

CONCENTRATIONS OF POLYBROMINATED DIPHENYL ETHERS (PBDEs) IN FISH FROM PUGET SOUND, WA, USA



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ABSTRACT

Concentrations of polybrominated diphenyl ethers (PBDEs) were measured in marine and anadromous fish species representing a variety of life history traits to give a broad overview of contamination in Puget Sound, WA. Our objectives were to determine to what extent PBDEs have infiltrated the benthic and pelagic food webs, and to assess potential exposure routes. To provide a coarse assessment of whether PBDEs are present in the benthic food web, we measured PBDE concentrations in muscle tissue of English sole (*Pleuronectes vetulus*) at urban, near-urban and non-urban sites throughout Puget Sound. At one of these urban sites, we also measured PBDEs in individual male quillback rockfish (*Sebastes maliger*) and individual female lingcod (*Ophiodon elongatus*) of varying ages to assess the degree that PBDEs biomagnify and whether PBDEs bioaccumulate in long-lived fish species like rockfish. We sampled whole bodies of Pacific herring (*Clupea harengus pallasii*) and resident chinook salmon (*Oncorhynchus tshawytscha*) from major oceanographic basins in Puget Sound to assess contamination in the pelagic food web. We also compared resident and migratory chinook to determine whether Puget Sound is a source of PBDEs. Our preliminary results show that PBDEs are found throughout benthic and pelagic food web with concentrations ranging from 1.3 to 57.7 ng/g wet weight. The highest levels were measured in Puget Sound chinook salmon, at concentrations considerably higher than those reported previously in farmed and wild salmon.

INTRODUCTION

Polybrominated diphenyl ethers (PBDEs) are flame-retardants that are added to furniture, upholstery, electronic components, computers and many other manufactured items. Exposure to these compounds has been associated with a variety of biological effects (e.g., thyroid disruption, neurobehavioral effects) in laboratory animals. Globally, the use of PBDEs is greatest in the Americas, followed by Asia and Europe (de Wit 2002), although several countries have banned their use or manufacture. As a result, higher concentrations of these compounds have been measured in samples of wildlife and humans from North America than in samples from other parts of the world.

Concentrations of PBDEs are increasing rapidly in marine organisms on the west coast of the U.S. (She *et al.*, 2002) and Canada (Ikonomou *et al.*, 2002) but little is known about the presence of these flame retardants in fish from the Puget Sound region. The objectives of our study were to determine the extent that PBDEs have infiltrated the benthic and pelagic food webs of Puget Sound (using a multi-species sampling design) and to assess potential exposure routes. In this study concentrations of PBDEs were measured in marine and anadromous fish species representing different life history traits to give a broad overview of contamination in Puget Sound, WA.

RESULTS AND DISCUSSION

PBDEs were measured in all species, with concentrations ranging from 1.3 to 57.7 ng/g (wet weight).

PBDEs were detected in all English sole samples and concentrations were highest in fish from urban areas (Figure 2). This pattern likely reflects localized sediment contamination because of this species' association with bottom sediments, its consumption of benthic invertebrates, and its relatively restricted movements.

Along the highly urbanized Seattle Waterfront, PBDE concentrations were highest in lingcod, a voracious predator, followed by English sole, and quillback rockfish (a demersal carnivore -- Figure 3).

PBDE concentrations were lower in rockfish than lingcod overall, however, levels in rockfish increased with age in males of this long-lived species (Figure 4). Previously, West *et al.* (1998) documented that male rockfish from this location accumulated PCBs with age but females did not. At this location, PBDEs did not accumulate with age in female lingcod (data not shown), however, we anticipate they would accumulate in male lingcod.

Whole-body concentrations of PBDEs in the pelagic Pacific herring were highest in stocks from urban basins (Figure 5).

In chinook salmon from Puget Sound, PBDE concentrations were higher in residents than those that migrate outside Puget Sound (Figure 6), suggesting that Puget Sound is a predominant source for this contaminant. Levels in both resident and migratory chinook from Puget Sound were considerably higher than those reported for wild chinook and farmed Atlantic salmon (Hites *et al.*, 2004).

Furthermore, the longer chinook reside in Puget Sound, the higher their PBDE concentration, as evidenced by the positive correlation of fish length with [PBDE] in resident chinook (Figure 7).

Overall, the ratio of PBDEs to PCBs is higher for pelagic than benthic species (Figure 8), suggesting that PBDEs may be accumulating to a greater degree in pelagic species.

Figure 1. Sampling locations of benthic species.

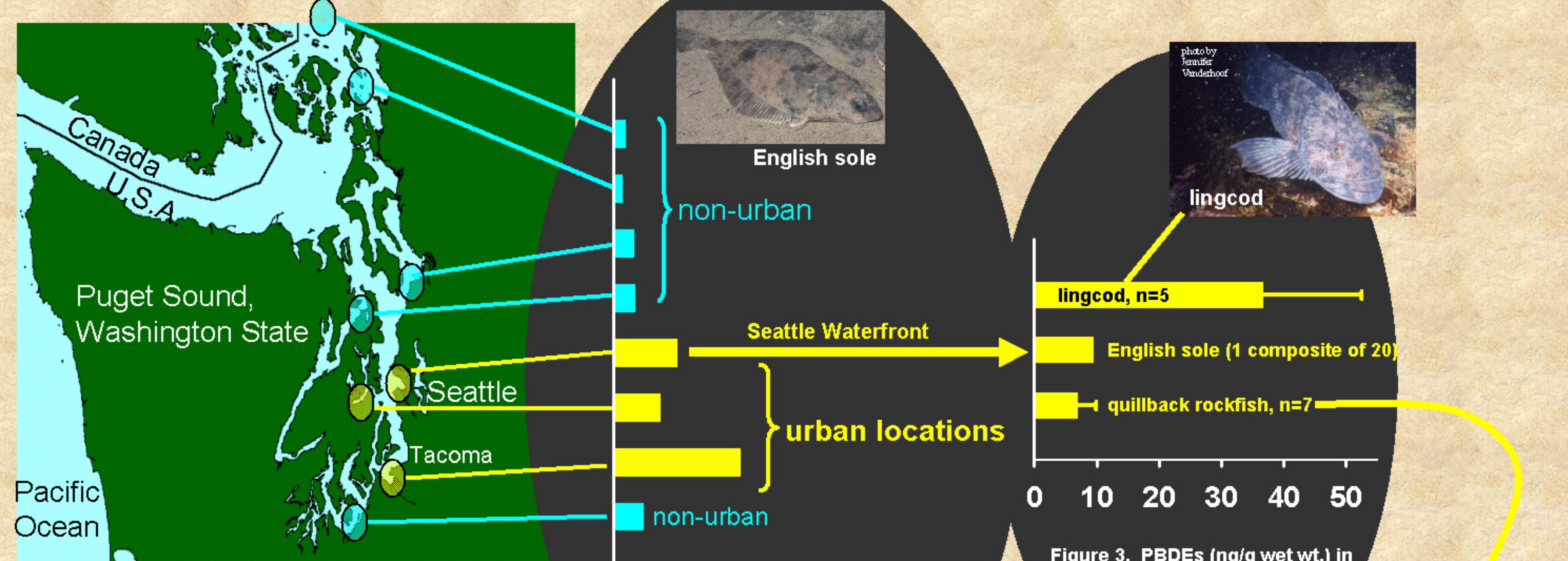


Figure 2. PBDEs (ng/g, wet wt.) in English sole from 8 Puget Sound locations. Each bar represents a composite sample of 20 skin-off muscle filets.

Figure 3. PBDEs (ng/g wet wt.) in muscle tissue of three benthic species from the highly urbanized Seattle Waterfront, Puget Sound, WA

MATERIALS & METHODS

Sampling Design

To assess contamination of the benthic food web, we measured PBDE concentrations in composite muscle tissue of English sole (*Pleuronectes vetulus*) at urban and non-urban sites throughout Puget Sound (Figure 1). At one of these urban sites we also measured PBDE concentrations in the individual muscle tissue of 10 male quillback rockfish (*Sebastes maliger*) and 5 female lingcod (*Ophiodon elongatus*) of varying ages. To assess contamination in the pelagic food web, we analyzed whole bodies of Pacific herring (*Clupea harengus pallasii*) from three major oceanographic basins in Puget Sound and four populations of resident or migratory chinook salmon (*Oncorhynchus tshawytscha*). We compared our results for resident chinook with those for migratory chinook to assess oceanic versus Puget Sound sources of PBDEs.

Chemical Analysis

All samples except the migratory chinook samples were analyzed for PBDEs at AXYS Analytical using EPA Method 1614. The samples were Soxhlet extracted with dichloromethane, cleaned by gel permeation chromatography and analyzed with gas chromatography/mass spectrometry (GC/MS). PBDE concentrations were reported as the sum of 34 congeners, although the majority of summed PBDEs although the predominant congeners were PBDE 47 followed by PBDEs 99, 100 and 49.

Migratory chinook salmon samples were analyzed at the Northwest Fisheries Science Center using a modified GC/MS method (Sloan *et al.*, in press). Briefly, whole salmon were extracted with dichloromethane using an accelerated solvent extractor. Size exclusion chromatography with high-performance liquid chromatography (HPLC) was used to collect the fraction containing the PBDEs and other persistent organic pollutants (POPs). The HPLC fraction was analyzed for PBDEs by capillary column GC/MS. Summed PBDEs were calculated by adding the concentrations of PBDE congeners 28, 47, 49, 66, 85, 99, 100, 153, 154, and 183 (predominant congeners bolded). The resident chinook samples were analyzed by both AXYS and to allow lab bias correction to us to pool samples from both labs.

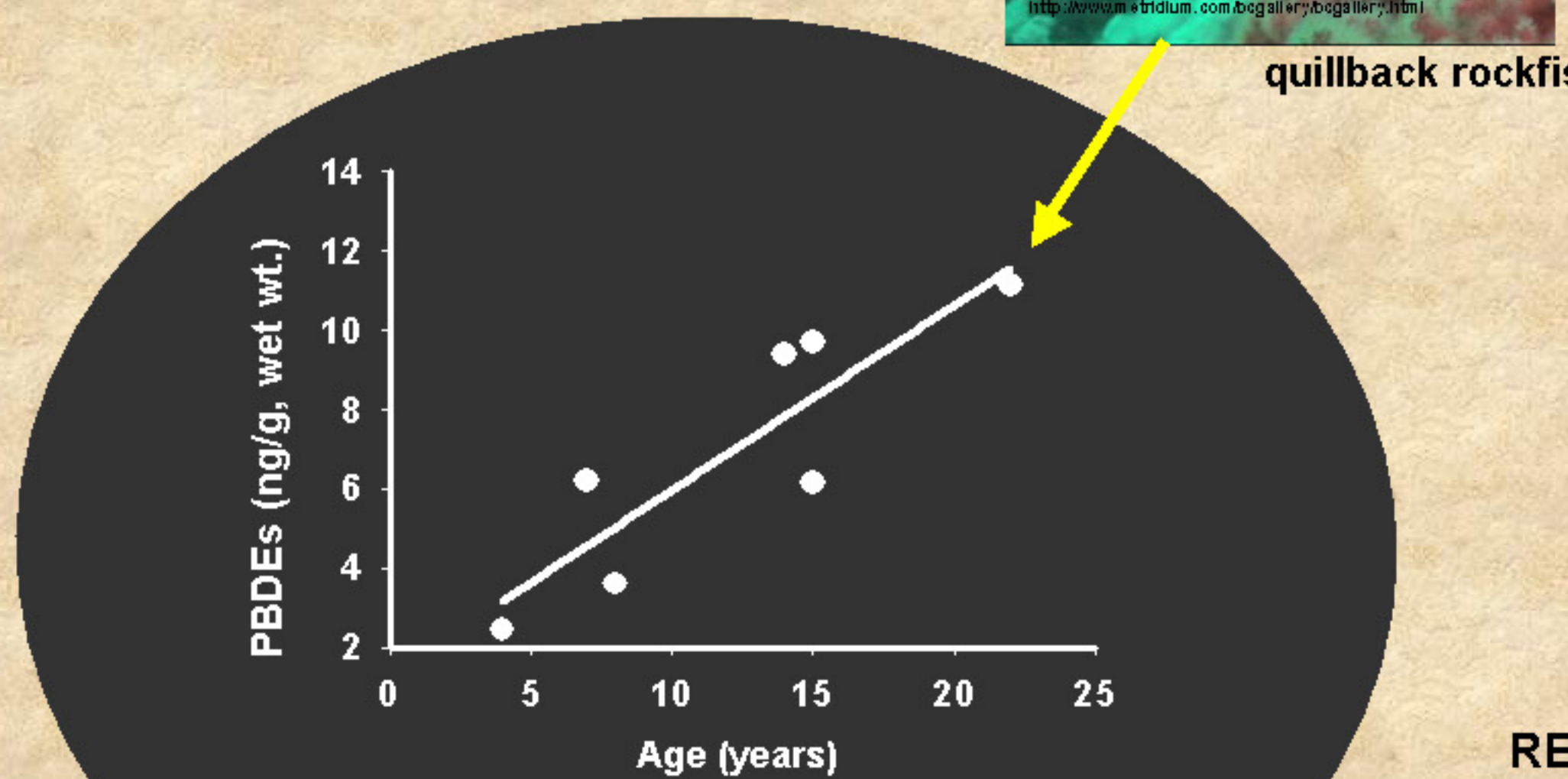


Figure 4. Accumulation of PBDEs (ng/g, wet wt.) with age in male quillback rockfish from the highly urbanized Seattle Waterfront.

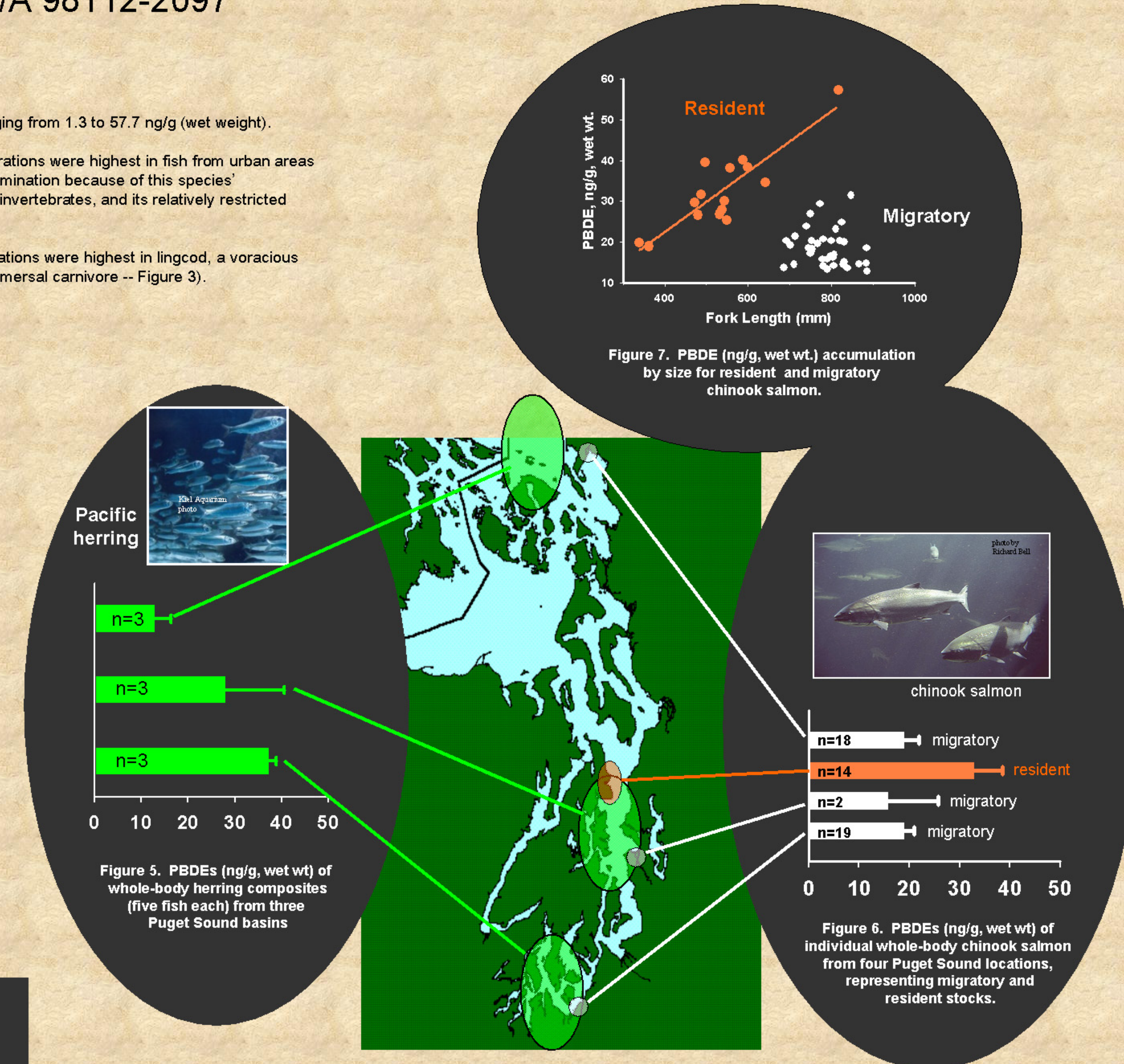


Figure 7. PBDE (ng/g, wet wt.) accumulation by size for resident and migratory chinook salmon.

Figure 5. PBDEs (ng/g, wet wt) of whole-body herring composites (five fish each) from three Puget Sound basins

Figure 6. PBDEs (ng/g, wet wt) of individual whole-body chinook salmon from four Puget Sound locations, representing migratory and resident stocks.

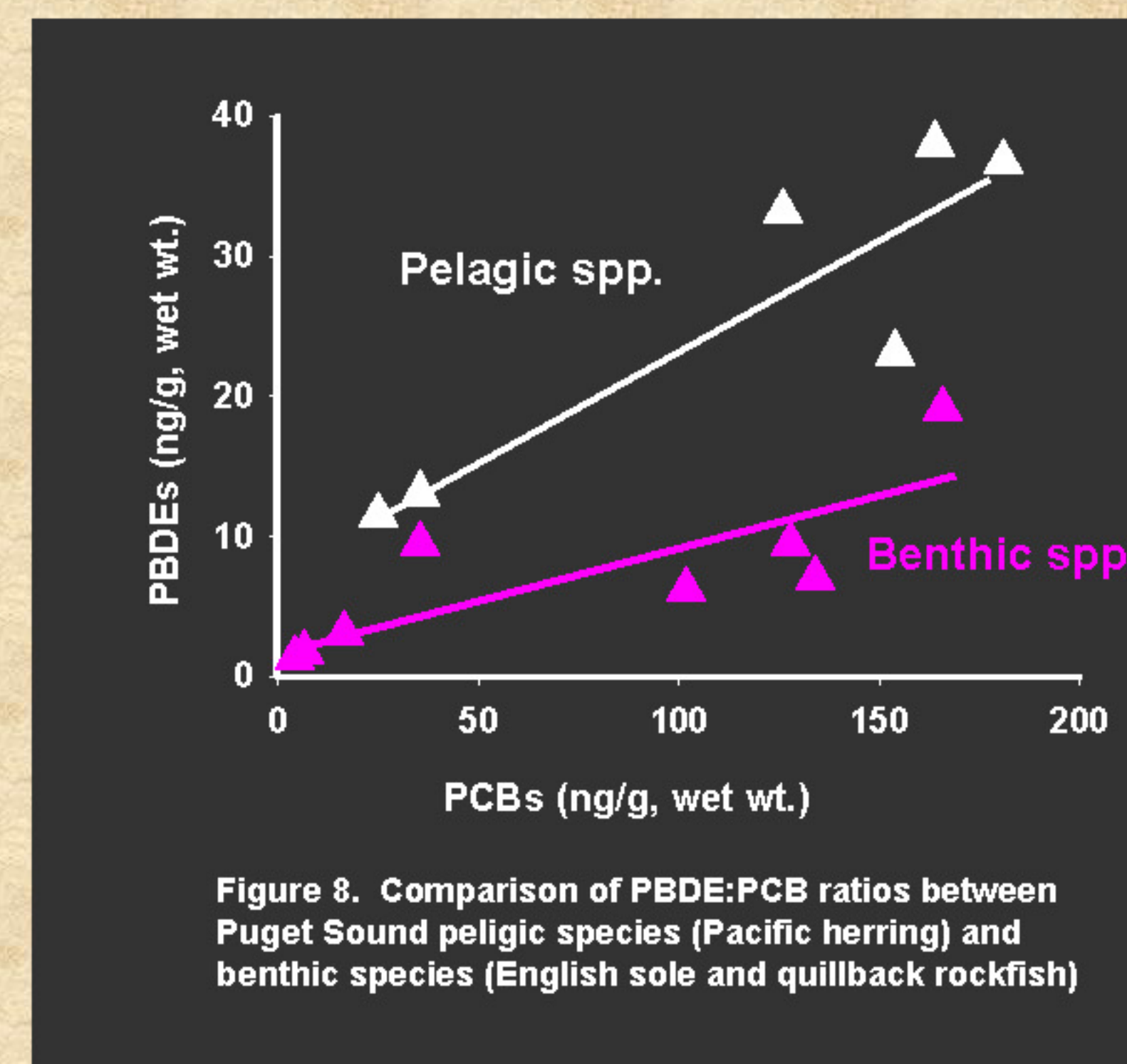


Figure 8. Comparison of PBDE:PCB ratios between Puget Sound pelagic species (Pacific herring) and benthic species (English sole and quillback rockfish)

CONCLUSIONS

- Puget Sound fish exhibited a wide range of PBDEs with the highest concentrations in more urbanized locations for both benthic and pelagic species.
- PBDEs bio-magnified in the benthic and pelagic food webs.
- PBDE accumulated in males for quillback rockfish, a long-lived demersal species.
- Residence time in Puget Sound appears to increase PBDE exposure for chinook salmon.
- PBDEs appeared to accumulate to a greater degree (relative to PCBs) in pelagic species than the benthic species we tested.

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