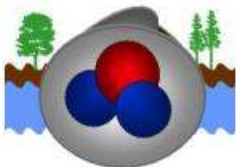


Pyrethroid Pesticides and PCBs in Bivalves from East Sound, San Juan County, WA



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Summary

Evidence of moderate but widespread pyrethroid pesticide contamination of lakes, streams, ponds and freshwater sediments in the San Juan Islands, WA (Barsh et al. 2008), suggested that these toxicants may be accumulating in nearshore trophic webs where they could affect threatened wildlife such as salmon as well as local human food supplies. At the same time, efforts to reduce chemical contamination in the San Juan Islands have met with a persistent local belief that the Salish Sea region's large cities (Seattle, Victoria and Vancouver) are responsible for any marine pollution of the San Juan Islands. The present study assesses the local-source contribution of the town of Eastsound, Orcas Island, to the pyrethroid pesticide and PCB loads of uncultivated bivalves harvested from four beaches in East Sound, including the beach into which the town discharges its storm water sewers, and beaches that we expected to be more influenced by wider regional marine circulation patterns.

While pyrethroid pesticides are associated with home and garden products, and therefore most likely to be discharged into East Sound by town storm sewers, PCBs were historically used only by a single electrical utility located near the town of Eastsound but on a separate drainage (Crescent Beach). We hypothesized that pyrethroid concentrations in bivalves would be greatest at Fishing Bay near the Eastsound storm sewer, while PCB concentrations would exceed regional marine background levels (if at all) nearer Crescent Beach and the electrical station.

Pyrethroid pesticide concentrations in commonly consumed local bivalves (butter clams, steamer clams, horse clams, chestnut clams, soft-shell clams; N = 99) were highest at Fishing Bay nearest the Eastsound storm sewer outfall, and PCB concentrations were greatest at Crescent Beach as predicted. Differences were statistically significant at the $p > .05$ level or better. There was no statistically significant correlation of toxic loads with bivalves' age. Butter clams (*Saxidomus giganteus*) tended to be accumulate greater toxic loads than other bivalve species.

The town of Eastsound appears to contribute significantly to pyrethroid pesticide contamination of bivalves in Fishing Bay, bringing them to levels above current federal food safety standards. In the absence of a correlation between pyrethroids and bivalves' age, and in the light of known rates of degradation of pyrethroids in the environment, it is probable that pesticide contamination of Fishing Bay is continuing, rather than a "legacy" of historic pesticide use.

Only wild-harvested bivalves were used in this study. Cultivated bivalves tend to grow faster, are kept out of the sediment, and are harvested younger—factors that would tend to make them less contaminated and safer to eat than uncultivated bivalves exposed to the same environment.

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We previously reported evidence of part-per-billion levels of pyrethroid pesticides in many San Juan County streams, lakes, and ponds (Barsh et al. 2008). The significance of this level of pyrethroid pesticides for local freshwater ecosystems, or for the far larger marine ecosystems into which they drain, is not well understood. A threshold question is whether local-source pyrethroid pesticides accumulate in nearshore invertebrates, through which they may enter the marine food web at even higher concentrations and impact fish and potentially also human consumers of seafood.

The San Juan Islands are surrounded by large cities including Seattle, Vancouver, and Victoria, and functionally form part of the estuary of the Fraser River, which drains the principal industrial areas of British Columbia. While it is tempting to attribute marine pollution exclusively to these large regional inputs, it is possible that local sources within the islands—in particular along urban waterfronts with large storm drain catchments—are contributing significantly to pollution of the islands' sensitive nearshore ecosystems, and threatening sensitive marine animals such as killer whales, Chinook salmon, herring, and rockfish. Toxic effects should depend not only on inputs from human activities, but also on dilution. Toxic inputs dissipate quickly in high-circulation bays but accumulate in low circulation environments, as we have shown with bioassays (Barsh 2009). Urban Growth Areas draining into low-circulation waters should be a focus of potential concern: Lopez village and Eastsound.

The goal of the present study was to determine whether the town of Eastsound has a significant chemical impact on nearshore ecosystems. We chose to measure pyrethroid pesticides because our previous research identified them in local watersheds, and because the immunoassay method we had used to test surface water and sediments can be adapted easily to methanol tissue extractions. Pyrethroids still dominate local pesticide sales, and therefore represent a current local toxic input that could be reduced or eliminated, if local consumers chose to do so.

While we expected the continued use of pyrethroid pesticides to make a relatively large local contribution to the contamination of bivalves in East Sound, we also measured polychlorinated biphenyls (PCBs) in bivalves because we did *not* expect to find evidence of a large local effect. Used in transformer insulating oils before their manufacture in the U.S. was banned in 1979, PCBs were associated with large electrical power systems and urban areas with networks of electrical substations. By comparison, San Juan County has a single historical transformer yard—a single legacy point source—in the Crescent Beach drainage, adjacent to but separated hydrologically from the town of Eastsound.

We chose five bivalve species (butter clams, steamer clams, horse clams, chestnut clams and soft-shell clams) as bioindicators because they are sessile filter feeders, they are abundant on East Sound beaches, and they form part of the local human food supply. Nutrient and contaminant loads in water can fluctuate daily if not hourly, whereas sessile

filter feeders accumulate toxic compounds over time, providing a more reliable integrated measure of total toxic inputs to the environment than water samples. By testing a number of locally abundant clam species, moreover, we hoped to ascertain differences in species' pyrethroid uptake rates, and thereby gain some insight into the best choice of species for future monitoring purposes. In addition, we collected and tested freshwater clams from a wetland upstream from the town storm sewer system, but influenced by the town airport, fire station, and some residences.

Materials and methods

Bivalves were collected by shovel from sandy sediments on four beaches in East Sound, the shallow fjord that bisects Orcas Island (Figure 1). Fishing Bay is immediately adjacent to the town of Eastsound, and bivalve were collected within 500 feet of the town storm sewer system outfall. Crescent Beach is located immediately east of the town, on a separate drainage with extensive wetlands. The regional power utility yard and a number of residences are located on the upstream fringes of the wetlands. Crescent Beach and its bay (Ship Bay) and Fishing Bay are separated into two shallow basins by Madrona Point, reducing but not eliminating mixing of their waters.



Figure 1:
Bivalve collection sites in
East Sound, Orcas Island, WA

For comparison with urban-influenced beaches, bivalves were also collected from two sites near the mouth of East Sound, where we expected a strong marine influence and more of a regional signal than a local one. White Beach is an exposed high-energy reach of the west shore of East Sound, with few residences and freshwater inputs in the form of runoff from hills and bluffs. Buck Bay is watered by Cascade Creek, and despite strong tidal flushing its substrates are relatively fine due to armoring, roads and residences on its

shores. However, the number of homes draining to Buck Bay are barely 10 percent of the number draining to Fishing Bay.

Five clam species harvested for human consumption were collected : butter clams (*Saxidomus giganteus*), chestnut clams (*Saxidomus nuttalli*), steamer clams (*Protothaca staminea*), horse or gaper clams (*Tresus capax*), and soft-shelled clams (*Mya arenia*). All were stored in cold seawater for up to 48 hours before being cracked, dissected from their shells with a surgical scalpel, and rinsed in nanopure water. Stomachs and their contents were carefully removed and the remaining tissue stored dry in sterile Nalgene jars at 4°C.

Freshwater clams (Sphaeridae) were collected by shovel and sieve from an area of approximately 25 square meters in a slough-sedge and alder wetland that drains towards Fishing Bay. A total of 104 clams were collected, averaging 3 mm in length and 5 years in age. All were homogenized, with their shells, as a single sample.

Figure 2: Age distribution of marine specimens (N=98 bivalves)

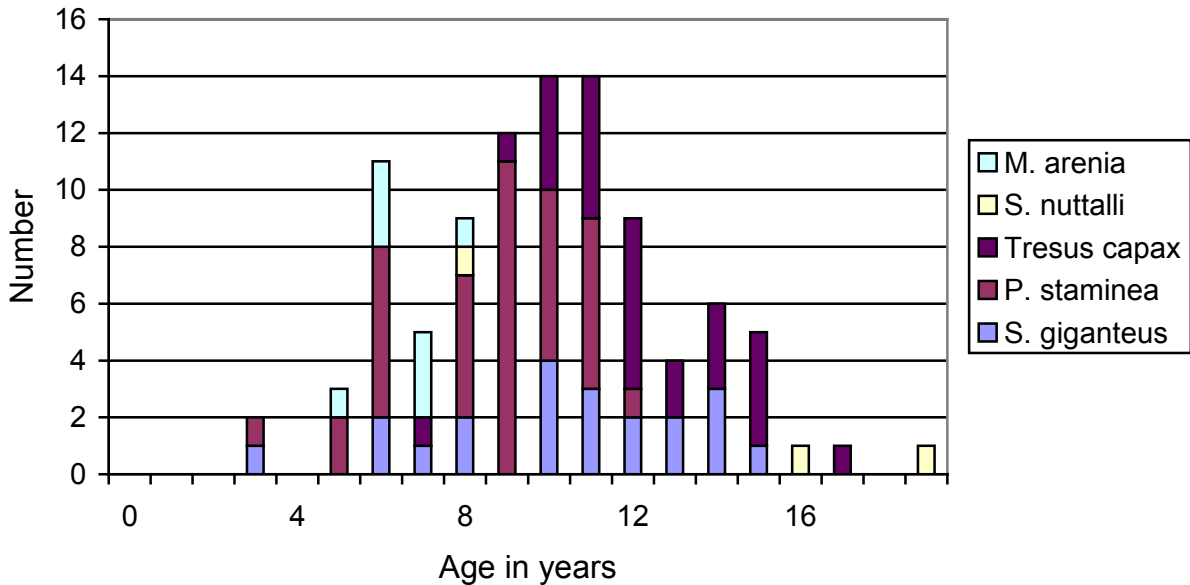


Table 1: Species composition of the marine sample (N=98 bivalves)

	Fishing Bay	Crescent Beach	Buck Bay	White Beach
<i>S. giganteus</i>	10	4		
<i>P. staminea</i>	16	4	6	13
<i>Tresus capax</i>	10	6	1	10
<i>S. nuttalli</i>	2	1		
<i>Mya arenia</i>			8	7
	38	15	15	20

Tissue samples were extracted within 72 hours by a matrix solid-phase dispersion (MSPD) method (Barker 2007; Zhao et al. 1999). For each extraction, we measured and combined 0.2g of tissue and 0.75g of Na₂SO₄ with 2.0g of 40-63 micron mesh silica in a ceramic mortar and ground them together.¹ This mixture was loaded into a 10-mL syringe with a 22-µm cellulose acetate membrane syringe filter. A 1.5-cm qualitative paper filter was placed on top of the tissue mixture, followed by another 2.0g of 40-63 micron mesh silica, and another paper filter. A total of 7.5mL spectro grade methanol was then added to the syringe in three equal aliquots over a period of 20 minutes. After adding the third aliquot of methanol, the plunger was inserted into the barrel of the syringe and depressed firmly, eluting 1-2 mL of extract. Each extract was re-filtered through a 22-µm cellulose acetate membrane syringe filter before use to remove any remaining silica particles.

The methanolic extracts were tested for pyrethroid pesticides and high-chlorinated PCBs using magnetic particle format ELISA immunoassay kits manufactured by Abraxis, LLC (Warminster, PA). Extracts were tested in batches of up to 12 specimens, each with positive and negative controls, an instrument blank and four standard solutions of varying concentrations of the target analytes to provide a calibration curve. Any anomalies in the calibration curve or results of the controls led to rejection of the batch and re-testing.

ELISA relies on the selectivity and sensitivity of antibodies produced by animals exposed to the target analytes. The refined antibodies are bonded to very small magnetic beads, which are mixed with the material to be tested in a methanolic solution; separated and washed; then reacted with an enzyme-tagged conjugate that reacts with all remaining unreacted antibodies on the magnetic beads. The beads are again washed; then combined with a reagent that reacts with the enzyme-tagged conjugate to produce a yellow color. A spectrophotometer is used to measure the color, and thereby the amount of conjugate that reacted with the bonded antibodies—which is inversely proportional to the concentration of target analytes in the sample. We modified the ELISA test procedures recommended by Abraxis in one respect: antibody reactions were incubated for 10 minutes rather than five minutes, which we have found by experiment to improve sensitivity and consistency of the test.

ELISA test systems for chemical families such as pyrethroids are standardized for a single chemical compound or “species” in each family. The Abraxis ELISA system for pyrethroids is standardized for permethrin, which is the pyrethroid pesticide most widely found in products currently sold in San Juan County (Barsh et al. 2008). Other pyrethroid species such as tetramethrin and deltamethrin react less strongly with the antibody, hence their presence is *underestimated*. Similarly, the Abraxis high-chlorinated PCB system is standardized for Arochlor 1254, a particular commercial mixture of PCB congeners.

At the same time, ELISA systems can cross-react with non-target compounds, and thereby *overestimate* the concentration of target compounds in the sample. For example, the Abraxis ELISA system for pyrethroids cross-reacts with PBA, a metabolite (or breakdown byproduct) of deltamethrin and other pyrethroids. The cross-reactivity is weak, so a very large concentration of PBA is required to mimic the effect of a small concentration

¹ In the case of freshwater clams, where separation of the thin shells from body tissue was impractical, we ground shells and body tissue together and used 0.4g of the combination in MSPD after estimating that the shells constituted approximately half of the total weight of each freshwater clam.

of amount of permethrin. The possibility that ELISA results are significantly influenced by non-target compounds can only be discounted by more specific analytical methods—in our laboratory, liquid chromatography and mass spectrometry (LC/MS).

ELISA systems function best within a relatively small range of concentrations of the target analyte, generally 0.1 to 15 parts per billion. Antibody reaction rates may also be depressed if samples contain high concentrations of large non-target molecules such as the lipids and polypeptides that presumably co-extracted with any pyrethroids or PCBs in our methanolic bivalve extracts. Results of our first batch of pyrethroid tests were mainly above the calibration curve. When we re-tested the same extracts at 50 percent dilutions, many of our results were still above the new calibration curve. Only when we diluted the first batch of extracts to 5 percent of their original concentrations did nearly all the results fall within the batch calibration curves. After back calculating to account for dilution, the results at 5 percent were up to several times higher than results at 50 percent, presumably due to the reduced concentration and diminished effect of co-extracted species.

For consistency, all samples were tested at 5 percent of original concentrations for pyrethroids, and 50 percent for PCBs, where we saw no evidence of interference from co-extracted compounds.

Results

Methanolic extracts of 100 marine bivalves and a single composite sample of 104 freshwater bivalves collected around East Sound were tested for pyrethroid pesticides and high-chlorinated PCBs using ELISA. Results are expressed in parts per billion (ppb) and shown by collecting site (graph colors) and the age of specimens (estimated from growth rings) in Figures 3 and 4.

Figure 3: Pyrethroid pesticides, by collecting site and age of bivalve (N=99)

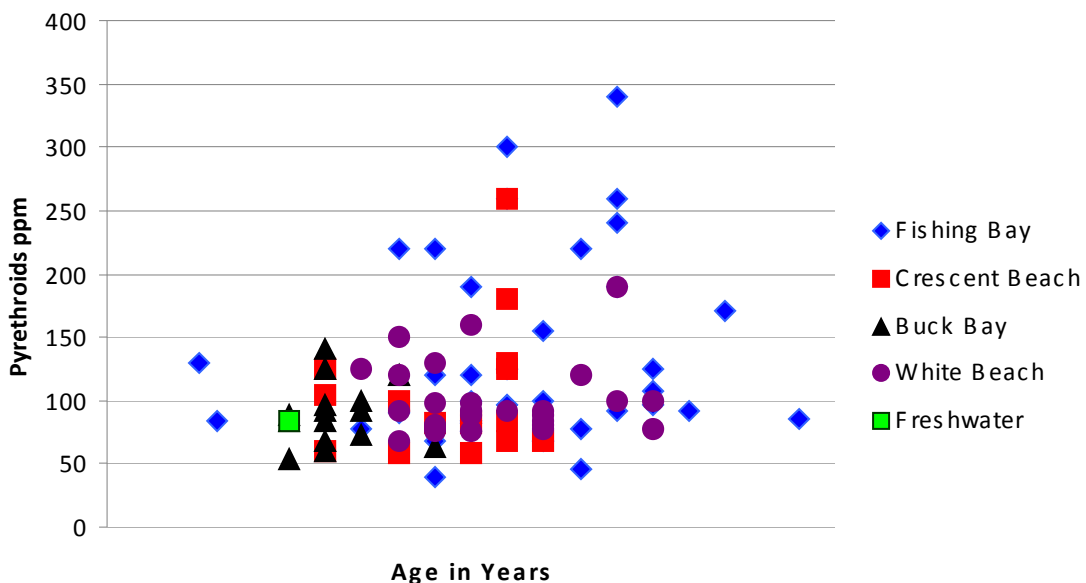
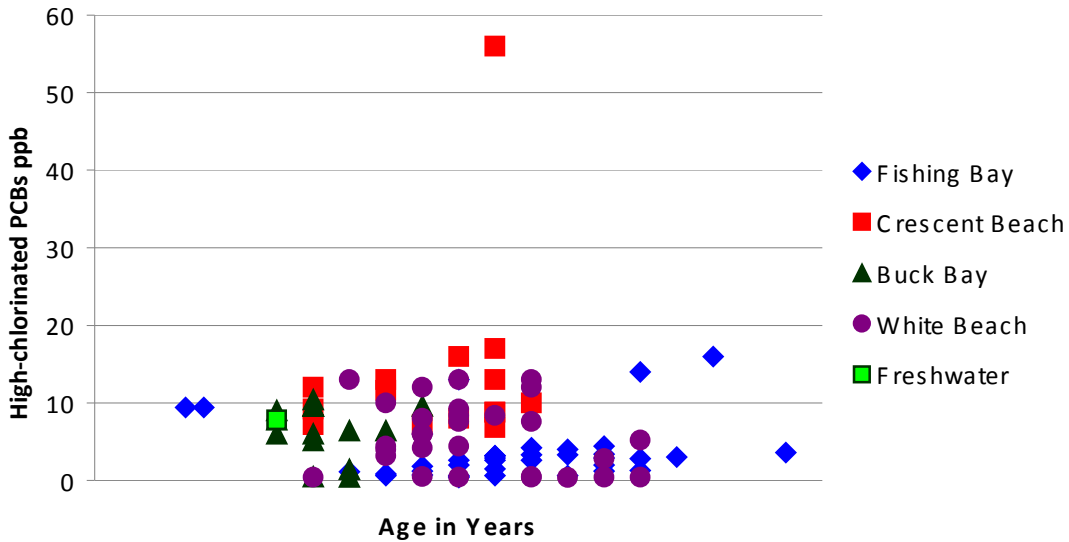


Figure 4: High-chlorinated PCBs, by collecting site and age of bivalve (N=99)



Pyrethroid concentrations appear to be greatest at Fishing Bay (Figure 3), whilst PCBs concentrations appear to be greatest at Crescent Beach (Figure 4), both consistent with predictions. We used the t-statistic to test these apparent differences; results appear in Table 2 (pyrethroids) and Table 3 (PCBs).

Table 2: Two-tailed t-statistic for pair-wise comparison of mean pyrethroid loads

	Fishing Bay	Crescent Beach	Buck Bay	White Beach
Fishing Bay		1.3878	<i>2.3254</i>	<i>2.5270</i>
Crescent Beach	1.3878		1.0231	0.4861
Buck Bay	<i>2.3254</i>	1.0231		1.1508
White Beach	<i>2.5270</i>	0.4861	1.1508	

Italics ($p < 0.05$), Bold ($p < 0.01$).

The observed greater pyrethroid loads at Fishing Bay, in comparison with loads in bivalves from Buck Bay and White Beach, are statistically significant at the $p < 0.05$ level; the probability that the observed differences in mean loads are due to chance alone is less than 5 percent. There was no statistically significant difference between pyrethroid loads in bivalves from Fishing Bay and Crescent Beach, however, or between Crescent Beach, Buck Bay, and White Beach. This is consistent with spillover of local-source toxics from Fishing Bay to Crescent Beach, as well as substantial mixing and dilution of toxics within East Sound as a whole. The local contaminant signal is strongest at Fishing Bay, and it is weakest at the entrance to East Sound, where oceanic mixing is greatest.

Table 3: Two-tailed t-statistic for pair-wise comparison of mean PCBs loads

	Fishing Bay	Crescent Beach	Buck Bay	White Beach
Fishing Bay		<i>4.6273</i>	<i>4.5824</i>	<i>2.0301</i>
Crescent Beach	<i>4.6273</i>		<i>2.7661</i>	<i>3.2678</i>
Buck Bay	<i>4.5824</i>	<i>2.7661</i>		0.7011
White Beach	<i>2.0301</i>	<i>3.2678</i>	0.7011	

Italics (p<0.05), Bold (p<0.01).

Statistical analysis of PCBs data poses more striking contrasts between collecting sites (Table 3). Crescent Beach is more contaminated than any other collecting site at the p<0.05 level or better. Fishing Bay—closest to Crescent Beach—is more contaminated than either of the two outer beaches (White Beach, Buck Bay). PCB loads do not differ significantly between the two outer beaches, however. This pattern is consistent with a local source of PCBs in the Crescent Beach drainage. PCB loads at Buck Bay and White Beach are not trivial, but presumably reflect chiefly the regional effect, i.e. pollution from the urban areas that surround the islands.

The local contaminant signal from the town of Eastsound boosts mean pyrethroid loads in bivalves by approximately 40 percent. The effect of legacy PCBs in the Crescent Beach watershed is slightly greater. We are assuming for this purpose that the pyrethroid and PCB loads we observed at White Beach and Buck Bay represent chiefly the regional background level of these contaminants within the Salish Sea. It would be useful to test this assumption by measuring contaminant loads in bivalves from other San Juan County beaches at a greater distance from, and less likely to be influenced significantly by runoff from Eastsound or other developed areas of the county.

A secondary question addressed by this study is bioaccumulation of contaminants. A brief glance at Figures 3 and 4 suggests that pyrethroid loads may be weakly correlated with the age of bivalves, at least at Fishing Bay. This appears more clearly when Fishing Bay results are isolated from the data for other sites (Figure 5). The apparent correlation is not statistically significant, however ($r^2 = 0.0024$). Similarly, we found no statistically significant correlation between PCB loads and age in our sample. This may partly be due to the small size of our Crescent Beach sub-sample, and relatively narrow range of ages it represents (6-12 years). As discussed below, pyrethroids gradually degrade in organisms as well as soils and water. More stable contaminants such as PCBs may slowly be shed when organisms excrete. Instantaneous body loads of contaminants reflect the net effects of uptake and excretion.

The same consideration led us to investigate possible differences between species in their uptake and excretion rates for pyrethroids and PCBs. Our random bivalve sample is not ideal for this purpose. Comparable numbers of each species could not be obtained by this means because species were not distributed randomly on all beaches, and some were present on only one or two beaches (*Mya arenia* and *Saxidomus nuttalli*). Figure 6

shows the relationship between age, species, and pyrethroid loads at a single site (Fishing Bay).

Figure 5: Indian Island pyrethroid loads as a function of age (N=38)

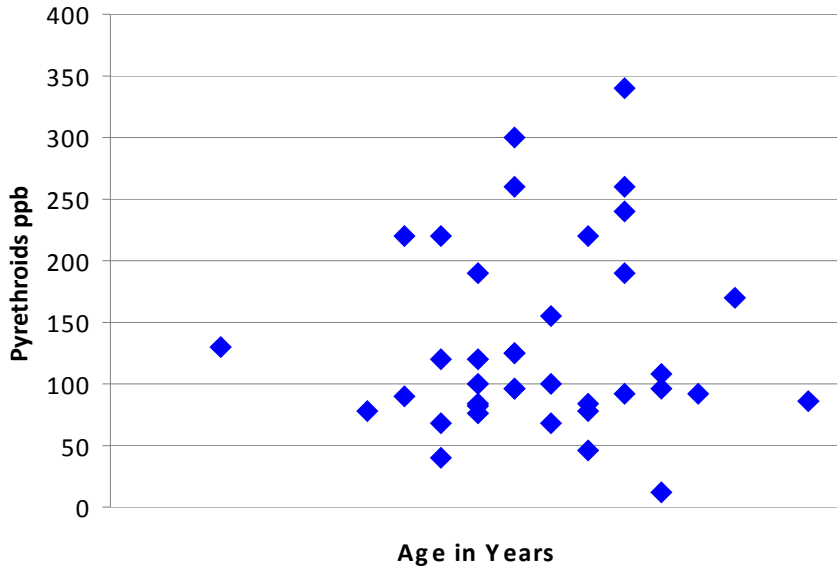
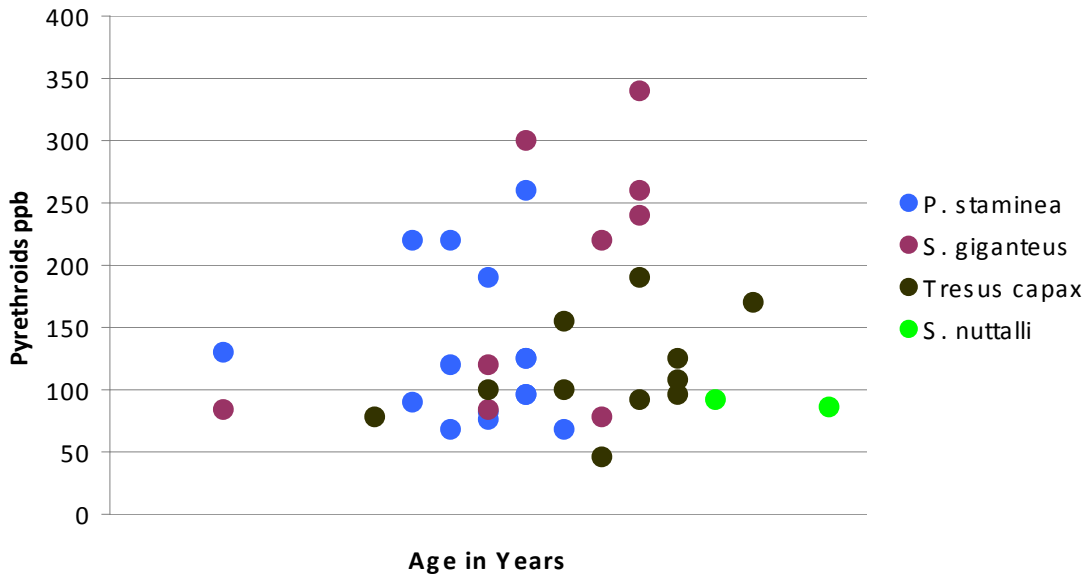


Figure 6: Pyrethroid loads by age and species, Fishing Bay (N=38)

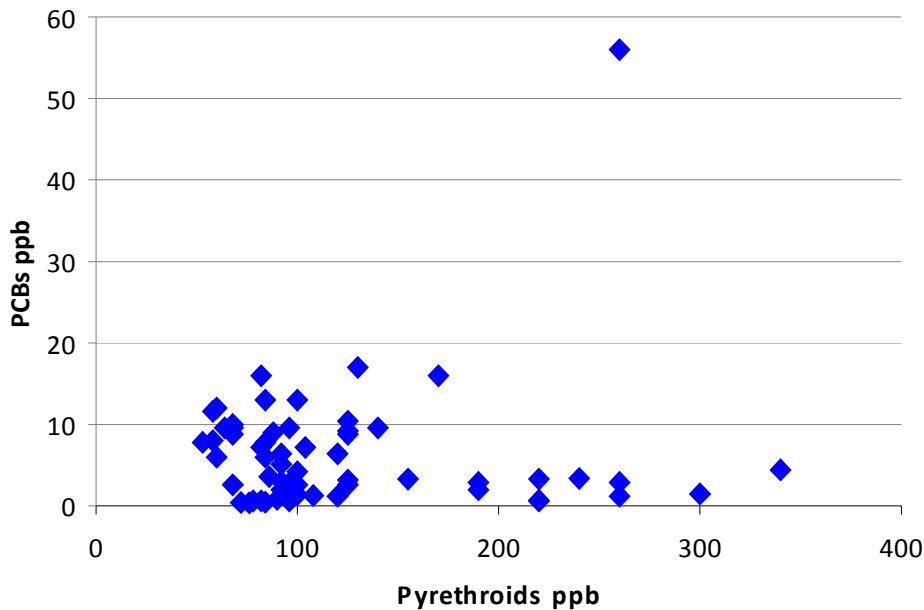


The mean pyrethroid load for the common butter clam *Saxidomus giganteus* (192 ppb) was greater than the mean for either *Protothaca staminea* (131 ppb) or *Tresus capax* (114 ppb). The difference between *S. giganteus* and *T. capax* is statistically significant at the $p < 0.05$ level ($t = 2.2979$), whereas the difference between *S. giganteus* and *P. staminea*

is not quite statistically significant ($t=1.8284$, $p=0.034$). That is, butter clams accumulate higher concentrations of pyrethroids per unit of body mass than horse clams, and perhaps more than steamers as well. A larger sample would clarify these relationships.

Since pyrethroid and PCB loads were elevated at different beaches, and neither of these contaminants was correlated with bivalves' age, it should not be surprising that we found no significant correlation between pyrethroid loads and PCB loads (Figure 7) ($r^2 = 0.007$). Pyrethroid and PCB loading reflect different sources and vectors.

Figure 7: PCB loads as a function of pyrethroid loads (N=99)



It should be noted that, notwithstanding the statistical tendency for Fishing Bay bivalves to be more contaminated with pyrethroids, and the Crescent Beach bivalves with PCBs, there was considerable individual variation in contaminant loads at all four marine collection sites. Even on the most contaminated beaches, some of the individual bivalves were no more contaminated than bivalves from the least contaminated beaches. Seven of the eight bivalves in the sample with the highest pyrethroid loads—all in excess of 200 ppb—were collected from Fishing Bay, however.

The composite sample of freshwater bivalves tested close to the sample mean for both pyrethroids and PCBs: 84 ppb pyrethroids (sample mean=112 ppb) and 8 ppb PCBs (sample mean=6.2 ppb). We have assumed that this reflects *only* the local signal whereas results from Buck Bay and White Beach *mainly* reflect the regional signal. Mean loads at Buck Bay were 85 ppb pyrethroids and 6.5 ppb PCBs. Combining the presumed regional signal with the presumed local-only signal yields 169 ppb pyrethroids and 14.5 ppb PCBs—somewhat more than our actual mean test results at Fishing Bay (136 ppb pyrethroids) and Crescent Beach (14 ppb PCBs).

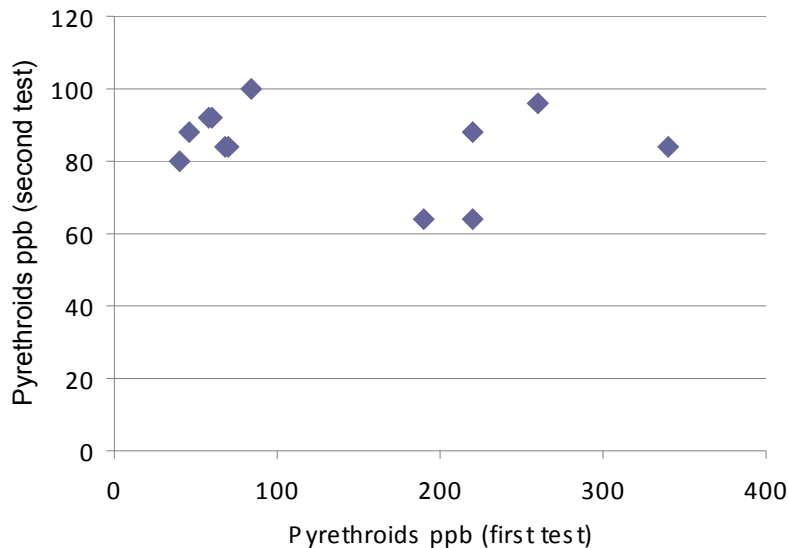
Comparison can be made with test results from three juvenile salmon collected in the course of the summer 2009 season in connection with another study. These fish were young-of-the-year and had lived their entire lives migrating down their natal streams and through the Salish Sea. Their mean pyrethroid load was 75.3 ppb, and their mean PCBs load was 8.5 ppb. This is close to bivalve loads at Buck Bay, which we have assumed to be primarily a regional signal. Additional sampling across sites and species in San Juan County would provide a more reliable measure of the relative magnitude of regional and local-source contamination of marine animals.

Quality assurance

Each test run or batch was independently calibrated to the pyrethroid (permethrin) standards supplied by the manufacturer of the ELISA test system. Any test run that failed to produce a logistic calibration curve, or that over- or underestimated the positive control (3.2-3.8 ppb permethrin) by more than 30 percent was rejected.

In addition we randomly selected and re-tested 12 bivalve extracts after 1-3 weeks to assess measurement error and sample degradation. Mean pyrethroid loads when these extracts were initially tested were higher (139 ppb) than when re-tested (86 ppb), but the difference in mean test results is not statistically significant ($t=1.753$, $p>0.1$), nor is there a statistically significant correlation between the initial results and re-test results (Figure 8) ($r^2 = 0.0569$).

Figure 8: Initial and re-test results for pyrethroids (N=12)



Variation in results between test runs does not invalidate our inferences about the local sourcing of pyrethroids and PCBs in East Sound, because the assignment of bivalve extracts to test runs was randomized. The observed level of variation weakens our ability to quantify the loads of contaminants in East Sound bivalves precisely, however; and thus

leaves us unable to state with confidence that individual bivalves or populations are “safe to eat” within current EPA guidelines. For the results in Figure 8, the standard error was 30 ppb. In other words, an ELISA result of 90 ppb should be understood as 90 ± 30 ppb (a range of 60-120 ppb).

As noted above, ELISA methodology is extremely sensitive and selective, but it is subject to error due to cross-reactivity of antibodies, as well as possible interferences with antibody reactions in complex biological matrices such as tissue extracts. Overestimation and underestimation of the target analyte are both possible. Independent verification by a different analytical method is required for confidence in results.

To corroborate the underlying validity of our immunoassay results we employed a Finnegan Duo electrospray ionization LC-mass spectrometer (LC/MS). We compared the ionization byproducts of selected bivalve extracts with two standards: deltamethrin, because it is the most persistent of the pyrethroids widely sold in San Juan County, and 3-phenoxybenzoic acid (3-PBA), a metabolic breakdown product of deltamethrin and other pyrethroids that cross-reacts with ELISA-system antibodies. With an ammonium chloride adduct, both deltamethrin and 3-PBA disintegrate into molecular fragments with distinct masses. We confirmed 3-PBA in the extracts tested, at concentrations consistent with our ELISA test results. (Complete details of tuning and spectrograms available upon request.)

Discussion

Pyrethroid pesticides are a family of naturally derived and synthetic neurotoxins that can persist in the environment for weeks to months, and can accumulate in the fatty tissues of both invertebrate and vertebrate animals (Todd et al. 2003; Shafer et al. 2005). Pyrethroids affect respiration, olfaction, and disrupt endocrine signaling in Pacific salmon (Tollefson et al. 2002; Wheelock et al. 2005; Tierney et al. 2006). Concentrations as low as 0.1 parts per billion can be toxic to rainbow trout (*Oncorhynchus mykiss*).

Pyrethroids also pose a threat to human health. Although they are much less toxic to mammals than to fish or invertebrates, pyrethroids are considered a health risk at very low levels. Federal law limits deltamethrin residues in meat and eggs to 20-50 parts per billion (EPA 2004). If we suppose that 120 ppb is the maximum acceptable ELISA result for pyrethroids in shellfish intended for human consumption, then more than one-third of the bivalves collected from Fishing Bay exceed this level of contamination (34 percent), as opposed to 11 percent at beaches farther from town (Buck Bay and White Beach).

Reducing this threat to food safety and human health in Fishing Bay is relatively easy. Pyrethroids break down in the environment within months; we saw no evidence of long-term accumulation in bivalves. Therefore the contamination we observed at Fishing Bay represents *current use* of home and garden pesticide products. Reducing local use of these products will translate immediately into safer shellfish.

Pyrethroids are considerably less toxic to mammals than the pesticides commonly used in agriculture or by licensed pest control operators, hence manufacturers prefer them in over-the-counter products intended for household use. Outdoor spraying of pyrethroid

pesticides produces an aerosol that can drift hundreds of feet and fall directly into nearby waters, or coat soil and dust particles that later wash off lawns and driveways into nearby streams or storm drains. Most pyrethroid products for lawn and garden use surfactants as emulsifiers,² and this facilitates their dispersal in soils, ponds, streams, and storm sewers. Potential sources of pyrethroid discharges to surface waters in Eastsound include homes, lawns, gardens, and businesses that have outdoor patios or decorative floral displays in the commercial center of the town. Drainage from yards, lawns, streets and sidewalks is collected by the town's storm sewers and discharged into Fishing Bay.

Although agriculture is widely perceived as the primary source of pesticides in the environment, cities and towns throughout the Puget Sound Basin contribute even larger loads of home and garden pesticides (Bortleson and Davis 1997). In an urbanizing region we should expect pyrethroids to overtake agricultural chemicals as a threat to wildlife and human consumers of wildlife.

The threat from residual polychlorinated biphenyls (PCBs) in East Sound is more difficult to address. PCBs were widely manufactured for use in electrical transformers until prohibited by Federal law in 1979. They are highly persistent and bioaccumulative. The elevated signal we observed in bivalves from Crescent beach presumably represents residual PCBs in the watershed and/or beach and bay sediments from the era when PCBs-filled transformers were stored outdoors at the OPALCO yard in the northeast quarter of this small drainage basin. Efforts to remove PCBs from the OPALCO property would not have removed PCBs from downstream wetlands or nearshore areas. We did not detect PCBs in this basin in a previous study (Barsh et al. 2007) that used colorimetric methods with a limit of detection of 100 ppb PCBs, compared with the 0.1 ppb ELISA LOD.

Federal law limits PCBs in drinking water to 0.5 ppb (EPA 2003), with somewhat higher cutoffs for PCBs in food residues. Infaunal bivalves wild-harvested from Crescent Beach, with ELISA test results averaging 6.5 ppb PCBs, should be treated with caution.

Pyrethroid and PCB loads in freshwater bivalves from the Eastsound slough were comparable to loads in bivalves at White Beach and Buck Bay. We suggested that this is an indication of the magnitude of the local-source signal, which is to say about as great as the regional signal from urban discharges into Puget Sound and the Gulf of Georgia that are greater initially, but have been much more diluted by the time they reach the islands.

Contaminant loads in the freshwater and marine bivalves we collected might also reflect airborne deposition. Both pyrethroid pesticides and PCBs can travel considerable distances as aerosols, and are detected in habitats where there are no possible waterborne vectors (Oono et al. 2008; Demers et al. 2007; Ohyama et al. 2004). Any airborne signal would contribute approximately equally to all freshwater and marine environments in the same airshed, however, and thus balance out of any local comparisons. It is most likely that the elevated levels of pyrethroids we observed at Fishing Bay and PCBs we observed at Crescent Beach, reflect the sum of the regional signal (airborne plus marine) and local-source pollution.

² Pyrethroid molecules are waxy and insoluble in water (hydrophobic). "Knock-down" products aimed at killing insects such as wasps instantly dissolve the active pyrethroid ingredient in a volatile organic solvent that evaporates quickly, leaving only very fine particles of the pesticide. Lawn and garden sprays suspend the active ingredient in water, which does not evaporate as quickly and can travel further.

Recommendations

Federal law limits deltamethrin residues in meat and eggs to 20-50 ppb (parts per billion) (EPA 2004). Using this benchmark, we recommend that human consumption of bivalves from Fishing Bay be suspended until such time as pesticide loads in Fishing Bay can be reduced to background levels through reduced local consumption of home/garden products containing pyrethroids.

This study did not examine cultivated bivalves, which are grown in bags or cages above the sediment, and are harvested at a younger age than the bivalves we collected for study. These factors should make cultivated bivalves safer for human consumption than infaunal wild-harvested bivalves living in the same bodies of water. Shellfish growers in East Sound will nonetheless face a growing threat from the development and build-out of the town of Eastsound, unless measures are taken now to reduce local use of pesticides in the developed area that drains or runs off into Fishing Bay and Crescent Beach.

Acknowledgments

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