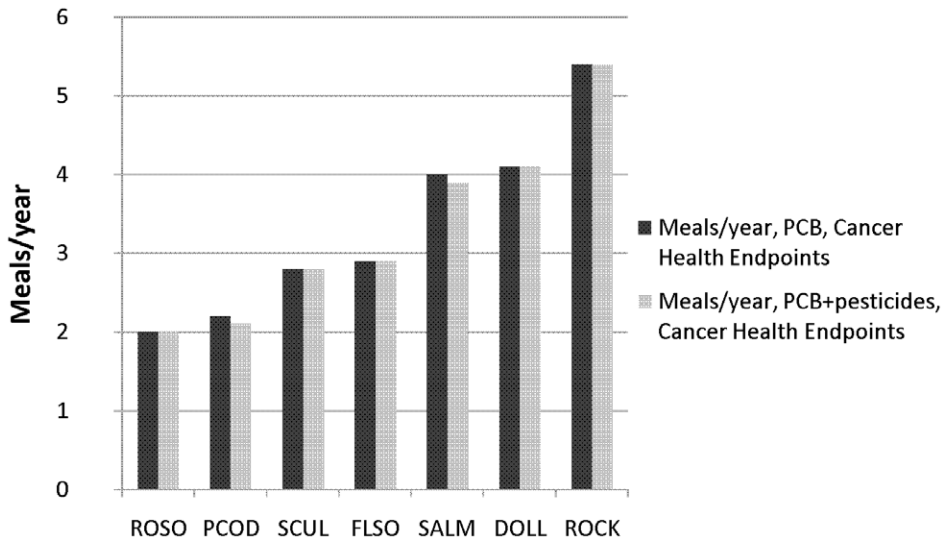


Figure 5. Consumption advisories for cancer health effects based on concentrations of PCBs and the mixture of PCB, HCB, and DDE+85, respectively. The advisories are in meals/year and are based on USEPA cancer slope factors (CSF). The species are arranged after recommended meals per year in ascending order. ROSO, rock sole. PCOD, Pacific cod. SCUL, great sculpin. FLSO, flathead sole. SALM, sockeye salmon. DOLL, dolly varden. ROCK, rock greenling. doi:10.1371/journal.pone.0012396.g005

outcomes [17,49]. Since humans normally are exposed to complex congener mixtures, it may be difficult to predict the health effects. Different congeners can even have opposing effects. One example is that co-planar congeners are anti-estrogenic, while most other congeners behave as estrogen agonists. The situation is further complicated by the fact that PCB metabolites can also have biological effects [17].

We found no statistically significant interspecies differences regarding total PCB levels, which might be attributed to the low sample number. However, our results are an indication of which species are most contaminated. Additional studies will need to be conducted to confirm the results. The highest PCB levels were found in rock sole, which is a bottom-feeding species that live in relatively shallow water bays [50]. This kind of fish is exposed to PCBs in the bottom sediment [51], and this could be one reason

for the high contaminant levels. The lowest PCB levels were found in rock greenling and dolly varden, which would be expected considering that these species feed relatively low on the trophic level [52]. It is difficult to compare the total PCB levels with previous studies, due to variations in the detection limits and the PCB congeners analyzed, in addition to other laboratory parameters. Similar difficulties have been encountered by other authors [40].

Due to the high lipid solubility of organochlorine compounds, they are generally well correlated to the lipid content in a sample [53]. Therefore, lipid adjusted values were calculated by relating the levels of the contaminants to the lipid content of each species. Again, rock sole had the highest PCB level, while dolly varden was among the least contaminated species. The lipid adjusted PCB level in Pacific cod was relatively high, which probably is

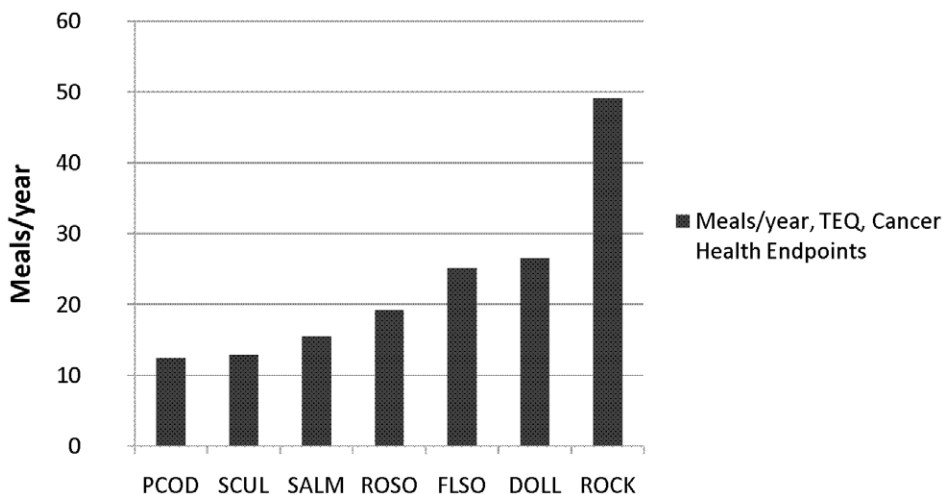


Figure 6. Consumption advisories for cancer health effects based on total TEQ. The advisories are in meals per year and calculated for the analyzed 5 out of 12 dioxin-like PCB congeners. The calculations are based on USEPA cancer slope factors (CSF). PCOD, Pacific cod. SCUL, great sculpin. SALM, sockeye salmon. ROSO, rock sole. FLSO, flathead sole. DOLL, dolly varden. ROCK, rock greenling. doi:10.1371/journal.pone.0012396.g006

attributable to the low lipid content in this species. The total amount of PCB is distributed in a smaller amount of lipid than for the other fish, leading to a higher concentration per gram lipid. Pacific cod eat at a higher trophic level, which is a likely reason for the higher levels of contaminant present [29,52]. The opposite situation is found for sockeye salmon, the fish with the highest lipid content (Table 1). Because of that, the lipid adjusted PCB value is low. The lipid content may vary between life stages of the fish, which can influence the concentration and composition of POPs in the muscle tissue [53]. The differences found between the lipid adjusted and wet weight values indicate that it is important to determine both these values when evaluating contaminant levels. It is important to emphasize, however, that we analyzed flesh, not skin or fat. Thus our samples reflect concentrations of POPs in the most edible part of the fish.

Among the pesticides the highest levels were found for *p*, *p'*-DDE. The concentrations we found are similar to the levels of total DDT measured in other Arctic fish [15]. While we measured *p*, *p'*-DDE together with PCB congener 85, the results can still be compared since previous studies show that *p*, *p'*-DDE typically is present at >100-fold higher serum levels than congener 85 [43] DDE is also the dominant and most persistent compound of total DDT [4]. It has also been reported that *p*, *p'*-DDE levels in human serum were the highest among the pesticides [14].

Our results show that all analyzed species contain substantial levels of PCBs. Due to the importance of fish in the diet and the high contaminant levels in the fish, the Aleuts are particularly at risk of high exposure to POPs. While the Alaska Division of Public Health recommends continued unrestricted consumption of traditional food for most people [33] the contaminant levels we found in the fish from the Aleutians evoke restrictive consumption advisories based on USEPA methods. For cancer health endpoints, the advisories calculated for the cumulative effect of PCBs and pesticides do not differ significantly from the advisories calculated for PCBs alone. This indicates that PCB contamination is the major risk for the people consuming the fish from the waters around the Aleutian Islands. None of the analyzed species should be consumed more than five times a year and the most contaminated species, rock sole, should be consumed no more than twice a year. A large proportion of the Aleutian people can be expected to consume these species in much higher amounts. The consumption advisories for the non-cancer health endpoints are less stringent. However, these restrictions likely recommend fewer meals per year of some of the fish species than what many Aleuts consume. In general, USEPA recommends that the more restrictive meal consumption limit (in this case the limits for the carcinogenic effect) serve as the basis for fish consumption advisory [44].

The calculated consumption limits based on TEQ values are less stringent than those based on total PCBs. However, these values should be interpreted with caution, since the calculations were based on only 5 of the 12 dioxin-like PCB congeners, and do not include the most potent congeners. Thus the TEQs values presented are significant underestimations of total TEQ.

A problem with using the USEPA guidelines is that the recommendations are based on analyses with higher detection limits for the contaminants than those of our analytical method. This might have affected the calculated consumption limits, as a lower detection limit results in higher total PCB levels, and thus likely results in more restrictive consumption advisories. The USEPA methods also are based on the assumption that the various effects are additive. However, synergistic effects have been reported for *p*, *p'*-DDE and PCBs, as well as for other contaminants [27]. It is also important to remember that fish

can contain additional contaminants to those measured in this study. Previous studies have shown that fish from the Aleutians contain mercury and other heavy metals in amounts sufficient enough to trigger consumption advisories. The mercury levels varied between species, and the highest mean level in muscle, 0.366 ppm, was found in great sculpin. A high proportion of fish triggered USEPA consumption advisories of less than 1 meal/week [29,36]. It can be assumed that the consumption limits calculated in this study would become increasingly stringent if heavy metals were included.

It is important to note that the health of the various fish species is also jeopardized by these PCBs concentrations. For example, lake trout embryos and fry are very sensitive to dioxin, and dioxin exposure was responsible for the dramatic decline in the lake trout population in the Great Lakes in the middle of the 20th century [54]. Exposure to aquatic PCBs (at concentrations as low as 1 ppb in water) during the early stages of development or as juveniles can disrupt developmentally appropriate behavior in Atlantic salmon and negatively impact endocrine parameters involved in smolting development [55]. PCB 153 has been found to increase estrogen concentrations in immature female rockfish [56]. These health effects can result in the reduction of the ability for these fish species to successfully reproduce. However, not all fish species are negatively affected by PCBs exposure. Flounder show no change in total cytochrome P450 concentration and only a slight induction in EROD activity when exposed to PCBs [57].

When assessing any risk of the traditional diet, however, it is also essential to consider its value for the indigenous people. There are many benefits from subsistence food concerning health, economy, and culture. The harvest of traditional food provides social interaction among the people and the food is normally shared between families. The lifestyle associated with the harvest also results in multiple forms of natural physical activity, which in itself is of great value for public health [12,31,32]. Subsistence food is a source of identity and self-respect in these cultures, and is an opportunity for meaningful productive work in regions where wage paying jobs are few. Traditional foods are also of great economic value, especially in remote areas, where other food items need to be imported by plane or boat and therefore are relatively expensive [31,32]. In addition it might be risky to rely on these deliveries, since they can be delayed by harsh weather conditions [30]. Furthermore, the imported food is often lower in nutrient value than the traditional food, and it is high in saturated fats and carbohydrates [34]. Fish is a good source of protein and omega-3 polyunsaturated fatty acids, and many studies have shown health benefits from eating a diet rich in fish [29]. The omega-3 fatty acids provide a variety of health benefits, including reduction of blood cholesterol levels, as well as lower incidences of cardiovascular disease, stroke, and pre-term delivery [31,58].

Historically, the mortality from coronary heart disease has been lower among Alaskan natives compared to nonnative Alaskans. Since the end of the 1980's the difference has disappeared and today Alaskan natives have a higher prevalence of many risk factors (hypertension, overweight/obesity, smoking, sedentary lifestyle) for coronary heart disease than nonnative Alaskans [59]. Also, the incidence of various tumors common in developed societies has previously been low in Arctic Native populations. During the second half of the 20th century, however, the risk of lifestyle-associated tumors, especially cancers of the lung, colon, breast, and uterus, increased considerably [60,61,62]. Dietary changes and related lifestyle changes are thought to be contributing factors to these patterns [12,58,60,62]. There has also been an increase in the prevalence of diabetes among Alaskan natives, a fact that also might be related to the change of diet [63].

If the people are advised not to eat fish from their traditional fishing waters, the dietary changes and the associated public health losses are likely to become even more pronounced.

The issue about risks versus benefits of fish consumption is not only a concern for Alaskan natives. Fish from the Aleutians are exported globally and are therefore consumed by people everywhere [64,65]. In addition, a number of studies from other parts of the world indicate that contamination of fish is a widespread problem. Levels of PCBs, dioxins, and mercury have been found to be sufficiently high to cause concern in fish from the Atlantic Ocean, the Pacific Ocean, and the Baltic Sea, as well as from other waters [27,66,67,68]. This raises the question of whether fish in general are a safe food to eat.

In conclusion, the fish caught in the Aleutian Islands (mainly from Adak) contain significant levels of POPs, in particular PCBs. On the basis of consumption advisories developed by USEPA for carcinogens, the PCB levels in these fish trigger very strict consumption recommendations. Therefore it is very important to conduct further studies of the contaminant levels in the Aleutian region, particularly around other inhabited villages. In particular it would be of great value to study interspecies differences in fish in more detail in order to be able to advise people on which species to avoid. To reduce the intake of contaminants it would also be wise to advise people to remove the skin from the fish. Previous studies have indicated that this can influence the PCB levels, but the results are not entirely consistent [31,69]. Further studies of how the preparation methods affect the contaminant levels are therefore important. In addition, there is a need of more human

biomonitoring data from the Aleutians, particularly concerning organochlorine contaminants and their health effects. One must also keep in mind that this is a problem not only for the Aleuts, but also for others outside of the region who consume fish caught here and sold elsewhere.

More importantly, most of the fish for the present study were collected at Adak which is on the National Priorities List for various contaminants including PCBs, dumped by the former military base over a period of many years [70]. Thus the results apply most strongly to the Aleuts living at Adak, and indicate that considerably more work is required to examine levels in fish from the other islands in order to determine the relative contribution of local as compared to global sources of PCBs.

Acknowledgments

We thank Michael Gochfeld, C. W. Powers, D. Kosson and C. Jeitner of the Consortium for Risk Evaluation with Stakeholder Participation (DE-FC01-06EW07053), NIEHS (P30ES005022) and EOHSI for sample collection and preparation, and Michael Carlberg, MSc, who assisted in the statistical calculations.

Author Contributions

Conceived and designed the experiments: SH HT GWS JB DOC. Performed the experiments: SH HT GWS. Analyzed the data: SH HT GWS. Contributed reagents/materials/analysis tools: JB DOC. Wrote the paper: SH HT GWS JB. Interpretation of data: DOC. Final approval of the article: DOC.

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