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Bioconcentration, Bioaccumulation, and Biomagnification in Puget Sound Biota: Assessing the Ecological Risk of Chemical Contaminants in Puget Sound

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Introduction:

Puget Sound has a large urban and rural human population, which currently exceeds 3 million, and many industrialized ports and shorelines that provide numerous sources of non-point and point source pollution to Puget Sound (Konasewich et al. 1982). Hundreds of potentially toxic chemicals are present in Puget Sound sediments (Malins et al. 1982, NOAA and WSDE 2000, Konasewich et al. 1982, and Lefkovitz et al. 1997). As of 1982, 183 organic compounds had been identified in Puget Sound sediments, biota, and water (Konasewich et al. 1982). Although chemical contaminants and heavy metals are present in sediments throughout Puget Sound, these pollutants are generally greatest in number and concentration within the sediments and embayments that are adjacent to the most populated and industrialized areas, such as Elliot Bay, Commencement Bay, and Sinclair Inlet (Malins et al. 1982, Lefkovitz et al. 1997, Konasewich et al. 1982, and NOAA and WSDE 2000). However, the distribution and concentrations of chemical contaminants and heavy metals in Puget Sound generally reflect their source. For example, the release of arsenic is relatively source-specific as compared to lead, which is released into the environment from many ubiquitous sources, e.g. tetraethyl lead in gasoline, and is therefore present throughout Puget Sound (Malins et al. 1982). Unfortunately, detailed coverage of the chemical contaminants and heavy metals in Puget Sound sediments and their concentrations, distribution, sources, and deposition trends are outside of the scope of this review. For information on these subjects, see Malins et al. (1982), NOAA and WSDE (2000), 2 Konasewich et al. (1982), and Lefkovitz et al. (1997). However, a brief list of chemical contaminants and heavy metals of concern, and their concentrations in Puget Sound sediments is provided in Table 1.

As a result of the degree of chemical contamination, recent studies have found that toxic chemicals are also present in many benthic and pelagic organisms within Puget Sound (See Tables 2 and 3) (Malins et al. 1982, Williams and C. Krueger 1988, NOAA and WSDE 2000, Konasewich et al. 1982, and Lefkovitz et al. 1997). Research has shown that certain chemicals have the ability to be bioconcentrated in organisms directly from the water, and bioaccumulated and biomagnified within food chains, causing higher trophic organisms to become contaminated with higher concentrations of chemical contaminants than their prey (Morrison et al. 1996, Gobas et al. 1999, Nakata et al. 1998, Bard 1999, Jarman et al. 1996, Konasewich et al. 1982 Williams, L.G. and C. Krueger 1988, Lee et al. 2000, Hayteas and Duffield 2000, and Hargrave et al. 2000).

The importance of understanding the mechanisms behind the bioconcentration, bioaccumulation, and biomagnification of toxic chemicals in biota is generally recognized among scientists. However, the mechanistic explanations for these processes are highly debated, and currently unresolved (Gobas et al. 1999). Furthermore, a review of the literature will quickly reveal that the terms bioconcentration, bioaccumulation, and biomagnification are used inconsistently (Konasewich et al. 1982). For the purposes of this review, the following definitions will apply to these terms: (1) Bioconcentration is the intake of chemical contaminants through an organism's epithelial tissues or gills, and the subsequent concentration of that chemical contaminant within the organism's tissues to a level that exceeds ambient environmental concentrations (adapted from Konasewich et al. 1982 and Gobas et al. 1999). (2) 3 Bioaccumulation is the process by which chemical contamination in organisms increases with each step in the food chain (Gobas et al. 1999). (3) Biomagnification is the process by which chemical contaminants are concentrated at levels that exceed chemical equilibrium from dietary absorption of the chemical (Gobas et al. 1999).

The bioconcentration, bioaccumulation, and biomagnification of chemical contaminants in marine biota are dynamic processes that involve many interconnected variables. For example, the potential of a chemical to bioconcentrate, bioaccumulate, or biomagnify in organisms and food webs is dependent upon the properties of the chemical (e.g. hydrophobicity, lipophilicity, and resistance to degradation), environmental factors (e.g. salinity, temperature, concentration of other organic chemicals, and redox potential), biotic factors (e.g. the organism's mode of feeding, trophic position, lipid concentration, and metabolism), and bioavailability (e.g. current chemical inputs, transport mechanisms, and degree of contamination) (Konasewich et al. 1982, Malins et al. 1982, Shin and Lam 2001, Gobas et al. 1999, Morrison et al.1996, and Lee et al. 2000). Although this is not a comprehensive list of variables, it serves to illustrate how dynamic these processes truly are, which makes determining the ecological risks associated with bioaccumulation potential in Puget Sound biota especially difficult.

Determining Bioaccumulation Potential in Puget Sound Biota:

Many researchers have attempted to determine the potential for bioconcentration, bioaccumulation, and biomagnification of chemical contaminants in Puget Sound biota with varying success. Research has shown that the bioaccumulation potential of a chemical contaminant is greatest for highly lipophilic chemicals, and increases with increased lipid content in aquatic organisms. In general, the bioaccumulation of organic chemicals is more significant than bioaccumulation of metals. This is due to the fact that most metal contaminants tend not to 4 be lipid-soluble in the aquatic environment. As a result, metals will more commonly accumulate in non-lipid rich tissues (e.g., the gills of fish). However, if the metal is incorporated into a lipophilic organic compound (e.g., methyl mercury compound) the accumulation of the metal is enhanced.

As a result of the relationship between lipophilicity and bioaccumulation potential, much research has focused on quantifying the lipid solubility of chemicals by determining the experimental partition coefficient (KOW) of the organic chemical between n-octanol (a surrogate for lipids) and water. As an example, PCBs have a KOW of 106, which means that PCBs are 106 times more soluble in n-octanol than water, and therefore highly lipophilic and very hydrophobic (Konasewich et al. 1982 and Malins et al. 1982). Generally speaking, chemicals with a KOW less than 105 are primarily bioconcentrated directly from the water, while those chemicals with a KOW greater than 105 - 106 are primarily bioaccumulated through dietary intake (Gobas et al. 1999). On the other hand, the American Institute of Biological Sciences Aquatic Hazards of Pesticides Task Group recommends that compounds with a KOW greater than 103 should be considered as having a high potential for bioaccumulation (Konasewich et al. 1982).

Although the KOW of a chemical can help predict a chemical's bioconcentration or bioaccumulation potential, there are several problems associated with relying on this measure alone, that may result in substantial error (Konasewich et al. 1982 and Gobas et al. 1999). As mentioned before, there are many environmental factors that affect a chemical's true bioaccumulation potential in the field that are not taken into consideration during the determination of KOW. For example, Konasewich et al. (1982)

report that increased salinity enhances bioconcentration and bioaccumulation rates by decreasing its solubility in water, and subsequently increasing its lipophilicity. Therefore, it can be assumed that areas with higher 5 salinities within Puget Sound will have higher bioaccumulation potential than areas with lower salinities, despite the calculated KOW (Konasewich et al. 1982).

In addition, researchers often attempt to calculate a bioconcentration factor (BCF), which is the ratio of chemical contaminants in an organism's tissues to ambient water concentrations (Hargrave et al. 2000 and Advanced Chemistry Development 2002), or the biomagnification factor (BMF), which is the ratio of a contaminant's concentration at one trophic level to that at the next trophic level calculated on a lipid weight basis (Bard 1999). Bioconcentration factors have been reported as arithmetic means for groups of organisms, e.g. fish, bivalves, and shrimp (Office of Environmental Health Hazard Assessment 1999). However, the reliance on these generalized BCFs in practical applications, such as for policy setting and contamination monitoring, may produce substantial error given that BCFs vary greatly with biotic factors (e.g., species, sex, and season) and physical factors (e.g., pH, temperature, salinity, and redox potential).

To incorporate more variables and interactions in the determination of bioaccumulation potential in Puget Sound biota, biologists and ecologists often use models. The equilibrium partitioning model (EPM) is a model that is most often used by researchers to predict bioaccumulation in benthic invertebrates (Morrison et al. 1996). This model assumes that chemical accumulation of contaminants is in thermodynamic equilibrium - an assumption that would normally be reasonable. However, Gobas et al. (1999) and other researchers (cited in Gobas et al. 1999) found that chemical contaminants are being transported against their thermodynamic gradient with each step in the food chain, otherwise known as biomagnification. This biomagnification produces concentrations of contaminants that are higher than that which can be explained by the chemicals physical properties alone. Gobas et al. (1999) proposed that 6 this biomagnification is the result of an increase in the chemical's fugacity, or thermodynamic potential, as a result of gastrointestinal digestion. Gobas et al. (1999) indicate that biomagnification at each step in the food chain can lead to considerably higher concentrations at the top of the food chain, even when changes in lipid content are taken into consideration.

Also, the EPM assumes that the biota sediment accumulation factor (BSAF) is constant and independent of the chemical, organism, and sediment properties. Research has shown that these assumptions can produce errors in bioaccumulation prediction of up to 5 orders in magnitude in PCB congeners. While this discrepancy may be an exceptional case, other studies have found smaller yet considerable degrees of variation in BSAFs. Furthermore, the EPM does not sufficiently distinguish between the diversity of feeding strategies (e.g., filter feeding vs. detritus feeding) which may result in significant differences between BSAFs in organisms (Morrison et al. 1996). For example, Lee et al. (2000) found that feeding strategy differences significantly affected the bioaccumulation of heavy metals. These potential sources of error are of particular concern given that this model has been adopted by United States agencies to establish sediment quality guidelines (Morrison et al. 1996). To more accurately predict bioaccumulation potential, models that account for differences in feeding strategies, such as the one created by Morrison et al. (1996), should be used.

In addition to models, researchers have relied on bioindices and bioassays to assess the level of contamination in Puget Sound biota (Rice et al. 2000, Roubal et al. 1978, Yunker et al. 2002, NOAA and WSDE 2000, and Malins et al. 1982). Surveying species diversity, benthic mortality, and changes in trophic composition is yet another approach used by researchers to obtain a more accurate picture of the effects of sediment contamination (Shin and Lam 2001). The use of bioindices has been proposed for Puget Sound, in which surveying the relative 7 abundances of species that are determined to be tolerant, sensitive, and intolerant can be used to assess the relative level of chemical contamination in sites. As an example, a study area in British Colombia used all scallops, sea cucumbers, sponges, and sea urchins as intolerant species (Reish et al. 1999).

The Washington State Department of Ecology's Sediment Management Unit has created a Sediment Quality Information System Database (SEDQUAL) that includes records for over 658,000 chemical, 138,000 infaunal benthic invertebrate surveys, and 36,000 bioassays from over 12,000 sample collection stations in Puget Sound. This database has been used to identify sites that exceed state Sediment Quality Values (SQS), Puget Sound Marine Sediment Cleanup Screening Levels (CSL), and Sediment Management Standards (SMS). Of the 2,063 SEDQUAL samples from central Puget Sound surveyed by NOAA and WSDE (2000), over half (1,034) registered contaminant levels that exceeded at least one SQS or CSL.

In a study by NOAA and WSDE (2000), use of the SEDQUAL, bioassays, species diversity surveys, toxicology studies, and sediment sampling were combined in an attempt to determine the spatial distribution of contamination and ecological risks in Puget Sound. However, reports for the Northern and Southern Basins of Puget Sound have not yet been published. The completion of these reports will provide researchers with a more accurate assessment of the distribution of contamination and its potential ecological impacts.

Ultimately, the collective goal of the previously mentioned research is to develop a model, indices, or monitoring system in which environmental factors, biotic factors, and chemical factors can be used to predict the impacts of a chemical contaminant on an ecosystem, which can then be used to develop policy and mediation strategies. However, as mentioned before, the processes of bioconcentration, bioaccumulation, and biomagnification are very 8 dynamic. Logically, it is necessary to review how these processes work in Puget Sound food webs to obtain a better understanding of the dynamics involved in determining the associated ecological risks.

Bioconcentration and Bioaccumulation in Puget Sound Food Webs:

Puget Sound's rich abundance of phytoplankton and varying physical characteristics produce an ecosystem that is rich in biological diversity and complex ecological interactions. While it is not possible to address every ecological interaction and pathway of bioconcentration and bioaccumulation in Puget Sound ecosystems, the diversity of literature on chemical contamination in Puget Sound biota can be used to address this issue on a general level.

Although the greatest accumulations of chemicals will typically be found in those organisms that are at the top of the food chain, e.g. marine mammals (Hayteas and Duffield 2000), it is important to understand how contaminants first enter and then accumulate through the food web. Puget Sound's rich diversity and abundance of phytoplankton form the base of a biologically diverse ecosystem. Heavy metals and organic pollutants are absorbed by plankton at the base of food webs and biomagnified to significant concentrations at higher trophic levels (Bard 1999). Hargrave et al. (2000) indicate that planktonic primary producers take up chemicals directly from the water through bioconcentration. Bioaccumulation Factors (BAFs) in phytoplankton have been found to be 104 - 106 on a wet weight basis (104 - 108 lipid weight), thus indicating that phytoplankton are bioconcentrating chemical contaminants to concentrations 104 - 106 times higher than ambient water concentrations. Moving one step up the food chain to zooplankton, bioaccumulation becomes the primary means of chemical uptake, although bioconcentration still occurs through the outer integument (Hargrave et al. 2000).

Filter-feeding species, e.g. some bivalves and polychaetes, consume large quantities of plankton from the water they filter. The National Oceanic and Atmospheric Administration's National Mussel

Watch Program has found that Puget Sound mussels are bioaccumulating organic pollutants, such as PAHs and PCBs, to higher concentrations than anywhere else in the United States, while heavy metals, such as copper, lead, silver, and mercury, are present at lower levels than elsewhere in the country (Dowty and Redman. 2002). Bioaccumulation in filterfeeding bivalves, and other taxa, is an important pathway in the bioaccumulation of chemicals in higher trophic levels given that they are important prey items for many species, such as decapods, asteroids, and gastropods. See Table 2 for chemical contamination in Puget Sound bivalves.

Although plankton form the base of many food webs, they are not always the primary means by which chemical contaminants enter the food web. As mentioned before, hundreds of potentially toxic chemicals are present in Puget Sound sediments, such as PCBs, DDT, and Arsenic (Malins et al. 1982). Puget Sound has an exceptional diversity of benthic invertebrates, which are at high risk of chemical bioconcentration because of their intimate contact with potentially contaminated sediments. As in many ecosystems, these benthic invertebrates are important prey items for many taxa, and create a pathway by which chemical contaminants are bioconcentrated from sediments and subsequently bioaccumulated in higher trophic levels (Morrison et al. 1996).

The ability of chemical contaminants to be bioaccumulated in higher trophic levels, e.g., top predators such as salmonids and pinnipeds, is usually dependent upon the level of bioconcentration and bioaccumulation in benthic invertebrates (Morrison et al. 1996). Therefore, benthic invertebrates are often used to conduct bioassays for sediment contamination 10 levels. Decapods, such as the Dungeness crab (Cancer magister), are often used as bioindicators of chemical contamination levels because they typically mirror sediment concentrations, due to their inability ability to metabolize chlorinated contaminants quickly (Yunker et al. 2002). In addition, the polychaete Armandia brevis has been used for sediment bioassays in Puget Sound for the last 10 years (Rice et al. 2000).

Bottom fish, such as the English sole (Pleuronectes vetulus), starry flounder (Platichthys stellatus), and rock sole (Lepidopsetta billineata), are also commonly used as biomarkers or bioindices of bioaccumulation in Puget Sound due to their intimate contact with sediments. The frequency and distribution of lesions in English sole is significantly correlated with bioaccumulation factors of PAHs and PCBs (Myers et al. 1998a, Myers et al. 1998b, and Rice et al. 2000). Malins et al. (1982) found that between 4 and 20 % of the English sole they collected had lesions, abnormal blood cell counts, and severe organ dysfunctions. Also, Roubal et al. (1978) found that starry flounder (Platichthys stellatus) accumulated 104 times the concentration of hydrocarbons in their tissues as was present in the water column after only one week of exposure (Roubal et al. 1978 cited in Malins et al. 1982). However, it is important to realize that both invertebrate bioassays and bottom fish bioindices are useless in the determination of bioaccumulation potential unless ecological interactions are taken into consideration.

In a study by Rice et al. (2000), the commonly used sediment bioassay invertebrate, Armandia brevis, was exposed to sediments that would typically not be identified as toxic using most invertebrate bioassays. When these Armandia brevis were fed to English sole, reduced growth and increased hepatic adducts were observed. These results indicate that sediment contamination levels that would normally be identified as non-toxic through typical invertebrate bioassay analysis can cause significant adverse effects at higher trophic levels.

Typically, demersal fish species, such as English sole, are found to have much higher concentrations of chemicals than more pelagic species, like salmonids. The difference in chemical concentrations may be related to the fact that demersal species live in close contact with sediments, which typically have much higher chemical concentrations than the seawater that surrounds pelagic species. Also, the differences in prey selection among pelagic and demersal species may partly explain the observed differences (Malins et al. 1982). It has been found that juvenile Chinook salmon (Oncorhynchus tshawytscha) (Collier et al. 2000 and Stein et al. 1995) and chum salmon (Oncorhynchus keta) (Collier et al. 2000) bioaccumulated PAHs, PCBs, and other chemical contaminants while residing in contaminated urbanized Puget Sound estuaries. See Table 2 for data on contamination in Puget Sound fish.

Puget Sound's pelagic fish species are important prey for many of Puget Sound's top predators, e.g. marine mammals and piscivorous seabirds. Specifically, concentrations of PCBs and DDE in the extensively studied Puget Sound harbor seals have been found to be significantly higher than in the fish they eat, 56 - 110 times greater on a lipid weight basis for Hood Canal and southern Puget Sound, respectively (Calambokidis et al. 1984). Puget Sound's marine mammal populations have been found to have high levels of organic chemicals (See Table 3) because they (1) readily accumulate highly lipophilic organochlorines due to their high lipid content, (2) generally have poor metabolic and excretory capabilities for these chemicals, (3) have long life spans, (4) transfer significant amounts of organic chemicals to their young during gestation and lactation, and (5) feed at the top of the food chain (Hayteas and Duffield 2000, Ross et al. 2000, Nakata et al. 1998, Calambokidis et al. 1984, Bard 1999, and Jarman et al. 1996). In fact, PCB concentrations in killer whales and southern Puget Sound harbor seals are among the highest in the world (Ross et al. 2000, and Calambokidis et al. 1984). While PCBs 12 are the primary chemical contaminant of concern in marine mammals, causing reproductive problems, biological disorders, and death, many other chemical contaminants and heavy metals, including mercury and DDE, frequently bioaccumulate to significantly high concentrations in Puget Sound's marine mammals (Calambokidis et al. 1984).

Killer whales (Orcinus orca) are a top predator with both migrant and resident populations in Puget Sound (Ross et al. 2000 and Calambokidis et al. 1984). Transient killer whales often feed preferentially on marine mammals (Ross et al. 2000, Hayteas and Duffield 2000, and Jarman et al. 1996), such as harbor seals (Phoca vitulina), stellar sea lions (Eumetopias jubatus), Dall's porpoises (Phocoenoides dalli), and harbor porpoises (Phocoena phococena). These have been found to represent 53%, 13%, 12%, and 11% of their diet, respectively (Ross et al. 2000). On the other hand, resident killer whale populations preferentially feed on adult salmonids (Ross et al. 2000, Hayteas and Duffield 2000, and Jarman et al. 1996), e.g. Chinook (Oncorhynchus tshawytscha), which are estimated to comprise about 96% of their total diet (Ross et al. 2000). Research has shown that bioaccumulation of PCBs and DDE in transient killer whale populations is significantly higher than in resident populations, which may be related to their preferred diet of marine mammals (Hayteas and Duffield 2000, and Ross et al. 2000). Also, migrant pods may be exposed to more contaminated prey items during migration than resident populations (Hayteas and Duffield 2000).

Some gray whale (Eschrichitius robustus) populations annually migrate to Puget Sound, and spend a considerable amount of time in some of its estuaries. These whales have a unique feeding strategy in which they filter sediments to feed on benthic invertebrates. Given the transient nature of these whales, they consume benthic invertebrates from many locations. Increased strandings in Puget Sound, 22 between 1988 and 1991, are raising concerns about 13 possible bioaccumulation of contaminants as the cause (Varanasi et al. 1994). However, Varanasi et al. (1994) found that bioaccumulation was substantially lower in gray whales than reported levels in Puget Sound pinnipeds. This may be related to the fact that pinnipeds consume pelagic fish, which have higher lipid contents than the invertebrates gray whales consume. Also, Varanasi et al. (1994) found that PCB and DDE concentrations in gray whales were relatively similar among populations that summer in Alaska, Washington, and California. Comparing transient populations of killer whales, which feed primarily on lipid rich pinnipeds and marine mammals, and gray whales, which primarily consume benthic invertebrates, lends credence to the hypothesis that consumption of higher lipid content food increases bioaccumulation potential.

Like gray whales, sea otters (Enhydra lutris) feed on benthic invertebrates, e.g. echinoids and bivalves. Sea otters (Enhydra lutris) were once harvested to extinction on the outer coast of Washington and Puget Sound. However, sea otters populations have been reintroduced and are currently increasing size. Typically, sea otters feed on sea urchins (which are very sensitive to chemical contamination and quickly bioconcentrate organic chemicals (Bard 1999)) and filterfeeding bivalves, which can also bioconcentrate contaminants. Given the sea otter's documented susceptibility to chemical contamination (Nakata et al. 1998), and the feeding strategies mentioned above, they are especially prone to experiencing the negative effects of chemical contamination.

Although many studies have focused on chemical contamination levels in marine mammals, most of these studies only sampled dead or beached specimens. Insufficient data on healthy specimens are available, making the determination of present bioaccumulation levels in marine mammals difficult. Given that Puget Sound's marine mammals are at the top of the food 14 chain, it is important to understand the degree of bioaccumulation in these organisms. However, it is important to remember that these high concentrations of chemical contaminants are the result of bioconcentration and bioaccumulation at much lower trophic levels. Only recently have studies began to focus on the uptake of chemical contaminants in lower trophic levels, e.g. plankton (Hargrave et al. 2000).

Conclusions/Recommendations:

As mentioned before, Puget Sound's food webs and physical and chemical characteristics are extremely complex and tightly interconnected. Accurate determination of bioaccumulation potential and ecological risk in Puget Sound biota is not possible without adequately considering all variables and biotic interactions. Scientists are still searching for a cost-effective approach for assessing the ecological risk of sediment contamination that will save both time and money by eliminating the need for site-specific bioeffects testing. However, site-specific conditions and biological interactions significantly affect a chemical's bioavailability and bioaccumulation potential (Chapman and Mann 1999 and Konasewich et al. 1982). Therefore, it is unlikely that any reductionist approach will ultimately accomplish this goal. Despite this, the value of models and monitoring systems are generally recognized among scientists, and will continue to be used as a tool to better understand the potential for bioconcentration, bioaccumulation, and biomagnification of chemical contaminants in Puget Sound biota. Continued research into the dynamics of bioaccumulation under different environmental conditions, biological interactions, and at different trophic levels will ultimately improve our ability to predict and assess problems associated with bioaccumulation. Currently, there exists a need to synthesize research and data on chemical properties (e.g. toxicity, lipid solubility, and persistence in the marine environment), sediment contamination 15 (e.g. identified contaminants, concentrations, and distribution), biota contamination, bioaccumulation potential (e.g. bioavailability and metabolism potential), environmental conditions (e.g. salinity, pH, and redox potential), and biological interactions (e.g. trophic relationships and feeding strategy) for Puget Sound. Synthesis of this research will enable a better, but not perfect, assessment of the ecological risk associated with chemical contaminants in Puget Sound sediments and biota. From this synthesis, important tools such as a bioaccumulation risk index and predictive models for contaminated areas in Puget Sound could be developed, thus enabling the prioritization of remediation efforts.

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