

Conceptual Foundations for Modeling Bioaccumulation in San Francisco Bay

RMP Technical Report

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Conceptual Foundations for Modeling Bioaccumulation in San Francisco Bay

Regional Monitoring Program Technical Report

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EXECUTIVE SUMMARY

Bioaccumulation of contaminants is a significant water quality concern in San Francisco Bay. The Bay is included on the 303(d) list due to elevated concentrations of many contaminants (e.g., PCBs, mercury, and DDTs) present in sport fish, bird eggs, and small fish species.

Mechanistic bioaccumulation models can be valuable tools to support management of bioaccumulative contaminants. A food web bioaccumulation model for San Francisco Bay has been developed and validated for PCBs and organochlorine pesticides. However, this model has yet to be parameterized at the sub-embayment or site-specific scale, and has not been explicitly developed to make time-dependent predictions. Mechanistic models of bioaccumulation have not been developed for other pollutants of concern, such as methylmercury or dioxins. In addition, the modeling and monitoring performed to date have not provided quantitative mechanistic links between particular sources (e.g., contaminated sites and tributary loadings on the margins of the Bay) and local and Bay-wide contaminant concentrations in key target species. Also, the models developed to date have limited capacity to forecast the impact of localized management actions on contaminant concentrations and associated risks for wildlife and humans. Since these model capabilities could be of great value in choosing management actions, there is a need to adapt the modeling approach to develop a better description of the relationship between contaminants in the margins and bioaccumulation in biota on local and regional scales.

The objectives of this report are to summarize key datasets and current knowledge of bioaccumulation in the Bay, and to identify priorities for future monitoring and modeling to support management of bioaccumulative pollutants. The report is intended for a technical audience of water quality managers, stakeholders, and external peer reviewers responsible for guiding the RMP modeling strategy and modeling tools utilized in San Francisco Bay.

This report summarizes empirical information on bioaccumulation in the Bay, with a focus on support for development of bioaccumulation models. A review of information on the pollutants of greatest concern in the Bay (methylmercury, PCBs, selenium, dioxins, organochlorine pesticides, PBDEs, and PFCs) is presented in Section 2. Key attributes of the primary indicator species are reviewed in Section 3; these are critical in understanding the information they provide on bioaccumulation and in building models to predict how concentrations in their tissues will respond to management actions. Section 4 provides a discussion of key concepts related to bioaccumulation modeling and highlights priority information gaps.

The report provides a conceptual foundation for a path forward for RMP studies of bioaccumulation, including modeling and monitoring, in support of effective and efficient efforts to reduce concentrations of contaminants in the Bay food web. RMP fate models must be developed with a sharp focus on understanding and predicting bioaccumulation.

Models of fate in water and sediment must be efficiently coupled with bioaccumulation modeling.

A conceptual understanding of the patterns revealed in food web monitoring to date provides a foundation for future modeling efforts. The following highlights of these patterns have a major bearing on consideration of the next steps in bioaccumulation studies.

- Long-term monitoring of sport fish has indicated no trend in MeHg concentrations since the early 1970s and no trend in concentrations of PCBs and other organics since 1994.
- In contrast to the sport fish, concentrations of organics have declined significantly in bivalves in the open Bay based on time series covering 1980 to the present.
- MeHg in the food web varies spatially 1) at a regional scale, with highest concentrations in the South Bay, and 2) with additional finer-scale variation at a local scale that is not clearly associated with source categories.
- PCBs and other organics also vary spatially 1) at a regional scale, with highest concentrations in the Central Bay, and 2) with additional finer-scale variation at a local scale that is very distinct and clearly associated with sediment contamination from past industrial and military activities.
- MeHg and PCB concentrations in Bay sport fish are exceptionally high on a statewide and national scale, suggesting an unusual degree of contamination or fate processes that make the Bay unusually slow to recover from contamination.
- Recent extensive small fish monitoring has revealed a much higher degree of accumulation for organics than was expected, with concentrations equaling or exceeding those in sport fish species at higher trophic levels.
- Food web uptake of MeHg, PCBs, and other organics appears to be driven by sediment contamination, as indicated by spatial correlations and isotope tracer studies.

Persistent contaminant concentrations in low energy environments on the Bay margins appear to be very stable, and in contrast to slow declines that are occurring in waters of the open Bay. Fate modeling efforts should be focused on forecasting recovery in these margin habitats.

The information presented in the report supports the following recommendations.

- Thoughtful articulation of the management decisions to be made based on bioaccumulation model outcomes is an essential first step to successful model development.
- The second step would be developing a comprehensive plan for creating the linked models for fate in water and sediment and for bioaccumulation in species of particular management interest. Understanding the coupling of these different model components is critical to the efficient and effective design of the water and sediment fate model.
- The models developed for open water habitats should be largely applicable to modeling the margins, and could be adapted to time-dependent and individual-based applications as well. The models could also be adapted to accurately represent the

different habitat types present on the margins (open water; sloughs, channels, and mudflats; tidal marsh; and salt pond).

- Sufficient empirical data on contaminant concentrations in sediment, biota, and perhaps water will be needed to develop and validate linked fate and bioaccumulation models on the margins. Information will be needed on spatial patterns in sediment contamination, long-term bioaccumulation trends, seasonal variation in bioaccumulation in some cases, and life history information of indicator species (such as diet, movement, foraging range, and physiological characteristics such as growth rate).
- Simple empirical correlational bioaccumulation models can be a very useful first step in evaluating relationships between bioaccumulation and environmental variables. A simple empirical correlation model for PCBs in small fish and sediment revealed a significant relationship. A similar analysis for MeHg did not show a correlation. Future field studies with PCBs should be designed to confirm and clarify this relationship. Future studies of MeHg bioaccumulation should endeavor to explain the observed lack of correlation. Future studies of other bioaccumulative contaminants of concern should similarly begin with evaluation of simple correlations with environmental gradients, proceeding to more complex models as needed to address management decisions.
- As modeling and empirical data gathering proceed, it may be valuable to obtain a more precise understanding of where key indicator species, such as shiner surfperch, silversides, topsmelt, and northern anchovy, are acquiring their contaminant burdens. A variety of methods are available for quantifying movement of fish, including telemetry studies, tagging studies, and stable isotopes.
- A MeHg bioaccumulation model for the Bay has yet to be developed. Development of linked mechanistic fate and bioaccumulation models that could address the dynamic processes that drive MeHg uptake in the Bay food web should be considered. However, the heterogeneous, dynamic, and poorly understood nature of MeHg cycling poses a formidable challenge.

1. Introduction

During the past 160 years, extensive human activity in the San Francisco Bay watershed has resulted in significant contamination in the Bay (Venkatesan et al. 1999, Conaway et al. 2007). Methylmercury and numerous hydrophobic contaminants (e.g., PCBs, dioxins, PBDEs, and DDTs) are currently the chemicals of greatest concern, due to their toxicity and persistence. San Francisco Bay is included on the 303(d) list due to elevated concentrations of many contaminants (e.g., PCBs, mercury, and DDTs) present in sport fish, bird eggs, and small fish species (SFRWQCB 2008, 2009). Studies of toxicological effects on wildlife suggest that additional contaminants may also attain concentrations of concern for biota, including some endangered species (Thompson et al. 2007, Brar et al. 2010). Some less studied contaminants (e.g., PFCs) may also cause impairment, but data are currently lacking to make a comprehensive assessment. Legacy sources remain for many contaminants, resulting in continued loading into or redistribution within the Bay, and thus continued bioaccumulation is anticipated for decades to come (Davis 2004, SFRWQCB 2006, Connor et al. 2007).

The term “bioaccumulation” refers to the increase in chemical concentrations in an organism as a result of uptake from all possible routes of exposure in its environment (Mackay and Fraser 2000). Aquatic species bioaccumulate contaminants via dietary absorption, respiratory transport, and dermal exposure (Gobas et al. 1999). Bioaccumulation can be viewed as the outcome of all competing uptake and loss mechanisms affecting contaminant concentrations in an organism. In San Francisco Bay (the Bay), as in all aquatic ecosystems, organisms bioaccumulate multiple contaminants encountered in sediment, water, diet, and other pathways.

Two other terms are commonly used to describe specific kinds of bioaccumulation. Bioconcentration occurs when contaminants are absorbed by an organism solely through respiration and dermal exposure routes. This differs from bioaccumulation, which considers the accumulation from dietary sources as well. Information on bioconcentration is useful for evaluating the outcome of complex biological processes, including the interactions between particulate and dissolved contaminants in water, uptake of contaminants from water into organisms, and the transformation of contaminants in tissue residues. Biomagnification, a special case of bioaccumulation, occurs when the chemical concentration in a consumer exceeds that of its diet (Gobas et al. 1993b, Gobas et al. 1999). Due to biomagnification, higher trophic level species exhibit greater contaminant concentrations than organisms at lower trophic levels. Many contaminants of concern in the Bay biomagnify, including methylmercury, PCBs, PBDEs, dioxins, pesticides, and selenium. This has led to exposure and potential risk to higher trophic level fish, birds, marine mammals, , and humans (Davis et al. 2002, Stewart et al. 2004, Thompson et al. 2007, She et al. 2008, Eagles-Smith and Ackerman 2009, Greenfield and Jahn 2010).

Mechanistic bioaccumulation models can be valuable tools to support water quality and aquatic resource management. Previously, a food web model for San Francisco Bay was

developed and validated for PCBs and organochlorine pesticides (Gobas and Arnot 2010). The model was used in development of the PCB TMDL for the Bay (SFRWQCB 2008). Under various scenarios, the model has been used to predict chemical concentrations in several indicator species based on sediment and water concentrations. However, it has yet to be parameterized at the sub-embayment or site-specific scale, and has not been explicitly developed to make time-dependent predictions. Mechanistic models of bioaccumulation have not been developed for other pollutants of concern, such as methylmercury or dioxins.

The fate and bioaccumulation studies performed to date have provided a good understanding of the general behavior of certain contaminants (e.g., PCBs) in the Bay and have provided useful information for managers (e.g., PCB TMDL). Continuing this work for other contaminants like dioxins and methylmercury can be expected to have similarly fruitful outcomes for managers. However, the modeling and monitoring performed to date have not provided quantitative mechanistic links between particular sources (e.g., contaminated sites and tributary loadings on the margins of the Bay) and local and Bay-wide contaminant concentrations in key target species and associated risks in humans. Also, the models developed to date have limited capacity to forecast the impact of localized management actions on contaminant concentrations and associated risks for wildlife and humans. Since these model capabilities could be of great value in choosing management actions, there is a need to adapt the modeling approach to develop a better description of the relationship between contaminants in the margins and bioaccumulation in biota on local and regional scales.

The objectives of this report are to summarize key bioaccumulation datasets for the Bay and current knowledge, and to identify priorities for future monitoring and modeling to support management of bioaccumulative pollutants. The report is intended for a technical audience of water quality managers, stakeholders, and external peer reviewers responsible for guiding the RMP modeling strategy and modeling tools utilized in San Francisco Bay.

This report is one of three related reports that are contributing to RMP monitoring and modeling strategy development. An RMP modeling strategy is in development that will describe a plan to develop cost-effective, predictive models to support water quality management decisions in the Bay. Another report presents a conceptual model for the fate and transport of contaminants on the San Francisco Bay margins (Jones et al. 2011). The present report is the third document being developed in support of RMP modeling.

2. Target Chemicals: Bioaccumulation Processes and Biota Concentrations

In this section, nine classes of legacy or current-use contaminants are reviewed, describing their uptake processes and general patterns in biota concentrations for the Bay.

Methylmercury

Mercury (Hg) is a widespread pollutant that is impacting aquatic ecosystems across North America. There are many sources of total mercury to San Francisco Bay. These include historic mercury and gold mining areas, urban and industrial runoff, wastewater outflow from treatment plants, and atmospheric deposition (Hornberger et al. 1999, Conaway et al. 2004, SFRWQCB 2006). However, inorganic mercury is not the primary concern for biota. An organic form, methylmercury (MeHg), is the most toxic and bioavailable mercury species (Scheuhammer et al. 2007). MeHg levels in the Bay are primarily the result of in-Bay mercury methylation and demethylation processes (Table 1; Yee et al. 2011). However, the degree of net production that occurs in the Bay compared to the upstream watersheds has not been fully characterized.

Dietary uptake and direct waterborne exposure are primarily responsible for MeHg transfer to upper trophic levels of aquatic food webs (Pickhardt et al. 2006). Direct uptake from water is the most important pathway for organisms at the lowest levels of the food web, such as phytoplankton and zooplankton. MeHg uptake in these organisms occurs through passive diffusion across the cell membranes into the algal cytoplasm (Mason et al. 1995). Methylmercury accumulates due to its strong affinity for sulfhydryl groups on proteins and processes that lead to high retention (Morel 1983). This initial point of entry into the food chain can be many orders of magnitude over ambient aquatic concentrations (e.g., Watras and Bloom 1992). Subsequently, MeHg biomagnifies through trophic transfer to concentrations of up to four times that in the lower food web, leading to elevated concentrations in small fish, avian wildlife, sport fish, and many other aquatic species (Watras et al. 1998, Pickhardt et al. 2006). Potential adverse effects of MeHg in humans include neurological, reproductive and behavioral effects (U. S. EPA 1997).

Monitoring of mercury in aquatic biota throughout the Bay has indicated that concentrations are elevated Bay-wide, and are also relatively high at certain Bay-margin hot spots. Monitoring of total Hg (THg) and methylmercury (MeHg) in the RMP Status and Trends Program and other local studies has shown that significant, positive correlations exist between concentrations in sediment and distance to shoreline or percent land cover. Further discussion of these data is discussed in the companion report by Jones et al. (2011). Recent sediment contamination data suggest that the Bay margins represent a major pool of the Hg ultimately methylated and bioaccumulated (Gehrke et al. 2011b). Forage fish and wildlife known to ingest sediments or sediment-dwelling prey have elevated concentrations, particularly in the South Bay (Grenier and Davis 2010). In the southernmost portion of the Bay, the Guadalupe River, which has been heavily impacted by historic Hg mining activity in the region, has been shown to be an important Hg source over the years (Thomas et al. 2002, Conaway et al. 2003, SFRWQCB 2006). In contrast, Hg burdens in fish and bird species in the North Bay are relatively low (Ackerman et al. 2007, Greenfield and Jahn 2010), despite the occurrence of in-Bay sediment deposits from historic gold mining operations in this region (Hornberger et al. 1999). A possible hypothesis to explain these observations is that higher exposure of organisms to open water conditions has resulted in source dilution at the base of the food chain in the North Bay, causing lower net methylation rates than at South Bay sites.

However, some of the lowest MeHg concentrations occur in backwater ponds and sloughs, where flushing is likely to be limited (Ackerman et al. 2007, Slotton et al. 2007). Other factors affecting net methylation may also be driving this pattern.

The complex biogeochemical cycling of MeHg, as well as the effects of physical and biochemical factors on methylation, are typically considered in mechanistic models of MeHg fate and bioaccumulation (Knights et al. 2009, Sunderland et al. 2010). MeHg exhibits high assimilation efficiencies and slow depuration, thereby driving the Hg burdens in higher trophic level species (Pickhardt et al. 2006). Since MeHg is the form that biomagnifies in food webs, understanding the conditions that increase net methylation throughout the Bay is needed to predict Hg concentrations in biota. A MeHg fate and bioaccumulation model should consider the varying conditions that affect in-Bay methylation. For example, MeHg contamination within the Bay could be affected by seasonal or spatial variation in concentrations of dissolved organic carbon, salinity, nutrients, and suspended particulate organic matter.

Due to concerns for wildlife and human health exposure, considerable effort has been put towards characterizing MeHg concentrations in San Francisco Bay wildlife (Davis et al. 2002, Ackerman et al. 2007, Eagles-Smith et al. 2009, Greenfield and Jahn 2010, Grenier et al. 2010b). Data collected by the RMP indicate concentrations in many sport fish species, including striped bass (*Morone saxatilis*), leopard shark (*Triakis semifasciata*), and California halibut (*Paralichthys californicus*) are above guidelines for human health consumption (Davis et al. 2006c, Hunt et al. 2008, Davis et al. 2011). Indicators of wildlife exposure, such as forage fish and bird eggs have indicated strong spatial patterns. In a study by Greenfield and Jahn (2010), more than 50% of the variation in Mississippi silverside (*Menidia beryllina*) MeHg concentrations was explained by spatial location, which suggested that their exposure to MeHg was likely restricted to locations relatively close to the site of capture. Mercury stable isotopes were significantly correlated between forage fish and sediment, further supporting the inference of site fidelity (Gehrke et al. 2011b). Some bird species also exhibit strong fidelity to specific foraging locations, with consequent spatial differences in MeHg concentrations (Ackerman et al. 2007). These examples of site fidelity suggest that studies conducted over relatively small spatial scales would be useful to further understand driving factors of bioaccumulation.

Of the sport fish monitored in the Bay, shiner surfperch (*Cymatogaster aggregata*) exhibit the highest site-specificity in contaminant concentrations (Davis et al. 2011). Due to their relatively low trophic position (see Section 3), concentrations are often lower than those in other piscivorous sport fish. However, shiner surfperch have proven over the years to indicate significant spatial differences and relatively low variability within locations. In 2009, for example, the coefficient of variation for each site ranged from 2 – 10% among contaminants. Average Hg concentrations in shiner surfperch were highest (0.19 ppm wet weight) at Oakland Harbor (a Bay margin site), which was significantly greater than all other locations that were more distant from the Bay margin. South Bay was the second highest location in 2009 (0.13 ppm wet weight), which was also significantly higher than the average concentration at Berkeley (0.10 ppm), San Francisco (0.09 ppm), and San Pablo Bay (0.08 ppm) (Davis et al. 2011).

Statewide and national comparisons of fish data indicate that San Francisco Bay biota accumulate unusually high concentrations of methylmercury. These observations suggest that legacy contamination from mercury and gold mining contributes significantly to accumulation in the Bay food web, adding to the contributions from atmospheric deposition and other sources. A statewide survey of contaminants in sport fish found that shiner surfperch, the best statewide spatial indicator species for bays and harbors, exhibited distinctly elevated methylmercury concentrations in the Bay (Figure 1) (Davis et al. 2012). The large number of fish that went into these shiner surfperch averages (585 in total for five sites) makes for robust estimates. The five locations sampled in the Bay accounted for the five highest concentrations for this species statewide. Only two other locations (Humboldt Bay and San Diego South Bay) had concentrations above 0.07 ppm.

Several studies have been published on mercury in striped bass in U.S. estuaries, and the highest average concentration reported is from San Francisco Bay. Striped bass are a relevant and useful indicator species for comparing methylmercury contamination across USA estuaries due to several factors: their popularity for consumption (this is the most popular species for consumption in San Francisco Bay – SFEI, 2000); their dependence on estuaries (Able, 2005); their broad spatial integration across the estuaries in which they reside due to their variable use of fresh, brackish, and saline habitat (Secor and Piccoli, 2007); their wide distribution on the east, west, and Gulf coasts; and strong correlation between size and methylmercury concentration. The average concentration measured in 2009 in San Francisco Bay (a length-adjusted mean of 0.44 ppm at 60 cm – Davis et al., 2011) was higher than average concentrations recently reported for five other USA coastal areas (Figure 2). The New Jersey coast (Burger and Gochfeld, 2011) had the second highest average concentration (0.39 ppm – based largely on fish greater than 85 cm). Average concentrations in striped bass from other USA coastal areas ranged from 0.12 to 0.23 ppm (Mason et al., 2006; Piraino et al., 2009; Glover et al., 2010; Katner et al., 2010, and Burger and Gochfeld, 2011).

Birds and mammals also accumulate high MeHg concentrations in the Bay. Harbor seals (*Phoca vitulina*) have shown elevated mercury concentrations in blood and hair (Brookens et al. 2007). California Clapper Rails (*Rallus longirostris obsoletus*) have been a primary species of concern for MeHg effects, particularly in Bay wetlands, being the only remaining year-round habitat for the federally endangered species. Clapper Rail eggs frequently exceeded toxic thresholds for avian embryos, exhibiting an average concentration of 0.81 ppm wet weight in a recent study of fail-to-hatch eggs (Schwarzbach and Adelsbach 2003). Forster's tern (*Sterna forsteri*) eggs at salt ponds in the North and South bays similarly have been observed to exceed threshold effects levels (0.5 – 0.8 ppm wet weight) (Ackerman et al. 2008a, SFEI 2008). Generally, foraging habitat has been important for predicting MeHg exposure in waterbirds, as species more associated with marshes and salt ponds had relatively high concentrations compared to species more associated with open Bay and tidal mudflats.

The longest time series available for MeHg concentrations in the Estuary is for striped bass (*Morone saxatilis*) monitored from 1971 to 2009. These data have exhibited no

significant trend over nearly four decades (Greenfield et al. 2005). Similar results have been observed during the late 1990s to 2009 in other sport fish (Davis et al. 2011) and waterbirds (Grenier and Davis 2010). The lack of long-term trends likely reflects the legacy Hg remaining in Bay sediments, and continuing food web uptake of MeHg emanating from these sediments (Conaway et al. 2007, Grenier and Davis 2010, Gehrke et al. 2011b).

Selenium

Selenium (Se) is a naturally occurring trace element that is an essential nutrient at low doses (Ohlendorf 2003, Chapman et al. 2010b). Loading of Se to the Bay occurs predominantly through agricultural drainage from the Sacramento and San Joaquin rivers, discharges from petroleum refineries in the North Bay, and wastewater effluent from municipal treatment plants (Presser and Luoma 2006). Loads from the Sacramento and San Joaquin rivers have been estimated to be more than an order of magnitude greater than from wastewater effluent. In the North Bay, loads of dissolved Se from industrial facilities have been ~ 230 kg yr, petroleum refineries ~ 540 kg/yr, and ~ 3,940 kg/yr on average from the Delta (Tetra Tech 2008).

Se occurs in many forms in the aquatic environment with its speciation influencing the degree of bioaccumulation in aquatic organisms. Selenomethionine (SeMet) is the dominant form of Se detected at all levels of aquatic food chains. SeMet is one of the primary forms of organic Se leading to bioaccumulation and toxicity (Hamilton et al. 1990, Hamilton 2004, Chapman et al. 2010b). Although Se is an essential nutrient for most organisms, it may cause severe reproductive and developmental toxicity when concentrations exceed nutritional requirements (Fan et al. 1988, Van Derveer and Canton 1997, Teh et al. 2002). Selenium accumulates in the proteins of organisms. The process occurs when sulfur in sulfur-containing amino acids is substituted with selenium producing selenoamino acids, which are readily incorporated into protein structures (Bakke et al. 2010).

Bioaccumulation of Se occurs primarily from the base of food chains, where phytoplankton transform dissolved selenite into organoselenide (Schlekat et al. 2002, Chapman et al. 2010b). Concentrations in phytoplankton generally are 100 – 500 times above that in ambient water (Luoma and Presser 2009). Organoselenide is then transferred up the food chain and often biomagnified in filter-feeding invertebrates and certain fish. Se bioaccumulates in filter-feeding benthic invertebrates, including the bivalves, *Corbula amurensis* and *Macoma balthica*, as well as copepods (Stewart et al. 2004).

Biomagnification of Se in the Bay has often been greatest in fish species that primarily consume clams, and recent studies have suggested that the invasion of *Corbicula amurensis*, which bioaccumulates Se more than other resident benthic invertebrates, has increased risk to benthivorous piscivores (Linville et al. 2002, Presser and Luoma 2006). A bioaccumulation model used to characterize processes influencing Se uptake and tissue

concentrations in lower trophic level organisms has showed that slower rates of elimination in bivalves results in higher steady-state concentrations (Presser and Luoma 2010). Therefore, future work to examine Se bioaccumulation in upper trophic level species in the Bay should consider the differential uptake and elimination rates in lower trophic level species as a critical step to predicting bioaccumulation. Trophic level species that consume large amounts of *C. amurensis* and other filter-feeding invertebrates include white sturgeon (*Acipenser transmontanus*), Sacramento splittail (*Pogonichthys macrolepidotus*), and diving ducks. Recent data suggest that Se may reach levels sufficient to cause growth impairment, deformities, and mortality in early life stages of splittail and white sturgeon (Linville et al. 2002, Teh et al. 2002).

Bioaccumulation of Se is primarily of concern in the North Bay (particularly Suisun Bay) (SFEI 2008, Baginska 2011). This is based on elevated concentrations in clams, fish, and diving ducks that predominate in this region of the Bay (Linville et al. 2002).

Concentrations have been generally lower in shorebirds than in diving ducks, due to their preference for small benthic invertebrates, rather than clams (Takekawa et al. 2002).

Diving ducks consume benthic bivalves that generally accumulate more Se than aquatic invertebrates and small fish (Ackerman and Eagles-Smith 2009).

Time series of Se have been maintained by USGS since 1994 for bivalves and sediments at a site adjacent to a wastewater treatment plant in the South Bay. Results indicate no long-term trends in either bivalves or sediments, although concentrations are often elevated in the spring compared to other seasons (Lorenzi et al. 2008). The invasion of *C. amurensis* posed a risk of increased bioaccumulation for predators, when this species became abundant in the late 1990s. Nevertheless, there has been no indication of a long-term trend in concentrations in white sturgeon (Greenfield et al. 2005). The lack of isotope or dietary information, however, prevents these data from being put in context of potential shifts in sources of exposure that may have occurred over time. RMP monitoring of white sturgeon detected average Se concentrations of 1.5 ppm (wet weight) in 2009, which was very similar to concentrations observed from 1997-2006 (1.2 - 1.6 ppm, wet weight).

Polychlorinated Biphenyls (PCBs)

San Francisco Bay has a history of PCB contamination from point and non-point sources (Davis et al. 2007a). Urban runoff, drainage from the Central Valley, erosion of buried sediment, and in-Bay hot spots represent the major sources and pathways of PCBs to the Bay.

PCB contamination in the Bay is primarily associated with shoreline, urbanized areas and local watersheds (Davis et al. 2006a, Davis et al. 2007a). Areas with elevated PCBs include Oakland and Richmond Harbors, San Leandro Bay, Napa River near Mare Island, and the waterfront alongshore Hunter's Point. Therefore, food web models for PCBs should consider the Bay margins associated with urbanized areas as an area of potential concern (Jones et al. 2011). One Bay margin area of potential concern for biota exposure

is the Lower South Bay, as urban runoff from the Guadalupe River carries significant quantities of PCBs and other contaminants to the Bay (McKee et al. 2006). Elevated PCB concentrations have been observed in sediment, sport fish tissues, bird eggs sampled in Central and South Bay, in comparison to San Pablo Bay locations (Hunt et al. 2008, SFEI 2008).

PCBs are readily adsorbed onto particles, and the highest concentrations are frequently observed in fine-grained, organic-rich estuarine sediment (Davis et al. 2007a). Hydrophobic contaminants such as PCBs partition relatively easily between the water column and organic-matter-rich sediment, a process currently being evaluated for potential PCB remediation in contaminated hotspots using activated carbon amendment (Cho et al. 1999). Generally, PCBs will adsorb to sediment and suspended particulates; thus biota with sediment-associated life histories may be more exposed to PCBs than pelagic organisms.

Due to the lipophilic nature of PCBs, they predominantly accumulate in the fatty tissues of organisms (Niimi 1983, Kidd et al. 1998). In San Francisco Bay, PCBs are monitored in multiple species of regulatory concern for human health and wildlife exposure (Table 1). The importance of lipid as a covariate in bioaccumulation modeling is discussed further in Section 4. Tissue residues in multiple taxa, especially those with high fat content, have indicated PCB concentrations that pose health risks to humans and wildlife (Greenfield et al. 2005, Davis et al. 2007a). Median concentrations in white croaker (*Genyonemus lineatus*) and shiner surfperch (*C. aggregata*), two primary indicators of human health exposure, were 47 ng/g ww and 106 ng/g ww in 2009, respectively (Davis et al. 2011). These concentrations have been well above the human health fish tissue TMDL target in San Francisco Bay (10 ng/g ww) since RMP monitoring began in 1994 (Fairey et al. 1997, SFRWQCB 2008). Statewide monitoring of shiner surfperch at coastal locations in 2009 and 2010 (Davis et al. 2012a) illustrates that contamination of the Bay food web is relatively severe (Figure 3). Even the lowest concentrations from the Bay were much higher than the least contaminated coastal locations, and concentrations in Oakland Harbor were the highest observed in the state.

PCB concentrations in bird eggs have been monitored because bird embryos are among the most sensitive life stages to the adverse effects of PCBs (Davis et al. 2004, Davis et al. 2007a) and because of the value of bird eggs as indicators of temporal trends in food web contamination. In a study performed in 1995, PCB concentrations in Double-crested Cormorant (*Phalacrocorax auritus*) eggs from San Pablo Bay were correlated with reduced egg mass, reduced spleen mass, and induced cytochrome P450 and appeared to be above the threshold for causing embryo mortality (Davis 1997). The California Least Tern (*Sternula antillarum browni*) and Forster's Tern (*S. forsteri*) are the most highly contaminated of the bird species sampled in San Francisco Bay with regards to PCBs. Concentrations in these species have been estimated at 2-6 times greater than those observed in the omnivorous rails and two times greater than the much larger, Caspian Tern (*Hydroprogne caspia*). PCBs in Least Tern eggs from San Francisco Bay have been significantly higher than eggs from in other parts of the state (Davis et al. 2003, Davis 2004, Davis et al. 2007a). Differences may be related to local contamination since Least

Terns forage mostly within about 3 km of their nest sites on Alameda Naval Air Station during the incubation and chick-feeding stages (Elliott 2005).

Due to their position at the top of the aquatic food chain and their relatively long life-spans, marine mammals are also important indicator species for PCBs. Studies have found marine mammals to be particularly susceptible to contamination of PCBs and other hydrophobic substances (Kannan et al. 2000). In San Francisco Bay, PCBs in blood and blubber of harbor seals (*Phoca vitulina*) were two times higher than that known to cause adverse reproductive and immune effects (Young et al. 1998). Blood PCB concentrations in free-ranging San Francisco Bay harbor seals were associated with increased white blood cell counts (an indicator of immune response) (Neale et al. 2005). Laboratory exposure of cell cultures from harbor seal blood to elevated PCB concentrations resulted in a trend (not statistically significant) of reduced white blood cells, suggesting a compromised immune system response (Neale et al. 2002).

The phaseout of PCBs during the 1970s appear to have led to declines in biota concentrations during the 1970s and early 1980s, followed by a slower trajectory of decline from 1982 to the present. Transplanted mussels at northern Estuary locations have shown declines from approximately 4,000 ng/g lipid weight in 1982 to 1,000 ng/g lipid in 2008 (Davis et al. 2006b, SFEI 2010). In the southern portion of the Estuary, concentrations have declined from approximately 6,000 ng/g lipid weight in 1982 to 2,400 ng/g lipid weight in 2008 (Risebrough 1995, Davis et al. 2006b, SFEI 2010).

In spite of these declines in transplanted bivalves, PCB concentrations in the key sport fish indicator species (shiner surfperch and white croaker) have shown no evidence of decline on a lipid-normalized basis since routine sport fish monitoring began in 1994. A hypothesis to explain these apparently contradictory results is that the bivalves may be reflecting gradually declining concentrations in the open waters of the Bay, while the shiner surfperch and white croaker may be accumulating PCBs from foraging in relatively contaminated margin habitats with longer residence times for sediment particles and associated contaminants.

Recent unexpected findings of concentrations of PCBs in lower trophic level Bay fish (topsmelt and northern anchovy) that are comparable to the highest concentrations observed in sport fish, such as shiner surfperch and white croaker support this hypothesis. A small-scale RMP pilot study in 2007 found PCB concentrations in composite samples of topsmelt ranging from 110 to 420 ppb wet weight (SFEI 2009). In a larger effort in 2010, PCBs in small fish were sampled at 17 probabilistically selected sites throughout San Francisco Bay and 12 targeted sites near historically polluted locations. The highest concentrations (in wet weight) were from targeted Central Bay locations, including Hunter's Point Naval Shipyard (1,130 ppb; topsmelt), Stege Marsh (970 ppb; silverside), Oakland Inner Harbor (590 ppb; topsmelt), and Richmond Inner Harbor (340 ppb; topsmelt). Concentrations in both topsmelt and silverside were comparable to those of high lipid sport fish in the Bay. To determine association with legacy sediment contamination, these 2010 PCB concentrations were regressed against total PCBs in sediments, using sediment contamination data from 1991 to 2003 (N = 440 samples)

obtained from the Sediment Quality Objectives (SQO) program (Barnett et al. 2008) (Figure 4). Given that fish and sediment data were not collocated, the average sediment concentration was determined within a 4 km radius of each fish collection location, and the results were regressed against fish tissue concentrations. Fish and sediment PCB concentrations were positively related ($R^2 = 0.51$). Examining species individually, the relationship was stronger for topsmelt ($R^2 = 0.71$, $N = 23$) than silverside ($R^2 = 0.27$, $N = 11$), though this may have been at least partly a function of the greater number of topsmelt/sediment comparisons available.

The hypothesis that the persistent elevated concentrations in Bay sport fish are a function of bioaccumulation on the polluted margins of the Bay will be referred to below as the “bathtub ring” hypothesis.

Polybrominated Diphenyl Ethers (PBDEs)

PBDEs are a group of chemicals used in flame retardants and many synthetic plastic and textiles that are known to biomagnify in the aquatic environment (She et al. 2008). They are often included on lists of persistent organic pollutants (POPs) and potentially affect higher trophic level species. Primary sources and pathways of PBDEs to Bay are thought to be wastewater treatment plants, urban runoff from residential and industrial areas, and remobilization of buried sediment (Oram et al. 2008).

PBDEs are persistent compounds that bioaccumulate by a similar process to many other lipophilic chemicals (e.g., PCBs and DDTs) (Boer 2009). They can partition relatively easily into the fatty tissues of biota, though debate exists regarding the degree of biotransformation. Higher-brominated PBDE congeners (e.g., BDE 209) are known to debrominate, resulting in lower-brominated congeners, including some of the more toxic compounds (He et al. 2006).

Bird egg data collected from 2002 to 2009 indicate that the Lower South Bay is a hotspot for PBDEs. Forster’s Tern eggs from the Eden Landing area of the South Bay had average total PBDE concentrations of 62,400 and 63,300 ng/g lipid weight in 2001 and 2002, respectively (She et al. 2008), the highest concentrations ever reported at the time. PBDEs were also measured in tern and rail eggs from around the Bay. During 2000 to 2003, geometric means of PBDEs ranged from 3,700 to 4,800 ng/g lipid weight for Caspian, Forster’s, and Least Terns (She et al. 2008). California Clapper Rail has a diverse diet of fish, mollusks, and seeds, as a result of foraging in tidal marshes, and exhibit lower PBDE concentrations than the piscivorous tern species that forage in the open Bay and managed ponds. The median PBDE concentration in California Clapper Rail eggs measured in 2001 was 379 ng/g lipid weight (She et al. 2008).

PBDEs have also been found at substantial concentrations in sport fish sampled in the Bay (Oros et al. 2005). Average PBDE concentrations in 2009 were 4.3 ng/g wet weight for white croaker, 2.8 ng/g for white sturgeon, 8.3 ng/g for shiner surfperch, and 7.9 ng/g for northern anchovy (Davis et al. 2011). Brown et al (2006) measured PBDE compounds in fillets of several fish species caught along the California coast. PBDEs ranged from 0.04 ppb wet weight (in speckled sanddab, *Citharichthys stigmaeus*) to 7.1 ppb wet

weight (in white croaker, *G. lineatus*). The highest PBDE levels were found in urban areas (Yogui and Sericano 2009). Holden et al. (2003) quantified PBDEs in whole fish (minus head, tail and guts) collected from San Francisco Bay in 2002. Concentrations ranged from 5.7 ppb wet weight in jacksmelt to 44.1 ppb wet weight in white croaker (Yogui and Sericano 2009).

During the past two decades, many legacy contaminants have shown declines or no significant changes, while PBDE levels increased sharply in the 1990s and appear to be remaining constant or declining in more recent sampling. PBDE levels in peregrine falcon (*Falco peregrinus*) eggs from 38 sites throughout California (1986 – 2007) indicated a 3-fold increase in concentrations in each decade, whereas PCB levels had no significant changes (Park et al. 2009). A dramatic increase in PBDEs in San Francisco Bay harbor seals was also observed in the 1990s (She et al. 2002). PBDE data collected in the RMP prior to 2006 are not directly comparable to the newer data due to analytical differences. The most recent sport fish sampling found lower concentrations than in previous rounds. Continued monitoring of PBDEs is needed to determine temporal patterns in Bay sport fish.

Dioxins

Polychlorinated dibenzodioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are organic polyhalogenated chemicals commonly referred to collectively as dioxins. Dioxins are a group of chemicals that occur in the environment as by-products of combustion and the manufacture of organochlorines and chlorine-containing substances, such as polyvinyl chloride. Dioxins are transported to the Bay by urban runoff, atmospheric deposition, and erosion of buried sediment.

Dioxins have similar physical properties to PCBs, but are detected at much lower concentrations in the environment. The most readily biomagnified congeners have 4, 5, or 6 chlorine atoms. Dioxins have been associated with a wide variety of toxic effects, even at very low environmental concentrations. 2,3,7,8-tetrachlorodibenzo-dioxin (TCDD) exhibits the highest carcinogenicity and immunotoxicity, and thus dioxin-like effects are modeled using TCDD as the reference compound (Van den Berg et al. 1998). Dioxins can affect the immune, endocrine, and reproductive systems of fish, birds, and mammals (Kannan et al. 2000, Fisk et al. 2005). It should also be acknowledged that dioxins share the same toxic modes of action with some PCB and PBDE congeners. The total dioxin-like potency of samples can be assessed by estimating the sum of dioxin toxic equivalents (TEQs) of these and other dioxin-like compounds.

Dioxins have not been as extensively characterized in Bay biota as PCBs. However, there does appear to be a similar pattern of urban and industrialized margins having the highest concentrations. For example, shiner surfperch, white croaker, and tern eggs have similar spatial gradients in PCBs and dioxins (Davis et al. 2011) with Oakland and South Bay having significantly higher concentrations than other areas in Central or North Bays. Average dioxin/furan TEQs in shiner surfperch and white croaker from the most recent RMP monitoring (2009) were 0.89 pg/g and 0.44 pg/g wet weight, respectively.

Historically, average concentrations in these species across all Bay segments were in the range of 3 – 4 pg/g wet weight. The lower concentrations in 2009 are largely driven by the switch to analysis of fillets without skin for white croaker. Insufficient data have been collected over time to examine whether current concentrations differ significantly from previous years.

Organochlorine Pesticides (DDTs, Chlordanes, Dieldrin)

Legacy organochlorine (OC) pesticides have entered the Bay from urban and agricultural sources. Delta outflow from upstream watersheds, urban runoff, remobilization of legacy Bay sediment (including dredging and disposal of previously contaminated sediment) are the primary pathways to the Bay (Pereira et al. 1994, Connor et al. 2007).

DDTs, chlordanes, and dieldrin are hydrophobic, with most (97-99%) of the current mass available to the biota residing in the sediment rather than the water column (Connor et al. 2007). As with other organic pollutants, OC pesticides are generally not easily metabolized or excreted, and are stored in fatty tissues (Fairey et al. 1997). Once incorporated into the food web, concentrations of OC pesticides have been typically highest in species with high lipid content, such as shiner surfperch, white croaker, and harbor seals. RMP sport fish data collected since 1994 indicate that OC pesticides concentrations have been highest in white croaker and shiner surfperch, intermediate in jacksmelt, and relatively low in species lower in lipid (striped bass, leopard shark, halibut, and sturgeon) (Fairey et al. 1997, Davis et al. 2002). Striped bass, the most frequently taken and consumed sport fish in the Bay (SFEI 2000), has been shown to have lower lipid content and subsequently lower concentrations of OC pesticides (Connor et al. 2007).

Contaminant concentrations in biota across the Bay have not indicated any clear regional patterns in OC pesticides. However, some areas of the Bay margin reveal elevated concentrations of DDTs (Jones et al. 2011). One DDT hot spot, Lauritzen Channel in Richmond Harbor, has undergone cleanup as a Superfund site, but remediation has not notably reduced DDT bioavailability at the site (Weston et al. 2002, Connor et al. 2007) due to significant mass remaining after remediation. Dredging removed three tons of DDT from the canal, but also remobilized large amounts, leading to increase pesticide concentration in tissue of biota during the activity (Weston et al. 2002).

Trends in OC pesticides have been documented in bivalves. All nine locations monitored across the Bay have shown significant declines (Davis et al. 2006b, Jones et al. 2011). RMP sport fish data have been consistently collected since 1994 for the evaluation of trends. Like other contaminants, there have been no obvious trends apparent in white croaker or shiner surfperch, although 1997 was higher than other years (Davis et al. 2011).

Clapper Rail eggs collected from several sites in San Francisco Bay in 1975 exhibited concentrations of DDE that ranged from 0.38 to 2.1 ppm. These concentrations were relatively high, but below concentrations thought to induce reproductive defects

(Lonzarich et al. 1992). By the mid-1980s, the DDE concentration of Clapper Rail eggs had shown a significant decline. Residues of DDE in eggs collected in 1987 ranged from 0.14 to 0.63 ppm, with eggshell thickness comparable to eggs collected prior to 1940 (Lonzarich et al. 1992). In 2001, the maximum concentration observed for cormorants in San Francisco Bay was 3 ppm (Davis et al. 2004), below a 5 ppm threshold level for reproductive impairment (Blus 1996).

Perfluorinated Compounds (PFCs)

PFCs are long-chained carbon compounds with a high degree of fluorine substitution and varying functional groups. They have been widely used in a variety of applications including insecticides, refrigerants, surfactants, stain repellants, and fire fighting foams. Sources and pathways to the Bay include wastewater treatment, landfills, urban runoff, and remobilization of buried sediment (Higgins et al. 2005). Sediment is suspected as a major sink of PFCs emanating from municipal waste streams.

PFCs are stable, hydrophobic and oleophobic (oil repellant) compounds. Their behavior in the environment is the least understood of the classes of contaminants being considered in this report. Some PFCs appear to associate with specific proteins rather than accumulate in the lipids, but the exact process has yet to be documented. Since their bioaccumulation properties are known to differ from other lipophilic, organic contaminants, such as PCBs, future models will need to be adapted to describe their behavior in the environment.

PFC compounds with greater than eight carbon atoms (C8) tend to degrade to one or two terminal end products, while PFC chains with less than four carbon atoms are degraded entirely. Perfluorooctane sulfonate (PFOS) has a C8 based structure and has been the primary PFC detected in biota monitored in the Bay, including cormorant eggs, sport fish muscle, and harbor seal serum. PFOS concentrations in harbor seals have been an order of magnitude higher in the Bay compared to a reference site in Tomales Bay (Sedlak and Greig 2011). In 2009, RMP sampling of sport fish in the Bay found that PFOS was the only PFC detected, and only four samples had detectable PFOS concentrations. The highest concentration was 18 ppb in a leopard shark composite. The other samples with reportable concentrations were from northern anchovy and white sturgeon. Data have not been extensively collected to further evaluate spatial patterns in the Bay or detection of trends.

3. San Francisco Bay Indicator Species of Concern

San Francisco Bay supports a highly diverse array of habitat types and indicator species. Resident birds, fish, and mammals include a number of endangered or threatened wildlife species. Species considered here as candidates for bioaccumulation modeling include those that have previously been used in the validated PCB bioaccumulation model for the Bay, are indicators of beneficial use impairment, provide significant information

regarding spatial and temporal variation in bioaccumulation, and/or are representative of the key habitats of interest. Key habitats included in this report are the open waters, sloughs, channels, mudflats, and tidal marshes of the Bay. Information gaps relating to development of bioaccumulation models are also highlighted. Species below are organized by the mercury and selenium indicators first, followed by the indicators for organic contaminants.

Striped Bass

Striped bass (*Morone saxatilis*) is the primary indicator for human exposure to MeHg in San Francisco Bay and thus would be a potential candidate for a MeHg bioaccumulation model. They are generally representative of exposure in the open waters of the Bay. However, their use as indicators of local contamination risk is limited by their complex and wide movement range.

Striped bass is an anadromous fish, where young-of-the year and juveniles generally reside upstream of the Bay in lower salinity habitats, while adult fish exhibit seasonal migrations in and out of the Bay. During the fall of each year, adults generally move from the open ocean to the Delta, Lower South Bay and San Pablo Bay. They return to the Central Bay and the open ocean after spawning each spring (Moyle 2002). Evaluations of spatial patterns in striped bass Hg contamination have not found any significant differences among locations (Greenfield et al. 2005), consistent with their wide movement ranges and high variability among individuals.

Quantitative data on striped bass movements have not been well documented in the literature. Recently, stable isotope techniques have been employed to evaluate regional patterns in anadromous fish movement and foraging (Kiriluk et al. 1995, Godbout et al. 2010). Strontium isotopes in the otoliths of striped bass suggested that over the course of 4-6 year lifespan, striped bass spend less than a year in the Bay (Melwani, A.R. unpublished data). Isotope data point towards most of their time being spent in freshwater reaches of the Delta. However, considerable variability among individuals was observed. Considering their wide movement range and lack of time spent in the Bay, striped bass should be considered an indicator species for contaminant exposure over the entire Estuary region including the Bay, Delta, and ocean, rather than San Francisco Bay alone. Yet, they are included as a target species in the San Francisco Bay Hg TMDL, and thus are a priority indicator for managers.

Adult bass are upper trophic level, epibenthic and pelagic carnivores. They are highly opportunistic and primarily consume fish, depending on prey availability. Large adults (> 25 cm) consume fish and invertebrates. Important prey include threespine stickleback (*Gasterosteus aculeatus*), juvenile Chinook salmon (*Oncorhynchus tshawytscha*), Pacific staghorn sculpin (*Leptocottus armatus*), various smelts, and young of the year striped bass (Stevens 1966). However, diet is thought to vary greatly amongst individuals, consistent with the high variability in MeHg concentrations at a given age or length. Key covariates in Hg bioaccumulation are further discussed in Section 4. Diet

parameters for modeling striped bass bioaccumulation have yet to be developed for San Francisco Bay.

White Sturgeon

White sturgeon (*Acipenser transmontanus*) is the primary indicator for wildlife exposure to selenium in the Bay and the focus of the selenium TMDL that is in development. Like striped bass, white sturgeon is anadromous, spawning in large rivers during the spring and summer, and remaining in freshwater while young (Moyle 2002). Older juveniles and adults move seasonally between the open waters of the Bay and ocean. In contrast to striped bass that have maximum total length at < 50 cm and lifespan of less than 10 years, white sturgeon can grow to well over 150 cm total length and may live to 100 years old. White sturgeon is most abundant in the northernmost regions of the Estuary (particularly, Suisun and San Pablo bays, and the West Delta); however it has also been found in the Central and South bays (Baxter et al. 1999). Leidy (2007) reported that most individuals tagged in San Pablo Bay have been recovered outside the Estuary in open marine environments. Therefore, contaminant exposures of sturgeon more likely reflect regional variation rather than specific locations of the Bay.

White sturgeon movements have not been well studied in the Bay. Five individuals of a similar species, green sturgeon (*A. inediostriis*), were tagged in San Pablo Bay during 2001-02 (Kelly et al. 2007). Over a 12 day tracking period four individuals stayed within San Pablo Bay, while another individual moved > 45 km up the Delta. If white sturgeon is similar, they may be expected to move beyond specific Bay regions, with their exposure area a large source of uncertainty for modeling.

Adult white sturgeon are primarily exposed to contaminants through benthic dietary pathways. Adults are known to consume fish, shellfish, crayfish, and various aquatic invertebrates, including clams, amphipods, and shrimp. Clams tend to be the most consistent food item consumed by individuals collected in San Pablo Bay (McKechie and Fenner 1971). Stewart et al. (2004) suggested that the propensity for sturgeon to feed predominantly through benthic pathways may be the reason for higher selenium bioaccumulation in this species. In contrast, species that feed in the water column on crustaceans and other species are less prone to selenium bioaccumulation. This suggests that in the bioaccumulation modeling of selenium it is important to account for diets between benthic and pelagic exposure pathways. Diet parameters for white sturgeon have been developed in previous modeling efforts for selenium in San Francisco Bay-Delta (e.g., Presser and Luoma 2010).

Jacksmelt

Jacksmelt (*Atherinopsis californiensis*) is an indicator for human exposure to MeHg from fish consumption. Sub-adults and juveniles have also been employed as indicators of wildlife exposure risks (Ridolfi et al. 2010). Jacksmelt are a pelagic species that reside in the shallow waters of the Bay and near-shore coastal ocean (e.g., piers, rocky shores, and

jetties). They are particularly abundant in Central, South, and San Pablo bays (Emmett et al. 1991). Movement ranges are not well known for the species, but are expected to cross multiple regions of the Bay. Young usually remain in brackish water (i.e., San Pablo Bay), while adults can be found along the edge of most of the Bay and coastline. Jacksmelt are one of the most common fishes caught by pier anglers, and are also frequently caught in the surf zone (Frey 1971).

Jacksmelt is an omnivorous species that primarily consumes algae, detritus, and small, epifaunal crustaceans (mysids, copepods, decapod larvae). Larger adults switch towards feeding upon zooplankton and small fish (Bane and Bane 1971). Stomach content analysis in jacksmelt from the Bay indicated that amphipods comprise a significant contribution to their diet (Wang 1986). Diet parameters have been developed and validated for the Bay based on the PCB model of Gobas and Arnot (2005). They determined that the majority of the diet is comprised of phytoplankton (63%) and zooplankton (20%), and lesser contributions from bivalves (15%) and sediment (2%).

Mississippi Silverside

Mississippi silverside (*Menidia beryllina*) is an important indicator of wildlife exposure to MeHg in the Bay. Silversides are common in shallow, near-shore vegetated habitats of the Bay, particularly marshes and creeks where wildlife are abundant. They school in very large numbers, concentrated near protected areas with sand or gravel bottoms. Feeding is thought to occur in both deep and shallow water areas. Mississippi silversides foraging in marshes and tributaries may explain their elevated MeHg concentrations, as shallow habitats tend to exhibit greater Hg methylation and subsequent bioaccumulation than open waters (St Louis et al. 1994, Snodgrass et al. 2000, Marvin-DiPasquale and Agee 2003, Chumchal and Hambright 2009).

Silverside exhibited highly significant differences in THg concentrations across five capture locations in San Francisco Bay (Greenfield and Jahn 2010), and sometimes exhibited ten-fold differences in THg between Central Valley sampling locations within several kilometers of each other (Slotton et al. 2007). These findings, in combination with their restriction to nearshore habitats or mesohaline salinities, suggest relatively small movement ranges on the order of one to ten kilometers.

The diet of Mississippi silversides primarily consists of zooplankton (copepods and cladocerans), insects, and small, pelagic invertebrates. They are also known to prey upon benthic species when constrained to shallow water habitat (Jahn, A. 2010, pers. comm.). Gut content analysis of silversides collected from three margin sites in San Francisco Bay found that the species mainly consumed epibenthic crustaceans (specifically, corophiid amphipods), with relatively lower abundance of insects and planktonic crustaceans (Greenfield and Jahn 2010). Another study of Mississippi silversides in China Camp marsh (North Bay) found the species to primarily consume benthic species, and less utilization of zooplankton and insects (Visintainer et al. 2006). These dietary studies document the potentially higher exposure of small fish species along margin habitats of

the Bay relative to the open waters, and illustrate the importance of habitat type in modeling bioaccumulation in the Bay. Diet parameters have yet to be developed for modeling contaminants in silversides.

California Least Tern

The federally threatened California Least Tern (*Sternula antillarum brownii*) is the primary regulatory indicator for wildlife exposure and risk from MeHg in San Francisco Bay. The largest colony in Northern California is located in San Francisco Bay at the former Alameda Naval Air Station next to Oakland Harbor (Elliott et al. 2007). The Least Tern is a migratory species spending the winter along the Pacific Coast of Central America and the remainder of the year at nesting sites on the edge of San Francisco Bay or further south.

California Least Tern forage over shallow to deep open waters. Movements of individuals have not been documented in San Francisco Bay. Foraging ranges are however, expected to be similar to studies for Forster's and Caspian tern that have been found to have relatively small home ranges (see below for Forster's Tern data).

Least Tern are opportunistic avian piscivores (Elliott et al. 2007). Their diet primarily consists of juvenile jacksmelt (*A. californiensis*), topsmelt (*Atherinops affinis*), and northern anchovy (*Engraulis mordax*), although other prey up to approximately 15 mm in body depth may be consumed (Atwood and Kelly 1984, Elliott et al. 2004, Elliott 2005, Zuria and Mellink 2005). Diet parameters still need to be developed for Least Tern in San Francisco Bay.

Forster's Tern

Forster's Tern (*Sterna forsteri*) is an indicator for exposure to MeHg and risks to wildlife in salt pond habitats of the Bay. There are seven colonies distributed along the margins of the North and South bays (Strong et al. 2004). Eighty-percent of the breeding pairs reside on salt pond islands adjacent to the South Bay Salt Pond Restoration Area (Strong et al. 2004). Most of the remaining Bay population exists among salt marsh and tidal islands at the edges of San Pablo Bay, particularly near China Camp. Other habitats used less frequently are sewage treatment pond islands, salt pond levees, and freshwater lake islands.

Forster's Tern is known to have relatively small home ranges, which makes them particularly useful indicators at small spatial scales. However, similar to the California Least Tern, they winter outside of the Bay, and are therefore exposed to different conditions for a portion of the year. Ackerman et al. (2008a) found up to 87% of 72 Forster's Terns tagged in the South Bay salt ponds were re-located within the site of capture. On average, terns moved 2 – 8 km from the site of capture.

Forster's Terns forage in slough channels, marshes, and salt ponds, primarily consuming fish. In the Bay, the species is known to utilize small, forage fish (< 10 cm TL), which are abundant within wetlands (McNicholl et al. 2001) and salt ponds (Ackerman et al. 2008a). Prey fish generally less than 10 cm in body depth can be consumed, which include threespine stickleback (*Gasterosteus aculeatus*), longjaw mudsucker (*Gillichthys mirabilis*), various silversides (*Atherinopsidae*), and yellowfin goby (*Acanthogobius flavimanus*). Field observations of foraging habits in the South Bay salt ponds (Ackerman, J. unpublished data) suggest about half of Forster's Tern diet are stickleback and mudsuckers, comprising 36% and 25% of their diet, respectively. Due to well studied risks of MeHg to Forster's tern, sufficient quantitative diet exists to develop food web modeling parameters for the species. Furthermore, food web model parameters were developed and validated by Gobas and Arnot (2005).

California Clapper Rail

California Clapper Rail (*Rallus longirostris obsoletus*) is a federally endangered species that is an indicator of MeHg exposure and risk in Bay wildlife. They are most commonly found nesting in tidal marsh habitat of the Bay (Rigney et al. 1989), utilizing marshes in both the North Bay (San Pablo and Suisun bays) and specific patches in South Bay (Gill 1979).

Movements of the Clapper Rail have not been studied widely in the Bay (Albertson 1995, Overton 2007). Twenty-nine Clapper Rails were tagged from three marshes in the South Bay and found to range from 8 – 12 hectares. Annual core use areas did not vary greatly among the marshes but did show seasonal differences. Marshes with greater Clapper Rail density tended to exhibit smaller home ranges. In addition, the largest home ranges tended to be prior to breeding seasons and also towards the end of the breeding season. Generally, rails indicated reasonable site fidelity, with adjacent individuals exhibiting overlapping home ranges. Banding studies conducted by the U.S. Fish and Wildlife Service have similar indicated strong site fidelity, with individuals showing annual movement of less than 1 km (U.S. Fish and Wildlife Service 1991).

The diet of the Clapper Rail is not well known in San Francisco Bay. From studies in other areas, Clapper Rails are considered primarily opportunistic, foraging within emergent vegetation and along edges of marsh and mudflats (Clark and Lewis 1983, Meanley 1985, Eddleman and Conway 1998). They consume seeds, small crabs, spiders, amphipods, aquatic insects, clams, and minnows in their diet. In a single study reported for San Francisco Bay (Moffitt 1941), rails consumed ribbed horse mussels (57% of volume); spiders (*Lycosidae*, 15%); seeds and hulls of cordgrass (15%); *Macoma* clams (8%); mud crabs (3%); snails (*Ilyanassa obsoletus*; 2%); and insects, worms (*Nereis* spp.), and carrion (total 1.1%).

Song Sparrow

The Tidal Marsh Song Sparrow (*Melospiza melodia*) is a bird species common in San Francisco Bay that is a valuable indicator of MeHg bioaccumulation. The habitat of the Song Sparrow is limited to tidal marsh plain along the edges of the Bay, where they remain resident throughout the year (Grenier 2004). Quantitative bioaccumulation models have not previously utilized song sparrow species to evaluate contaminant risks.

Song Sparrow exhibit one of the smallest home ranges of any wildlife species, making them particularly useful indicator for monitoring and modeling contamination on relatively small spatial scales. They are known to forage over less than 1 hectare within marshes in San Pablo Bay and South Bay (Grenier 2004).

The diet of the song sparrow consists of seeds, insects, and small crustaceans. The vast majority of their diet is derived from small insects in marsh vegetation as they generally forage on the ground, in shrubs or in very shallow water (Grenier 2004).

Longjaw Mudsucker

Longjaw mudsucker (*Gillichthys mirabilis*) is another important indicator of MeHg patterns and risk in the aquatic food web. The species primarily inhabits shallow sloughs and tidal mudflats around the margins of the Bay. Due to their wide tolerances of salinity and temperature, the species also inhabits edges of salt ponds in the South Bay, where they are commonly associated with distributions of topsmelt, yellowfin goby, and staghorn sculpin.

Foraging ranges of longjaw mudsucker have not been extensively studied. They are, however, known to be territorial, spending their entire lifespan within the same marsh. Typically, longjaw mudsucker have been observed to live within a 30-50 m home range, though no quantitative information was found in the literature.

Diet of longjaw mudsucker consists of a wide variety of invertebrates and small fish. Their diet is known to shift seasonally, depending on what prey are available. Diet studies conducted in the South Bay salt ponds have indicated that longjaw mudsucker diet varies by salinity. In salt ponds with lower salinity (<40‰), mudsuckers consume polychaetes and amphipods. In the higher salinity (40-84‰) ponds, they consume primarily brine shrimp and backswimmers (Lonzarich 1989). During the winter, copepods are known to be an important prey item as brine shrimp are generally not available. In tidal sloughs and creeks draining salt marsh habitat, mudsuckers consume a higher degree of benthic species that include algae, isopods, amphipods, and small fish.

White Croaker

White croaker (*Genyonemus lineatus*) is a target species for current regional and statewide contaminant monitoring programs. The species is utilized as an indicator of organic contaminants, particularly PCBs, legacy pesticides, PBDEs, and dioxins (Davis et al. 2006c). White croaker is bottom-dwelling species that inhabits shallow near-shore areas of the Bay and San Francisco coastline.

Acoustic telemetry techniques can provide data on the home range area for target species. Using such techniques, recent and ongoing studies of white croaker have been successful in documenting their movement patterns on the Palo Verdes Shelf, as well as into and out of the nearby Los Angeles Harbor. Such studies indicate that croaker move broadly across 3 - 4 km of the shelf in an area of 1 km width (i.e., a 3-4 km² movement range). About 30% of the fish also moved into and out of Los Angeles Harbor, spending from a few hours to several weeks within the harbor and then returning to the shelf (C. Lowe, 2010, CSU-Long Beach, pers. comm.). Similar studies have yet to be conducted in San Francisco Bay. A lack of spatial information on white croaker foraging, and the potential for broad movement, is an impediment to development of site-specific models for this species.

White croaker are bottom-feeders, with a diet that predominantly consists of benthic invertebrates and fish. The most common food items are polychaetes, crabs, amphipods, and small fish. Several dietary studies in San Francisco Bay observed gut contents to include bivalves, polychaetes, crangonid shrimp, and small fishes (Sanchez 2001, Sigala et al. 2002, Jahn 2008). Food web model parameters for white croaker have been validated in case studies for San Francisco Bay (Greenfield et al. 2007, Gobas and Arnot 2010). The modeled diet largely includes benthic invertebrates: polychaete worms (40%), amphipod crustaceans (20%), and cumacean crustaceans (20% *Nippoleucon hinumensis*). Additional invertebrate prey included mysids (10%) and *Crangon* (shrimp) (5%). Sediment also often represents up to 5% of white croaker diets, because croaker are roving benthic grazers that siphon sediment to consume prey (C. Lowe, 2010, CSU-Long Beach, pers. comm.).

Shiner Surfperch

Shiner surfperch (*Cymatogaster aggregata*) is a small, abundant fish species in San Francisco Bay. Despite its small size, shiner surfperch is frequently caught by recreational fishers due to high abundance and easy capture at piers and docks (Bane 1970). Therefore, like white croaker, they have been frequently used in regional and statewide contaminant monitoring programs as indicators of human exposure to organic contaminants. However, they feed lower in the food web than other typical sport fish.

Shiner surfperch are most abundant in the shallow areas of South, Central and San Pablo bays. Shiner surfperch reside in deeper water during winter months and many adults also migrate to the ocean (Moyle 2002). During the spring breeding season, females

immigrate from near-shore coastal waters to the Bay prior to giving birth. Little is known about the movement and foraging range of shiner surfperch. They are assumed to stay relatively localized, except during seasonal migrations between deep and shallow waters. The extremely low variance observed in replicate samples at different sampling locations in the Bay (Davis et al. 2011) suggests a high degree of site fidelity, at least in the exposure period prior to capture.

Shiner surfperch are generally epibenthic feeders, primarily feeding off the sediment surface or on epifauna of hard structures. Shiner surfperch has been the subject of several diet studies (Odenweller 1975, Hobson and Chess 1986, Roberts et al. 2002, Sigala et al. 2002, Jahn 2008). Odenweller (1975) reported that for Anaheim Bay shiner surfperch, the primary food source was zooplankton and benthic organisms, including bivalves, gastropods, polychaetes, tunicates, and fish eggs. Several dietary studies in San Francisco Bay indicate particular reliance on benthic and epibenthic crustaceans, augmented by polychaetes and clams (Jahn 2008). Shiner surfperch are consumed by many species of larger marine fishes, including sturgeon, salmon, and striped bass. They are also consumed as prey by piscivorous birds and harbor seals (Elliott 2005, Froese and Pauly 2010). Diet parameters developed for shiner surfperch have been validated for PCBs and legacy pesticides (Gobas and Arnot 2005, Greenfield et al. 2007). They estimated that equal proportions of the diet is comprised of polychaetes, amphipods, and cumacea (20% each), and lesser contributions from mysids (15%), phytoplankton (10%), zooplankton (10%) and sediment (5%).

Topsmelt

Topsmelt (*Atherinops affinis*) are small, schooling fish, abundant in near-shore marshes and open waters of San Francisco Bay. Juvenile topsmelt are more likely to be found in freshwater and are less benthically focused. The species does not tend to make seasonal migrations but will move between shallow and deep water with the tide, in search of food.

Tag recapture or telemetry studies of movement distance are not available for topsmelt or similar California species. Although topsmelt are not expected to exhibit any kind of fidelity to a specific site or region, their small relative size and nearshore habitat would suggest relatively small home ranges. Studies of Hg and organic pollutants in topsmelt similarly indicate significant differences in contaminant concentrations among adjacent sampling locations (Battelle et al. 2005, Greenfield and Jahn 2010). For example, topsmelt exhibited significant differences in Hg concentrations across five capture locations in San Francisco Bay, though these differences do not explain as much variation as for Mississippi silverside (Greenfield and Jahn 2010). Topsmelt also exhibited PCB concentrations adjacent to the contaminated Hunter's Point Naval Shipyard, approximately three times higher than five other Bay-wide monitoring stations (SFEI 2009). Topsmelt sampled across different southern California mainland and coastal sites exhibit significant variation in body morphology (O'Reilly and Horn 2004). These findings of differences in contaminant concentrations and morphology suggest that

topsmelt populations are spatially distinct among regions, and that the species has a limited movement range. Given their low trophic position, the high PCB concentrations observed in topsmelt (up to 420 ppb near San Francisco) are surprising.

Topsmelt consume benthic and pelagic plants and invertebrates. Topsmelt usually feed by grazing the bottom of the water column. The species are considered omnivores and feed on diatoms, benthic algae, detritus, midge larvae, and amphipods (Marine Biological Consultants Inc. and SCCWRP 1980, Logothetis et al. 2001, Horn et al. 2006, Visintainer et al. 2006, Greenfield and Jahn 2010). Dietary studies indicate a combination of benthic and pelagic invertebrates in the diet. Gut analyses by Horn et al. (2006) indicate dietary adaptations for herbivory, suggesting that plant material constitute a primary component of topsmelt diets (O'Reilly and Horn 2004). Results from these studies suggest that bioaccumulation model input parameters should include moderate dietary proportion from both herbivory and benthic amphipods (i.e., 40% each); and minor contributions from zooplankton (10%), sediment (5%), and small, benthic invertebrates (mysids, polychaetes, cumaceans; 5%).

California Halibut

California halibut (*Paralichthys californicus*) is a large flounder that has been utilized as an indicator of human exposure to contaminants. They are observed in near-shore waters on the San Francisco Bay and northern California coastline (Eschmeyer and Herald 1983, Allen 1990). San Francisco Bay is the nursery ground for juvenile halibut less than eight inches, because it provides more protected and stable habitat for growth and survival than the offshore waters (Allen and Herbinson 1990). Halibut greater than eight inches migrate seasonally between the Bay and the open coast (Domeier and Chun 1995). Adults occur on sandy sediment but sometimes concentrate near rocks, algae, or Pacific sand dollar beds. As with other flatfishes, they frequently lie buried or partially buried in the sediment. Adults move inshore to spawn during the spring and summer and offshore during the winter (Haaker 1975). These movements may also be associated with schools of prey (e.g., California grunion, *Leuresthes tenuis*), which are abundant near the surf zone during the spring and summer (Feder et al. 1974).

There are several published studies of tag-recapture in California halibut to develop movement parameters for food web models. CDFG has conducted extensive tag and release studies of California halibut (Haaker 1975, Tupen 1990, Domeier and Chun 1995, Posner and Lavenberg 1999). Results have indicated that halibut less than 20 inches remained relatively localized and traveled less than 2 miles, although halibut greater than 20 inches traveled greater distances. The average overall distance traveled was eight miles. Results of some studies have also documented greater halibut movement parallel to the coastline than further offshore. California halibut analyzed in the RMP have been greater than 55 cm (22 inches) in total length (TL) suggesting they would be of the size expected to have wider movement patterns.

Adult California halibut greater than 20 cm TL are primarily piscivorous, with fish comprising the vast majority of their prey by mass, including topsmelt, California killifish, northern anchovy, and gobies (Plummer et al. 1983, Wertz and Domeier 1997). Other species in their diet include ostracods, snails, and cephalopods. Overall, the diet of the California halibut changes from harpacticoid copepods, amphipods, and gobies in small bay-living juveniles to primarily northern anchovy. The largest individuals consume croakers and other larger fishes. These shifts in diet and extensive migrations suggest California halibut are challenging for modeling site-specific bioaccumulation in the Bay. Based on the available information, a bioaccumulation model using halibut should parameterize a diet of 98% forage fish, including both benthic and pelagic prey fish and minor (2%) contribution of sediment.

Double-crested Cormorant

Double-crested Cormorant (*Phalacrocorax auritus*) is a species of special concern in California. They are primarily utilized as indicators for exposure to organic contaminants in the open water habitats of the Bay.

Cormorants forage and roost throughout the Bay on bridges, electrical towers, and pier pilings. Breeding cormorant colonies only exist at a few locations around the Bay, with the most stabilized being present in the North Bay at the salt evaporators near Napa, in the Central Bay at the Richmond-San Rafael and Oakland-San Francisco bridges, and in the South Bay at the Dumbarton Bridge (Goals Project 2000). Cormorants in these colonies are resident year round and are known to eat fish from 20 or more miles away from these areas.

Cormorants reflect regional conditions due to their relatively wide foraging ranges. Bird egg monitoring studies conducted by the RMP have indicated significant spatial variation among regions in several contaminants (PCBs, Hg, and dioxins) (Davis et al. 2007b). Similarly, Schwarzbach and Adelsbach (2003) observed that cormorant eggs were indicators of regional variation in mercury concentrations, with relatively high concentrations in Suisun Bay and San Francisco Bay compared to the Delta.

Cormorants generally feed in shallow, open waters and close to shore. Their diet primarily consists of fish (Goals Project 2000). During the winter, herring is an important prey item. Plainfin midshipmen are utilized in their diet during spring and summer (Ainley et al. 1981). Dietary information collected from Cormorants residing on the Richmond-San Rafael Bridge in 2000 indicated that yellowfin goby, Bay goby, Cabezon, and sculpins represent the major prey items (Rauzon et al. 2001). Quantitative diet data from these various studies need to be compiled to develop model parameters for Cormorants.

California Mussel

The California mussel (*Mytilus californianus*) is observed in dense aggregations on rocks in the middle intertidal zone, where they are exposed to the strong action of the surf. California mussels can grow up to 20 cm shell length and may live for more than 20 years. However, mortality in the intertidal zone is often high, due to physical disturbance, predation, desiccation, and disease.

Several agencies have used the California mussel (*Mytilus californianus*) to measure the bioavailability of chemicals in the water column (Stephenson et al. 1995, O'Connor and Lauenstein 2006, Hunt and Slone 2010). Mussels are efficient integrators of water column concentrations of bioaccumulative substances over time. Thus, they have been utilized in San Francisco Bay for identifying hotspot areas needing further investigation and monitoring long-term trends. Mussels are sessile organisms distributed widely along the coasts and Bay margins. Their life history minimizes the problems of spatial integration inherent in most other mobile, indicator species.

Diets of the California mussel have not been studied extensively. California mussels are filter-feeding bivalves that siphon phytoplankton from the water column for food. Therefore, it is safe to assume that > 90% of their diet is obtained from the water-column and this likely is comprised of phytoplankton, zooplankton, and detritus.

Harbor Seal

San Francisco Bay supports a year-round resident population of Pacific harbor seals (*Phoca vitulina*). They are the only marine mammals that are permanent residents in the Bay. Although infrequently sampled, due to their relatively large size and high trophic position, they are frequently utilized as indicators of organic contaminants in the Bay.

Pacific harbor seals are distributed at several haul-out sites associated with Central Bay and South Bay feeding areas. Haul-outs usually have gently sloping terrain, adjacent to deep water for feeding, and are free of disturbance (Allen 1991). The Central Bay feeding areas include Castro Rocks, Brooks Island, Angel Island, Yerba Buena Island, and at the Oakland breakwater entrance into Alameda Harbor (Allen 1991, Harvey and Torok 1994). The South Bay feeding area consists of 14 haul-out sites that include Coyote Point, Belmont Slough, Bair Island, Greco Island, Hayward Slough, Dumbarton Point, Newark Slough, Mowry Slough, Calaveras Point, and Guadalupe Slough. Pacific harbor seals often use several haul-out sites throughout the year, moving between them seasonally (Thompson et al. 1989). However, in San Francisco Bay, seals tend to collect on the same haul-out sites year after year (Yochem et al. 1987, Kopec and Harvey 1995).

Pacific harbor seals generally exhibit considerable site fidelity, but individuals are known to range up to 500 km. The majority of tagged seals examined by Nickel and Grigg (2002) were observed to use foraging areas within 20 km of a known haul-out site. Most foraging occurred within 1-5 km of the site; however they noted substantial variability

among individuals. In some instances, individuals moved 50-100 km between foraging locations and a few made long-distance trips outside of the Bay. Differences in an individual seal's tendency to travel over long distances may be due to sex or age. Similar variation among individuals has been observed elsewhere (Yochem et al. 1987).

Harbor seals are opportunistic predators, feeding primarily on small, benthic and pelagic fish in the deeper waters of the Bay (Grigg 2003 and references cited there-in). In 215 fecal samples, Harvey and Torok (1994) identified 14 species of fish and one cephalopod collected from harbor seals at seven haul-out sites around the Bay in 1991-92. Diet was observed to change seasonally and regionally, with yellowfin goby and staghorn sculpin consumed during the fall and winter, and plainfin midshipman, white croaker, jacksmelt, and yellowfin goby predominant during spring and summer. The goby, sculpin, and croaker were primarily identified in samples from the Lower South Bay. In the Central Bay, the plainfin mid-shipman predominated, comprising 91% of the diet. The PCB food web model of Gobas and Arnot (2005) estimated diet parameters for juvenile and adult harbor seals.

4. Key Concepts Supporting Bioaccumulation Modeling

Bioaccumulation integrates chemical, physical, and biological processes that vary both spatially and temporally. Mechanistic bioaccumulation models quantify and predict the net effect of these processes and thus must be based on conceptual models that describe the important processes. This section of the report discusses three categories of topics that must be understood in order to model bioaccumulation in the San Francisco Bay ecosystem: 1) factors influencing bioaccumulation; 2) uptake into the food web; and 3) mechanisms for contaminant uptake and elimination. The present state of knowledge on these topics with respect to supporting model development is reviewed, and key gaps are highlighted.

Factors Influencing Bioaccumulation

There are many co-varying factors that can influence the degree of bioaccumulation that occurs in an organism. Here, we discuss those that are important for quantitative modeling of bioaccumulation in the Bay, including the distribution of contaminants, the influence of management actions, habitat types, and spatial and temporal trends in concentrations.

Distribution of Contaminants

Contaminant concentrations in San Francisco Bay biota and sediment vary both spatially and temporally (discussed in Section 2). A key factor driving variation in patterns of biota contamination in the Bay is the heterogeneous spatial distribution of water and sediment contaminant concentrations. PCB concentrations, for example, are relatively high in sediments of specific Bay regions, such as Lower South Bay and Oakland Harbor (Figure 5). Spatial variation in contamination patterns is caused by variation in legacy

pollution sources, hydrological processes (e.g., flow and sediment deposition), or biogeochemical processes (e.g., *in situ* methylation). Historic industrial areas are associated with elevated bioaccumulation in certain Bay margin locations (Jones et al. 2011). For example, shiner surfperch collected from the industrialized Oakland Inner Harbor have distinctly elevated PCB concentrations (Davis et al. 2011).

Methylmercury (MeHg) is an example of a contaminant with spatial patterns in biota concentrations that result from a combination of patterns in both historic sources and biogeochemical processes. Elevated MeHg in water and sediment is associated with elevated mercury concentrations in bird eggs and forage fish at locations in closer proximity to the Guadalupe River and Alviso Slough pond complex of the Lower South Bay (Ackerman et al. 2008b, Greenfield and Jahn 2010, Grenier and Davis 2010). This region exhibits elevated THg and MeHg in water and sediments, influenced by historic mercury mining operations, industrial activity, and local biogeochemical processes (Thomas et al. 2002, Conaway et al. 2004, Conaway et al. 2008, Grenier and Davis 2010, Gehrke et al. 2011a). In this region, sediment percent MeHg correlates with Hg in forage fish and song sparrows (Grenier et al. 2010b, Gehrke et al. 2011b), suggesting a key role for mercury methylation in bioaccumulation.

Spatial variation in MeHg bioaccumulation is often driven by production in sediment and the water column (Mason and Lawrence 1999, Hammerschmidt and Fitzgerald 2004). The MeHg mass balance developed by Yee et al. (2011) identified *in situ* production of MeHg as the likely largest source of MeHg to the Bay. However, current estimates of production rates are highly uncertain. Data on methylation rates are limited, particularly in habitats with relatively high methylation potential (Marvin-DiPasquale and Agee 2003, Yee et al. 2007, Windham-Myers et al. 2009, Grenier et al. 2010b).

The amount of wetland area and the occurrence of wetting and drying cycles have also been shown to be positively correlated with biota MeHg contamination (Snodgrass et al. 2000, St Louis et al. 2004). Positive correlations of MeHg concentrations in biota with sediment organic matter (Benoit et al. 1998) and dissolved organic carbon (Driscoll et al. 1995, Watras et al. 1995) may reflect effects of methylating bacterial activity that are often enhanced in such situations. A number of studies have observed correlations between the percentage of wetlands in a watershed and concentrations of MeHg in waters draining the watershed (St Louis et al. 1994, Hurley et al. 1995). Marvin-DiPasquale (unpublished data) observed MeHg concentrations between 0.41 ng/g and 25.2 ng/g in tidal marsh sediment in the Bay, with percent MeHg ranging between 0.1% and 6.6%. As percent MeHg is considered an indicator of the rate of MeHg production (Gilmour et al. 1998), the relatively high maximum percent MeHg measured in some Bay tidal wetlands indicates potentially high net methylation rates in some of these ecosystems.

Detailed information on the spatial distribution of contaminants is needed for quantitative estimation of exposure in mechanistic bioaccumulation models. Bioaccumulation modeling can begin with a simple, static representation of environmental contamination (Gobas and Arnot 2010). When more spatial and temporal detail is needed, empirical data can be developed in parallel with contaminant fate and bioaccumulation models to

simulate important dynamic processes for comparing management alternatives. Extensive regional monitoring in San Francisco Bay has provided some information that can be used to drive spatially-explicit bioaccumulation models. However, information on local scale patterns and processes (e.g., methylation rates) are lacking in many areas, especially on the Bay margins.

Management Actions

On a Bay-wide scale, the mercury and PCB TMDLs call for load reductions that are expected to reduce bioaccumulation in key target species. The mercury TMDL is based on a simple assumption that food web contamination will decline in proportion to projected decreases in sediment contamination. On the other hand, bioaccumulation modeling was conducted as part of development of the PCB TMDL, but is still dominantly influenced by projected trends in sediment contamination.

Tidal marsh restoration is another principal type of environmental management activity that could affect bioaccumulation in the Bay food web. Environmental managers and regulators in the Bay area are concerned that tidal marsh restoration may increase the amount of MeHg entering the food web and elevate risks to wildlife (Davis et al. 2003). Tidal wetland restoration could create conditions within the wetlands that favor net MeHg production (Wiener et al. 2003).

One way to evaluate this concern is to monitor and model MeHg bioaccumulation in the food web under pre- and post-restoration scenarios. A recent South Bay study evaluated the effects of restoration actions (Grenier et al. 2010b). The study employed indicator species to compare existing managed salt pond habitat to restored tidal marsh habitat. Three indicator species (longjaw mudsucker, *Gillichthys mirabilis*; brine fly, *Ephydra riparia*, and Song Sparrow, *Melospiza melodia*) were used to represent different parts of the tidal marsh (marsh channel, marsh panne, and marsh plain, respectively). No differences in MeHg concentrations were observed in these species under the pre-restoration conditions compared to the projected restored, tidal marsh conditions. These findings suggest that in some circumstances restoration of managed habitat to tidal marsh may not increase MeHg exposure to wildlife.

However, different types of restoration actions may have different effects on local MeHg bioaccumulation. Flooding of upland areas is known to increase net MeHg production and potentially increase exposure in the lower food web (Slotton et al. 2002, Mailman et al. 2006). Altered hydrological regimes may change rates of delivery of Hg to areas where net MeHg production is high. It follows that changes in elevation, vegetation, and productivity resulting from wetland creation can possibly create conditions favoring enhanced net MeHg production.

Extensive contaminant remediation efforts underway at multiple legacy polluted Bay margin sites (Jones et al. 2011) are also expected to influence patterns of bioaccumulation. These activities are intended to reduce biota exposure to pollution at hotspots of contamination. Combinations of dredging and capping are in various stages

of application at a few locations, and have met with varying degrees of success (e.g., Weston et al. 2002, U. S. EPA 2004). Activated carbon amendment to contaminated sediments has been the subject of extensive local study at Hunters Point Naval Shipyard and Luritan Canal in Richmond Harbor (Cho et al. 1999, Cho et al. 2007, Tomaszewski et al. 2008). Activated carbon tightly binds hydrophobic sediment-associated pollutants (e.g., PCBs, DDTs, PAHs), and is intended to reduce bioavailability and bioaccumulation (Hale et al. 2009, Sun et al. 2009, Janssen et al. 2011). Linked pollutant fate and bioaccumulation modeling is needed to guide these efforts through forecasting their likely effectiveness.

Overall, management actions in the Bay are expected to affect future accumulation of contaminants in biota. Models of bioaccumulation should thus account for differences in water and sediment quality as a result of actual and anticipated management actions, especially in cases where they affect chemical mobility and bioavailability.

Seasonal Trends

Seasonal variation in bioaccumulation of pollutants is observed in primary producers, invertebrates, and fish of many aquatic ecosystems (e.g., Harding et al. 1997, Cleckner et al. 1998, Gorski et al. 1999, Stapleton et al. 2002, Slotton et al. 2007), including San Francisco Bay. Eagles-Smith and Ackerman (2009) recently documented pronounced seasonal variation of MeHg in forage fish from two salt ponds in the Lower South Bay. The authors observed a 40% increase during the summer in length-adjusted Hg of longjaw mudsucker (*Gillichthys mirabilis*) and threespine stickleback (*Gasterosteus aculeatus*). These results illustrate the importance of short-term variation in forage fish in modeling MeHg exposure to wildlife. However, seasonal patterns also vary among locations. Spring peaks have been observed in forage fish captured within tributaries adjacent to Suisun and San Pablo Bays after large storm events (Slotton et al. 2007). In North Bay salt ponds, seasonal variation in forage fish Hg vary among sites (Grenier et al. 2010a). RMP data collected at Martin Luther King Jr. Regional Shoreline in San Leandro Bay also indicate differences in seasonal variation among species; while arrow goby peak concentrations occur in summer, topsmelt peak concentrations occur in winter (Rauzon et al. 2001).

Given differences among sites and species in seasonal contamination patterns, and limited data, there remains uncertainty regarding seasonal variation in risk to piscivorous wildlife. The only published evaluation of seasonal variation in bioaccumulation of organic pollutants in San Francisco Bay was for PCBs and legacy pesticides in white croaker (Greenfield et al. 2005). In this study, white croaker PCBs (fillets with skin) were lower during the spring (mean = 115 ng/g ww) compared to concentrations measured in the summer (277 ng/g) and fall (314 ng/g). Similar observations were made for DDTs and chlordanes. These patterns corresponded with similar variation in lipid concentrations. Therefore, observations of seasonal differences were hypothesized to result from seasonal differences in condition (measured as percent lipid) and diet. Previously, white croaker collected from southern California water bodies has indicated lower body condition during spawning (Love et al. 1984). This suggests that energy

transferred to reproductive organs and allocated toward spawning explain the reduced muscle and skin lipid content and contaminant concentrations of samples collected during the spring. Alternatively, reduced feeding during the winter months may have resulted in the observed decline of lipid content in the sampled muscle tissue and repartitioning of the contaminants to lipid storage areas.

In some cases, short-term seasonal variation may need to be characterized to understand exposure at sensitive life stages, and to explain differences both within and among sites at different times of the year. This includes circumstances where management actions could cause variable exposure throughout the year, such as hydrological modifications affecting MeHg production. In these circumstances, time-dependent mechanistic models of contaminant bioaccumulation may aid in forecasting exposure during periods that are critical for health risks.

Long-term Trends

Since the bioaccumulative pollutants of greatest concern are extremely persistent, interannual trends over the long-term are of primary interest to managers. Empirical information on long-term trends is an essential benchmark for bioaccumulation model development.

RMP sport fish bioaccumulation data have not indicated long-term trends over the past 15 years (Davis et al. 2011). The longest time series for trace organics exist for PCBs and legacy pesticides in shiner surfperch and white croaker. On a wet-weight basis, PCBs have exhibited interannual variation among some years but have not indicated a consistent pattern of declining concentrations. Interannual variation is largely associated with variation in lipid content among years (Davis et al. 2011). Examining RMP data in combination with historical data from the 1960s indicates approximately a 90% decline in PCBs and a 97% decline in DDTs (Risebrough 1995, Davis et al. 2011), consistent with a general decline in these pollutants in biota since domestic PCB production was banned in the 1970s (Borgmann and Whittle 1992, Stephenson et al. 1995, Lauenstein and Daskalakis 1998, Greenfield et al. 2004). It should be acknowledged, however, that significant improvements in analytical techniques for measuring organic contaminants since the 1960s may in part explain these large declines.

In the case of Hg, a relatively extensive historical dataset exists for striped bass in the Bay-Delta Estuary, which has not indicated a decline from the early 1970s to the present (Davis et al. 2011).

In addition to sport fish, the RMP continues to monitor contaminant accumulation in bivalve tissue at nine sites distributed throughout the Bay. Many of these sites were historically monitored by the State Mussel Watch Program and the time series were subsequently continued by the RMP. All nine sites have indicated declines in PCBs, DDTs, and other legacy contaminants (Gunther et al. 1999). These patterns suggest that concentrations of organic contaminants on the suspended matter consumed by these

filter-feeders in open Bay locations has declined. These declines are in contrast to the lack of trends observed in sport fish from 1994 to the present.

The large foraging range of some indicator species such as striped bass and sturgeon make them suitable only for regional scale (e.g., Delta to Gulf of the Farallones) trends monitoring. Sedentary or resident species, species such as bivalves or small fish, can provide indications of long-term trends within Bay segments, as long as potential causes of seasonal and interannual variability are adequately accounted for in the analysis (Gunther et al. 1999, Greenfield et al. 2005, Greenfield and Jahn 2010).

Overall, the long-term trend data that are presently being collected by the RMP provide a good foundation for bioaccumulation model development.

Habitat Types

One aspect of spatial pattern is the type of habitat under consideration. This section reviews four basic habitat types that are prevalent in San Francisco Bay. Depending on the contaminant and species under consideration, processes specific to these habitats may require consideration in local bioaccumulation model development.

Open Waters

Open waters are important habitat for many aquatic species considered in this report. Important indicator species that spend much of their time in the open waters include striped bass, white sturgeon, California halibut, topsmelt, jacksmelt, and Double-crested Cormorant. Pacific harbor seals predominantly reside on haul-out sites along the edges of the Bay, but forage primarily in the open waters (Harvey and Torok 1994). Generally, within the open waters of Central Bay and South Bay, organisms that are more representative of the coastal ocean predominate. Surfperches, sculpins, and gobies are common fish in this habitat. Additionally, dynamics of the broad, shallow, brackish waters of the North Bay (e.g., Suisun Bay) that historically produced abundant annual diatom blooms differ from other parts of the Bay. These blooms used to support abundant populations of zooplankton. However, filtration by the invasive Asian clam (*Corbula amurensis*) has effectively eliminated the phytoplankton blooms in this region (Alpine and Cloern 1992, Jassby et al. 2002). White sturgeon, which are more abundant in these waters, prey heavily on the Asian clam (Stewart et al. 2004).

Sloughs, Channels, and Mudflats

Near-shore habitats, particularly sloughs, channels, and mudflats of the Bay, are important for many organisms. Diverse assemblages of benthic invertebrates live on or just below the surface of the sediment present in nearshore habitats (Nichols et al. 1988). During high tides, the mudflats and edges of sloughs and channels provide foraging habitat for many species of fish and birds (Cohen 1990, Goals Project 2000). Sloughs and channels are habitat for forage fish, like the longjaw mudsucker, Mississippi silverside, and topsmelt (Goals Project 2000, Visintainer et al. 2006, Mejia et al. 2008). Mudflats are

also vital foraging habitat for many shorebirds, like Clapper Rail that forage on invertebrates and small fish (Goals Project 2000). Infrequently, harbor seals may also forage in the larger sloughs present near haul-out sites (Grigg 2003).

Salt Ponds

Salt ponds, now primarily owned by state or federal agencies and managed as wildlife habitat, surround the southern end of the South Bay, the northeast shore of San Pablo Bay, and the lower reaches of the Napa River. Active salt ponds are also present along the shallow margins on either side of the South Bay. Salt ponds differ from other major habitats of the Bay, being disconnected to the Bay and devoid of vegetation. Salt ponds offer isolation from the dynamic, open waters of the Bay, yet still provide plentiful food, which attract many bird species in significantly greater concentrations than are observed in any other habitats of the Bay. During certain times of the year, over 200,000 shorebirds and 75,000 waterfowl have been reported on the South Bay's salt ponds. Of the indicator species being considered in this paper, Forster's Tern is particularly known to concentrate in the South Bay salt ponds, as they nest on dikes and islands among the ponds that are not found in many other regions of the Bay. Fish species residing in salt ponds comprise a subset of Bay species that is tolerant of extreme conditions, including elevated salinity (Mejia et al. 2008). For example, the longjaw mudsucker is able to withstand the higher salinities and warmer temperatures.

Tidal Marsh

Tidal marshes represent important habitat for fishes and aquatic and riparian birds of San Francisco Bay. Tidal marshes are hydrodynamically connected to the Bay, with vegetation typically consisting of species of vascular plants, dominated by common pickleweed (*Salicornia pacifica*) and California cordgrass (*Spartina foliosa*). Small fish including longjaw mudsucker and Mississippi silverside forage in the smaller channels and among the fringes of lower tidal marshes (Goals Project 2000, Visintainer et al. 2006, Mejia et al. 2008). Many birds prey upon the concentrated abundance of fish carried with the tide. Along channel banks and slough edges, California Least Tern is frequently observed foraging at high tide. Tidal marsh Song Sparrows peck the mud beneath the pickleweed canopy at low tide for small worms and snails to augment their diet of seeds (Grenier 2004). California Clapper Rails and Black Rails are other tidal marsh species that have been used as biosentinels.

Summary of Habitat Issues and Previous Model Development

San Francisco Bay contains a diverse array of habitats, each of which is used by multiple taxa. To understand the relative importance of various sources of contamination, the habitats used by indicator species as well as those of their prey items must be understood. Habitats to be considered will depend upon the foraging ranges and life histories of indicator species of interest and upon the specific management questions to be answered. Restoration plans, sea level rise, and other morphological changes will alter the areas of

these habitats, which in turn will affect contaminant bioaccumulation in future (long-term) modeling scenarios.

Despite the importance of San Francisco Bay margin habitats for management, they have been the subject of almost no mechanistic modeling activity to date. All bioaccumulation modeling development has been focused on food webs and processes within the open water habitats. Examples include a PCB food web model (Gobas and Arnot 2010), bioenergetic models of mercury bioaccumulation in sport fish (Pickhardt et al. 2006, Greenfield et al. 2009b), and bioenergetic models of selenium in clams and pelagic invertebrates (Schlekat et al. 2004, Lee et al. 2006). Other San Francisco Estuary habitats (including salt ponds, sloughs, channels, mudflats, and tidal marshes) have received increasing attention for contaminant monitoring in biota, focused almost exclusively on methylmercury (Ackerman et al. 2007, Best et al. 2007, Best et al. 2009, Greenfield and Jahn 2010, Grenier et al. 2010b). To our knowledge, there have been no attempts at developing mechanistic models for bioaccumulation of any contaminants in these habitats. The modeling platforms and processes used for open water habitats (Pickhardt et al. 2006, Greenfield et al. 2009a, Gobas and Arnot 2010) should be largely applicable to these other habitat types. The primary effort would lie in assembling parameters (e.g., food web structure and abiotic input variables) specific to these habitats.

Spatial Scale and Movement

Due to the spatial variation in sediment and biota contamination, spatial scale must be considered in species selection and other aspects of bioaccumulation model development. Species with relatively narrow foraging ranges (e.g., longjaw mudsucker, Song Sparrow) will be expected to have tissue concentrations most reflective of local site conditions. Concentrations in wider ranging species will reflect contamination at broader spatial scales.

Bioaccumulation models have been developed to incorporate space dependent parameters. In these models, elements related to species behavior that pertain to exposure and risk from contaminants include foraging area size, habitat preferences, and characteristics (size and quality) of the study site (Freshman and Menzie 1996, Menzie and Wickwire 2001, Hope 2005). Accounting for spatial movement can change estimated risks and bioaccumulation calculations compared to spatially independent models (i.e., Bay or region wide) (Linkov et al. 2002, von Stackelberg et al. 2002, Oram and Melwani 2006, Burkhard 2009, Melwani et al. 2009). The bathtub ring hypothesis described above suggests that spatial patterns in contamination and foraging of key indicator species may be fundamentally important in explaining spatial and temporal patterns of bioaccumulation. The purpose of this section is to review methods for determining spatial movement by bioaccumulation indicator species, and how this information may be incorporated into conceptual models of bioaccumulation.

Indicator species vary widely in their movement and foraging patterns. At the high end of the scale, anadromous striped bass reside in both estuarine San Francisco Bay, as well as a substantial portion of time in the ocean and the Delta (Calhoun 1952). On the opposite

extreme, some species of bottom fish, as well as shellfish, are sedentary, effectively residing at a single location for their entire adult life. Similarly, individuals within a given species may vary widely in their movement, with some individuals remaining in a relatively small locale, and others captured in a different Bay region entirely. Therefore, wildlife can exhibit significant spatial and seasonal variation in bioaccumulation depending on their foraging area, size of the habitat, and distribution of contaminants. This variation in contaminant exposure must be addressed in modeling bioaccumulation.

Home Range

Home range is an important concept in characterizing spatial movement of wildlife. Home range can be defined as the estimated spatial area that an animal covers during its adult lifetime foraging activity (Hope 1995). There are many factors that influence home range. As an example, Lowe and Bray (2006) developed a conceptual model for the home range of marine and estuarine finfish based on studies to date. Based on this conceptual model, five species attributes affect home range: body size, diet, foraging strategy, territorial behavior, and habitat. Larger fish will tend to have larger home ranges (Minns 1995). Foraging strategy will also influence home range; ambush predators have relatively small home ranges, as they do not actively move in seeking prey. In contrast, active foragers that search for areas of prey availability will tend to have larger ranges. Territorial fishes will tend to have smaller ranges than non-territorial fishes, as they inhabit and defend a discrete location. Finally, fishes that inhabit structurally complex habitats (e.g., eelgrass, rocky reefs, and human-made piers and other structures) will have smaller home ranges. Due to higher prey density, these areas tend to require less movement to obtain sufficient prey areas with limited structural complexity (e.g., soft sediment). The Lowe and Bray model forms a basis to extrapolate across otherwise similar species, based on differences in these factors. For example, if a species exhibits general life history characteristics intermediate between two previously studied species, than an intermediate estimated home range would be appropriate.

Benthic fish species tend to exhibit less variation in home range relative to pelagic species. Linkov et al. (2002) examined three fish species (eel, flounder, and bluefish) that represented different foraging strategies in New York-New Jersey coastal areas. Eels and flounder generally forage on the benthos in the same area, while bluefish are known to forage in the water column over large areas. Observed PCB concentrations in eel varied by up to three orders of magnitude in six sampling areas, while the range for PCBs in bluefish was an order of magnitude or less. Winter flounder caught within the same general area also exhibited spatially variable PCB concentrations, while individual bluefish did not show as much variation. These differences support the hypothesis that benthic fish with small foraging areas are likely to reflect local sediment contamination. However, the converse is also true – benthic fish with large foraging areas can have a weaker connection to local contamination. For example, in San Francisco Bay, adult California halibut are understood to undertake large migrations from the Bay, and exhibit weaker linkages to local contamination than shiner surfperch, which have high site fidelity (Melwani et al. 2009).

Some large pelagic fish are known to forage over broad areas, and to consume prey that forage over various ranges. As a result, these large and mobile fish reflect an average degree of contamination from across a broad area. This has been observed for several indicator species in the Bay, including striped bass, white sturgeon, and white croaker, and suggests that foraging area may be a significant source of uncertainty in bioaccumulation models. Additionally, contaminant concentrations in these species are likely to be relatively insensitive to local patterns in water or sediment contamination.

Methods to Quantify Home Range Scale and Biota Movement

Quantitative studies have not been performed on home range for most of the commonly modeled species that inhabit San Francisco Bay. Several methods can be employed to estimate home ranges of species of interest. These include using direct observations from telemetry studies, results from tagging or contamination studies from which home range could be estimated, or home range information for similar California species combined with knowledge of local wildlife experts. The preferred method is direct measurement and recording of movement of individual species. Estimating home range based on spatial patterns in contaminant concentrations, stable isotope ratios, or other tissue measurements is a secondary alternative. For example, rough estimates of home range can be developed based on the spatial association between fish and sediment contamination (Burkhard 2009, Melwani et al. 2009). Alternatively, if large spatial datasets exist on contaminant patterns within individual species, spatial statistics (e.g., kriging) may be employed to help estimate home range.

Simple empirical models have previously been applied to evaluate relationships between tissue chemistry and environmental variables. Melwani et al. (2009) presented a simple statistical method for optimizing fish-sediment correlations for PCBs and organochlorine pesticides, where exposure may occur over broad spatial scales. The spatial scales identified for pairing biota and sediment observations generally corresponded to the known exposure ranges of the species examined. The limitation of simple empirical models is that they generally use linear relationships, and are most relevant for scenarios with similar conditions (e.g., food web structure, sediment/water column concentration quotients, chemical bioavailability, and diets of the organisms) (Burkhard 2009). This can be problematic in areas with strong fine-scale spatial gradients or other highly heterogeneous conditions. If an individual species shows a lack of correlation to ambient contaminant concentrations despite strong gradients in the latter, it may suggest that the contaminant exposure occurs over a wider region (for motile and migratory species) or via a different matrix (e.g., from more uniform water column concentrations despite strong spatial gradients in sediment). Nevertheless, these procedures may be useful first steps for future RMP efforts to explore appropriate species and spatial scales for mechanistic modeling. Simple empirical models can provide an indication of the range of variation that models would need to explain before embarking on extensive efforts to parameterize more complex models only to find that they can explain just a minor portion of the variation.

Recently, the Long-Term Management Strategy for the Placement of Dredged Material in the San Francisco Bay Region (LTMS) has supported extensive acoustic telemetry monitoring to characterize movement patterns of green sturgeon, Pacific herring, white sturgeon, chinook salmon, and coho salmon (Kelly et al. 2007, Stanford et al. 2009, Chapman et al. 2010a). Similarly, radiotelemetry has been performed to determine site use for American avocets and black-necked stilts (Ackerman et al. 2007), and harbor seals (Grigg 2003). Telemetry data on movement in the Bay are not available for shiner surfperch, white croaker, striped bass, or other key contaminant indicator species.

There are a range of possible approaches to address spatial scale and movement (i.e., home range) in bioaccumulation models. These include: 1) use of generic species information representative of conditions throughout a site or region, 2) use of a guild approach in which spatial variation is represented by multiple indicator species, or 3) development of site-specific model parameters for local indicator species. These different approaches embody the tradeoff between ease and accuracy in addressing scale and movement. The use of generic species information may not be adequate to represent localized conditions, while detailed dietary and movement characterization of local species is prohibitively costly in many circumstances.

To overcome issues related to spatial scale, bioaccumulation models are usually applied at a regional scale, assuming steady-state conditions and equal biota exposure to all locations (Hope 2006). Less work has been performed on the characterization of the contribution of a localized contaminated area (e.g., “hotspot”) to the contaminant exposure of mobile organisms. This type of characterization is needed to evaluate the potential impact of localized remediation activities on exposure to humans and to sensitive fish and wildlife. Prior evaluations have emphasized statistical assessments of the overall probability distribution of exposure on a large regional scale, including the application of steady-state assumptions about lifetime exposure of individual organisms (Linkov et al. 2002).

The validated bioaccumulation model equations developed in other efforts (Arnot and Gobas 2004, Gobas and Arnot 2010) could be converted from their current steady-state formulation to a time-dependent and individual-based form. Models based on individuals are appropriate to track the variable movement, consumption, and consequent exposure of individual animals, based on information on life history and migration patterns (Jaworska et al. 1997). Model runs can then generate predicted statistical distributions of exposure. A statistical distribution of home range parameters for each indicator species can be generated to account for variability and uncertainty. This distribution is estimated based on differences among individuals within a species. The shape of the distribution can be inferred based on patterns in home ranges across estuarine and nearshore marine species, such as based on ecological niche. Application of the model to several spatially explicit contaminant data sets could then be performed. Final validation would be performed against observed biota concentrations.

The appropriate temporal and spatial scales to model and monitor (and thus the appropriate species and habitats to consider) will depend on the management questions to

be answered. Local impacts of cleanup actions taken at a specific hotspot or restoration of one salt pond would call for a different modeling and monitoring strategy than would be developed for evaluating benefits of stormwater source reduction across broad regions. Likewise the need to simulate seasonal patterns in individual organisms versus population averages or other aggregates would depend on whether there is a need to manage for the overall population or the individual and seasonal life stage most at-risk. For example, aggregate methods would be acceptable to determine long-term trends in population-level results for tern eggs. In contrast, seasonal and interannual variability in individual egg concentrations could be best characterized using time-dependent individual-based modeling approaches.

Uptake into the Bay Food Web

Bioaccumulation is the net result of exposure of species to chemicals in their aquatic environment. Uptake by primary producers represents the initial, and often substantial, step increase of chemical concentrations entering the food web and the key route for exposure. This section describes the role of primary producers and relevant efforts to model their uptake.

Bioconcentration into aquatic primary producers (algae and macrophytes) primarily occurs by passive diffusion through the cell walls of tissue exposed to surface water or porewater. This initial transfer between water and algae represents the greatest step increase in biomagnification of MeHg, selenium, and chlorinated organic pollutants. For most legacy pollutants of concern in San Francisco Bay, contaminant uptake into primary producers (e.g., phytoplankton) results in concentrations several orders of magnitude higher than in water (Krabbenhof 1996, Krabbenhof and Rickert 1997).

Mechanistic models describing contaminant bioaccumulation in primary producers are parameterized with contaminant concentrations in water, rate constants for chemical uptake and elimination, and growth rates (Gobas et al. 1991, Arnot and Gobas 2004). Rapid increases in phytoplankton biomass can cause reduced tissue concentrations in plankton and higher trophic level organisms, a process referred to as biodilution (Pickhardt et al. 2002). In addition to changes in phytoplankton uptake as a result of elevated phytoplankton abundance (i.e., bioconcentration or biodilution), dissolved water column metal concentrations, microbial oxidation, and reductive dissolution have also been inferred to alter tissue concentrations (Luoma et al. 1998, Luengen et al. 2007, Luengen and Flegal 2009).

Despite the important role of phytoplankton in affecting water column contaminant concentrations, modeling efforts for trace metals and organic pollutants in the Bay have not focused on phytoplankton or other primary producers. The primary reason for this is that variability is typically observed at shorter time scales than those of interest for managing long-term trends and impacts to wildlife and humans (i.e., years to decades). On time scales of days to weeks, phytoplankton biomass can change dramatically (Cloern 1991, 1996). Nevertheless, long-term trends also occur in plankton dynamics, potentially resulting in changes in water column concentrations and food web biomagnification. In

San Pablo Bay, South Bay, and Central Bay, phytoplankton populations have reported to have increased in the last decade due to increasing transparency and heightened predation on bivalves, reducing the consequent rate of filter feeding (Cloern et al. 2007, Cloern et al. 2010). The effect of these trends has not been quantified for food web uptake of water column-associated contaminants, such as selenium and waterborne MeHg.

Mechanisms of Contaminant Uptake and Elimination

Bioaccumulation in an organism can be modeled by quantifying the mechanisms governing the uptake and loss of contaminants. The primary processes leading to uptake of contaminants in animals are diet and respiration, while chemicals can be eliminated via excretion and egestion, metabolism, growth dilution, and reproduction.

Once incorporated into phytoplankton, many legacy contaminants continue to biomagnify up the food web. For example, MeHg increases approximately three-fold with each additional trophic level. Species at high trophic positions in the aquatic food web, such as predatory fish, ultimately attain concentrations that are approximately a million times higher than concentrations in water (Watras et al. 1994).

The rate of food web biomagnification depends on a combination of uptake and loss processes occurring within each organism. A variety of mechanistic models have been developed to describe contaminant bioaccumulation at the individual scale in aquatic animals (Norstrom et al. 1976, Thomann 1981, Thomann et al. 1992, Gobas 1993, Trudel and Rasmussen 2001, Connolly and Glaser 2002, Arnot and Gobas 2004, Trudel and Rasmussen 2006, Barber 2008, Gobas and Arnot 2010). As reviewed in Barber (2008), these models generally describe net accumulation of body weight (W) and contaminant mass (C):

$$dW/dt = D - E - R - EX - SDA - S$$

$$dC/dt = F + G - T - S$$

In essence, these models characterize change in an animal's weight as a function of energy uptake in the diet (D) and energy loss due to fecal egestion (E), respiration (R), urinary excretion (EX), and specific dynamic action (SDA; i.e., the additional respiratory expenditure required to assimilate food). Simultaneously, contaminant body burden (C) reflects net contaminant exchange across the gills (G) and the digestive tract (F, which includes both uptake from food and elimination via feces), and loss due to biotransformation (T). Finally, both weight and contaminant burden can be reduced periodically by spawning or other reproductive activity (S). Contaminant concentration at a given time point may then be portrayed as C/W.

At a finer level of detail, modeling and experimental studies have characterized specific uptake processes, such as the mechanisms of intestinal absorption. Empirical studies have demonstrated that contaminants are primarily accumulated in fish by direct uptake of contaminated water through the gills and absorption from contaminated prey through the

gastrointestinal tract (Gobas 1993, Gobas et al. 1993a). The remainder of this section reviews dietary uptake and elimination processes that are of particular importance in modeling bioaccumulation, and relevant covariates that should be considered in future modeling efforts.

Dietary Uptake

Dietary uptake is the predominant exposure pathway for most bioaccumulative contaminants (Pickhardt et al. 2006). Subsequently, trophic transfer is the process that results in the increased exposure of upper trophic level organisms. Models have shown that fish in contaminated water bodies attain elevated burdens of persistent contaminants largely through dietary uptake, rather than waterborne exposure (Rasmussen et al. 1990, Pickhardt et al. 2006, Trudel and Rasmussen 2006).

For fish, dietary uptake rate has been considered a significant source of uncertainty in contaminant mass balance models. Food consumption rates (grams per gram per day) can vary two to five-fold among species, potentially causing differences in contaminant bioaccumulation (Trudel et al. 2000, Trudel and Rasmussen 2001). Uncertainty associated with the estimation of fish feeding rates is typically on the order of 15–25% (Trudel et al. 2000). However, the uncertainty in uptake should also be examined with regard to elimination rates. If variability in uptake and elimination rates is observed to be similar, these parameters are unlikely to be strong drivers of model performance (Frank Gobas, Simon Fraser University, 2011, pers. comm.)

Dietary parameters used in mechanistic models have often been assumed to be constant through time (Gobas 1993, Arnot and Gobas 2004). Some authors have questioned the validity of this assumption when modeling over an extended period of time (e.g., a year), because dietary consumption rates, growth rates, and elimination rates vary as a function of body size, temperature, and life stage (Trudel and Rasmussen 1997, Barber 2008). One method of addressing this is to model bioaccumulation using time-dependent models which characterize contaminant uptake and loss processes on daily or monthly time-steps (Trudel and Rasmussen 2006, Barber 2008, Greenfield et al. 2009b).

Elimination Processes

Metabolism, excretion, egestion, growth dilution, and reproduction are the primary processes that govern the loss of contaminants in organisms. Many organic chemicals are metabolized very slowly relative to other elimination parameters; therefore, mechanistic models often apply the simplifying assumption of zero metabolism. When examined in sensitivity analysis, metabolism has had generally low influence, unless the rate of metabolism in a particular species is very high relative to elimination rates (Gobas and Wilcockson 2002, Gobas and Arnot 2005). Therefore, data on metabolism is not a high priority information need for organics. The influence of metabolism on the bioaccumulation of MeHg may be of somewhat greater significance, although studies often report only net depuration loss from organisms (Headon et al. 1996, Wang and

Wong 2003), not distinguishing metabolism (i.e., demethylation) from other elimination processes.

Excretion (including gill ventilation and exhalation of air), egestion, and growth are important processes that require parameterization in bioaccumulation models. Elimination of contaminants in birds and harbor seals is generally due to the same processes as fish, except that excretion occurs by respiratory exhalation of air rather than gill ventilation of water. Additionally, molting has been shown to affect contaminant burdens in seals and birds. Harbor seals are known to fast and molt at particular times of the year, which can reduce their PCB and Hg burden (Hoenicke et al. 2007). Similarly, feather growth and molting can lower blood MeHg concentrations in birds (Evers et al. 2005).

Females can also depurate contaminants during reproduction. Fish transfer contaminants to their gonads for egg production and spawning, female birds transfer contaminants into their eggs and offspring, and female harbor seals can transfer contaminants into their offspring and milk. To model these processes, food web models have assumed that the contaminant is evenly distributed throughout the organism's tissues (Gobas and Arnot 2010). This is a relatively safe assumption for lipophilic contaminants that partition strongly to lipid tissues that are distributed throughout the organism. The main impact that producing eggs/offspring/milk has on the female is the change in body mass that is associated with the process. Therefore, although reproduction may cause a decline of contaminant concentration in females, this is compensated by intake of additional contaminants with the diet during development of eggs or offspring.

Models of MeHg dynamics in fish have assumed that the elimination rate due to spawning (egg and sperm production) was small relative to the MeHg loss from other elimination processes (Trudel and Rasmussen 2006). Furthermore, such models have not observed reproductive elimination to have significant effect on model results. This may not be the case for modeling of organic contaminants, however (Vander Zanden and Rasmussen 1996).

Growth

In addition to the processes described above, changes in growth rate with size and activity level influence contaminant concentrations. Growth dilution of contaminants can occur as a result of the increased tissue biomass relative to uptake. This pseudo-elimination process is strongly related to growth rate in organism regardless of the contaminant accumulation (Simoneau et al. 2005). Increased rates of foraging or shifts in foraging mode as well the amount of available food will often induce changes in growth rate.

Growth rates can thus be a sensitive parameter in modeling bioaccumulation. This is particularly the case in species with relatively high growth rates, like phytoplankton, or those that disproportionately increase in size during their lifespan (plankton and fish). Growth rate effects may require specific focus in future models given the recent data

from Cloern et al. (2010) that showed significant changes in phytoplankton bloom dynamics in San Pablo, Central, and South bays. Since the late 1990s, larger phytoplankton blooms have occurred in the Bay and a progressive increase in the annual minimum chlorophyll concentration. Since phytoplankton cells can absorb and concentrate dissolved substances, including toxic contaminants, these observations could have the reverberating effect of reducing contaminant flux to the sediment due to scavenging by newly abundant phytoplankton. The quantitative information on phytoplankton bloom dynamics collected by the USGS are a valuable source of information for modeling the phytoplankton growth parameters in future models. Growth rates for certain fish and bird species considered in Section 3 of this paper are also available. The growth rate of phytoplankton as well as fish and mammals were sensitive parameters in the PCB bioaccumulation model of Gobas and Arnot (2005).

Although the specifics of the relative importance of these various uptake and elimination processes to net bioaccumulation will differ among the contaminants and species considered, a general conceptual framework incorporating all the known environmental and dietary exposures for a species with its life history and metabolic characteristics can serve as the basis for a mechanistic model for all species and contaminants.

Ecological Attributes

Ecological attributes, such as dietary mode, trophic position, habitat association, and body size, can strongly influence chemical uptake in biota. These factors must be considered in efforts to model bioaccumulation at both the organism and population level. In this section, the importance of these factors in modeling bioaccumulation is discussed.

Life History, Dietary Mode and Trophic Position

Life history characteristics of individual species can influence spatial and temporal patterns in bioaccumulation. Types of life history events that may be important to consider in food web modeling include: changes in dietary mode with size or age; shifts in consumption patterns and lipid with temperature or season; sexual dimorphism in physiologic parameters (e.g., growth or energy); and differences in movement as a function of age, size, or gender.

Diets of most aquatic species are known to change with size and age (Petersen and Chen 1998, Vander Zanden et al. 1998), as well as seasonally depending on available food (Nakashima and Leggett 1978, Storck 1986). These shifts in diet can cause changes in concentrations of MeHg, PCBs, and other pollutants that are associated with trophic position (Kiriluk et al. 1995, Vander Zanden and Rasmussen 1996, Kidd et al. 1998). Dietary mode and trophic position strongly influence bioaccumulation. Dietary preferences in bioaccumulation models are often inferred from stomach contents data. Additionally, stable isotope ratios of carbon, nitrogen, and sulfur can be used to categorize an organism's trophic position and carbon source (Stewart et al. 2004, Stewart et al. 2008). Stable isotopes reflect the assimilated food, rather than simply the material

ingested, and thus provides integrated dietary information over a long time period (Peterson and Fry 1987). This offers advantages over dietary studies that can be time-consuming, show highly variable results, and offer a single snapshot of intake that may not reflect the long-term exposure conditions. However, isotopic markers can also be difficult to interpret, especially as dissimilar species can show very similar C/N ratios. Recently, Hg stable isotopes in forage fish from the Bay have revealed mine-impacted sediments to be a likely source of contaminant burden (Gehrke et al. 2011b). However, similar data have yet to be measured in other Bay indicators. This kind of information could be sought to better characterize dietary exposure pathways.

Trophic position strongly drives the degree of bioaccumulation among species in the food web, both within and among species. Due to contaminant biomagnification, higher trophic position organisms tend to exhibit higher body burdens of MeHg, PCBs, and other bioaccumulative pollutants (Vander Zanden and Rasmussen 1996, Kidd et al. 1998). Furthermore, contaminants tend to increase significantly in organism tissues between trophic positions but much less between organisms that occupy a similar trophic level. It is thus possible to “lump” species of similar trophic guilds. For the 16 target indicator species considered in this paper, trophic information has been summarized to distinguish species based on and dietary mode and trophic position (Table 2). Relevant trophic guilds include phytoplankton and algae, zooplankton, filter feeding invertebrates, benthic detritivores, benthic and pelagic fish, fish eating birds and mammals. In addition to the information detailed in Section 2, consideration of guilds may aid in selecting appropriate species for future models.

Modeling is more feasible for species known to have more restricted dietary modes or sedentary life histories, as this reduces uncertainties about spatial variability in exposure. Shiner surfperch potentially have great value as a finer-scale indicator of patterns in contamination on the margins. Lower trophic level prey items for indicator species of concern will often have limited mobility and should also be considered for inclusion in bioaccumulation models. Largely sessile benthic invertebrates are excellent indicators of fine-scale spatial patterns in contaminant concentrations. Small fish, which have been sampled extensively in the last few years by the RMP, are also valuable indicators of finer-scale spatial and temporal patterns due to their limited ranges (Greenfield and Jahn 2010). These more localized species are important prey items consumed by fish and wildlife indicator species and can help link water and sediment contamination in margins to bioaccumulation at a regional scale.

Benthic Versus Pelagic Habitat

In San Francisco Bay, contaminants are often considered to be in dynamic flux between sediment and the water column (Davis 2004). Therefore, bioaccumulation of contaminants from sediment and the water column are largely interdependent. Nevertheless, models that quantify the biological uptake of contaminants from bed sediments (e.g., via deposit feeding) versus the water column (e.g., via respiration, and filter feeding) separately provide a first order approximation of their relative importance to biota exposure. Mechanistic bioaccumulation models have previously been used to

examine the relative importance of these different exposure compartments (Gobas 1993, Greenfield et al. 2007).

Potential pathways of exposure from the sediment include direct ingestion of sediment, respiratory exposure to porewater contaminants (modeled assuming equilibrium partitioning), and food web trophic transfer from prey organisms that consume or respire sediment associated contaminants (Arnot and Gobas 2004). Exposure to water column contaminants occurs via respiratory uptake (transpiration in phytoplankton), consumption of suspended particles and associated contaminants via filter feeding, and food web trophic transfer from prey organisms employing these pathways (Arnot and Gobas 2004).

Using a mechanistic food web bioaccumulation model developed for San Francisco Bay (Arnot and Gobas 2004, Gobas and Arnot 2010), Greenfield et al. (2007) contrasted the contribution of sediment versus water column to biotic concentrations of legacy organic pollutants. This was achieved by separately changing sediment and water column concentrations from their empirical observed values to zero, and comparing the resulting change in modeled tissue concentrations. Model results for p,p'-DDE indicated that at relatively high water column concentrations, biota exposure primarily results from the water column, resulting in very high bioaccumulation factors (BAF; here defined as [wet weight concentration in biota/dry weight concentration in sediment]). The model also illustrated the importance of contaminant partitioning properties (e.g., K_{ow}) and biota life history for contaminant uptake from sediments versus water. For example, the relative sediment contribution of p,p'-DDE was lower than for PCB 118, due to the lower K_{ow} (and consequent lower sediment association). For p,p'-DDE, water column exposure contributed 50-100% of modeled body burden in all fish species and benthic invertebrate species (Figure 6). In contrast, PCB 118 exhibited a generally higher overall sediment contribution to biota exposure (Figure 7), due to the higher ratio of sediment/water column concentration, stemming from the higher K_{ow} of PCB 118. Additionally, a greater contribution of legacy sediment sources, in comparison to watershed loading, may play a role. These results are consistent with the conceptual model that sediment versus water column disequilibrium is one of the factors that affects empirical observed BAFs (Burkhard et al. 2003).

Comparing model results across species illustrates the importance of biota life history (i.e., dietary and respiratory activity, Burkhard et al. 2003) for sediment versus water column contribution. Water column sources were most important for those species that prey predominantly on plankton, including *Mysis* sp., *Crangon* sp., and jacksmelt, while sediment exposure was greatest for the sediment dwelling and annelid, *Neanthes succinea* (Figures 6 and 7) (Greenfield et al. 2007). Scenarios to examine water versus sediment exposure to PCBs in four San Francisco Bay locations by Gobas and Wilcockson (2002) indicated that sediments contribute largely to contaminant burdens of higher chlorinated PCBs in benthic dwelling species, such as white croaker. Recently, Gehrke et al. (2011b) also observed strong correlations between sediment and forage fish Hg isotope values ($r^2 = 0.83$), in nearshore habitats Bay-wide. In combination, these findings suggest that both sediment and water can both be important sources of contaminant exposure to the nearshore aquatic food web. The water versus sediment association of a species will

influence measured and modeled uptake from different pollutant pools and can be assessed through a mechanistic modeling framework.

A model simulating food web uptake of MeHg could be used to evaluate the role of legacy sediments versus watershed loading and other sources of this pollutant. Yee et al. (2011) observed in-situ methylation in the sediment and demethylation in the sediment and water column to be primary drivers of the MeHg mass balance in the Bay. This is consistent with the empirical linkage between sediment and forage fish Hg stable isotopes (Gehrke et al. 2011b). These findings in combination suggest the legacy sediment to be an important pool of Hg for biota MeHg exposure. In contrast, a mechanistic fate and food web model application to the nearshore coastal Bay of Fundy observed water column exposure to be largely decoupled from the sediments, suggesting the legacy sediment pool to be relatively unimportant for pelagic food webs (Sunderland et al. 2010). Resolving the relative role of sediments versus the water column in the MeHg uptake in the Bay margins would aid in forecasting the relative benefits of loads reduction versus factors that affect within-Bay methylation or demethylation. A MeHg bioaccumulation model coupled with a site-specific model of loading and speciation could help to define the uncertainties and forecast impacts of different management actions.

Another contaminant where water column dietary exposure can predominate is selenium. Selenium is known to accumulate to levels of concern in filter-feeding benthic bivalves (Stewart et al. 2004). Selenium is highly bioavailable to benthic invertebrates from ingested particles (Lee et al. 2006). A mechanistic selenium fate and transport model for North San Francisco Bay has been developed in support of development of a numeric TMDL target (Baginska 2011). The modeling framework includes a numeric estuary component (salinity, total suspended material, chlorophyll a, dissolved and particulate selenium) and a dynamic bioaccumulation model (referred to as DYMBAM) to simulate first order transformations and biological uptake processes in the North Bay. Future efforts to model selenium bioaccumulation may seek to employ the framework developed for the North Bay selenium TMDL to answer management questions of broader interest.

Lipid Content

Organic contaminants bioaccumulate due to fugacity (equilibrium partitioning) gradients created in the gut of predator species that favor high absorption of lipids and associated contaminants (Gobas et al. 1993b). Therefore, hydrophobic organic chemicals are predominantly associated with the lipids in an organism's tissue (Niimi 1983, Kidd et al. 1998). Not surprisingly, lipid content and lipid absorption efficiency were documented as sensitive state variables in the previous bioaccumulation model developed for PCBs in the Bay (Gobas and Arnot 2005, Gobas and Arnot 2010). Empirical sport fish data indicate lipid content to be associated with variation in organic pollutant concentrations among species (Davis et al. 2002), as well as seasonal and inter-annual variation within species (Greenfield et al. 2005). Of the species being considered in this report, white croaker, shiner surfperch, and harbor seal are most likely to exhibit variation in organic contaminant burdens associated with variable lipid content. This is largely due to the

relatively high lipid content and relatively high variation in lipid among individuals. At the highest extreme, the whole body lipid content of harbor seals in the Bay has been shown to vary between 36 and 50%, resulting in some of the highest concentrations of organics contaminants observed in Bay wildlife (Lydersen et al. 2002).

Age, Body Size, and Sex

Age and size are covariates that can influence bioaccumulation. Many empirical studies indicate increases in MeHg and organic pollutants with fish size and age (Harding et al. 1997, Davis et al. 2002, Johnston et al. 2002, Greenfield and Jahn 2010), although exceptions are found for organic pollutants (Larsson et al. 1993). Concentrations of these contaminants in biota increase with age because of the relatively slow rate of elimination relative to uptake. Mechanisms for the increase can include higher trophic position (i.e., consumption of more contaminated prey) and increased metabolic costs with activity for larger organisms (Trudel and Rasmussen 2006). Due to these factors, long-lived predatory fish species tend to accumulate high MeHg concentrations, and this is a primary driver of spatial variation in MeHg contamination at the top of the food chain across the state (Davis et al. 2012). To evaluate this, models can be parameterized for different age classes (Connolly and Glaser 2002, Gobas and Arnot 2010).

Studies of MeHg in fish have observed statistically significant relationships between size and concentrations (Greenfield et al. 2005). Many species with elevated concentrations illustrate linear relationships between size and MeHg. Some species, such as striped bass, have indicated a polynomial relationship in very large adults (> 500 mm). This may be due to the fact that fish growth rates subside in older fish, while consumption rates remain the same. Similarly, the size of small fish can vary greatly seasonally due to considerable growth throughout the year. Small prey fish, such as shiner surfperch, Mississippi silverside, topsmelt, or longjaw mudsucker, typically grow rapidly during the summer doubling their biomass or concentrations. Total body burden of contaminants in individual fish thus increase substantially during summer months (Hobson and Chess 1986).

In contrast, modeling of bioaccumulation in bird species does not generally need to focus on size effects. This is largely due to the fact that birds normally reach full size during the first year of life, and as such do not exhibit perpetual growth during their lifetime like many fish species.

Sexual dimorphism, including differences in size and reproductive output (MacInnis and Corkum 2000), can be responsible for large differences in contaminant concentrations. In finfish, female fish can release a substantial portion of their contaminant burden through the transfer of energy and lipids to egg production and spawning, thereby incurring a substantially lower contaminant burden than male fish of the same size (Niimi 1983). Such a life history trait is likely most influential in species that spawn multiple times within their lifespan (Larsson et al. 1993), and may explain some of the high degree of variability in contaminant concentrations observed in regional bioaccumulation studies. Ng and Gray (2009) modified the bioaccumulation model of Arnot and Gobas (Arnot and

Gobas 2004) that integrated life history scenarios to examine changes in PCBs through the lifespan of the round goby (*Apollonia melanostoma*). The authors observed that lipid utilization associated with reproductive output can substantially affect bioaccumulation. Rather than simply depurating PCBs through egg production, some females exhibited greater average lipid content, presumably as a result of higher consumption rates. Therefore, some females had higher tissue burdens than were expected. Though differences in contaminant burdens between sexes can be substantial for PCBs, DDTs and other organic pollutants, loss of MeHg by females is relatively unimportant due to the relatively low concentrations in reproductive tissues (Trudel and Rasmussen 2001, 2006).

These biological covariates are often important to understanding differences between individuals or populations in contaminant bioaccumulation. In some cases, the subpopulations of greatest concern have certain characteristics (e.g. age, gender), requiring explicit consideration of the covariates. Interpretation of monitoring data and modeling efforts thus needs to address or at least acknowledge these differences.

Abiotic Covariates

Similar to the biological covariates, there are abiotic variables that may affect the uptake or loss of contaminants in biota. In this section, three abiotic covariates that are essential components in modeling sediment-associated contaminants are discussed: the influence of bioavailability, organic carbon content, and redox conditions.

Bioavailability

Contaminants present in aquatic media (sediment or water) are never completely available for uptake by organisms. Some proportion of the total concentration is sequestered in dissolved and/or particulate organic and inorganic matter. The concentration of the chemical that can be absorbed by the organism from the water or sediment is referred to as the “bioavailable” fraction. Describing bioavailability is one of the most important components of bioaccumulation models (Gobas 1993). Bioavailability is principally estimated by the ratio of the dissolved: total concentration using chemical partition coefficients between the sorbed and dissolved state. Generally, highly hydrophobic chemicals (e.g. PCBs) have high partition coefficients, and thus exhibit smaller dissolved fractions, reducing their bioavailability.

The bioavailability of many chemicals can be generally estimated based on the interaction of the chemical with particulate organic matter. Organic matter and sulfide concentrations are the most important factors controlling the bioavailability of MeHg in surface sediment (Mason and Lawrence 1999, Conaway et al. 2003, Hammerschmidt and Fitzgerald 2004). Sediment redox conditions (discussed further below) have also been shown to play a significant role in Hg bioavailability due to influence on the nature and concentration of the binding phases in the sediment. Furthermore, sediment resuspension can induce a change in sediment redox status, which can be an important factor in controlling Hg methylation in sediment, and can also affect MeHg bioavailability.

Total Organic Carbon (TOC)

Organic carbon (OC) is the amount of material derived from decaying matter, bacterial growth, and metabolic activities of organisms. OC in water and sediment comprise both particulate and dissolved forms. Particulate organic carbon refers to the proportion of organic carbon sorbed to particles in water, while dissolved organic matter form is freely dissolved form, not attached to particles. Environments enriched with organic carbon tend to accumulate higher concentrations of contaminants. For this reason, it is imperative that OC information to be incorporated into bioaccumulation models of contaminants such as PCBs, MeHg, and others. Sediment organic content plays an important role in Hg methylation as it controls the distribution of Hg between particulate and dissolved phases, determining Hg bioavailability to sulfate-reducing bacteria.

Particulate organic carbon content was identified as a key abiotic state variable in the PCB bioaccumulation model of Gobas and Arnot (2005). POC contributes to the amount of chemical mass in water available for uptake. The model results indicated that a reduction in POC content in water resulted in higher concentration of chemical in freely dissolved form. This had the effect of increasing the mass of chemical that could be directly absorbed from water by phytoplankton, invertebrates, and fish and subsequently biomagnified to higher trophic levels. Organic carbon content in certain organisms can also have a strong influence on model outcomes. Some models also consider different forms of carbon with greater affinities for hydrophobic organics, e.g., “black” or “soot” carbon which can affect solubility and thus bioavailability compared to models considering all organic forms of carbon together.

Redox Potential

Redox potential is another abiotic parameter that can be an important factor in modeling bioaccumulation. Redox potential provides an estimate of whether sediment is under aerobic or anaerobic conditions, and whether certain elements are in their reduced or oxidized forms, with important implications for their solubility and mobility. Redox potential is especially important with regard to MeHg (Mason and Lawrence 1999). Sulfate reducing bacteria are thought to drive MeHg production in coastal sediment. These microorganisms primarily occur at the interface between oxygenated and anoxic conditions in sediment (Gilmour et al. 1992). Therefore, relationships between redox potential and MeHg in sediment have frequently been demonstrated (Mason and Lawrence 1999). Redox potential may also be important for modeling of sediment-associated organic contaminants due to changes in speciation of chemicals as a function of oxidation state.

5. Summary and Recommendations

The purpose of this report is to provide a conceptual foundation for a path forward for RMP studies of bioaccumulation, including modeling and monitoring, in support of effective and efficient efforts to reduce concentrations of contaminants in the Bay food web. Biotic exposure and risk are ultimately the regulatory endpoints of concern for bioaccumulative contaminants, as reflected in the tissue targets that are the centerpieces of the Hg and PCBs TMDLs. RMP fate models must be developed with a sharp focus on understanding and predicting biotic exposure. Models of fate in water and sediment must be efficiently coupled with bioaccumulation modeling. Water and sediment fate models should provide the input data needed to drive the bioaccumulation models; anything less would be insufficient, and anything more should be carefully scrutinized as to whether it is really necessary.

A conceptual understanding of the patterns revealed in food web monitoring to date are a strong foundation for future modeling efforts. The documented patterns have been described in this report. Critical aspects of these patterns that have a major bearing on consideration of the next steps in bioaccumulation studies are briefly reiterated here. This summary focuses on MeHg and PCBs, the two contaminants of greatest concern in the Bay food web.

- Long-term monitoring of sport fish, key indicator species in TMDLs, has indicated no trend in MeHg concentrations since the early 1970s and no trend in concentrations of PCBs and other organics since 1994.
- In contrast to the sport fish, concentrations of organics have declined significantly in bivalves in the open Bay based on time series covering 1980 to the present.
- MeHg in the food web varies spatially 1) at a regional scale, with highest concentrations in the South Bay, and 2) with additional finer-scale variation at a local scale that is not clearly associated with source categories
- PCBs and other organics also vary spatially 1) at a regional scale, with highest concentrations in the Central Bay, and 2) with additional finer-scale variation at a local scale that is very distinct and, in the case of PCBs, clearly associated with sediment contamination from past industrial and military activities.
- MeHg and PCB concentrations in Bay sport fish are exceptionally high on a statewide and national scale, suggesting an unusual degree of contamination or fate processes that make the Bay unusually slow to recover from contamination.
- Recent extensive small fish monitoring has revealed a much higher degree of accumulation for organics than would be expected, with concentrations equaling or exceeding those in sport fish species at higher trophic levels.
- Food web uptake of MeHg, PCBs, and other organics appears to be driven by sediment contamination, as indicated by spatial correlations and isotope tracer studies.

For PCBs and the organics, these observations support a hypothesis that the exposure of small fish, sport fish, and much of the Bay food web is derived from a “bathtub ring” of sediment contamination along the margins of the Bay. Contaminant concentrations in these low energy environments appear to be very stable, in contrast to declines that are

occurring in waters of the open Bay. If this hypothesis is correct, then fate modeling efforts should be focused on forecasting recovery in these margin habitats, particularly in relation to species of management interest in the Bay.

For MeHg, spatial and temporal patterns in food web uptake appear to be driven by variation in biogeochemistry. Developing a capacity to predict future trends and the effect of management actions will depend on understanding and modeling the factors that drive MeHg production and degradation, and subsequent uptake and bioaccumulation in the food web.

These observations and hypotheses support the following recommendations.

- As mentioned above, bioaccumulation is the ultimate endpoint of concern for many of the highest priority contaminants in the Bay, and modeling efforts should focus on the goal of forecasting changes in bioaccumulation in response to management actions. Thoughtful articulation of the management decisions to be made based on bioaccumulation data is essential to successful model development. In addition, accurate forecasts of future management actions are important in designing appropriate models and monitoring. The appropriate temporal and spatial scales to model and monitor (and thus the appropriate species and habitats to consider) will depend on the management questions to be answered.
- After the management questions have been articulated, the next key step will be developing a comprehensive plan for creating the linked models for fate in water and sediment and for bioaccumulation in species of particular management interest. Understanding the coupling of these different model components is critical to the efficient and effective design of the water and sediment fate model.
- Patterns in bioaccumulation support the notion that a focus on fate and uptake in the Bay margins is appropriate. Mechanistic bioaccumulation modeling has been very limited to date on the margins. The modeling platforms and processes used for open water habitats (Pickhardt et al. 2006, Greenfield et al. 2009a, Gobas and Arnot 2010) should be largely adaptable to modeling the margins, and could be adapted to time-dependent and individual-based applications as well. As needed to address management decisions requiring understanding of finer scale processes, the models could also be adapted to accurately represent the different habitat types present on the margins (open water; sloughs, channels, and mudflats; tidal marsh; and salt pond).
- Sufficient empirical data on contaminant concentrations in sediment, biota, and perhaps water will be needed to develop and validate fate models on the margins. Indicator species with high site fidelity will be best suited for these studies. Shiner surfperch appear to be valuable in this regard: they are the most important sport fish indicator species for organics, and also have shown great potential as an indicator of relatively fine-scale spatial variation. Small fish may also be valuable indicators on the margins. Along with the spatial dimensions of exposure, information on the diets of the indicator species will be needed. This information will help to identify lower

trophic level indicators with even greater site fidelity that may also be useful in monitoring and modeling fine-scale processes on the margins. Other key forms of empirical information that may be needed include data on long-term bioaccumulation trends, seasonal variation in bioaccumulation in some cases, and life history information of indicator species (such as movement, foraging range, and physiological characteristics such as growth rate).

- Simple empirical correlational bioaccumulation models represent a low cost technique that can be a very useful first step in evaluating whether mechanistic modeling is warranted. Correlation methods can be used to establish relationships between environmental variables (e.g., sediment chemistry, chlorophyll, DOC) and biota concentrations using existing data. They can also be used to evaluate appropriate spatial scales for associating abiotic and biotic data. However, they are crude, limited in their capability to link sources and forecasting, and may only work for a subset of variables and species. Significant linear relationships are most likely to be observed in chemicals and biota with relatively large sample sizes and where a wide range in values is present in both environmental (independent) variables and tissue chemistry (dependent variables) (Burkhard 2009). Even where significant correlations are not observed, such explorations can describe the magnitude of variation that would need to be explained by any more complex models, to identify whether (and which) available models are even capable of reproducing such variation before embarking on efforts to parameterize more complex models, or to identify additional factors to incorporate in models that could account for the observed variations. A simple empirical correlation model for PCBs in small fish and sediment revealed a significant relationship, even using sediment data that were not closely matched in time and space to the fish data. A similar analysis for MeHg did not show a correlation. Future field studies with PCBs should be designed to confirm and clarify this relationship. Future studies of MeHg bioaccumulation should endeavor to explain the lack of correlation between concentrations in small fish and concentrations in sediment. Future studies of other bioaccumulative contaminants of concern should gather similar data to allow evaluation of simple correlations with environmental gradients.
- As modeling and empirical data gathering proceed, it may be valuable to obtain a more precise understanding of where key indicator species, such as shiner surfperch, silversides, topsmelt, and northern anchovy, are acquiring their heavy contaminant burdens. A variety of methods are available for quantifying movement of fish, including telemetry studies, tagging studies, and stable isotopes (Griffin and Valiela 2001, Lowe and Bray 2006, Ackerman et al. 2007). The potential application of these methods should be evaluated in consultation with fish biologists. With a solid understanding of habitat use, development of bioaccumulation models for organics will be relatively straightforward, applying the framework already developed for the Bay by Gobas and coworkers. Obtaining this information will also likely be important for understanding and modeling MeHg bioaccumulation, although the lack of correlation between environmental and biota MeHg concentrations suggest other factors that may be more critical to resolve.

- A MeHg bioaccumulation model for the Bay has yet to be developed. Development of a mechanistic bioaccumulation model that could address the dynamic processes that drive MeHg uptake in the Bay food web (Figure 8) should be considered. The model architecture for a MeHg model could be based upon the Gobas and Arnot (2005) bioaccumulation model that has been successfully applied to PCBs, but substantial revision of the model would be required to model uptake and accumulation of a non-lipophilic contaminant. An adaptation of the PCB model would need to account for THg and MeHg sediment depth profiles (Yee et al. 2010), local loading rates (Lewicki and McKee 2009, McKee et al. 2009), and spatial and temporal trends (SFEI 2010). Subsequently, a contaminant sub-model for MeHg would need to be developed, which incorporates Hg methylation and MeHg demethylation. This could be initiated using recently developed Hg speciation and mass balance approaches (Knights et al. 2009, Sunderland et al. 2010, Yee et al. 2011), graduating as needed to a more complex fate model such as that developed for the New York/New Jersey Harbor (Hydroqual 2007). The heterogeneous, dynamic, and poorly understood nature of MeHg cycling poses a formidable challenge to development of a fate and bioaccumulation model for MeHg.

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8. Tables

Table 1. Summary information on the major contaminants being considered for food web models in San Francisco Bay

Chemical Group	Largest Pathways to Bay	Primary Source of Contamination to Biota	Bioaccumulation Mechanism	Indicator Species of Concern	Spatial Complexity in Biota Concentrations
Methylmercury	• In-situ production	Sediment	Sulfhydryl groups on proteins	<ul style="list-style-type: none"> • Striped bass • Jacksmelt • Clapper Rail • Least Tern • Forster's Tern • Song Sparrow • Mississippi silverside • Long-jaw mudsucker • Topsmelt 	Elevated Bay-wide; South Bay, Oakland Harbor, and other Bay-margin hot spots (relatively high)
Selenium	<ul style="list-style-type: none"> • Delta outflow • Wastewater 	Water	Sulfhydryl groups on proteins	<ul style="list-style-type: none"> • Splittail • White sturgeon • Clams/mussels 	San Pablo and Suisun Bays (high)
PCBs	<ul style="list-style-type: none"> • Delta outflow • Urban runoff • Remobilization 	Sediment	Lipid-based	<ul style="list-style-type: none"> • White croaker • Shiner surfperch • Topsmelt • Least Tern • Forster's Tern • Caspian Tern • Double-crested Cormorant • Harbor seal 	Urban/Industrialized margins (high)
PBDEs	<ul style="list-style-type: none"> • Wastewater • Urban runoff 	Sediment	Lipid-based	<i>Same as PCBs</i>	Urban/Industrialized margins (high)
Dioxins	<ul style="list-style-type: none"> • Atmospheric dep • Urban runoff • Remobilization 	Sediment	Lipid-based	<i>Same as PCBs</i>	Urban/Industrialized margins (high)
OC Pesticides	<ul style="list-style-type: none"> • Delta outflow • Urban runoff • Remobilization 	Sediment	Lipid-based	<i>Same as PCBs</i>	No clear regional patterns, some elevated margin sites
PFCs	<ul style="list-style-type: none"> • Urban runoff • Remobilization • Landfills • Wastewater • Military facilities • Airports 	Sediment and Water	Protein-based (specificity unknown)	<ul style="list-style-type: none"> • Least Tern • Forster's Tern • Caspian Tern • Double-crested Cormorant • Harbor seal 	Lower South Bay (high)

Table 2. Dietary guild categories for indicator species of concern. Guild species were selected from the list of species examined in Section 3.

Dietary guild	Description	Guild species
Piscivore	The majority of the diet is fish. Large predatory invertebrates (e.g., cephalopods, decapod crustaceans, and echinoderms) are also consumed to some degree.	California halibut Double-crested Cormorants Forster's Tern California Least Tern Pacific harbor seal
Benthic diet <i>with piscivory</i>	Diet regularly includes a mixture of benthic invertebrates and forage fish.	Striped bass White sturgeon
Benthic and pelagic diet <i>with piscivory</i>	Diet includes a combination of benthic invertebrates, pelagic invertebrates (e.g., zooplankton, shrimp, and mysidae), and forage fish.	Longjaw mudsucker Clapper Rail
Benthic diet <i>without piscivory</i>	Diet largely composed of small benthic invertebrates, such as amphipods and other crustaceans, bivalve mollusks, and polychaete worms.	White croaker
Benthic and pelagic diet <i>without piscivory</i>	Diet includes a mixture of epibenthic and pelagic invertebrates (e.g., zooplankton, shrimp, and mysids).	Shiner surfperch
Benthic diet <i>with herbivory</i>	Largely consumes benthic invertebrates, benthic algae, and aquatic plants.	Song Sparrow
Benthic and pelagic diet <i>with herbivory</i>	Diet consists of benthic and pelagic invertebrates and plant material, including benthic algae and phytoplankton.	Topsmelt Mississippi silverside Jacksmelt
Pelagic diet with benthic herbivory	Diet includes largely pelagic invertebrates and benthic algae. This includes a substantial component of benthic algae and attached plants, likely as floating detritus.	California mussel

9. Figures

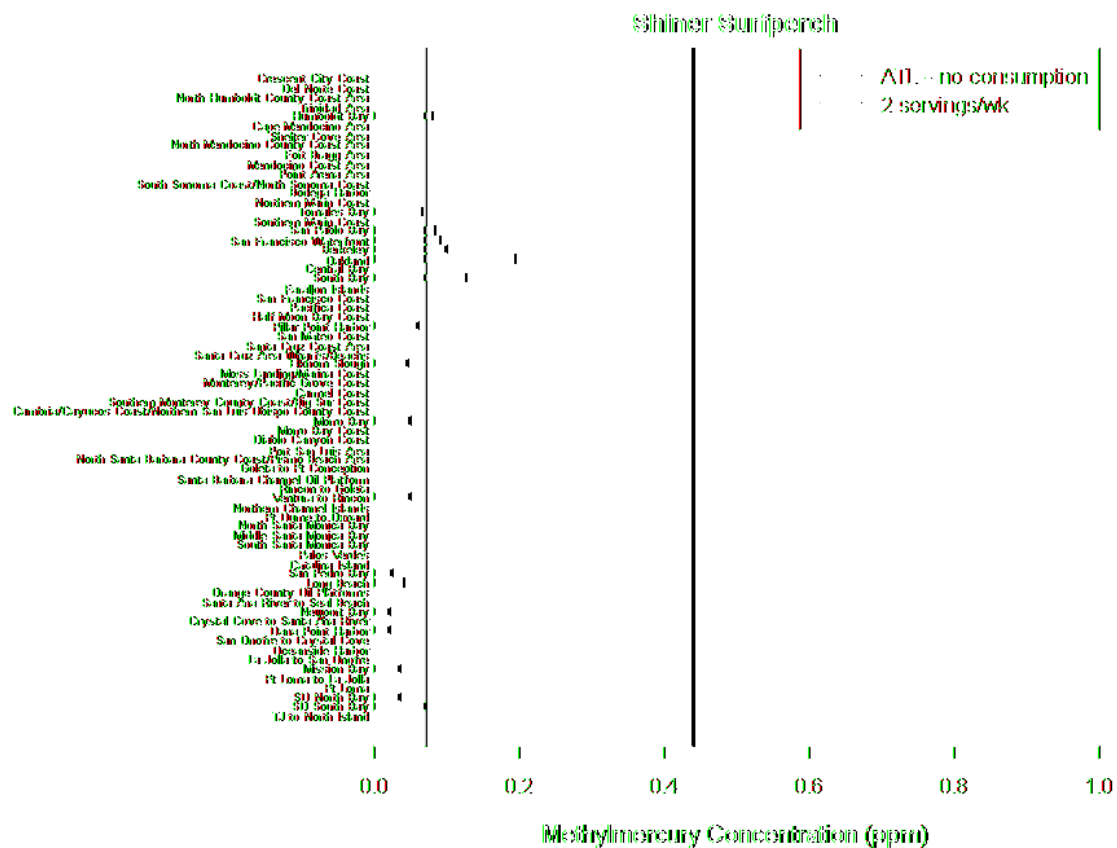


Figure 1. Mercury concentrations in shiner surfperch from locations throughout the California coast. Shiner surfperch were not collected from locations without bars. From Davis et al. (2012).



Figure 2. Mercury concentrations measured in striped bass in U.S. estuaries. From data compiled in Davis et al. (2012b)

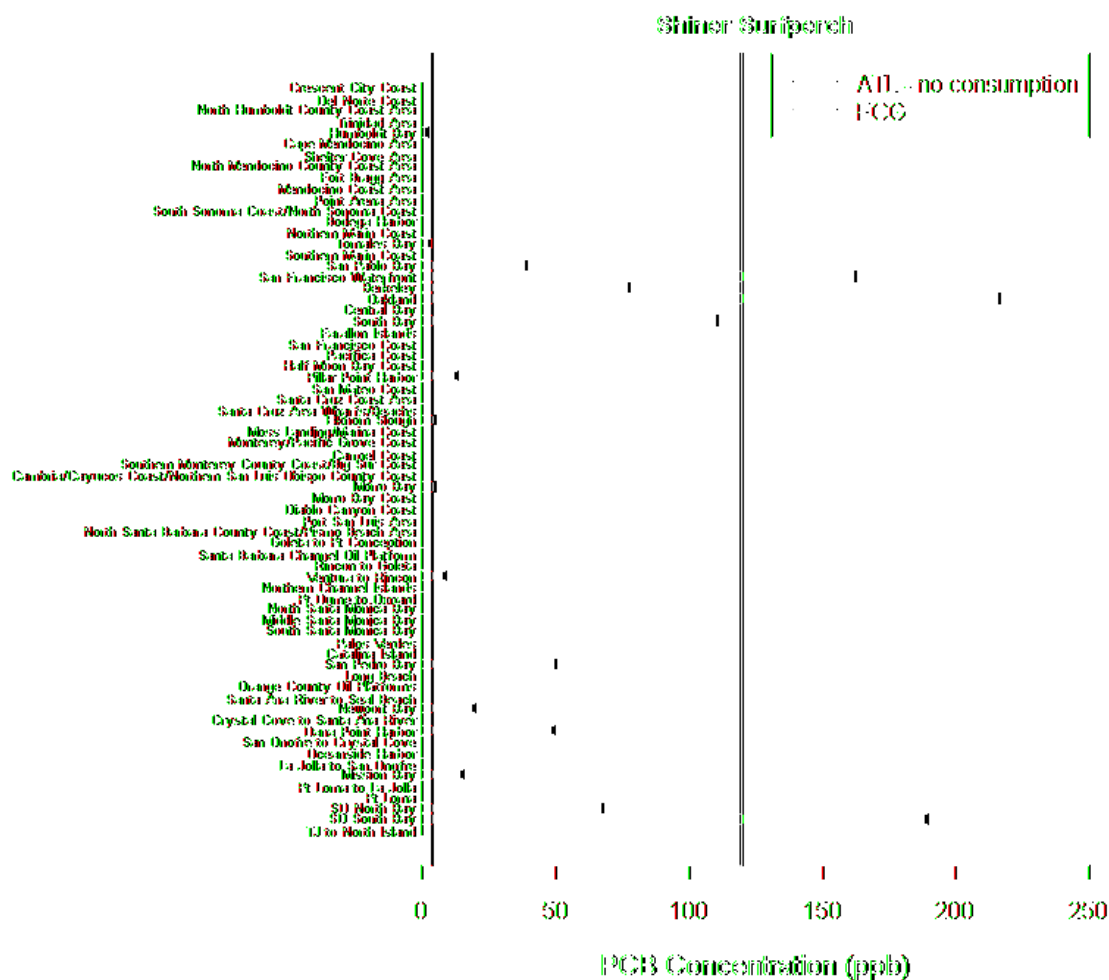


Figure 3. PCB concentrations in shiner surfperch from locations throughout the California coast. Shiner surfperch were not collected from locations without bars. From Davis et al. (2012).

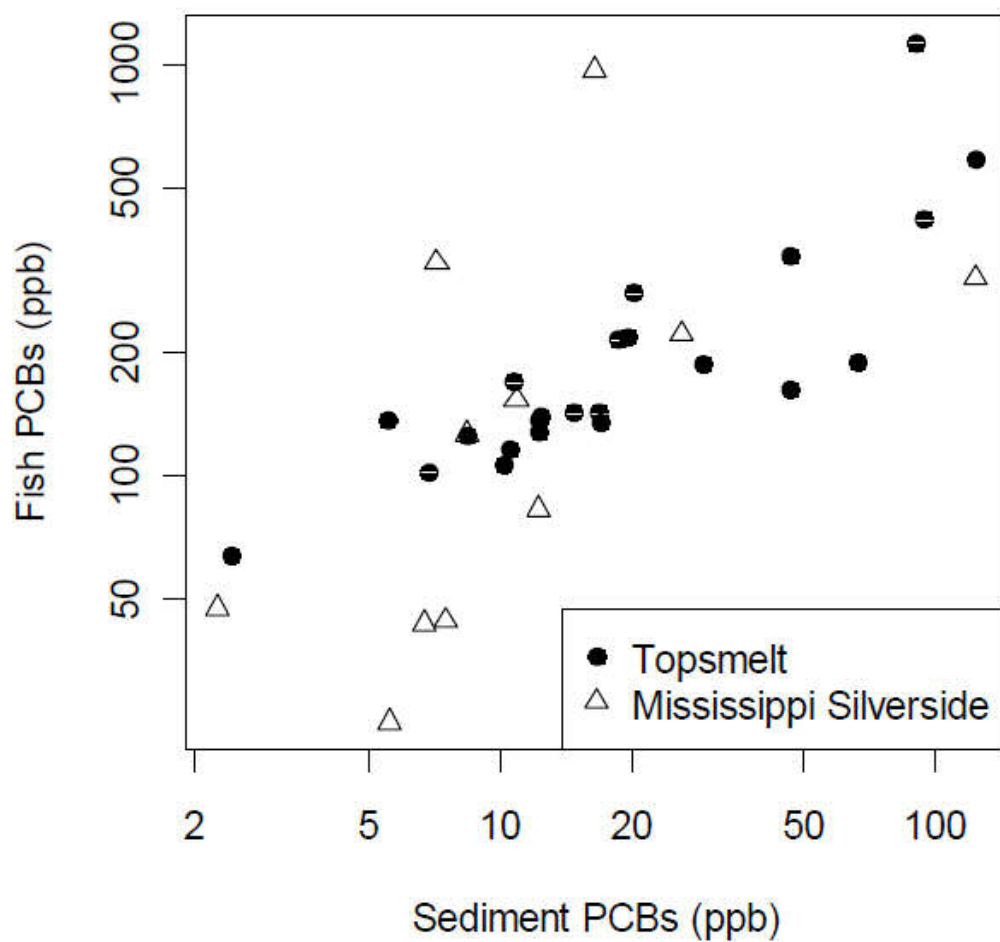


Figure 4. Concentrations of PCBs in small fish (2010) versus concentrations in nearby sediment. From Greenfield et al. (2012).

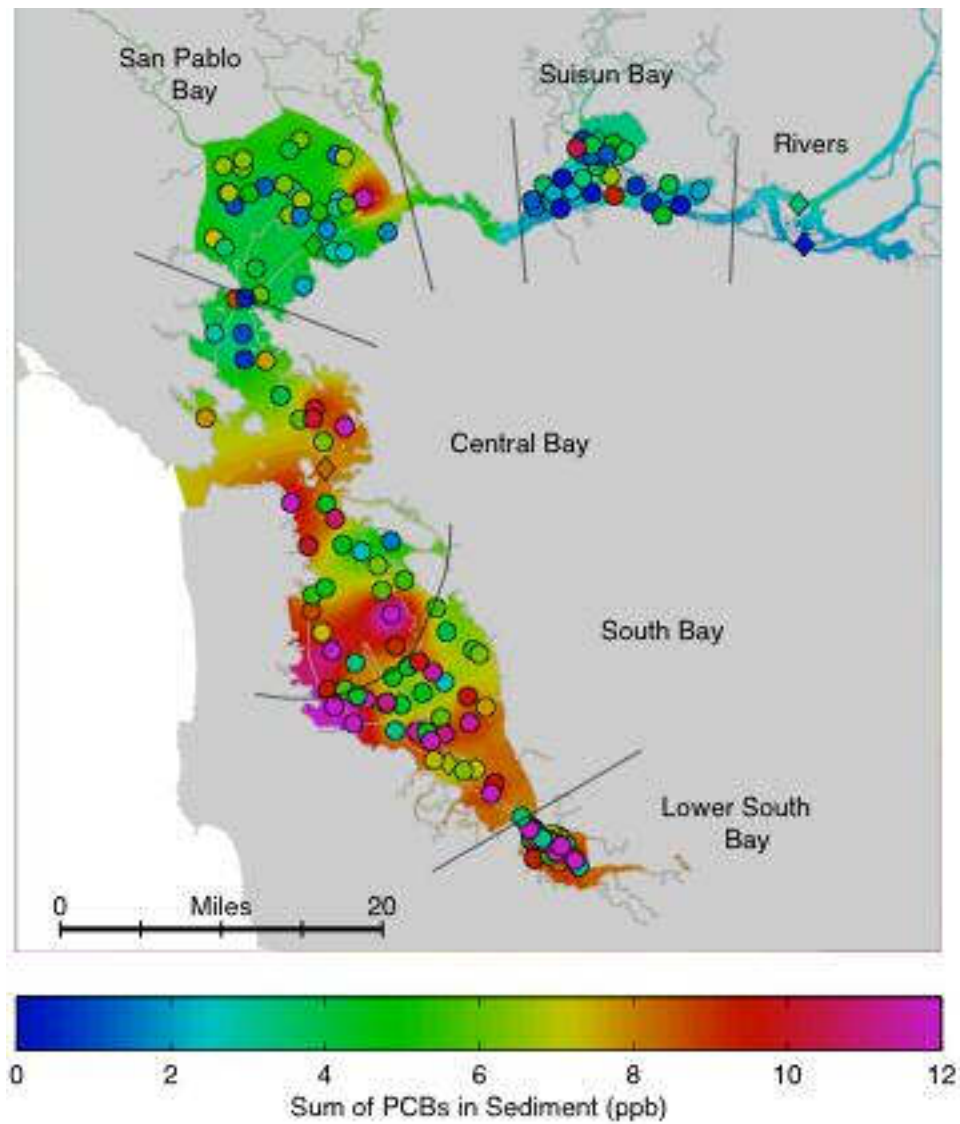


Figure 5. Bay-wide Sum of PCBs in sediment. Data are from samples taken between 2004 and 2008 by the Regional Monitoring Program (RMP).

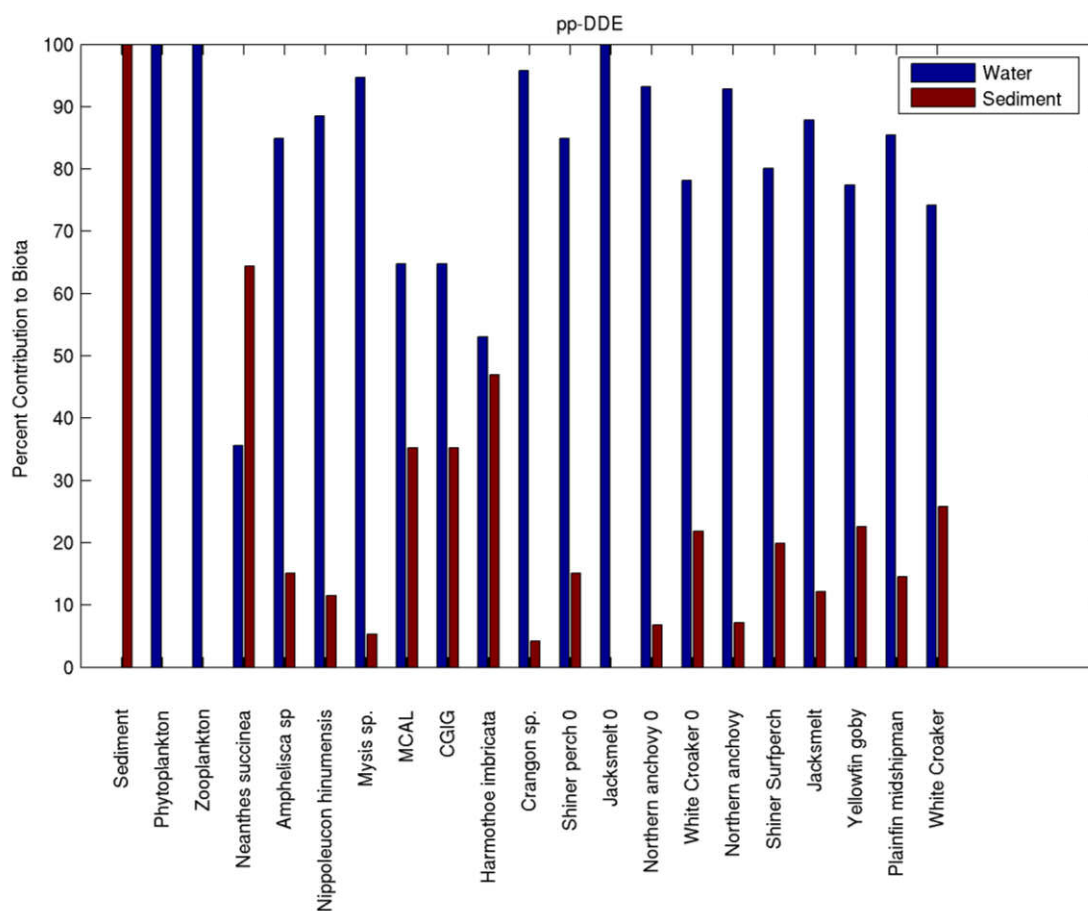


Figure 6. Relative contribution of water and sediment to p,p'-DDE bioaccumulation. Results are based on mechanistic model application to multiple species using parameters developed for San Francisco Bay.

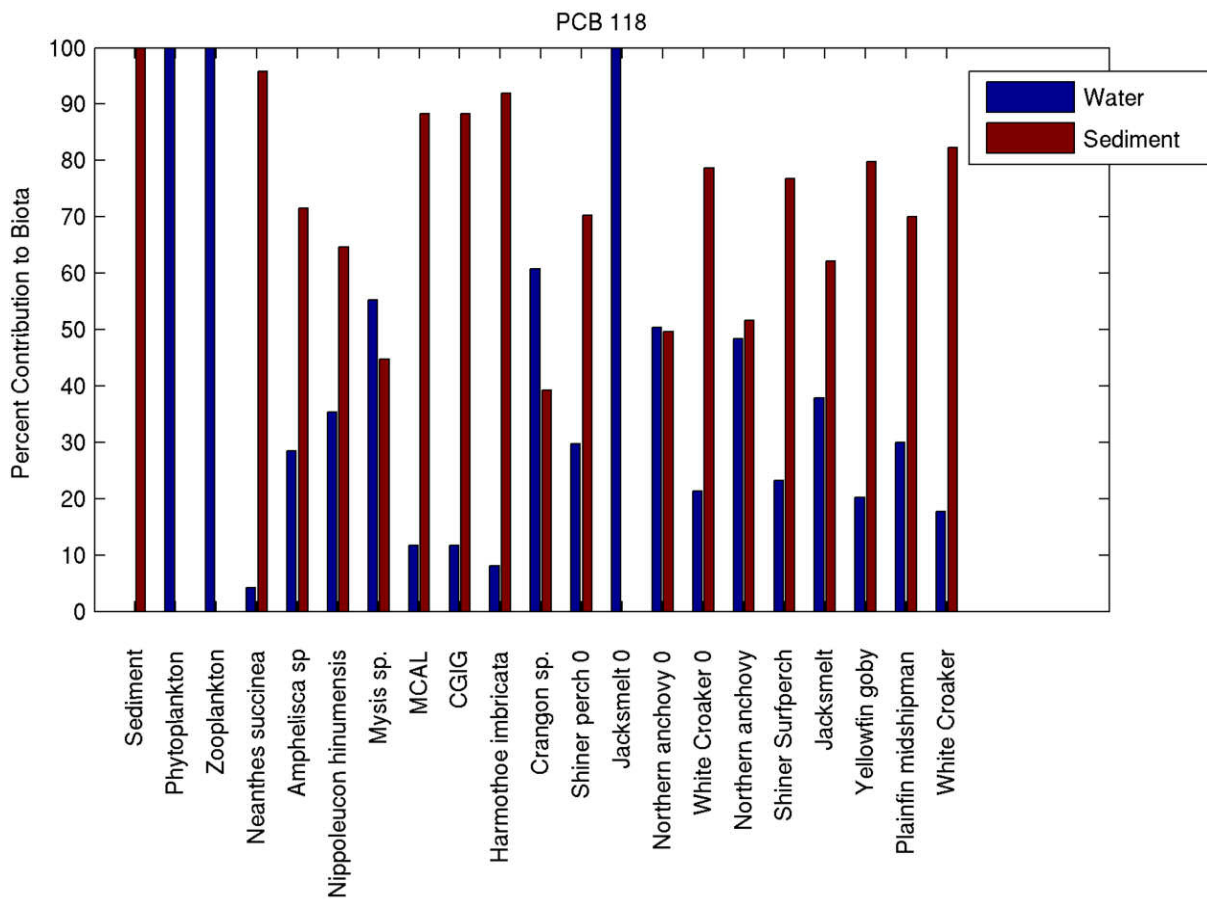


Figure 7. Relative contribution of water and sediment to PCB 118 bioaccumulation. Results are based on mechanistic model application to multiple species using parameters developed for San Francisco Bay.

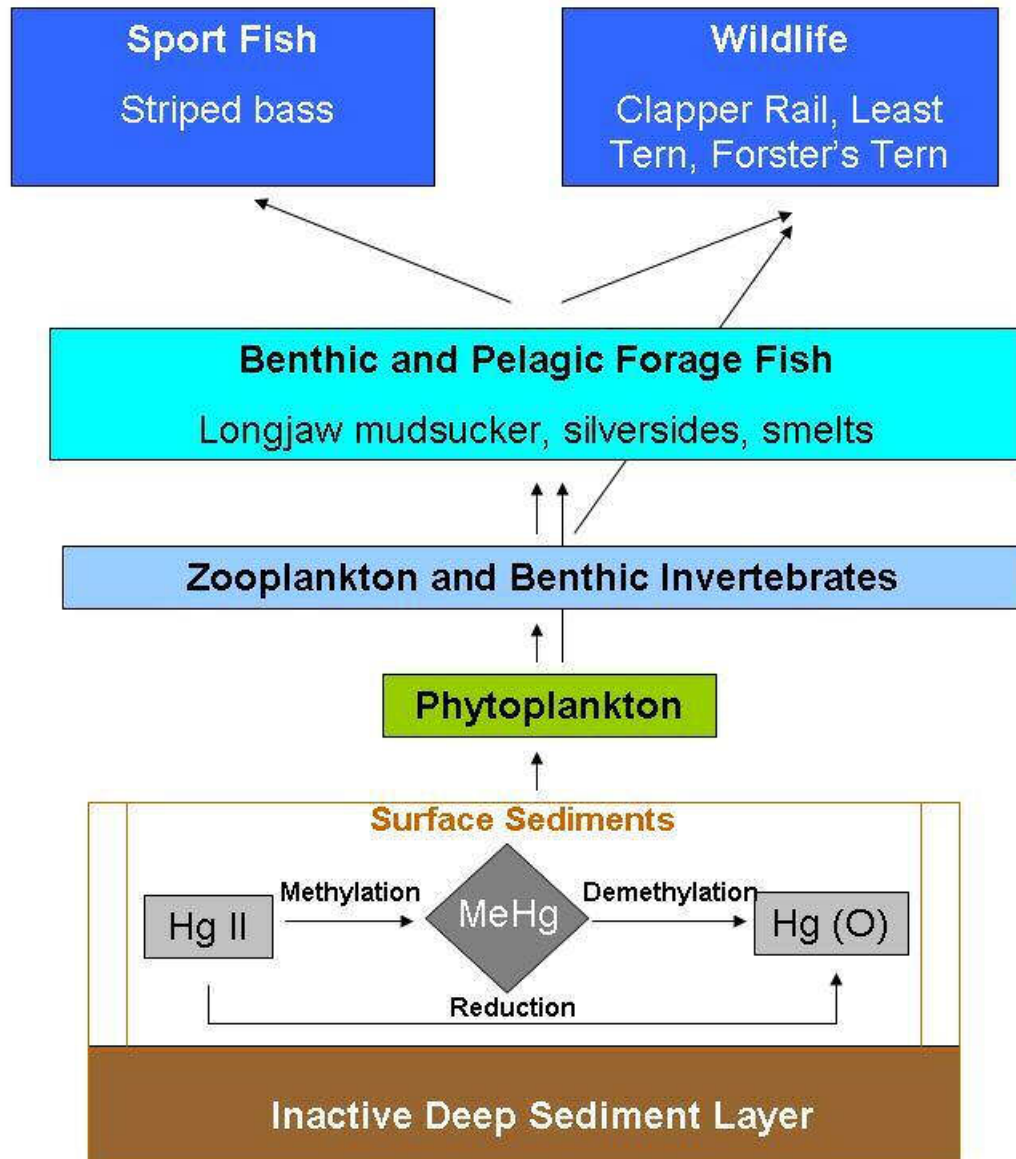


Figure 8. Generalized food-web linked to methylmercury bioaccumulation in San Francisco Bay