Toxic Cleanup Sites

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The contribution of mercury loads from in-situ sediments within the San Francisco Bay has not been adequately evaluated and is very difficult to quantify. Because bedded sediments throughout the Bay contain elevated levels of mercury at varying concentrations, it is likely that in-situ sediments constitute a significant source. However, limited opportunities exist for large-scale source reductions because it is not feasible, cost-effective, or desirable to dredge or cap large areas of the Bay. Consideration of potential control actions for contaminated sediments should focus on “hot spots”, or locations with a large mass of mercury in a small volume of sediment. The purpose of this evaluation was to identify sites that contain high concentrations of mercury in sediments and recommend an approach for evaluating site-specific contributions in terms of mass loading and Bay-wide risk.

IDENTIFICATION OF SITES WITH ELEVATED MERCURY

The first task in this evaluation was to identify sites of potential concern, review data, and summarize mercury concentrations and estimate mass of mercury at each site. This section summarizes the data sources that were used to identify sites with elevated concentrations of mercury in sediment and describes the screening approach used to determine whether a site constitutes a potentially significant source of mercury to the Bay. A summary of the data collected from the identified sites and the calculations developed from these data, are also provided.

Data Sources

In order to identify sites with elevated concentrations of mercury in sediments, data were compiled from various sources, including:

- San Francisco Estuary Regional Monitoring Program
- Bay Protection and Toxic Cleanup Program
- California State Mussel Watch Program
- Individual site investigation reports
- Data collected with CALFED Science Program funding

The Regional Monitoring Program (RMP) monitors contaminant concentrations in water, sediments, and fish and shellfish tissue in the San Francisco Bay and Delta. The Bay Protection and Toxic Cleanup Program (BPTCP) also conducted sampling of sediment, water, and fish and invertebrate tissue, but the purpose of this sampling was focused on areas where sediment contamination was suspected. The California State Mussel Watch Program (SMWP) conducts analysis of resident and transplanted mussels and clams, and also targets areas with known or suspected water quality impairment.

In addition to these monitoring programs, many individual sites have been investigated under other regulatory programs such as the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA). Sites evaluated under CERCLA include all of the former military bases in the San Franscisco Bay area, as well as privately owned sites. Other relevant regulatory programs include Site Cleanup Orders issued under the state Water Code, RWQCB enforcement actions under the BPTCP, and dredging projects under Clean Water Act Section 401 and 404. Remedial investigation and risk assessment reports were reviewed as part of this evaluation in order to identify those sites with significantly elevated mercury concentrations in sediments.

Some additional potentially valuable sources of data not included in this evaluation because of time, budgetary and data availability constraints include:

- Studies conducted by United States Geological Survey (USGS)
- Other studies conducted under the CALFED science program
- Mercury and methylmercury data collected by SFEI
- Hunters Point Shipyard Remedial Investigation/Feasibility Study reports
- Mare Island Remedial Investigation/Feasibility Study reports
- Dow Pittsburgh reports
- Port of Oakland data
- Port of Richmond data
- RWQCB SLIC sites adjacent to the Bay
- Additional reports for individual site investigations

It is recommended that data for these sites be reviewed. For example, a substantial amount of sediment, water and tissue data have been collected for Hunters Point Shipyard. All data for this site and other Navy sites are available in an electronic database that the RWQCB could request access to. SFEI has collected data on total mercury and methylmercury concentrations that would be useful for identifying hot spots as well as evaluating site-specific methylation potential. However, these data are not yet publicly available because of quality assurance problems.

Site Screening Criteria

In order to identify sites with “significantly elevated” mercury concentrations, a screening criterion was selected. Bay sediments have been influenced by natural and anthropogenic influxes of toxic chemicals over time, with a significant increase since the 1800s, when mining and industrial activities in the region became widespread. The San Francisco Bay Regional Water Quality Control Board (SFBRWQCB) performed a statistical analysis of available sediment chemical data. The results of this study are reported in two documents: the *San Francisco Bay Sediment Criteria Project Ambient Analysis Report* (Smith and Reige 1998), which contains the full data set; and *Ambient Concentrations of Toxic Chemicals in San Francisco Bay Sediments* (SFBRWQCB 1998), which summarizes and discusses the results. The objective of the study was to determine what the SFBRWQCB should consider as "ambient" upper bound threshold levels of various contaminants in the Bay. The Bay Ambient Threshold levels are defined as the 85th percentile of concentrations typical in sediment affected by anthropogenic sources, as opposed to background levels that are defined as naturally occurring concentrations. The Ambient Threshold for mercury (<100% fines) is 0.43 mg/kg (dry weight).

The National Oceanic and Atmospheric Administration (NOAA) has generated numerical sediment quality guidelines to help estimate the possible toxicological significance of chemical concentrations in sediments (Long 1995). These guidelines are not intended as regulatory standards or criteria. Effects Range-Low (ER-L) is the concentration for a specific chemical at which biological adverse effects in benthic organisms are seen in 10 percent of the cases. Effects Range-Median (ER-M) is the concentration of a specific chemical at which biological adverse effects are seen in 50 percent of the cases. The NOAA guideline concentrations for mercury in sediments are 0.15 mg/kg (ER-L) and 0.71 mg/kg (ER-M).

Because the Ambient Threshold is based on the 85th percentile of data collected in areas not directly affected by point sources, the statistics indicate that 15% of samples will exceed this threshold, even for a site that is representative of sediment concentrations throughout the rest of the Bay. The intent in this evaluation is to identify sites with *significantly* elevated (not just slightly elevated) mercury concentrations. Therefore, the effect range-median (ERM) was selected as the screening criterion for inclusion of sites. If at least one sample exceeded the ERM, the site was included in Table 1.

Some sites were reviewed and not included in Table 1 because the available data did not indicate that sediment concentrations exceeded the ERM for mercury, or because the sites have recently been remediated. These sites included:

- Peyton Slough (Rhodia)
- San Francisco International Airport (proposed platform area for expansion)
- Shearwater/US Steel
- Selby Slag

Summary of Site Data

For all sites identified with at least one sediment sample exceeding the ERM for mercury, available data on mercury in sediment, water and tissue are summarized in Table 2. For each site, an attempt was made to estimate the aerial extent of contamination when multiple samples were collected. Similarly, the depth of contamination was estimated when sufficient core data were available. It should be noted that all estimates of aerial and vertical extent are very rough estimates based on limited data. To calculate a rough estimate of the volume of contaminated sediment, the area was multiplied by the depth of contamination. To calculate the mass of sediment (dry weight), a unit weight of 20 kg/ft^3 was assumed (corresponding to a moisture content of 60%). To estimate the total mass of mercury at a given site, the mean mercury concentration was multiplied by the mass of contaminated sediment. These are preliminary estimates only that may be used for a rough comparison of the relative contribution of mercury from each site. Additional data review should be conducted to more thoroughly evaluate and quantify the concentrations and masses of mercury at each site. For many sites, little or no data on depth of contamination were available, so a depth of 2ft was assumed based on the estimated "active layer" in which bioturbation and other mixing processes occur. In addition, it was often very difficult to locate spatial extent of "hot spots", either because too few samples were collected or because high concentrations were found at various locations.

MODELING CONTAMINANT FLUX FROM SEDIMENT TO WATER

This section of the report describes approaches for modeling the flux of contaminants between bedded sediments in San Francisco Bay and the water column. First, some general characteristics of models are described. Later, the various characteristics of different model types are discussed, from simplest to more complex, that can be used to model contaminant flux from Bay sediments.

Important issues to consider in model selection are:

- Spatial Scale
- Temporal Scale
- Model Complexity
- Model Type

Each of these model characteristics is briefly described below.

Spatial Scale

Spatial scale is the size of the geographic area that can be resolved by the model. The largest spatial scale would be represented by a single-box model. In this case contaminant concentrations and fluxes would be represented by a single value that applies to the whole Bay. At the other end of the spectrum are two and three-dimensional models where thousands of grid cells (boxes), each representing an area as small as hundreds of square feet or a few acres, are used to represent the Bay. For TMDL modeling small scale models with thousands of boxes may be impractical and single or multi-box (few or at most hundreds of boxes) models are the most likely to be used. The spatial scale of the model should be at least sufficient to distinguish the variation in sediment concentrations observed in the Bay.

Temporal Scale

Temporal scale is the time step used in the modeling. Generally, three time scales could be considered for sediment flux modeling;

- Tidal time scale (minutes, hours)
- Tidally averaged (daily)
- Annual

Modeling at a tidal time scale allows for resolution of details of the tidal processes such as erosion/deposition (sediment exchange) due to tides and wind wave resuspension. These processes are important in determining instantaneous concentrations of contaminants in the water column since sediment is a major source for many contaminants. However, short time scales may be impractical for long term TMDL modeling. When using tidally averaged (or daily) models some processes have to be simplified since the mechanisms for erosion and deposition respond at tidal time scales. For example, erosion and deposition are governed by peak tidal velocities or wind speeds and deposition occurs during slack tide or low wind conditions. However, other processes such as pore water diffusion and long term changes in erosion/deposition can be represented at tidally averaged time scales. Processes such as erosion and deposition cannot be represented at annual time scales but long term changes in bathymetry can be included, based on empirical data. Processes such as pore water diffusion usually occur at a slow rate and can be represented using any of the above time scales. However, recent observations of porewater gradients by the USGS and others indicate sediments in the Bay can be both a sink and a source of dissolved chemicals to the water column, depending on the time of year. Therefore, modeling of longer time scales will require an estimate of the average flux over the time scales (or steps) of interest.

Another important aspect of choosing an appropriate timescale is the intended predictive purpose of the modeling. For example, if a model is desired to estimate the change in mercury concentration in fish tissue then a time scale that is comparable to the rates of the physical, chemical, and biological processes that govern the accumulation should be chosen. For example, if the mercury dupuration time in fish is on the order of months than a model that is able to predict changes in the mercury concentration over time on shorter timescales in the primary sources of mercury to fish (dissolved methyl mercury, methyl mercury in primary and secondary producers) may be appropriate. However, if the goal is to model the change in mercury concentrations in the sediment of the Bay timescales reflecting the processes important in the mercury sediment cycle (net sedimentation rate, sediment residence time) may be more appropriate.

Model Complexity

Model complexity describes the number and detail of the physical processes that are included in a model. Simple models may not include any physical processes or might use very simple relationships to incorporate them. Complex models may include many physical mechanisms/processes and interactions between them. The physical mechanisms can be described by very complex relationships. For example, erosion and deposition in a simple model may be represented by an average erosion or deposition rate. In a more complex model erosion may be represented by comparing bottom shear stress due to currents or winds to soil strength.

Model Type

Models can be divided into several types:

- Deterministic (mechanistic). These models are based on physical laws and relationships. A well formulated and calibrated deterministic model will provide the most reliable predictive capability but is often the most difficult to implement.
- Empirical. These models are based upon measured data. A common empirical model is a regression relationship between a physical parameter and observed data.

For example, particle size and concentration on sediment. An empirical model can be used for prediction but is generally only valid for the conditions that existed when the data used to develop the relationships were collected.

· Stochastic. Stochastic models attempt to account for the inherent variability or uncertainty in the model results by representing model inputs using statistical relationships. The relationships between the inputs are usually simple but they do not have to be. However, the model complexity or scale has to be such that either an analytical solution can be derived or many models runs can be conducted in a reasonable time period.

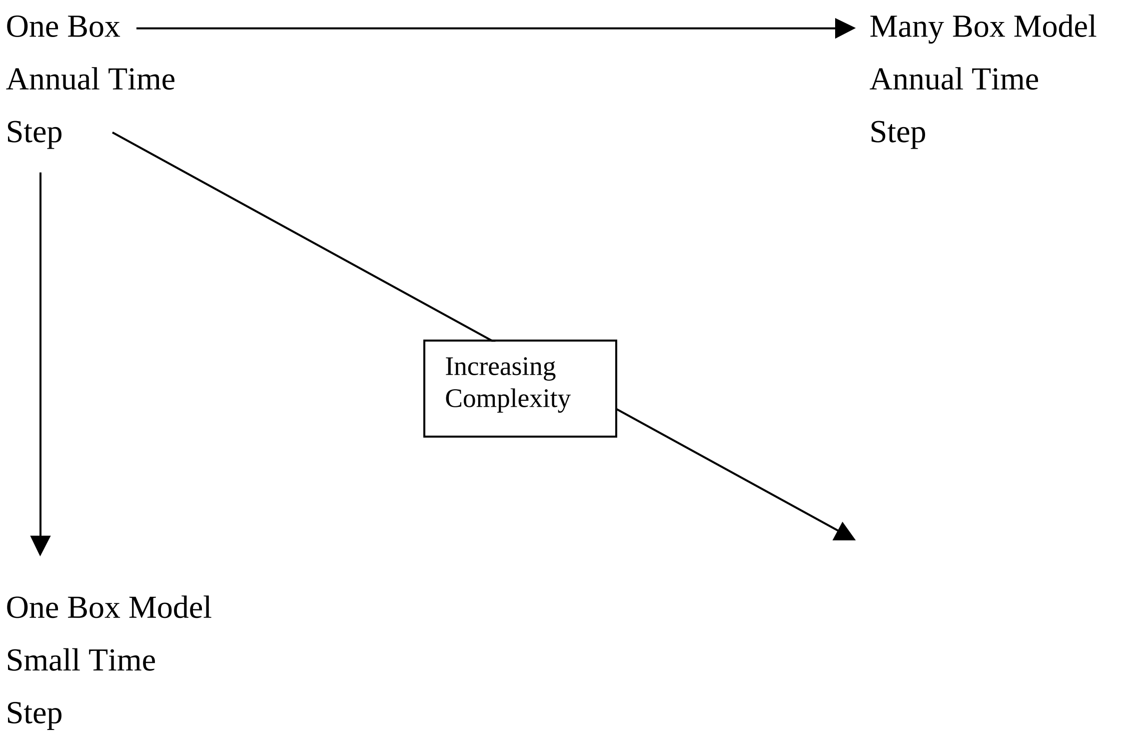
Models of Contaminant Flux between Sediment and Water

Major processes that need to be included in a model of sediment water interaction are sediment exchange between water and bed (i.e., sediment erosion and deposition), contaminant flux from pore water into or from the water column, chemical exchange between benthic sediment and pore water, and chemical exchange between suspended sediment and the water column. Erosion and deposition are very important for predicting small time scale (instantaneous, hourly) concentrations in the water column. To get accurate concentration predictions on these short time scales a complex sediment model may be needed. For long term simulations (e.g., decades) that will be used for TMDL analysis these short time scale processes are less important. However, erosion and deposition provide a primary mechanism for the removal of contamination from sediment in the Bay so some relationship is needed to represent these processes. For example, if the source of contamination from external sources to the south Bay were eliminated, erosion (i.e., resuspension of sediment) and transport of sediment would be the primary mechanism for removing contaminated sediments. Therefore, even simple models need to include some description of the erosion processes.

The flux of contaminants from Bay sediments and pore water is usually a slow process and can be represented using relatively simple relationships. Therefore, simple low resolution models can be used to describe contaminant flux adequately. Also, chemical exchange between sediment and pore water is usually assumed to be at equilibrium, in which case simple relationships can be used.

In developing the appropriate model for sediment water interactions for TMDL analysis it is important to specify the type of results the model needs to provide. No model will be able to answer all questions. For example, a simple, one box model can be used to easily simulate many decades of contaminant transport. It could provide information on the relative merits of various implementation options on a Bay wide comparison. However, it would provide no useful information on the benefits of removing toxic hot spots. A complex, high resolution model could be used to analyze the benefits of removing toxic hotspots but would be difficult to use for modeling decades of contaminant transport.

Three levels of models that could be used to model sediment water interactions are described below.



Simple, One Box Model, Annual Time Step

A one box model has been developed for PCBs and mercury. These models do not include any detailed physical processes and are based on mass balance and empirical data. They provide a general impression of what will occur with general changes in inputs but do not provide enough detail to be useful for TMDL implementation. For example, cleaning riverine sources may have large local benefits but small overall benefits. A single box model would not show the local benefit. Also, since they rely on empirical data and do not include a description of physical process they are of limited utility for future predictions under changed conditions (e.g., global warming, change in delta outflow).

Multi-box model, Annual Time Step

This is an improvement over the one box model since it will provide more geographic detail. For example, sediment concentrations in different parts of the Bay could be included in the model (e.g., Lower South Bay vs. San Pablo Bay). This would provide a method for comparing implementation options that only apply to a portion of the Bay, e.g., clean Guadalupe River versus Napa River. However, since it only uses an annual time step erosion/deposition would rely on empirical or very simple relationships. Therefore, calculations such as sediment residence time would be speculative and highly sensitive to model input. On the other hand, exchanges between the water column and pore water (through partitioning between sediment and pore water and pore water diffusion) could be modeled adequately. In this case the number of boxes would only need to be sufficient to represent the variation in sediment concentrations in the Bay.

Multi-box model with Daily Time Step

Going from an annual time step to a daily time step would allow for more resolution in the annual variability in concentrations. For receptors for which shorter term (less than yearly) exposures are important, use of a time step less than annual may be necessary. For example, if six month exposure is sufficient to cause an impact to a receptor than the time step used in the analysis should be less. This could be important since the source of many contaminants is seasonal, e.g., from winter storms. The exchange between contaminated bed sediments and new sediments in the water column is mainly seasonal and therefore, it may be necessary to use a smaller time step to adequately represent this.

Multi-box Model with Tidal Time Scale

A multi-box model with a tidal time scale could be used to model specific processes with greater detail but may not be practicable for long term TMDL modeling. To predict the exchange of sediment in the bed with water column sediment it may be necessary to model at this scale. To predict the exchange of contaminates due to pore water diffusion a longer time scale could be used.

Field Measurements

Field measurements can also be used to identify site-specific model input parameters and to calibrate and validate models. Benthic flux chambers can be used to measure sediment-water exchange fluxes for mercury and methylmercury. Flux chamber results give short-term determinations which may not be in steady-state conditions relative to the sampling interval. It may be difficult to obtain good agreement between flux chamber results and model results.

Recommended Model Approach

No single model approach will be sufficient to answer all questions relating to sediment water exchange for TMDL implementation. To pick the best approach it is necessary to answer several questions, including:

- What is the exposure time scale of interest? If it is months then an annual model will not be sufficient.
- What implementation strategies are to be compared? If removal of toxic hot spots is to be compared to removing contaminated sediments from rivers then a spatially detailed model is needed.
- What processes are important to include? If pore water diffusion and the exchange of contaminants between sediments and pore water is important then large time scales may be adequate. If the exchange of sediment between the bed and water column is important than short time scales may be required.

- What types of predictive capabilities are needed? If predictions under different sets of conditions are required then a more complex model is needed. If extrapolation of existing data is sufficient than a simple model may suffice.
- How is uncertainty in model predictions to be included in the results? If uncertainty and variability are important then a stochastic approach may be considered.

If the purpose of the contaminant flux modeling is to estimate the long term load from sediment sites to the Bay, a multi-box model with an annual time step might be sufficient. If the purpose is to use the model results in predicting fate, transport, and exposure of mercury within the system, a shorter time scale would be necessary.

Regardless of the type of model used, the contribution of mercury flux from each contaminated sediment site must be evaluated in context with the contribution from Bay-wide sediments. As stated earlier, mercury concentrations in sediments throughout the Bay are elevated, and it is likely that the load of mercury from one 10-acre site with concentrations of 10 mg/kg would be insignificant compared to the load of mercury from the rest of the Bay, where concentrations are lower but cover a much larger area.

In order to estimate the loading contribution from a site (or group of sites), the model could be run assuming current site concentrations are present. The model could then be run assuming that concentrations of mercury at the site (or group of sites) are typical of ambient conditions elsewhere in the Bay. The model results under both scenarios could then be compared to determine the magnitude of change.

In addition to mass loading questions, bioavailability and risk assessment questions should be considered when selecting and designing a model. If the ultimate goal of TMDL is to reduce mercury concentrations in fish tissue to levels that are protective of human health and the Bay ecosystem, then mercury loads must be linked to fish tissue concentrations. The following section discusses a risk assessment approach that could be used to help determine the contribution of mercury concentrations at individual sites to Bay-wide risk.

RISK ASSESSMENT APPROACH

Many of the sediment sites with elevated mercury concentrations identified in Table 2 are currently being investigated and considered for remedial action. In most cases, risk assessments have been or will be conducted to evaluate potential impacts to human and/or ecological receptors potentially affected by contamination at the site. However, in general these risk assessments are site-specific and do not evaluate the contribution of mercury-contaminated sediments a each site in the context of Bay-wide risk to human and ecological receptors. This section lays out a framework for developing a process to estimate the contribution of site-specific mercury-contaminated sediments to overall Bay-wide risk. The process includes consideration of applicable TMDL targets for establishing site-specific cleanup levels and selecting remedial alternatives.

The following section provides a background of the regulatory vehicles that commonly facilitate the risk assessment process at sites that warrant investigation.

Current Site-Specific Risk Assessment Approaches

CERCLA

Human health and ecological risk assessments play a major role during the remedial investigation phase for a CERCLA site. For human health risk assessment, EPA has developed standard tables and worksheets to clearly and consistently document important parameters, data, calculations, and conclusions from all stages of human health risk assessment. During the feasibility study, remedial options are evaluated using the baseline human health and ecological risk assessments, as well as the potential for the remedial actions themselves to cause adverse ecological impacts. The Record of Decision for a site has several purposes, including documentation of the selected remedy, providing a rationale for the remedy, and establishing performance standards or goals for the site or operable unit. The purpose of the remedy selection is to reduce, or control site risks to acceptable levels.

The baseline risk assessment data that must be described in the ROD include:

- contaminated media and chemicals of concern;
- time frame for risk (current or future);
- land-use assumptions
- receptor populations and location of receptors (on-site or off-site); and
- exposure routes to receptor populations that trigger the need for cleanup.

As part of the feasibility study, applicable and relevant or appropriate requirements (ARARs) are identified. CERCLA provides that any standard, requirement, criteria, or limitation under any federal or state law may be an ARAR. Remedial actions taken under CERCLA must require a level or standard of control which at least obtains such requirements. The human health and ecological risk assessments should identify threshold contamination levels for adverse effects on assessment endpoints. These threshold values provide a yardstick for evaluating the effectiveness of remedial options and can be used to set cleanup goals as appropriate. Following the Record of Decision and implementation of a remedial design, monitoring programs are implemented to evaluate the success of a remedial action.

For military sites addressed under the CERCLA regulations, the Department of Defense is the lead implementing agency and EPA is the lead regulatory agency. All military sites in the San Francisco Bay area have been closed and are currently undergoing cleanup and being transferred to other land uses. For other Superfund sites, EPA is the lead regulatory agency. EPA Region 9 has established a group of scientists from state and federal regulatory and natural resource agencies to assist site managers with ecological risk assessments and remedial action planning. This group is known as the Biological Technical Assistance Group (BTAG), and includes representatives from USFWS, NOAA, CDFG, RWQCB, DTSC.

BPTCP
California Water Code, Division 7, Chapter 5.6 established a comprehensive program to protect the existing and future beneficial uses of California's enclosed bays and estuaries. Water Code Section 13394 requires that each RWQCB complete a toxic hot spot cleanup plan. Each cleanup plan must include: (1) a priority listing of all known toxic hot spots covered by the plan; (2) a description of each toxic hot spot including a characterization of the pollutants present at the site; (3) an assessment of the most likely source or sources of pollutants; (4) an estimate of the total costs to implement the cleanup plan; (5) an estimate of the costs that can be recovered from parties responsible for the discharge of pollutants that have accumulated in sediments; (6) a preliminary assessment of the actions required to remedy or restore a toxic hot spot; and (7) a two-year expenditure schedule identifying State funds needed to implement the plan.

Process to Develop a Consistent Bay-wide Risk Assessment Approach

Although the mandate of the BTAG is to provide assistance specifically for CERCLA sites, the risk assessment and management issues dealt with at CERCLA sites are essentially the same as those encountered at contaminated sediment sites addressed under other regulations or evaluated for other purposes. Because the BTAG consists of ecological risk assessments experts from all or most of the relevant state and federal agencies, the BTAG would provide a good forum for review and refinement of a Bay-wide risk assessment approach that could be incorporated into the existing risk assessment process for sediment sites. Although the BTAG was set up to address ecological risk assessment issues, many of the BTAG members are also familiar with issues regarding risk to human health via fish consumption, and have dealt with these issues in conjunction with site-specific risk assessments.

As many of these sites are being addressed under other regulatory programs, it is important to determine how TMDL load allocations will fit into the existing risk assessment/feasibility study process. It should be noted that in order for TMDL targets, load allocations and/or implementation plan control measures to be considered as ARARs in the CERCLA process, the TMDL must be approved by EPA and promulgated as a regulatory requirement. Additionally, the Basin Plan must be amended to include TMDL requirements. In the meantime, an interim guidance could - be incorporated into the feasibility study process as criteria “to be considered” or TBCs, which include federal or state advisories, criteria, or guidance that may be useful in developing CERCLA remedies and determining cleanup levels.

The primary guidance documents used to conduct ecological risk assessments for contaminated sediment sites in the Bay include:

· (*Ecological Risk Assessment Guidance for Superfund: Process for Designing and Conducting Ecological Risk Assessments (*USEPA 1997),

· *Guidelines for Ecological Risk Assessment* (USEPA 1998),

· *Guidance for Ecological Risk Assessment at Hazardous Waste Sites and Permitted Facilities* (Cal/EPA 1996), and

· *The Role of Screening-Level Risk Assessments and Refining Contaminants of Concern in Baseline Ecological Risk Assessments* (USEPA 2001).

The primary guidance documents used to conduct human health risk assessments for contaminated sediment sites in the Bay include:

· *Risk Assessment Guidance for Superfund. Volume I: Human Health Evaluation Manual* (USEPA 1989 et seq.),

· *Supplemental Guidance for Human Health Multimedia Risk Assessments of Hazardous Waste Sites and Permitted Facilities* (DTSC 1992), and

· *Application of Risk-Based Screening Levels and Decision Making to Sites With Impacted Soil and Groundwater, Interim Final* (SFBRWQCB 2001).

These documents provide a good framework for conducting site-specific risk assessments, and the same framework can be used to develop regional or Bay-wide risk assessments. In order to develop a Bay-wide risk assessment approach to address sources of mercury from contaminated sediment sites, the following questions should be addressed:

<u>Human Health</u>

· What is the potentially exposed population that should be evaluated?
· What are the appropriate exposure parameters?
· What is the appropriate risk threshold?
· Based on the exposure parameters and risk threshold, what is the Bay-wide fish tissue concentration threshold?
· What is the contribution of site-related mercury contamination to Bay-wide fish tissue concentrations, and is this contribution significant compared to other sources?
· How would potential remedial actions taken at the site affect Bay-wide fish tissue concentrations?

<u>Ecological Risk Assessment</u>

· What are the most sensitive and highly exposed species that should be evaluated on a regional (rather than a site-specific) basis?
· What are the typical home ranges of these receptors and their prey?

- Based on current conditions, are impacts to these receptors now occurring?
- If so, how can site-specific measurement endpoints be linked to impacts to these receptors?
- To what degree does mercury contamination at a site contribute to risk posed to these receptors, and is this contribution significant compared to other sources?
- How would potential remedial actions taken at the site affect regional or Bay-wide risk to these receptors?

Many of the above questions (especially the human health ones) are already being addressed in the TMDL process, and this work can be incorporated into the sediment risk assessment approach. It is important to keep in mind that the ecological assessment endpoints most appropriate for site-specific risk assessments may be different than those that are most appropriate for a regional risk assessment, because of the difference in scale. For example, harbor seals would not be considered in a site-specific risk assessment if they are not believed to use the area for foraging, but they would be an important receptor to consider for a Bay-wide risk assessment. Similarly, the human population considered under a site-specific risk assessment may be different than the population considered under a regional risk assessment. For example, if shellfish collection is known to occur at the site but no fishing is believed to occur in the immediate vicinity, the site-specific risk assessment might only consider consumption of shellfish as the exposure pathway. However, the regional assessment would also consider the exposure pathway through consumption of sport fish with a large home range that may feed at the site but are caught in another area of the Bay.

Although the BTAG would provide a good forum for review and refinement of the risk assessment approach, members of the BTAG do not have the time or resources to develop the approach. The approach should be developed and funded as part of the TMDL Implementation Plan, which should include a schedule for evaluation of each site identified as a potentially significant source of mercury.

Technical Approach for Bay-Wide Risk Assessment

Conceptual Exposure Model

For effects to occur, a receptor and a complete exposure pathway must be present. An exposure pathway is only considered complete when all four of the following elements are present: source of a chemical, a mechanism of release of the chemical from the source to the environment, a mechanism of transport of the chemical to the ecological receptor, and a route by which the receptor is exposed to the chemical.

The exposure routes associated with the complete pathways to ecological receptors for mercury-contaminated sediments include sediment ingestion, water ingestion, direct contact, and food-web exposure. A conceptual exposure model should be developed to provide a schematic representation of the links between sources, release and transport mechanisms, affected media, exposure routes, and potentially exposed ecological receptors. Although several complete exposure pathways may exist, not all pathways are comparable in magnitude or significance. The significance of a pathway as a mode of exposure depends on the identity and nature of the chemicals involved and the magnitude of the likely exposure dose. For birds and mammals, ingestion of prey is generally the most significant exposure pathway and, to a lesser extent, incidental ingestion of sediment and water. For humans, consumption of fish and shellfish is the main pathway considered.

Human Health Risk Assessment

The threat to human health comes from mercury that has bioaccumulated in fish and shellfish and can be consumed by humans. In order to minimize the threat of adverse effects to human health resulting from mercury contamination, various agencies have set total mercury concentration guidelines for water and fish tissue.

The USA EPA CTR mercury water quality criterion (0.051 ug/L) for protection of human health from consumption of aquatic organisms is based on the “practical bioconcentration factors” (PBCFs) that were derived in 1980 for fresh water, estuarine water, and open oceans. These PBCFs relate the mercury concentration in water to the concentration in fish. A weighted average of these three PBCFs was calculated to take into account the average consumption of organisms from the three types of waters.

The U.S. Food and Drug Administration (FDA) action level of 1 µg/g for fish tissue is widely accepted as a regulatory guideline for the protection of human health. The FDA advises that fish with mercury concentrations in excess of 1 µg/g should not be consumed.

The San Francisco Estuary Institute (SFEI) developed a screening level of 0.23 µg/g to be used when assessing fish tissue mercury concentrations. Although not a human health action level, the SFEI screening level can be used to determine whether continued monitoring should be performed. RWQCB has also published this value (0.23 µg/g) as a fish tissue threshold of concern. Consensus appears to be that for an individual on a high fish consumption diet, risk of toxicity starts in the range of about 0.16 to 1.0 µg/g mercury in edible portions of fish (Lee and McFarland 2000).

Identifying the Population to be Evaluated

Although the water quality criterion is generic and may be more conservative (or less conservative) than necessary to protect human receptors that consume fish from San Francisco Bay, it is unlikely that a more site-specific water quality criterion will be developed and promulgated. However, fish tissue numbers are guidance numbers only and it would be appropriate to develop a fish tissue target level specific to the Bay, based on local fish consumption data. Recreational anglers and/or subsistence fish harvesters are the primary human receptors that would require consideration for the purposes of this type of evaluation. Studies on consumption habits of anglers in the San Francisco Bay area have recently been conducted. For example, survey data indicate that anglers of Asian ethnicity represent 63 percent of the total consumers of fish at Point Pinole (DHS 2001). The survey also revealed that these anglers tend to fish for subsistence purposes, rather than as a means of recreation, and, therefore, consume fish more regularly than anglers of other ethnic backgrounds.

Exposure Parameters

The types of fish that are present in the vicinity of a site may also be used to develop a more realistic understanding of fish consumption and site-specific contribution to risk. A study conducted by the San Francisco Estuary Institute (1999) on popular sporting fish in San Francisco Bay may be consulted for information regarding sport fish species caught near a specific site. For example, a site near the San Pablo Bay fish sampling location would be expected to harbor the fish that were documented as present in San Pablo Bay. Based on this information, a site-specific lipid content in fish tissue could be used to estimate chemical concentrations in fish tissue for purposes of food-chain modeling.

At a minimum, the following documents should be consulted for evaluation of the fish consumption pathway in a human health risk assessment:

- *San Francisco Bay Seafood Consumption Report* (California Department of Health Services 2001).

- *Assessing Human Health Risks from Chemically Contaminated Fish and Shellfish* (USEPA 1989).

Effects Assessment

For noncarcinogenic chemicals, a hazard quotient (HQ) of one represents the cumulative target acceptable health risk level. The HQ is defined by the ratio of the estimated chemical intake to the reference dose (RfD). The RfD is an estimate of a daily chemical intake per unit body weight that is likely to occur without deleterious effects during a lifetime or a portion of a lifetime (USEPA, 1989). HQs of 1 or below indicate no adverse health effects are expected. HQs (or HIs) above 1 indicate a potential for adverse effects, and that further evaluation of exposure conditions and toxicity is warranted in determining the need for remedial action.

The following sources of RfDs should be used:

- Cal/EPA Office of Environmental Health Hazard Assessment (OEHHA) – California Cancer Potency Factors and Toxicity Criteria Database (OEHHA 1994, 2000).
- USEPA Integrated Risk Information System (IRIS) Database (USEPA 2000A).
- USEPA Health Effects Assessment Summary Tables (HEAST) (USEPA 2000b).

- USEPA Region 9 Preliminary Remediation Goals (USEPA 2000).

Ecological Risk Assessment

Highest levels of methylmercury (the most toxic form) occur in long-lived, slow-growing, higher-trophic-level aquatic species, typically those that feed on fish. Methylmercury is a neurotoxin, mutagen, teratogen, and carcinogen.

Assessment and Measurement Endpoints

USEPA defines an assessment endpoint as an “explicit expression of an environmental value to be protected” (USEPA 1997). Assessment endpoints define both the valued ecological entity and a characteristic of the entity to protect, such as individual survival, population success, production per unit area, or changes in species distribution in an ecosystem community. Generally, each assessment endpoint includes a guild or a functional group within an ecosystem, rather than one particular species. However, for purposes of evaluation, a representative or surrogate species is selected. When threatened or endangered species are present, it is often appropriate to define assessment endpoints based on effects to an individual. Other assessment endpoints are typically defined on the basis of effects to a population or a community.

Ecological risk assessment involves multiple species that are likely to be exposed to differing degrees and to respond differently to the same contaminant. It is not practical or possible to directly evaluate risks to all of the individual components of the ecosystem that could be adversely affected by contaminants. Instead, assessment endpoints focus the risk assessment on components of the ecosystem that are judged to have high potential for adverse effects. The selection of assessment endpoints depends on the following:

- The contaminants present and their concentrations
- Mechanisms of toxicity of the contaminants to different groups of organisms
- Ecologically relevant receptor groups that are potentially sensitive if highly exposed to the contaminant, and attributes of their natural history
- Potentially complete exposure pathways

USEPA defines measurement endpoints as “a measurable ecological characteristic that is related to the valued characteristic chosen as the assessment endpoint and is a measure of biological effects (e.g., mortality, reproduction, growth)” (USEPA 1997). Measurement endpoints can include measures of exposure or effect and are frequently numerical expressions of observations that can be compared statistically to a control, reference site, or scientific study to predict adverse responses to a site-specific COPEC. Each measurement endpoint correlates directly with one of the defined assessment endpoints.

Bay-wide measurement endpoints can be used to evaluate regional impacts to upper trophic level receptors that occur as a result of the sum of mercury sources throughout the Bay. For example, Bay-wide measurement endpoints may include monitoring of mercury concentrations in bird eggs, blood of mammalian and avian receptors, reproductive success, etc. While these types of measurements are useful for evaluating cumulative Bay-wide impacts, they do not provide information that can be tied to mercury sources at a particular site. Site-specific measurements typically include analysis of mercury concentrations in sediment, water, and tissue of organisms with a very small home range, such as benthic invertebrates. For many sites identified in Table 2, some or all of these types of measurements have already been collected. These site-specific measurements can be used to assess the contribution of mercury from an individual site to Bay-wide risk from mercury, as discussed later in this section.

Selection of Representative Ecological Receptors

For compounds such as mercury that biomagnify in the food chain, it is generally more appropriate and conservative to evaluate piscivorous receptors than benthic-feeding receptors. Pollutant concentrations in fish tissue are likely to be higher than concentrations in tissue of benthic invertebrates because invertebrates serve as prey for most adult fish species. Consequentially, piscivorous receptors are likely to receive higher exposure.

Representative receptors should be selected on the basis of their occurrence, habitat and dietary preference, trophic level, sensitivity to mercury, and availability of toxicity and natural history data. At a minimum they should include a piscivorous bird and a marine mammal, based on the tendency of mercury to biomagnify. While it would also be useful to include a piscivorous fish, there may not be sufficient data available to evaluate food-web toxicity of mercury to fish. Although direct mercury toxicity can affect aquatic and benthic organisms, this type of effect is localized and would be evaluated as part of a site-specific risk assessment rather than a regional risk assessment.

Some suggested representive species include the following:

Double-crested cormorant. The double-crested cormorant (*Phalacrocorax auritus*) is a state species of special concern and year-round resident of the coastal salt marshes of northern California. It nests in high marsh vegetation and has a home range of approximately 200 km^2 (Zeiner et al. 1990). Its primary food source includes small fish and occasionally benthic invertebrates. The fish include both trophic level 2 forage fish (primary consumers that feed on plankton) and trophic level 3 fish (secondary consumers that feed on invertebrates). The double-crested cormorant is suggested as a representative species because of its dependence on forage fish as a food source, year-round presence in the Bay Area, relatively small home range, and societal value as a protected species. It represents a semiaquatic, avian tertiary-trophic-level piscivore. Exposure to mercury for the cormorant is expected to occur by ingestion of forage fish and water and by incidental ingestion of sediment and water during foraging activities and preening.

Pacific harbor seal. The Pacific harbor seal (*Phoca vitulina*) is a pinniped (an aquatic carnivorous mammal with limbs modified as flippers) protected by the Marine Mammal Protection Act of 1972 and a year-round resident in the Bay. The harbor seal uses the

Bay for foraging and breeding. The home range of the Pacific harbor seal may be as small as 80 km^2, based on a tracking study conducted in a bay in California (USEPA 1993). However, radio tracking of harbor seals in the Bay has shown that individual seals tend to move around the Bay fairly frequently. It is not unusual for harbor seals to forage in the South Bay and move to the North Bay several weeks later (Harvey, pers. comm., 2002). Harbor seals are opportunistic feeders, eating a variety of fish, cephalopods, crustaceans, and mollusks that live near the ocean floor. In the Bay, harbor seals feed primarily on Pacific herring, plainfin midshipmen, yellowfin goby, white croaker, Pacific staghorn sculpin, and northern anchovy (San Francisco Bay Seal Project 1989). The harbor seal is recommended as a representative species because of its dependence on forage and commercially important fish as a food source, its likelihood to occur in the affected areas, and its societal value as a protected species. It represents a semiaquatic, mammalian tertiary-trophic-level carnivore. The harbor seal's exposure to mercury is expected to occur by ingestion of forage and predatory fish, benthic invertebrates, and incidental ingestion of sediment and water during foraging activities. Although dermal contact with water is a potential route of exposure, it is expected to be insignificant compared to dietary exposure.

Exposure Assessment

For higher-trophic-level receptors, the exposure dose can be estimated as a function of mercury concentrations in the water, sediments, and prey. If data are not available for mercury concentrations in prey, these concentrations can be modeled using empirical bioaccumalation factors (BAFs) or biota-sediment accumulation factors (BSAFs), or mechanistic models that predict biotransfer through the food web, such as kinetically-based bioenergetic models. Food-chain models often use a BSAF multiplied by the contaminant concentration in sediment to estimate concentrations in invertebrates and fish. This approach assumes that uptake is dependent on the concentration in bottom sediments as the primary source rather than in overlying water, which is likely to be the case for mercury. Doses to upper trophic level receptors can be estimated using parameters such as dietary composition, food ingestion rates, etc. Exposure doses for representative receptors can be estimated by a series of models to represent mercury concentrations as they transfer from the water through the biotic compartments of the food web and into the dietary components of the receptors.

Effects Assessment

A no-observable-adverse-effects-level (NOAEL)-based toxicity reference value (TRV) represents a chronic dose corresponding to an estimate of no adverse effects to an organism. This dose may be thought of as a chronic toxicity threshold below which adverse effects would not be expected to occur. The goal of the effects assessment for food-web receptors is to identify TRVs that are appropriate to the representative receptors and assessment endpoints that have been selected. TRVs are expressed as mg/kg body weight/day.

TRVs are developed from toxicological data, and several steps are used to convert a laboratory-administered dose to a chronic toxicity threshold. These steps include a review of the duration of the study, test species involved, exposure endpoints measured, whether sensitive life stages were included in the study duration, and method of dose application. Uncertainty factors are developed to extrapolate from high-dose studies and laboratory species to the low-dose conditions typical of environmental exposures and wildlife species.

The California Department of Toxic Substances Control TRVs, i.e., Region 9 BTAG TRVs, (Cal/EPA 2000) for mammals and birds were developed by an interagency group on behalf of the U.S. Navy for potential general use in ecological risk assessments in California. These TRVs include both low-dose and high-dose values, representing doses at which toxic effects would rarely be expected and doses at which toxic effects would always be expected. These TRVs are currently in draft form, have not been peer reviewed, and represent generally conservative values drawn from a review of the toxicological literature. Rather than selecting TRVs from individual toxicological studies that are relevant to the ecological receptors and exposure routes at a particular site, the Region 9 BTAG TRVs represent a more general approach whereby a single representative TRV is allometrically adjusted (i.e., based on the difference between the body weight of the test species upon which the TRV was derived versus that of the ROI) to a site-specific receptor.

Although general use of the Region 9 BTAG TRVs has the advantage that each TRV may be drawn from an individual study that meets the criteria for high-quality data, it suffers from the disadvantage that the form of the chemical administered, the exposure duration and routes, the phylogeny of the test species, and other factors may have little or no relevance to the conditions and receptors selected for a particular evaluation. Therefore, the Region 9 BTAG TRVs should be used with caution and a recognition of their assumptions and limitations. These TRVs are currently used for most ecological risk assessments conducted on sediments sites in the San Francisco Bay.

Because little or no data are available on toxicity of mercury to many of the receptor species of concern, toxicity data for other related species must be extrapolated. For example, a mammalian TRV for mercury based on reproductive effects to rats may be applied to evaluate potential impacts to harbor seals. Although the TRV can be allometrically converted to account for large differences in body weights, this type of assessment essentially assumes that mercury will affect seal reproduction in a manner similar to the way it affects rat reproduction. In addition, evaluation of effects in this type of assessment is based on an average daily dose of a mercury. Although this approach generally assumes that exposure is over a period of at least several months, it does not consider the potential cumulative effects that may occur due to long-term accumulation of pollutants in the tissue of upper-trophic-level receptors. For example, the life span of harbor seals is generally 20-30 years (USEPA 1993), and the accumulated body burden of mercury in an older seal is likely to be significantly higher than it would be in a younger seal. Mercury tends to accumulate mainly in muscle tissue. Long-term accumulation of mercury may have additional effects that are not accounted for in the daily dose approach. However, insufficient data currently exist to estimate bioaccumulation factors in seals and to link tissue concentrations with adverse effects.

Risk Estimation

The hazard quotient (HQ) provides a mathematically derived index that expresses the relationship between predicted exposure concentrations or doses and derived toxicity doses (TRVs). If the HQ is greater than 1, i.e., the predicted dose exceeds the TRV, this is an indication that the receptor might be at risk of adverse effects from the particular COPEC. If the HQ is less than 1, then the predicted exposure is less than the toxic dose, and adverse effects are not expected. In other words, the HQ is simply the ratio of the exposure estimate to an effects dose considered to represent a “safe” environmental dose.

Factors Affecting Bioavailability and Bioaccumulation

This section discusses the factors that affect bioavailability and bioaccumulation of mercury in the aquatic system, and provides a foundation for estimating or modeling invertebrate and fish tissue concentrations on both a Bay-wide and site-specific basis. These estimated tissue concentrations could then be used for both human health and ecological risk assessment purposes.

Bioavailability

As mercury is transported within the Bay, mercury species undergo transformations and form compounds with other chemical components of the aquatic environment including chlorine, hydroxide, sulfide, and organic material. The types of compounds that mercury forms can influence the availability of mercury for bio-geochemical reactions and thereby govern the cycling of mercury between species. Mercury occurs naturally in the environment in a variety of valence states and conjugations, such as Hg^0 (elemental mercury), Hg^{2+} (dissolved), HgS (the ore cinnabar), and as an organometal such as methyl mercury (CH_3Hg and $(CH_3)_2Hg$) (Jones and Slotten 1996). Chemical speciation terms commonly include total, inorganic, organic, and methyl-mercury. Inorganic mercury includes elemental mercury and some complexes of the mercurous (Hg^{1+}) and mercuric (Hg^{2+}) oxidation states. Organic mercury includes organomercury salts (complexes with humic/fulvic acids and amino acids) and other compounds such as methylmercury, dimethylmercury, and phenylmercury. Some of the most common forms of mercury in the Bay are briefly discussed below.

Elemental mercury. Elemental mercury occurs as a liquid at room temperature, but has a high vapor pressure and a portion occurs as mercury vapor. Elemental mercury in sediment and water is thought to be unavailable for methylation, although under certain conditions can be oxidized to Hg^{2+}. This can occur through chemical reactions with chlorine, and through photo-oxidation. Elemental mercury may readily diffuse across the air-water interface (Bale 2000), while other chemical forms of mercury diffuse across this interface at a comparatively slower rate.

Monomethylmercury (MeHg). MeHg (CH_3Hg^+) is formed by microorganisms in both sediment and water through the methylation of inorganic mercuric ions Hg^{2+}. MeHg is a charged lipophilic, organic form of mercury that is highly toxic and readily accumulated by aquatic organisms. MeHg generally occurs in seawater environments as the chloride salt (CH_3HgCl). The average MeHg percentage (0.77%) of total mercury found by Kannan et al. (1998) in south Florida estuarine sediments is similar to that observed by Bartlett and Craig (1981) in British estuarine sediments (0.46%). This percentage is much higher in freshwater sediments (up to 37%) (Bechvar et al. 1996).

Dimethylmercury. Dimethylmercury ($(CH_3)_2Hg$) is also formed by microorganisms through the methylation of inorganic mercuric ions Hg^{2+}. Dimethylmercury is uncharged and highly volatile and is generally not persistent in aquatic environments (Beckvar et al. 1996). For this reason, any reference to MeHg within this report refers to monomethylmercury, unless otherwise stated.

Speciation of mercury is affected by environmental conditions such as pH, dissolved oxygen, sulfide concentration, chloride concentration, organic matter content, redox potential, and microbial activity. Mercury methylation is the biologically mediated transformation of Hg^{2+} to MeHg. In order for mercury methylation to take place, there must be a bioavailable source of mercury, and conditions within the system must be conducive to mercury methylating bacteria.

Most mercury is released into the environment as inorganic mercury, which is primarily bound to particles and organic substances and may not be available for direct uptake by aquatic organisms (Beckvar et al. 1996). Inorganic forms of mercury are a source for reactive Hg^{2+}, which is readily methylated by bacteria to methylmercury. Nearly all mercury found in fish and wildlife is methyl mercury. Biological activity near the anoxic-oxic interface of the sediment bed produces methyl mercury at rates that depend upon ambient conditions. The methyl mercury produced by benthic methylation disperses into overlying waters and may be available for uptake by organisms. As a first approximation, species of mercury may be determined at representative locations in the Bay by applying chemical equilibria models to observed or assumed concentrations of dominant water chemistry constituents.

Sulfate-reducing bacteria in anaerobic sediments have been implicated as primary methylators of inorganic mercury (King et al. 1999). Microbial inhibition studies utilizing molybdate, an inhibitor of sulfate reduction, effectively reduced the methylation of mercury in sediments by 95% (Compeau and Bartha 1985). As explained previously, sulfate reduction is the primary pathway for organic carbon decomposition in salt marshes. In freshwater systems, sulfate reducers are also present, but generally limited in activity by lower sulfate concentrations. Sulfate reduction occurs primarily in the sub-oxic zone of sediment, occurring just below the sediment water interface.

In order for sulfate reducing bacteria to methylate mercury they require an available source of mercury, sulfate, and organic matter. In addition, sediment chemical parameters including redox potential, oxygen concentration, and salinity need to be conducive to sulfate reduction. In general, methylation rates have been found to be higher a) under anoxic conditions, b) in freshwater compared to saltwater, and c) in low pH environments (Beckvar et al. 1996).

Although sulfate feeds the mercury methylation process, the resultant sulfide produced by sulfate reduction has significant effects on the bioavailability of mercury. Sulfide binds with mercury to form mercury sulfide (HgS), which is largely unavailable to biota. Field studies have generally shown a decrease in methylmercury production as sulfide concentration increases (Gilmour and Henry 1991). Gilmour et al. (1998) found MeHg concentration and production were inversely related to sulfate reduction rate and pore water sulfide, further showing how increased sulfide concentrations can result in decreased mercury methylation. Pak and Bartha (1998) also found mercury methylation rates far lower in high-sulfate estuarine sediments, as opposed to low-sulfate freshwater sediments. In addition to repressing mercury methylation by limiting the amount of bioavailable mercury, high sulfide concentrations may also be toxic to certain sulfate reducing bacteria, further reducing methylation rates.

In inter-tidal systems, there is a constant input of sulfate from ocean waters. Sulfate in marine systems is normally around 28 mM, and therefore is not a limiting or controlling parameter in saltmarsh sulfate reduction.

Since sulfate reducers are primarily responsible for mercury methylation, one would expect a positive correlation between sulfate levels and mercury methylation rates. King et al. (1999) developed a model to predict mercury methylation rates based on sulfate reducing rates and inorganic mercury concentrations in a saltmarsh ecosystem. The model provided a reasonable estimate of mercury methylation rates observed in the initial 12 hours of slurry incubations, but it was concluded that other factors affecting the bioavailability of mercury need to be included in future models. Choi and Bartha (1994) also found a correlation between mercury methylation rates and sulfate reducing rates. However, this correlation occurred at a constant salinity of 7 ppt, and mercury methylation rates were found to correlate more closely with the concentration of sediment organic matter at higher salinities (7-20 ppt).

Concentrations of total mercury and MeHg were found to increase with increasing concentrations of DOC in remote Adirondack lakes (Driscoll et al. 1995). Similarly, Ravichandran et al. (1998) found that hydrophobic (a mixture of both humic and fulvic) acids caused a dramatic release of mercury from HgS. Total mercury and MeHg have been shown to have different distribution patterns in the suspended particulate, colloidal, and dissolved phase of organic carbon. Cai and Jaffe (1999) found the majority of total mercury to be associated with colloidal forms, while MeHg was present almost entirely in the lower molecular weight fraction of the colloids and in the truly dissolved fraction. Increased amounts of DOC promotes microbial respiration, however, as discussed earlier, increased sulfate reduction rates have varying effects on the amount of MeHg produced. Increased DOC levels may inhibit methylation due to the binding of free mercury ions (Winfrey and Rudd 1990).

Sulfate reduction, and therefore mercury methylation, occurs primarily under anaerobic conditions, below the point where oxygen from overlying waters is present. The amount of oxygen in sediment and water affects the redox potential, which dictates whether methylating or demethylating bacteria will dominate. The addition of dissolved oxygen to sediment and water could inhibit methylation. Also, during bioturbation or dredging, when formally anoxic sediment is exposed to dissolved oxygen, sulfide can be oxidized to sulfate, releasing inorganic mercury to the system.

Lee and McFarland (2000) described how conditions of low dissolved oxygen (DO) in overlying waters can increase Hg bioavailability during bioturbation. In waters with low DO, oxidation of disturbed sediments proceeds more slowly, allowing more time for inorganic mercury to stay available for methylation. In waters with higher levels of DO, any mercury released from insoluble HgS will become re-immobilized by sorption with iron and manganese oxides (Hammer et al. 1988).

Mercury methylation appears to decrease as salinity increases in estuarine environments (Compeau and Bartha 1984, Gilmour and Capone 1987). This may be due the bicarbonate component of seawater slowing methylation (Compeau and Bartha 1985) and the increased chloride ions binding with mercury to decrease the amount of reactive mercury (Craig and Moreton 1985).

The growth cycle of certain plants within a saltmarsh can have an effect on mercury methylation rates. Marsh grasses release dissolved organic carbon (DOC) and dissolved oxygen (DO) through rhizomes during certain times of year, which can affect mercury bioavailability as well as redox potential in the surrounding sediments. The increase in DOC promotes microbial respiration in general, while the increase in DO to the system could help create an oxic environment to aid in the survival of non-sulfate reducing species. In addition, there is a potential for increased sediment DO to oxidize mercury compounds, providing additional sources of bioavailable mercury.

Seasonal changes in mercury methylation rates are related to temperature, hydrodynamics, and inputs from vegetation. A review of multiple mercury methylation studies by Bechvar et al. (1996) revealed that mercury methylation rates may peak with high temperature and biological productivity in the summer months and decrease during winter months when biological productivity and temperature are low.

Because both inorganic and organic species of mercury may be securely bound to sediment materials, the sediment bed is often a repository for mercury discharged or released into the aquatic environment (Bale 2000). As sediment burial depth increases, the sediment becomes compacted and further away from the biologically active sediment-water interface. Re-suspension of the sediments by dredging, bioturbation, or as a result of winter storm discharges, can often disrupt this burial. However, natural sediment burial can be an effective way of reducing bioavailable mercury if the incoming sediments are not contaminated, and preventive measures are taken to limit resuspension.

A model study by Bale (2000) indicated that the mercury burden in Clear Lake, California is steadily declining due in great part to burial of surficial sediments.

Bioaccumulation.

Invertebrates accumulate mercury in a similar fashion to fish. However, the concentrations in invertebrates have been found to be highly variable, and the percentage of methylmercury has been found to be lower than with fish or mammals (Beckvar et al. 1996). This may be due to the fact that invertebrates are generally tested whole and may have inorganic mercury in their stomach and attached to portions of their body. The percentage of methylmercury, as compared to total mercury, increases with age in both fish and invertebrates (Beckvar et al. 1996). Some of the highest mercury tissue concentrations of all marine organisms investigated have been found in fish-eating marine mammals (Andre et al. 1991). Gardner et al. (1978) found that the percentage of mercury present as the organic form increased with trophic level and was essentially 100% in most birds and mammals.

Data on mercury and methyl mercury concentrations in sediment, water, and invertebrate and fish tissue have been collected in various areas throughout the Bay, under several types of investigations. Much of these data have been compiled and are summarized in Table 2, but additional data exist that are not yet included in the table. These data can be used to develop empirical BAFs and/or BSAFs. However, it should be noted that these factors are site-specific, and can vary dramatically based on the factors described above. When possible, BAFs and/or BSAFs should be developed for each sediment site that has been identified as a significant source of mercury. Bioaccumulation data will help to determine the contribution of mercury from that site to overall risk within the Bay. Even though a given site may be a significant contributor in terms of mass loading, the impact of that site in terms of risk may not be significant if the bioavailability of mercury is low. When site-specific data are not available, the potential for mercury methylation and bioaccumulation should be estimated by evaluating the factors that influence methylation, and identifying empirical BAFs or BSAFs developed for similar sites.

Laboratory studies of methylation and demethylation are always an option, but they are frequently difficult to carry out at environmentally relevant mercury levels and may be difficult to interpret. Alternate approaches which focus on net methyl Hg production and bioaccumulation, have the advantage of also providing data necessary for loading calculations and determination of localized human and wildlife health risk. These data also provide a set of relevant baselines for use in the assessment of any future remedial work. By linking localized bioaccumulation to corresponding aqueous and sediment chemistry, site-specific driving parameters may be identified for net Hg methylation, bioaccumulation, and export. Meaningful reductions, based on this information, may be obtained through specific load reductions and/or habitat manipulations.

It should be noted that most of the fish tissue data collected in the San Francisco Bay has been collected for human health risk assessment purposes, and may not be appropriate for use in ecological risk assessment. One reason is that it is often only the filet portion of the fish that is analyzed, and concentrations in this portion may not be representative of the whole body concentrations. Another reason is that sport fish are often large fish that are not normally consumed by receptors such as birds and marine mammals. The type of fish tissue data that is generally most useful for evaluating ecological risk to birds and mammals is whole body tissue analysis of small fish, preferably resident species with small home ranges.

Site-Specific Contribution to Bay-Wide or Regional Risk

As previously stated, most upper trophic level receptors have large home ranges and are likely to forage in multiple locations. Mercury concentrations in sediments throughout San Francisco Bay are elevated, and the portion of the daily dose to an individual receptor that can be attributed to a specific site may be very small. The contribution of mercury contamination at a specific site to Bay-wide or regional ecological risk can be estimated based on the size of the receptor's home range. For example, assuming that the cormorant has a home range of approximately 200 km^2 and the size of a particular contaminated site is 5 km^2 (or 2.5% of the total home range), the fraction of the daily dose of mercury attributed to that site would be calculated as:

(0.025 X Hg conc. in prey tissue at site + 0.975 X Hg conc. in ambient prey tissue) X daily ingestion rate

The resulting daily dose can be compared to the daily dose that would result if the contaminated sediment site were not there. This daily dose would be simply:

Hg conc. in ambient prey tissue X daily ingestion rate

The daily doses with and without the contaminated site included could be compared to evaluate the magnitude of change to the daily dose. This calculation assumes that the cormorant obtains equal amounts of food from all regions of its home range. However, some habitats may be more suitable than others for foraging, and this can also be taken into account if sufficient foraging data are available. If these data are available, factors can be applied to the dose equation for a receptor to account for the expected duration spent at each suitable foraging habitat.

A similar method could be used to evaluate the site-specific contribution of each site to human health risks in terms of fish tissue concentrations. The home ranges of different species of sport fish could be used to determine how tissue concentrations would change if mercury contamination at a given site is present or not present. Sediment and/or invertebrate prey tissue concentrations could be spatially averaged over the home range of the fish species with and without inclusion of a specific site, and the relative change could be evaluated.

REMEDIATION OF CONTAMINATED SEDIMENT SITES

The information presented in Table 1 provides some rough estimates of the relative amount of mercury at each sediment site in terms of total mass. While this information is useful to get an idea of the magnitude of mercury at each site, it does not provide information about the mass loading or flux from sediment into the water column. Of equal importance to the loading numbers are questions of (1) relative bioavailability of the specific inorganic mercury pools for methylation and (2) localized and regional mercury bioaccumulation patterns (indicators of net mercury methylation and food web transfer).

It is likely that if mercury concentrations at a given site are high enough over a large enough area to have a significant effect on Bay-wide mass loading and risk, there would also be significant site-specific risk that would drive cleanup under other regulatory programs. However, TMDL requirements could help to establish cleanup levels and select remedial alternatives for some sites. In addition, the TMDL process could help to identify additional sites with high concentrations of mercury that are not currently being addressed under any regulatory program.

RECOMMENDATIONS

Although the contribution of mercury loads from bedded sediments within the San Francisco Bay is believed to be significant, less effort has gone into quantifying this input as compared to other sources. A more thorough evaluation of the spatial variability of mercury concentrations in sediments, as well as spatial patterns in the relative bioavailability of mercury, would provide valuable information to support the assessment of potential control actions considered for the TMDL Implementation Plan. The following steps are recommended:

1. **Review Data**. Compile additional existing data on mercury and methylmercury concentrations in sediment, water, and tissue, and conduct a thorough assessment of these data. As stated earlier, the information reviewed for this report was limited by the available time funding, and the estimates provided in Table 1 are very rough.

2. **Identify Ongoing Studies**. Various studies are in progress throughout the Bay to investigate mercury processes in sediment and water. For example, SFEI is starting work on a food web bioenergetics model for mercury. Gary Gill of Texas A&M University and Sam Kuabara of USGS - Menlo Park are conducting benthic flux studies that may be useful as a means of obtaining diffusive flux terms in irrigated benthos for complex or simple models.

3. **Develop Bioaccumulation Factors**. Utilize the available data to identify spatial patterns in mercury methylation and bioaccumulation. This should include investigation of how various factors (redox potential, habitat types, organic content, etc) quantitatively affect these processes. Using the data compiled in Step 1, bioaccumulation factors could be calculated for different micro-environments within the Bay.

4. **Target Potential Problem Sites**. Use the available data to estimate the contributions of various areas throughout the Bay in terms of methylmercury production and bioaccumulation potential. Use this information to target sites that: (a) contain high concentrations of mercury in sediments; (b) provide an environment conducive to methylation and bioaccumulation; and (c) are of significant magnitude in terms of mass of mercury.

5. **Develop Mass Loading Model**. Determine the modeling approach to estimate the mercury load from in-situ sediments within the Bay. Flux modeling should not be done just for individual hot spots, but must be put into context with the loads from Bay-wide sediments containing ambient concentrations of mercury. Model results should provide information that can be used: (a) to determine the magnitude of the total mercury load from in-situ sediments, for comparison to other sources of mercury; and (b) to evaluate and compare contributions of mercury from individual sites or areas within the Bay.

6. **Assess Risk**. Develop a risk assessment approach to predict impacts to human and ecological receptors. Use this approach to estimate Bay-wide risk, and compare contributions from various sites or areas within the Bay, including those sites identified under Step 4.

7. **Identify Significant Sites and Potential Control Actions**. Use results of mass loading and risk assessment to identify sites that are significant contributors in terms of both mass loading and Bay-wide risk. For sites currently under investigation or remedial action, evaluate cleanup alternatives in terms of load reductions and reduced risk. For sites for which no actions are planned, identify regulatory processes that can be used to facilitate remediation.

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Table 1: Preliminary summary of contaminanted sites						Mercury Concentration Sediment (mg/kg)					
Site Name	Sample Dates	Regulatory/ Remedial Status	Approximate Aerial Extent (ft^2)	# of Samples	Estimated Depth (ft)	Max	Min	Mean	Estimated Volume (ft^3)	Estimated Mass of Sediment (kg)	Estimated Mass of Hg (kg)
Naval Station Treasure Island - Area B	1993-2001	no remediation planned	1.92E+06	13	4	1.2	ND	0.62	7.68E+06	7.68E+06	4.76
Naval Station Treasure Island - Area E	1993-2001	no remediation planned	1.00E+06	10	2	1	ND	0.51	2.00E+06	2.00E+06	1.02
Hamilton Army Airfield	1991-2002	ongoing remediation	2.50E+06	119	2	8.4	0.096	0.6	5.00E+06	5.00E+06	3.00
UC-Berkeley Richmond Field Station	2001	remediation 2002-2004	1.00E+06	84	8	430	< 0.19	16	8.00E+06	8.00E+06	128.00
Zeneca - Stege Marsh	1992-2000	remediation 2002-2004	5.40E+05	49	8	73	0.19	5.2	4.32E+06	4.32E+06	22.46
Alameda Seaplane Lagoon	1993-1997	feasibility study underway	7.00E+06	99	5	2.6	0.03	1.04	3.50E+07	3.50E+07	36.40
Castro Cove (Chevron data)		remediation plan to be submitted Oct 2002	9.60E+05	25	2	17	0.5	2.3	1.92E+06	1.92E+06	4.42
Point Potrero	1995	?	1.30E+05	3	5	9.1	0.33	4.7	6.50E+05	6.50E+05	3.06
Pacific Dry Dock (BPTCP data)	1995	?	unknown	4	4	1.75	0.85	1.3	unknown	unknown	unknown
San Leandro Bay	1995	no action currently planned	1.94E+06	2	2	0.87	0.68	0.77	3.88E+06	3.88E+06	2.99
SF Airport (BPTCP data)	1995	no action currently planned	unknown	1	2	1.87	1.87	1.87	unknown	unknown	unknown

Notes:

= Not Detected

Table 2: DATA ON MERCURY AND METHYLMERCURY IN SEDIMENT. WATER. AND TISSUE

					Mercury Concentration											
					Sediment (mg/kg)			Water (mg/L)			Bivalve Tissue (mg/kg)			Fish Tissue (mg/kg)		
Site Name	Sample Dates	Type of Mercury	# of Samples	ER-M (mg/kg)	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean
ALAMEDA																
Mussel Watch - Alameda Yacht Harbor	1985-1988	Mercury	5	0.71							1.348	0.282	0.7428			
RMP - Alameda	2000	Methyl Mercury	2	0.71				8.50E-08	1.00E-08	4.80E-08						
RMP - Alameda	1993-2000	Mercury	20	0.71	0.41	0.11	0.284	1.60E-06	4.00E-07	1.00E-06						
Alameda NAS - Seaplane Lagoon	6/1/01	Mercury	208	0.71	2.6	0.03	1.04							0.313	0.0594	0.142
CASTRO COVE																
Mussel Watch - Castro Cove Bridge	1988	Mercury	2	0.71							0.6625	0.532	0.5973			
Castro Cove - Chevron		Mercury	25	0.71	13	0.5	2.3									
Bay Protection - Castro Cove	1995	Mercury	1	0.71	2.9	2.9	2.9									
SOUTH BAY																
Mussel Watch - Dumbarton Bridge/Channel Marker 14	1980-1991	Mercury	14	0.71							0.842	0.187	0.4569			
RMP - Dumbarton Bridge	2000	Methyl Mercury	1	0.71	6.08E-04	6.08E-04	6.08E-04									
RMP - Dumbarton Bridge	1993-2000	Mercury	15	0.71	0.472	0.13	0.326	6.40E-06	9.00E-07	2.00E-06	0.58	0.199	0.282			
Mussel Watch - San Mateo Bridge/ 8B	1982-1993	Mercury	12	0.71							0.628	0.12	0.3829			
RMP - South Bay Bridges	2000	Methyl Mercury	1	0.71	1.52E-03	1.52E-03	1.52E-03	7.00E-09	7.00E-09	7.00E-09						
RMP - South Bay Bridges	1993-2000	Mercury	12	0.71	0.51	0.12	0.314	3.80E-06	1.10E-06	2.00E-06				1.05	0.08	0.44
CalFED Dust Marsh, SFB/Coyote Hills Slough	12/7/99	Methylmercury	1	0.71	4.4E-04	4.4E-04	4.4E-04									
CalFED Dust Marsh, SFB/Coyote Hills Slough	12/7/99	Mercury	1	0.71	0.1	0.1	0.1									
CalFED SFB Marsh/ Coyote Hills Slough South	12/6/1999	Methylmercury	1	0.71	1.1E-04	1.1E-04	1.1E-04									
CalFED SFB Marsh/ Coyote Hills Slough South	12/6/1999	Mercury	1	0.71	0.1	0.1	0.1									
CalFED SFB Mudflat	12/6/1999	Methylmercury	2	0.71	3.5E-04	2.2E-04	2.9E-04									
CalFED SFB Mudflat	12/6/1999	Mercury	2	0.71	0.3	0.2	0.3									
CalFED SFB Marsh/Mudflat	12/6/1999	Methylmercury	1	0.71	1.2E-04	1.2E-04	1.2E-04									
CalFED SFB Marsh/Mudflat	12/6/1999	Mercury	1	0.71	0.1	0.1	0.1									
CalFED South SFB Marsh	12/10/1999	Methylmercury	1	0.71	5.3E-04	5.3E-04	5.3E-04									
CalFED South SFB Marsh	12/10/1999	Mercury	1	0.71	0.3	0.3	0.3									
CalFED SFB near Cooley Landing/Mayfield Slough	12/10/1999	Methylmercury	1	0.71	6.8E-04	6.8E-04	6.8E-04									
CalFED SFB near Cooley Landing/Mayfield Slough	12/10/1999	Mercury	1	0.71	0.3	0.3	0.3									
CalFED SFB/ South Bay Central channel	12/10/1999	Methylmercury	1	0.71	6.3E-04	6.3E-04	6.3E-04									
CalFED SFB/Coyote Creek Mouth	12/10/1999	Methylmercury	1	0.71	7.4E-04	7.4E-04	7.4E-04									
CalFED SFB/Coyote Creek Mouth	12/10/1999	Mercury	1	0.71	0.3	0.3	0.3									
CalFED Guadalupe Slough mouth	12/10/1999	Methylmercury	1	0.71	7.9E-04	7.9E-04	7.9E-04									
CalFED Guadalupe Slough mouth	12/10/1999	Mercury	1	0.71	0.4	0.4	0.4									
CalFED Guadalupe Slough @ Sunnyvale	12/10/1999	Methylmercury	1	0.71	1.1E-03	1.1E-03	1.1E-03									
CalFED Guadalupe Slough @ Sunnyvale	12/10/1999	Mercury	1	0.71	0.3	0.3	0.3									
CalFED Alviso Slough	12/10/1999	Methylmercury	1	0.71	1.3E-03	1.3E-03	1.3E-03									
CalFED SFB/ South Bay Central channel	12/10/1999	Mercury	1	0.71	0.3	0.3	0.3									
CalFED Alviso Slough	12/10/1999	Mercury	1	0.71	0.5	0.5										
Selby Slag Sites 2&	17/94 - 5/4/	Mercury	20		1.70	0.32	0.07				0.06	0.25	0.89			
OAKLAND																
Mussel Watch - Oakland Back Harbor	1986-1999	Mercury	4	0.71							0.779	0.262	0.5393			
Mussel Watch - Oakland Inner Harbor West	1986-1987	Mercury	2	0.71							0.475	0.14	0.3075			
Mussel Watch - Oakland Harbor/Embarcadero	1986-1993	Mercury	7	0.71							0.8083	0.37	0.563			
RMP - Oakland	1997	Mercury	10	3.71	na	na	na	na	na	na				0.25	0.13	0.17
RICHMOND																
Mussel Watch - Point Pinole	1980-1993	Mercury	15	0.71							0.5	0.165	0.3438			
RMP - Pinole Point	2000	Methyl Mercury	1	0.71	2.30E-04	2.30E-04	2.30E-04	3.10E-06	4.00E-07	1.00E-06	1.936	0.106	0.385			

Table 2: DATA ON MERCURY AND METHYLMERCURY IN SEDIMENT. WATER. AND TISSUE

					Mercury Concentration											
					Sediment (mg/kg)			Water (mg/L)			Bivalve Tissue (mg/kg)			Fish Tissue (mg/kg)		
Site Name	Sample Dates	Type of Mercury	# of Samples	ER-M (mg/kg)	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean
RMP - Pinole Point	1993-2000	Mercury	15	0.71	0.56	0.1	0.275									
Mussel Watch - Richmond Inner Harbor Basin	1985-1988	Mercury	4	0.71							0.575	0.361	0.4578			
Mussel Watch - Richmond/San Rafael Bridge	1980-1982	Mercury	5	0.71							0.342	0.149	0.2704			
Bay Protection - Potrero Point - Warm Water Cove	1995	Mercury	1	0.71	0.33	0.33	0.33									
Bay Protection - Point Potrero Site 1	1995	Mercury	1	0.71	4.6	4.6	4.6									
Bay Protection - Point Potrero Site 2	1995	Mercury	1	0.71	9.14	9.14	9.14									
Zeneca	1992-2000	Mercury	49	0.71	73	0.19	5.2									
RMP - Point Isabel	2000	Methyl Mercury	1	0.71	4.26E-04	4.26E-04	4.26E-04									
RMP - Point Isabel	1993-2000	Mercury	15	0.71	0.33	0.13	0.267	2.10E-06	3.00E-07	1.00E-06						
San Bruno/South SF Area																
Mussel Watch - San Francisco Airport	1982	Mercury	1	0.71							0.317	0.317	0.317			
RMP - Oyster Point	2000	Methyl Mercury	1	0.71	4.46E-04	4.46E-04	4.46E-04									
RMP - Oyster Point	1993-2000	Mercury	15	0.71	0.281	0.12	0.211									
RMP - San Bruno	2000	Methyl Mercury	1	0.71	1.78E-03	1.78E-03	1.78E-03									
RMP - San Bruno	1993-2000	Mercury	13	0.71	0.307	0.1	2.42									
RMP - San Bruno Shoal	2000	Methyl Mercury	1	0.71				2.00E-09	2.00E-09	2.00E-09						
RMP - San Bruno Shoal	1993-2000	Mercury	23	0.71				5.80E-06	3.00E-07	1.00E-06						
RMP - Oyster Point	1993-2000	Mercury	1	0.71				2.20E-08	2.20E-08	2.20E-08						
RMP - Oyster Point	1993-2000	Mercury	23	0.71				6.30E-06	4.00E-07	1.00E-06						
Bay Protection - S.F. Airport - Central	1995	Mercury	1	0.71	1.87	1.87	1.87									
CENTRAL BAY																
Mussel Watch - San Francisco Bay/Angel Island	1980-1982	Mercury	5	0.71							0.304	0.086	0.2164			
Mussel Watch - San Francisco Bay/ Hunter's Point	1981-1993	Mercury	10	0.71							0.385	0.21	0.3258			
Mussel Watch - San Francisco Bay/ near Redwood Cr.	1981-1993	Mercury	13	0.71							0.654	0.2	0.4408			
Mussel Watch - San Francisco Bay/ Treasure Is.	1980-1993	Mercury	17	0.71							0.471	0.196	0.303			
RMP - Yerba Buena Island	2000	Methyl Mercury	1	0.71				1.10E-08	1.10E-08	1.10E-08						
RMP - Yerba Buena Island	1993-2000	Mercury	23	0.71				1.90E-06	4.00E-06	1.00E-06	0.37	0.19	0.26			
Mussel Watch - San Francisco/ Islais Channel	1987-1988	Mercury	2	0.71							0.714	0.709	0.7115			
Bay Protection - North India Basin - Site 2	1995	Mercury	1	0.71	0.89	0.89	0.89									
Bay Protection - Central Basin - Inner	1995	Mercury	1	0.71	0.74	0.74	0.74									
RMP - Yerba Buena Island	2000	Methyl Mercury	1	0.71	4.69E-04	4.69E-04	4.69E-04									
RMP - Yerba Buena Island	1993-2000	Mercury	15	0.71	0.346	0.12	0.229									
RMP - SF Waterfront	1997	Mercury	9	4.71										0.31	0.07	0.14
SAN LEANDRO BAY																
Mussel Watch - San Leandro Bay/Damon Channel	1999	Mercury	1	0.71							0.212	0.212	0.212			
Mussel Watch - San Leandro Bay/ Elmhurst Channel	1999	Mercury	1	0.71							0.168	0.168	0.168			
Bay Protection - San Leandro Bay Site -Site 1	1995	Mercury	1	0.71	0.87	0.87	0.87									
Bay Protection - San Leandro Bay Site -Site 2	1995	Mercury	1	0.71	0.68	0.68	0.68									
NORTH BAY																
RMP - San Pablo Bay	1993-2000	Methyl Mercury	1	0.71	2.80E-04	2.80E-04	2.80E-04									
RMP - San Pablo Bay	1993-2000	Mercury	15	0.71	0.17	0.394	0.326									

Table 2: DATA ON MERCURY AND METHYLMERCURY IN SEDIMENT. WATER. AND TISSUE

					Mercury Concentration											
					Sediment (mg/kg)			Water (mg/L)			Bivalve Tissue (mg/kg)			Fish Tissue (mg/kg)		
Site Name	Sample Dates	Type of Mercury	# of Samples	ER-M (mg/kg)	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean
RMP - Richardson Bay	1993-2000	Methyl Mercury	1	0.71	3.38E-04	3.38E-04	3.38E-04									
RMP - Richardson Bay	1993-2000	Mercury	15	0.71	0.408	0.11	0.247	3.00E-06	4.00E-07	1.00E-06						
RMP - San Pablo Bay	1993-2000	Mercury	23	0.71	na	na	na	3.30E-06	3.00E-07	1.00E-06	0.493	0.102	0.22	1.13	0.08	0.35
RMP - Red Rock	1993-2000	Methyl Mercury	1	0.71				1.13E-07	1.13E-07	1.13E-07						
RMP - Red Rock	1993-2000	Mercury	20	0.71	0.1	0.015	0.041	1.80E-06	2.00E-07	1.00E-06	0.351	0.196	0.287			
RMP - Berkeley	1997	Mercury	18	5.71										0.88	0.62	0.26

Note: This table includes only sampling locations for which methylmercury and/or tissue data are available. It does not include many additional sampling locations for which only total mercury data for sediment and/or water are available.