Global Assessment of Polybrominated Diphenyl Ethers in Farmed and Wild Salmom

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Introduction

Despite their societal benefits, polybrominated diphenyl ethers (PBDEs) seem to be migrating from the products in which they are used and entering the environment and people. PBDEs are now ubiquitous; they can be found in air, water, fish, birds, marine mammals, and people, and in most cases, the concentrations of these compounds have increased dramatically over the last 20 years.[1] One source of PBDEs in people is their food supply, and an increasingly important food is salmon, which is nutritious and high in beneficial fats.

Between 1987 and 1999, salmon consumption increased annually at a rate of 10-20% (*2*). Currently, over half the salmon sold globally is farm-raised in Northern Europe, Chile, Canada, and the United States, and the annual global production of farmed salmon (predominantly Atlantic salmon) has risen from ~27,000 to over 1 million metric tons during the past two decades.[2] The health benefits of eating fish such as salmon have been well documented. However, salmon are relatively fatty, carnivorous fish that feed high in the food web, and as such, they bioaccumulate contaminants. In a previous study, we reported that levels of persistent, bioaccumulative contaminants (such as PCBs, dioxins, and several chlorinated pesticides) are significantly higher in farm-raised salmon than in wild salmon and that salmon raised on European farms have significantly greater toxic contaminant loads than those raised on North and South American farms.[2] In this report, we provide results on polybrominated diphenyl ether concentrations in salmon purchased from farms in eight major salmon farming regions, in five species of wild Pacific salmon, and in salmon fillets purchased at supermarkets in North America and Europe.

Materials and Methods

PBDEs were measured in about 700 farmed and wild salmon (totaling ~2 metric tons) collected from around the world. Farmed Atlantic salmon (*Salmo salar*) were purchased from wholesale suppliers in the United States, United Kingdom, Norway, and Canada between March and

December, 2002. These suppliers provided farmed salmon from eight major salmon farming regions: Norway, Chile, Scotland, British Columbia, Eastern Canada, the Faroe Islands, Maine, and Washington State. One to three wholesale suppliers provided fish from two to ten farms in each region for a total of 51 different salmon farms, with representation from each of these eight regions. Suppliers provided information on the origin of the fish (region and farm) by including the original labels from the farm source. The farms chosen reflect those from which suppliers could obtain farmed Atlantic salmon of the appropriate size within the sampling period. Ten fish were obtained from each farm, nine of which were randomly grouped into three composites of three fish each. Most individual fish weighed ~4-6 kg. A total of 459 farmed salmon from wholesalers was used to produce 153 samples for analysis. Viscera and gills from all whole fish were removed before they were shipped; the heads were left on.

Between September 2001 and August 2002, other suppliers provided 135 wild fish representing five wild species of Pacific salmon: chum (*Oncorhyncus keta*), coho (*O. kisutch*), chinook (*O. tshawytscha*), pink (*O. gorbuscha*), and sockeye (*O. nerka*). Samples of each species were purchased from different geographic regions, including Kodiak, Alaska; Southeast Alaska; British Columbia; and Oregon. Three composites of three fish for each species at each location resulted in a total of 45 samples for analysis.

We purchased an additional 144 salmon fillets (three whole fillet samples from each of three retail outlets) from supermarkets in Boston, Chicago, Denver, Edinburgh, Frankfurt, London, Los Angeles, New Orleans, New York, Oslo, Paris, San Francisco, Seattle, Toronto, Vancouver, and Washington DC. Purchasers specifically asked for farmed salmon at fish counters and were instructed not to purchase any other type of salmon. Fillets from supermarkets were composited by the retail outlet where they were purchased. Composite samples consisted of three fillets per retail outlet, for a total of 48 samples.

All samples came to the analytical laboratory (AXYS Analytical in Sidney, British Columbia) fresh or frozen on ice or gel-packs. Fish were thawed and inspected by a fisheries biologist to verify species. Each fish was weighed, its length measured, and filleted to give two skin-on fillets. We analyzed skin-on fillets because most salmon are sold at retail outlets with the skin on. In each case, the fillets from three fish were ground and re-ground together to make a homogenous composite. Ten grams of wet fish tissue were spiked with known amounts of nine individual fully ^{13}C-labeled brominated diphenyl ethers before extraction, ground with anhydrous Na_2SO_4, and Soxhlet extracted with dichloromethane for 16 hours. The extract was cleaned up by gel permeation chromatography on Biobeads SX-3 and fractionated on Florisil, silica, and alumina. Gas chromatographic mass spectrometric analysis of PBDEs was accomplished using a Micromass Autospec Ultima magnetic sector high resolution mass spectrometer equipped with a Hewlett Packard 6890 gas chromatograph. Chromatographic separation was achieved using a Durabond DB-5HT high temperature column (30 m × 250 µm i.d., 0.10 µm film thickness). The PBDE concentrations were obtained by isotope dilution quantification using the ^{13}C-labeled internal standards. All analyses were conducted in accordance with AXYS' accredited QA/QC program.

Results and Discussion

Figure 1 shows that total PBDEs in the farmed salmon were significantly more concentrated as a group (red bars) than in the wild salmon (green bar) [$F = 30.33$, $p < 0.0001$, with $df = (1, 64)$]. PBDE concentrations were significantly higher in farmed salmon from Europe than in those from North America, in farmed salmon from North America than in those from Chile, and in farmed salmon from Chile than in wild salmon [$F \geq 126.55$, $p < 0.0001$ with $df = (1, 62)$ for all]. PBDE concentrations in the salmon purchased from retail outlets (yellow bars) in Europe were significantly higher than in salmon purchased in stores in North America [$F = 6.12$, $p = 0.0268$ with $df = (1, 14)$], but both types of store-bought samples had average PBDE concentrations higher than in wild salmon.

PBDE concentrations were highest in wild chinook from British Columbia and in farmed salmon from Scotland and western and eastern Canada and lowest in farmed salmon from Chile and Washington State. Salmon fillets purchased from supermarkets in Edinburgh, London, and San Francisco were generally the most contaminated with PBDEs, and those purchased in New Orleans, Washington DC, Denver, and New York were the least contaminated of the store-bought samples. Most of the salmon sold in European stores comes from European farms, which produce the more contaminated salmon, while most of the salmon sold in US stores comes from Chile and Canada.

The relatively high PBDE concentration in the chinook samples from British Columbia was interesting. In fact, an analysis of variance for total PBDE concentrations in the 45 samples of wild salmon, treating the three composites from each source as replicates, showed significant differences among the species [$F = 3.92$, $p = 0.0363$, $df = (4, 10)$], with the chinook elevated relative to the other species. The average total PBDE concentration in the 9 chinook samples was 2.29 ± 0.56 ng/g compared to 0.18 ± 0.025 ng/g in the 36 others. The elevated PBDE levels found in the wild chinook may be related to their feeding behavior and trophic level. Among all of the wild species we studied, chinook tend to feed higher in the food web throughout their adult life stage and to grow to be larger individuals on average.

The congener profiles of PBDEs in these salmon are similar: BDE-47 dominates, just as it does in almost all ambient environmental samples and in people.[1] The next most abundant congeners, BDE-99 and BDE-100, are also relatively abundant in most other environmental samples. The PBDE congener profile of the wild salmon is similar to that of the farm-raised samples, indicating that all salmon, farmed or wild, are ultimately getting their dose of PBDEs from similar sources. Of course, the absolute PBDE dose is much higher for the farmed relative to the wild salmon.

This study further demonstrates the importance of labeling salmon as farmed and identifying the country of origin. Additional studies of contaminant sources, particularly in feeds used for farmed, carnivorous species such as salmon, are needed. This conclusion is consistent with the recommendations of a recent panel of the Institute of Medicine,[3] which called for major efforts to reduce the content of dioxin-like compounds in food given to animals and fish that are used for human consumption.

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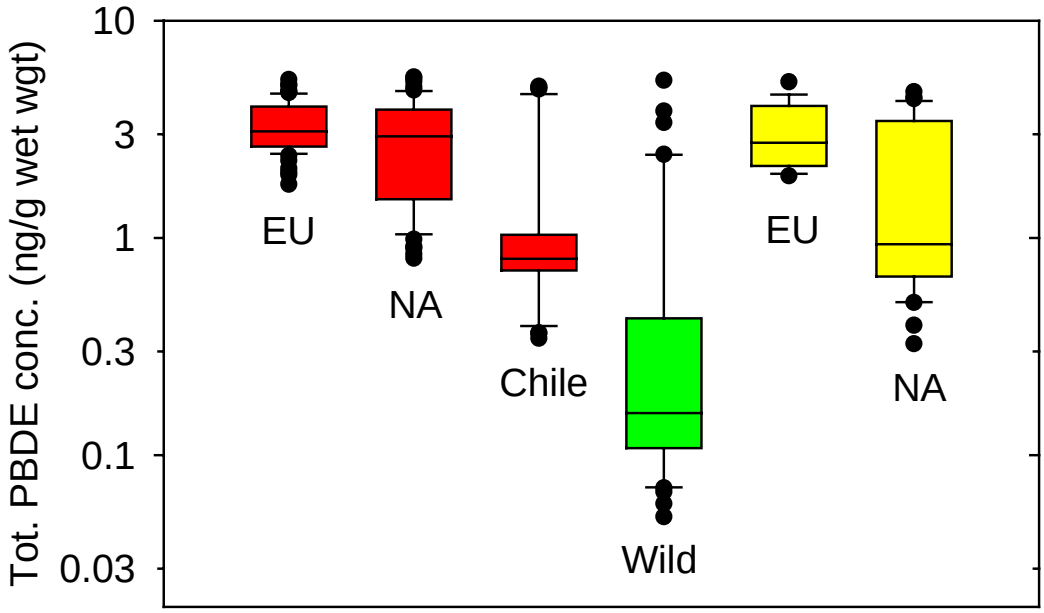

Figure 1. PBDE concentrations (in ng/g wet weight) found in farm-raised (red bars), wild (green bar), and supermarket-purchased (yellow bars) salmon. The horizontal lines represent the 10th, 50th, and 90th percentiles, and the boxes represent the 25th to 75th percentiles; outliers are shown as individual points. Abbreviations: EU, Europe; NA, North America.