

# Conceptual Model and Working Hypotheses of Mercury Cycling and Transport in the Bay- Delta Ecosystem and its Tributaries

**By**

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## **SUMMARY OF MAJOR FEATURES**

Figure 1 depicts a conceptual overview of the transport and biogeochemical cycling of mercury in the Bay-Delta ecosystem and its watersheds. This cycle is based upon recent environmental mercury research; particularly the findings from the Calfed Mercury Project entitled "*Assessment of Ecological and Human Health Impacts of Mercury in the Bay-Delta Watershed*". The cycle also includes current working hypotheses describing Hg behavior in the Bay-Delta ecosystem. The major sources, losses, internal cycling processes, and environmental impacts depicted in the figure are summarized below.

### **Major inputs of total mercury to the Bay-Delta include:**

- Riverine input from two distinct sources, the Sierra and Coastal mountain ranges
- Atmospheric deposition

### **Major loss (or export) terms for mercury in the Bay-Delta include:**

- Estuarine mixing and transport to San Francisco Bay
- Freshwater export to Southern California
- Evasion of volatile Hg species, especially elemental mercury
- Particle settling and burial in sediments

### **Major internal biogeochemical cycling processes for mercury include:**

- Bioaccumulation of mercury (as monomethyl mercury) into the aquatic food chain
- Sediment-water exchange
- Production and destruction of monomethyl mercury (MMHg) in surficial sediments

### **Primary environmental impacts include:**

- Human health concerns from consumption of fish with elevated mercury content
- Reproductive impairment of birds which feed from the aquatic environment

## **PRIMARY WORKING HYPOTHESES**

Our work to characterize the major reservoirs and flows of mercury (Hg) and monomethyl mercury (MMHg) and biogeochemical processes in the Delta and San Francisco Bay estuary to date have lead us to develop the following working hypotheses. Primary evidence in support of these hypotheses follows.

### **Mercury Sources, Sinks, and Biogeochemical Processes**

1. Monomethyl mercury is removed from the water column along the transport pathway across the Delta as water flows from the Sacramento River to the export pumps in the southern Delta and to San Francisco Bay during summer months (low flow).
2. The riverine and estuarine transport (mass balance) of particulate Hg in the Delta dominates the flows of total Hg within the Delta during all river flow conditions; The Delta appears to be losing total Hg, perhaps from sediment erosion processes.
3. Rivers in both the Coastal and Sierra Ranges contribute to total Hg and MMHg in the Delta. Sediment concentrations of MMHg and MMHg/Hg<sub>T</sub> concentration ratios are different in the Coast and Foothill watersheds. The coast streams have higher Hg<sub>T</sub>. Both coastal and Sierra rivers have equivocal MMHg levels. The MMHg/Hg<sub>T</sub> ratio is higher in the Sierras.
4. *In situ* production of MMHg within surficial sediments and it's exchange flux into the water column from the sediments is the major source of MMHg to the Delta only during low river flow conditions.
5. Within the Delta, MMHg in sediments varies with season, geographically and by habitat type. Wetland and marsh regions appear to be major sites of MMHg production and are habitats of significant MMHg production (on a per square meter basis) compared to open water sites.

***Hypothesis 1: Monomethyl mercury is removed from the water column along the transport pathway across the Delta as water flows from the Sacramento River to the export pumps in the southern Delta and to San Francisco Bay during summer months (low flow)***

A special study was conducted in the summer of 2001 to ascertain the location where the decrease in MMHg concentration occurred. Three transects were run down the Sacramento River and out toward Suisun Bay, the water path from the main source to the main sink of methyl mercury. The largest drop in concentration occurred on all three cruises in the vicinity of, or immediately downstream of, Rio Vista (see Table 1). The drop in concentration was between 30 and 60 percent. There are several possible, but not mutually exclusive, explanations. First, electrical conductivity also began to increase in the vicinity of Rio Vista and the decrease in MMHg may be related to the increase in salinity (see Table 1). Second, the Sacramento River joins the Deep Water Ship Channel a mile upstream of Rio Vista. At this point the River abruptly changes from being narrow and shallow (<10 ft) to wide and deep (35 ft). Increased channel cross-section results in an increase in water residence time and a decrease in travel rate. The longer travel time may facilitate some new process or may make on-going rate dependent processes more efficient.

This hypothesis is also supported by mass balance estimates for the Delta. Average MMHg sources and sinks in the Bay-Delta estuary are depicted in Figure 2. Average river loads of MMHg for the study period were about 10 g/day (Task 1A). In contrast, while it is clear that exports were less than imports, a precise estimate of the export rate is more difficult as MMHg concentrations in 30 percent of the water column measurements were below detection. An unidentified loss of about 10 g/day is needed for mass balance. Mechanisms contributing to the loss are not known.

***Hypothesis 2: The riverine and estuarine transport (mass balance) of particulate Hg in the Delta dominates the flows of total Hg within the Delta during all river flow conditions; The Delta appears to be losing total Hg, perhaps from sediment erosion processes in Suisun Bay.***

Illustrated in Figures 3 and 4 are the mass balances of (total) Hg in the unfiltered and filtered phases, respectively. Taken together, these figures illustrate the importance of the suspended particulate fraction as a transport pathway for Hg in the Bay-Delta estuary. In fact, it appears that there is more total Hg leaving the system than can be accounted for by input

sources. The source of this Hg leaving the system is not known, but may come from sediment erosion processes in Suisun Bay.

***Hypothesis 3: Rivers in both the Coastal and Sierra Ranges contribute to total Hg and MMHg in the Delta. Sediment concentrations of MMHg and MMHg/Hg<sub>T</sub> concentration ratios are different in the Coast and Foothill watersheds. The coast streams have higher Hg<sub>T</sub>. Both coastal and Sierra rivers have equivocal MMHg levels. The MMHg/Hg<sub>T</sub> ratio is higher in the Sierras.***

The Sacramento River was the principal river source of filtered and total Hg, transporting 162 kg of total Hg or 58 percent of all imports (see Tables 2 and 3). Most of the material entered the Estuary in winter storm runoff (January through March). About 15 percent of the entire load (24 kg) was in a filter-passing form. Prospect Slough, which receives storm runoff from Hg mines in Putah and Cache Creeks as well as flow from the upper Sacramento Basin, transported the largest amount of Hg of any of the river systems in a single month (70 kg in March 2000).

The Sacramento River was also the major river source of MMHg to the estuary. The only exception was in March 2000 when the Yolo Bypass was flooded and Prospect Slough became the major source of organic mercury. In all other months the Sacramento River accounted for 60-85 percent of the total load. The load of total (or unfiltered) MMHg was highest in winter and spring (January to May). This was because both flow and unfiltered MMHg concentrations (0.08-0.34 ng/l) were largest then. Total MMHg concentrations averaged half this during the rest of the year (about 0.08 ng/L). The filter-passing MMHg fraction in the Sacramento River was about 45 percent of the unfiltered value. The major sources of total and MMHg to the Sacramento River are not known.

There was a higher methylation potential in sediment from the mountain streams in the Sierra area than in the mountain streams in Cache (Figure 5). This is primarily because the total Hg concentrations near the mine site stations in the coast range are relatively high at most stations (ie. > 3,000-35,000 ppb) which forces the methyl to total ratios to be very low. By the time the mercury is deposited to the valley streams and rivers the total Hg concentrations are almost always below 1000 ppb and are primarily <500 ppb. The valley streams of both mountain ranges contain equivalent concentrations of total and methyl Hg, and the methyl to total ratios are also equivalent. Therefore, the methylation potential starts out very different in the two mountain ranges but becomes similar as the mercury and methyl mercury is transported to the valley streams and Delta.

***Hypothesis 4: In situ production of MMHg within surficial sediments and its exchange flux into the water column from the sediments is the major source of MMHg to the Delta only during low flow conditions.***

As noted previously, the average river loads of MMHg for the study period were about 10 g/day; the average MMHg export rate from the Delta to both San Francisco and Southern California was about 6 g/day; and the average sediment-water exchange flux of MMHg in the Delta and Suisun Bay was estimated at about 6 g/day (Figure 2). During high flow conditions the riverine sources are about 6-fold higher than the benthic input (~ 35 g MMHg/day) and during low flow conditions the riverine dissolved MMHg sources are about one quarter that of the benthic input (~ 1.6 g MMHg/day). This assessment is based on the assumption that the dissolved load is about half the total.

***Hypothesis 5: Within the Delta, MMHg in sediments varies with season, geographically and by habitat type. Wetland and marsh regions appear to be major sites of MMHg production and are habitats of significant MMHg production (on a per square meter basis) compared to open water sites.***

This hypothesis is supported by several lines of independent evidence, including geographical sediment Hg concentration and time series data; MMHg bioassay studies; surface water transect studies; and time series measurements.

Six sites within the Bay-Delta system were selected for monthly or bi-monthly monitoring for MMHg concentrations in sediments (Figure 6). Solid phase MMHg showed distinct seasonal patterns at locations within the central Delta and Cosumnes River (Figure 6). MMHg concentrations at Franks Tract, Connection Slough, and Cosumnes River were elevated twice per year (Figure 6 panels c,d,f). The largest peak in magnitude occurred during summer and the peak lesser in magnitude occurred over winter.

The production and bioaccumulation of MMHg in aquatic environments can vary widely. Wetlands in particular are often considered to be regions of high MMHg production potential (St. Louis et al., 1994, 1995; Hurley et al. 1995, Krabbenhoft et al. 1999; Rudd, 1995). Three wetland areas in the Delta were investigated to determine if they have high levels of methyl

mercury in their sediments. At each of three areas a transect was conducted that consisted of three stations, one inner station furthest into the interior of the wetland, one midway from the inner station to the outside of the wetland, and one just outside of the wetland. Sediments were collected from the top two cm. of a core. Transect results are shown in Figures 7 and 8. Webber Pt., Mandeville, and 14 Mile Slough wetland areas showed a consistent trend of higher MMHg, methyl/total mercury, and sediment LOI levels in the inner part of the wetlands and lower in the outside channels.

Dr. Mark Marvin (USGS) and his research group sampled five sites during February/March for Hg-methylation potential rates, 4 sites were in the Delta (Sherman I., Connection Slough, Franks Tract, and Prospect Slough,) and one was in a tributary (Cosumnes R.) (Figure 9). Of these four Delta sites, the organic rich (28% loss on ignition) Franks Tract site had the highest rates of Hg-methylation, compared to the lower organic sites (5-14% loss on ignition). This suggests that organic rich zones within the delta generally have higher rates of MMHg production. However, for this same sampling event, the tributary site (Cosumnes R.) exhibited the highest MMHg production potentials, even though the organic content was comparatively low (7% LOI). Thus, organic content alone can not explain the spatial variations observed in MMHg production rates when both delta sites and tributary locations are considered. Microbial sulfate reduction (SR) rates were a much stronger explanatory variable, across all sites, during this period. The positive relationship between SR and Hg-methylation was linear ( $r^2 = 0.70$ ,  $P < 0.17$ ,  $n = 4$ ) for 0-2 cm surface sediment although it was not significant. So to the extent that microbial sulfate reduction is higher in marsh sediments than non-marsh sediments, it can be speculated that the hypothesis is supported, at least during late winter/early spring, the period prior to build up of increased concentration of reduced sulfur, which can actually inhibit Hg-methylation. There was a strong seasonal signal in both MMHg production and degradation potentials (Figure 9). MMHg production was highest during February/March and decreased in May and October. The reverse trend was observed for MMHg degradation potentials. A non-linear decrease in MMHg production (assumed net production) was observed with increasing MMHg degradation, suggesting a complex relationship between these two opposing processes.

Surface water transect studies were conducted in Frank's Tract moving from near shore to open water to investigate the importance of near shore marsh areas as habitats of particular importance to the production of MMHg. Unfiltered and filtered water samples were collected from the Franks Tract marsh site and three transect stations in Franks Tract (Figure 10). Total (unfiltered) Hg was elevated only in the open water of Franks Tract (station

FT3) and was fairly constant in other stations. Filter- passing Hg did not show any clear evidence of a surface water concentration gradient between near shore and open waters within Franks Tract. However, MMHg revealed a significant surface water concentration gradient within Franks Tract. The highest total and filter-passing MMHg concentrations occurred at the marsh site. This result supports the hypothesis that sediment-water exchange of MMHg in near shore marsh environments influences water column MMHg concentrations in Frank's Tract, and probably other similar habitats in the Delta as well.

Time series measurements of Hg and MMHg were conducted at the entrance to Frank's Tract Marsh (Figure 11) in May 2001 to monitor changes in Hg concentration associated with water entering and leaving the marsh during a tidal event. Both Hg and MMHg concentrations in unfiltered and filtered surface waters changed with time as water flowed into and out of the marsh, driven by a tidal cycle. Mercury concentrations were lowest during high tide and highest between high and low tides when the water flow rate was maximum. MMHg concentrations were highest during low tide and lowest during high tide. As the water level dropped from high tide to low tide, MMHg-enriched Franks Tract marsh water was exported through the inlet to Franks Tract, resulting in a maximum MMHg concentration during low tide. Conversely, as the water level rose from low tide to high tide, Franks Tract water with a low MMHg concentration flowed through the inlet, resulting in the lowest MMHg concentration occurring during high tide. This suggests not only that the benthic flux of MMHg is significant enough to influence the overlying water column concentrations, but also that the Franks Tract marsh site could be a major source of MMHg.



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**Table 1. Change in water quality seaward in the Sacramento River during the summer of 2001 (Data From C. Foe, CRWQCB)**

Station	Date	River Mile	Chlor (µg/l)	Phaeo (µg/l)	TSS (mg/l)	Sp. Cond (mS)	Raw THg (ng/l)	Filt. THg (ng/l)	Raw MMHg (ng/l)	Filt. MMHg (ng/l)	THg/TSS (ppm)	MMHg/Chlor <sup>2</sup> (ng/µg <sup>2</sup> )
Freeport	29 May	120	5.06	5.28	14.0	177	2.39	0.50	0.09	0.04	0.17	0.09
Greene Landing	29 May		6.12	6.16	21.0	192	3.63	0.5	0.10	0.03	0.17	0.10
Walnut Grove	29 May		4.7	4.63	13.0	180	2.19	0.44	0.10	0.03	0.17	0.10
Rio Vista	29 May		4.47	4.5	26.0	245	5.87	0.68	0.06	0.02	0.23	0.06
Brannon Is (X1)	29 May		3.12	4.42	37.0		13.00	0.93	0.05	0.01	0.35	0.05
Grizzly Bay (X3)	29 May		2.49	2.87	34.0	5723	10.20	0.31	0.04	0.01	0.30	0.04
Grizzly Bay (X5)	29 May		1.39	2.21	24.0	8421	6.53	0.37	0.05	0.01	0.27	0.05
Freeport	26 June	120	2.79	1.86	14.0	151	2.55	0.41	0.07	0.04	0.18	0.07
Green Landing	26 June		2.99	2.79	21.0	145	4.46	0.15	0.09	0.01	0.21	0.09
Walnut Grove	26 June		3.18	2.69	22.0	155	4.71	0.38	0.08	0.03	0.21	0.08
Rio Vista	26 June		3.63	3.61	32.0	310	6.90	0.71	0.06	0.02	0.22	0.06
Brannon Is (X1)	26 June		3.04	4.12	47.0	1800	9.21	0.42	0.03	0.02	0.20	0.03
Sherman Is (X3)	26 June		1.83	3.74	34.0	5400	11.00	0.43	0.04	0.01	0.32	0.04
Freeport	31 July	120	2.93	2.50	19.0	136	1.21	0.21	0.06	0.03	0.06	0.06
Greens Landing	31 July		2.77	2.18	13.0	159	1.31	0.28	0.11	0.02	0.10	0.11
Walnut Grove	31 July		2.89	1.72	14.0	172	1.34	0.21	0.06	0.01	0.10	0.06
Rio Vista	31 July		4.67	3.02	30.0	179	4.69	0.28	0.06	0.03	0.16	0.06
Sherman Is (X1)	31 July		3.04	3.61	31.0		11.10	0.45	0.04	0.01	0.36	0.04
Pittsburg (X2)	31 July		2.14	2.38	11.0		3.95	0.26	0.04	0.01	0.36	0.01

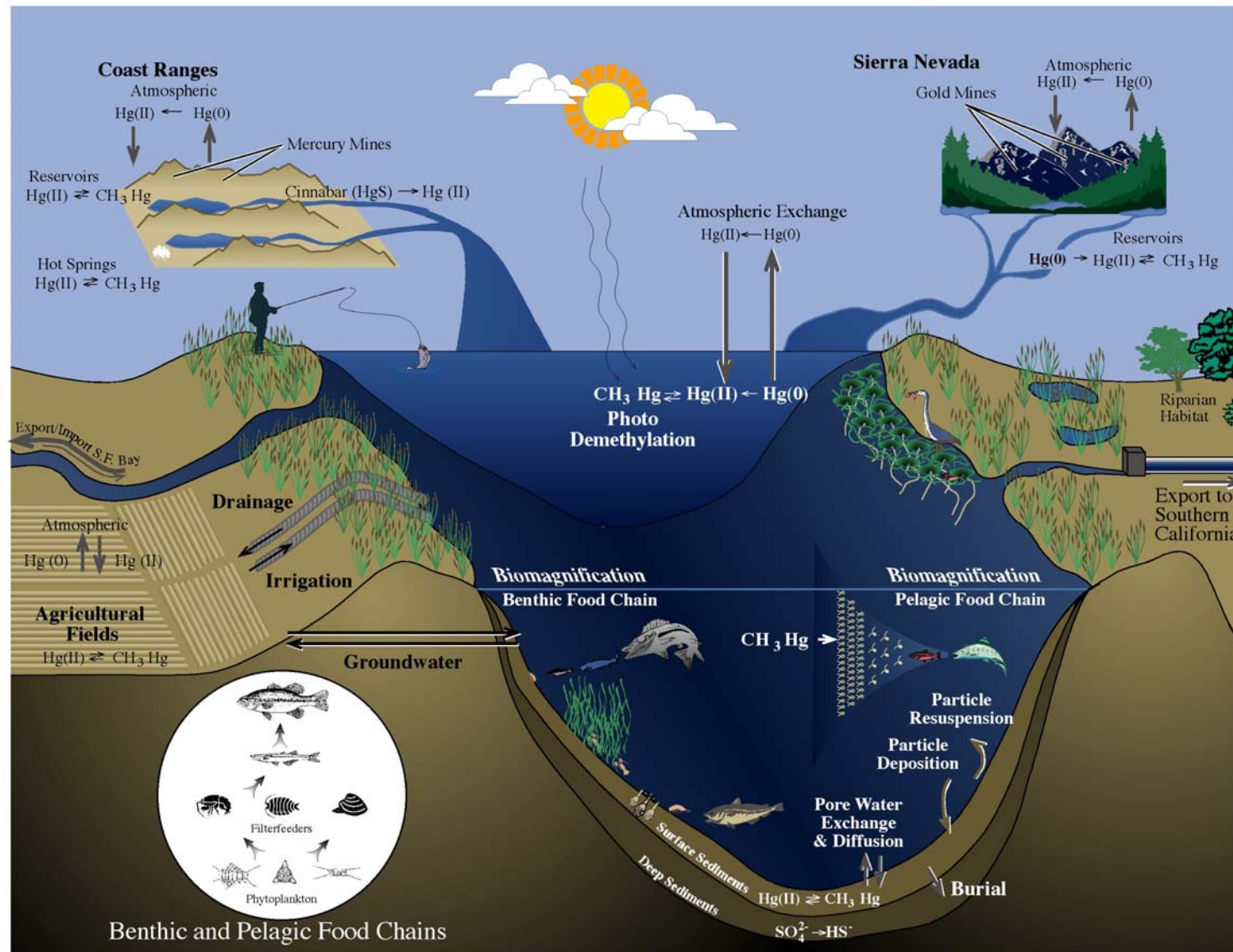
**Table 2. Raw total mercury mass balance (kg) for the Bay-Delta Estuary between March 2000 and September 2001. Negative import-export values indicate that the estuary is exporting more total mercury than is entering it (Data From C. Foe, CRWQCB).**

Date	IMPORTS					EXPORTS				Imports-Exports
	Sacramento River	San Joaquin River	Mokelumne and Consumnes Rivers	Prospect Slough	Total Imports	DMC	SWP	X2	Total Exports	
Mar-00	39.20	8.03	1.49	70.50	119.2	1.07	1.16	279.54	281.8	-162.5
Apr-00	12.88	4.60	0.64	0.74	18.9	0.43	1.60	102.44	104.5	-85.6
May-00	5.20	2.75	0.48	0.19	8.6	0.57	0.49	87.45	88.5	-79.9
Jun-00	4.05	1.82	0.27	0.13	6.3	0.59	0.71	13.89	15.2	-8.9
Jul-00	4.86	1.07	0.17	0.14	6.2	0.89	0.85	19.32	21.1	-14.8
Aug-00	4.58	2.03	0.16	0.07	6.8	1.50	1.04	4.77	7.3	-0.5
Sep-00	3.38	1.06	0.08	0.07	4.6	1.33	0.82	3.62	5.8	-1.2
Oct-00	3.41	1.53	0.13	0.11	5.2	1.27	0.94	3.41	5.6	-0.4
Dec-00	3.17	0.52	0.08	0.06	3.8	0.72		4.78	5.5	-1.7
Jan-01	30.33	1.32	0.29	0.62	32.6	0.65	0.49	14.94	16.1	16.5
Feb-01	21.12	2.29	0.00	0.00	23.4	0.98	0.79	19.99	21.8	1.7
Mar-01	6.47	2.31	0.24	2.13	11.1	0.63	1.52	42.28	44.4	-33.3
Apr-01	4.15	1.60	0.29	0.13	6.2	0.33	0.27	10.06	10.7	-4.5
May-01	2.57	2.28	0.21	0.21	5.3	0.22	0.11	7.72	8.1	-2.8
Jun-01	4.09	1.19	0.06	0.13	5.5	0.77	0.00	6.58	7.3	-1.9
Jul-01	4.91	0.58	0.04	0.20	5.7	0.89	1.85	1.51	4.3	1.5
Aug-01	5.64	0.63	0.02	0.13	6.4	0.90	0.48	2.65	4.0	2.4
Sep-01	1.96	0.61	0.03	0.03	2.6	1.28	0.54	4.11	5.9	-3.3
<b>TOTAL</b>	<b>162</b>	<b>36.2</b>	<b>4.69</b>	<b>75.59</b>	<b>278</b>	<b>15.0</b>	<b>13.7</b>	<b>629</b>	<b>658</b>	<b>-379</b>

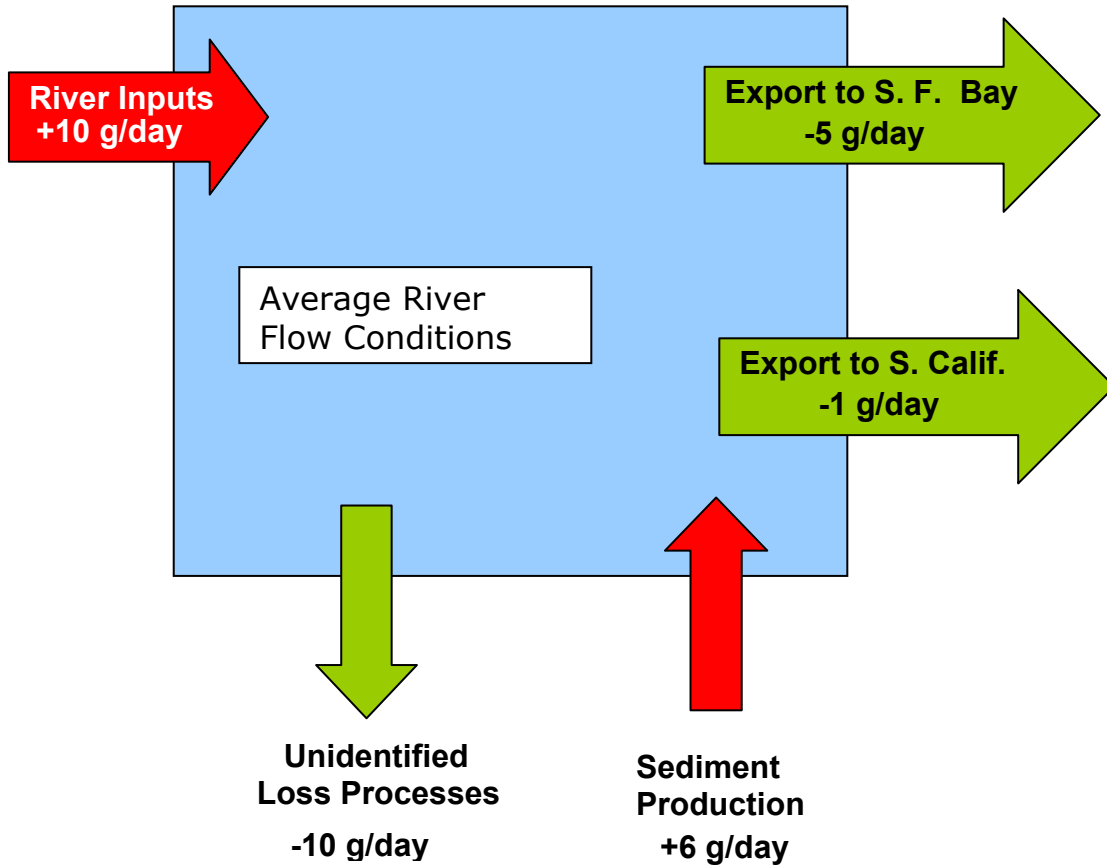
**Table 3. Raw methyl mercury mass balance (g) for the Bay-Delta Estuary between March 2000 and September 2001. Positive import-export values indicate that the estuary is importing more methyl mercury than is leaving it. Values with an asterisk were calculated from a water concentration that was below detection. Half the detection limit was used in calculating loads. The detection limit was 0.022 and 0.011 ng/l in 2000 and 2001, respectively (Data From C. Foe, CRWQCB).**

Date	IMPORTS					EXPORTS				Imports-Exports
	Sacramento River	San Joaquin River	Mokelumne-Consumnes River	Prospect Slough	Total	DMC	SWP	X2	Total	
Mar-00	657.0	153.6	37.8	1280.3	2128.7	39.2	58.5	1601.8	1699.5	429.1
Apr-00	228.7	58.7	23.8	14.0	325.3	1.8*	10.5	170.5	182.8	142.5
May-00	520.4	49.6	23.1	2.9	596.0	16.4	18.7	428.4	463.4	132.6
Jun-00	86.2	47.7	7.2	1.0	142.1	16.5	3.5*	79.3	99.3	42.8
Jul-00	82.5	17.6	0.5*	0.8	101.4	3.6*	4.9*	8.2*	16.7	84.8
Aug-00	146.8	22.4	4.9	0.6	174.7	3.7*	5.2*	5.4*	14.3	160.4
Sep-00	57.2	16.8	0.3*	0.4	74.7	3.4*	27.7	8.5	39.6	35.1
Oct-00	77.0	33.6	2.7	1.1	114.5	3.5*	4.2*	4.8*	12.5	102.0
Dec-00	94.2	17.1	2.8	1.1	115.2	18.5	18.0	31.5	68.0	47.2
Jan-01	323.3	44.5	8.2	9.6	385.6	29.9	33.6	113.0	176.5	209.1
Feb-01	256.2	39.3	15.0	36.9	347.4	2.7	24.7	84.9	112.2	235.2
Mar-01	159.1	48.1	10.5	59.2	276.7	13.2	24.5	290.7	328.4	-51.6
Apr-01	103.8	21.1	9.8	0.9	135.5	3.8	7.1	6.4*	17.3	118.2
May-01	69.9	33.6	6.2	1.1	110.8	3.6	2.1	30.9	36.6	74.1
Jun-01	80.5	30.5	1.7	0.7	113.3	13.4	0.0	22.1	35.5	77.9
Jul-01	123.1	15.6	1.0	1.2	141.0	20.2	5.7	26.9	52.8	88.2
Aug-01	72.1	19.7	0.3	0.6	92.8	9.9	2.1*	14.4	26.4	66.3
Sep-01	87.9	16.5	1.0	0.2	105.6	2.1*	8.4	2.4*	12.9	92.7
<b>TOTAL</b>	<b>3230</b>	<b>686.0</b>	<b>156.7</b>	<b>1410</b>	<b>5480</b>	<b>205.3</b>	<b>259.5</b>	<b>2930</b>	<b>3390</b>	<b>2090</b>

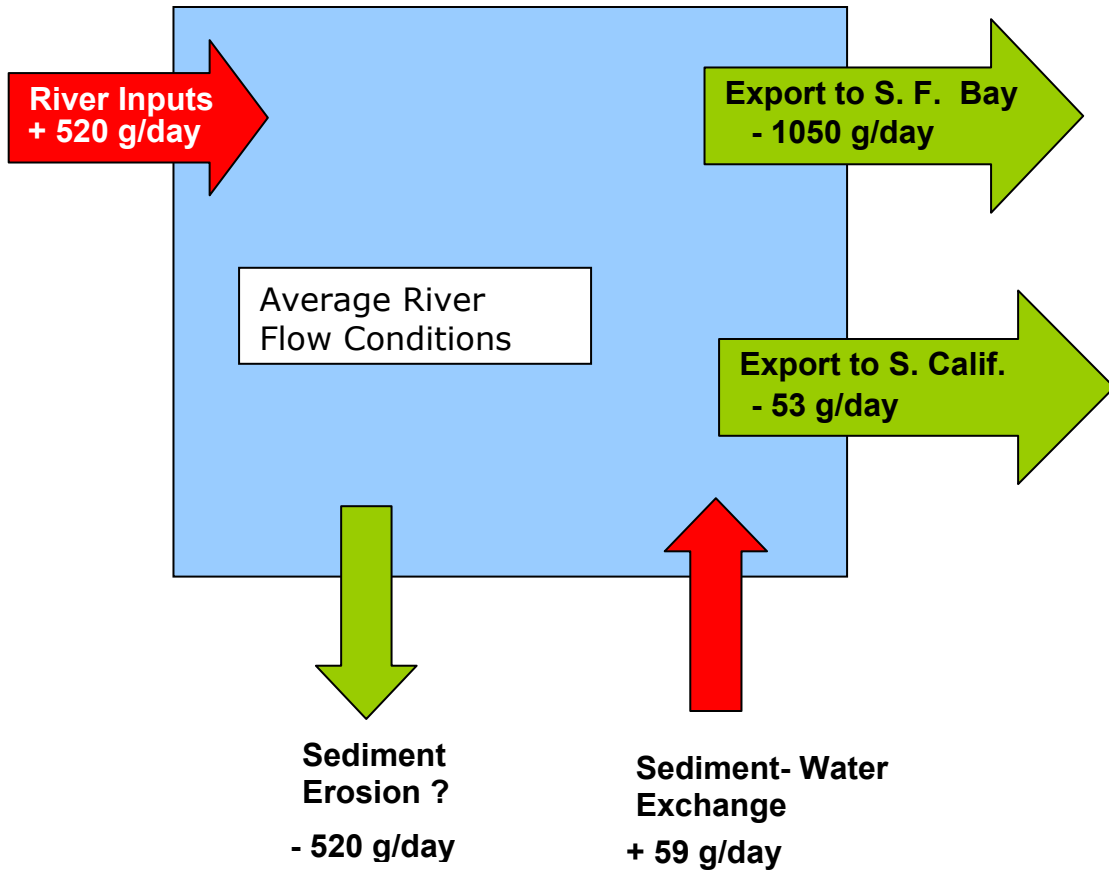
**Figure 1. Delta Conceptual Model.**



**Figure 2. Monomethyl mercury fluxes in the Bay-Delta Estuary. River input and export rates to the San Francisco Bay are from Foe et al. (2002)(Task 1A) and sediment-water exchange flux is from Gill et al. (2002) (Task 4A). The flux associated with the unidentified loss processes was determined by mass balance.**

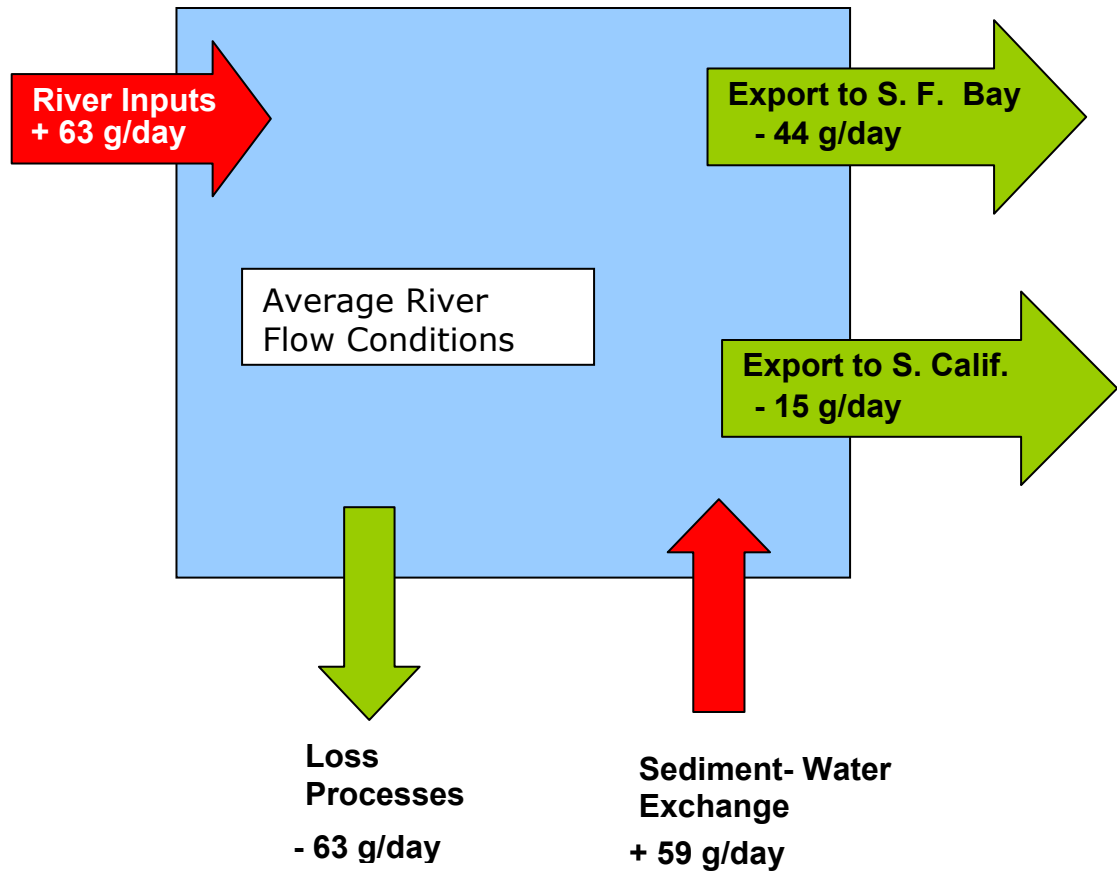


**Figure 3. Total Hg fluxes in the Bay-Delta Estuary. River input and export rates to the San Francisco Bay are from Foe et al. (2002)(Task 1A) and sediment-water exchange flux is from Gill et al. (2002) (Task 4A). The flux associated with the loss is assumed to be sediment erosion, but has not been identified.**

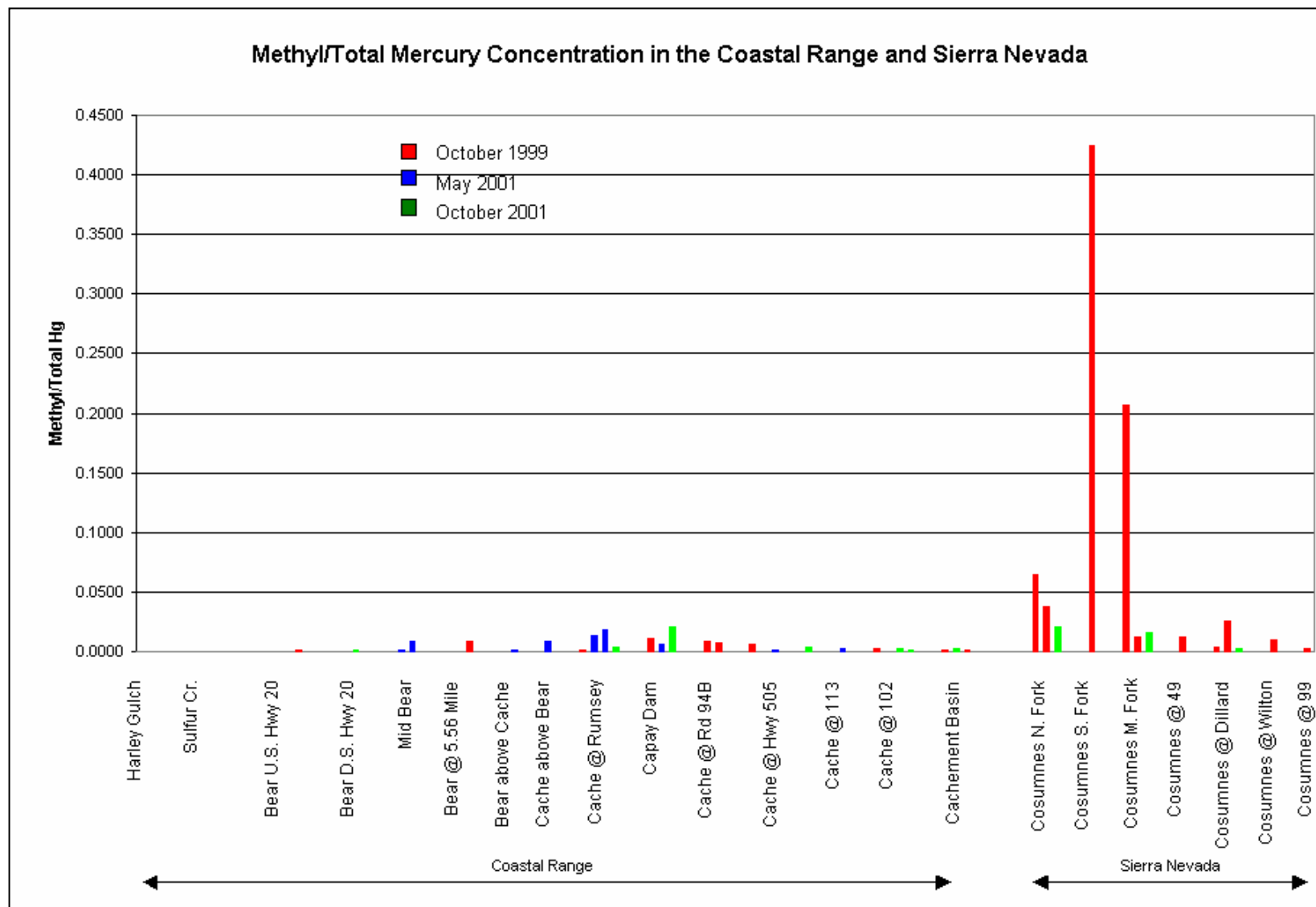




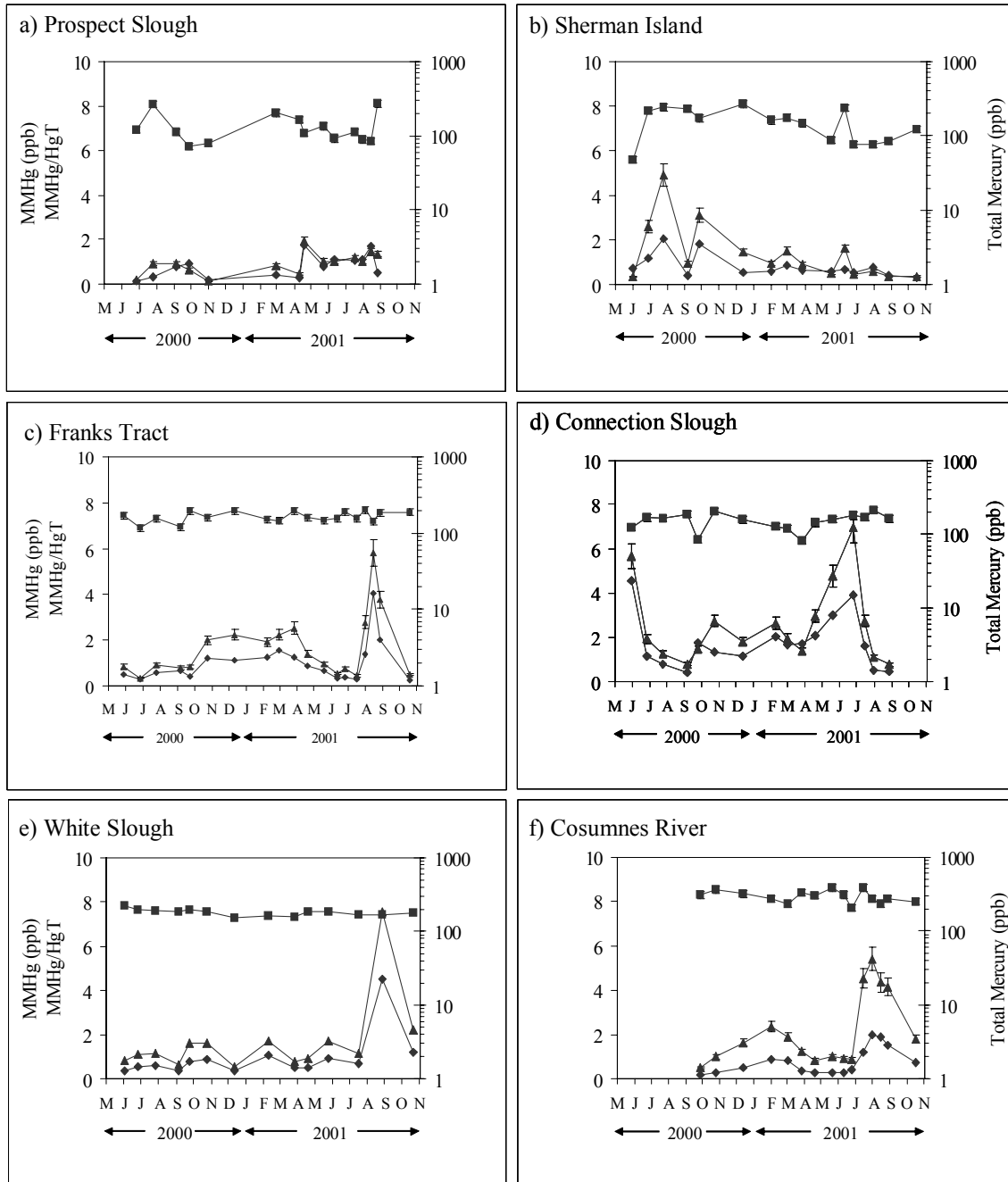
**Figure 4. Dissolved Hg fluxes in the Bay-Delta Estuary. River input and export rates to the San Francisco Bay are from Foe et al. (2002)(Task 1A) and sediment-water exchange flux is from Gill et al. (2002) (Task 4A). The flux associated with the has not been identified.**



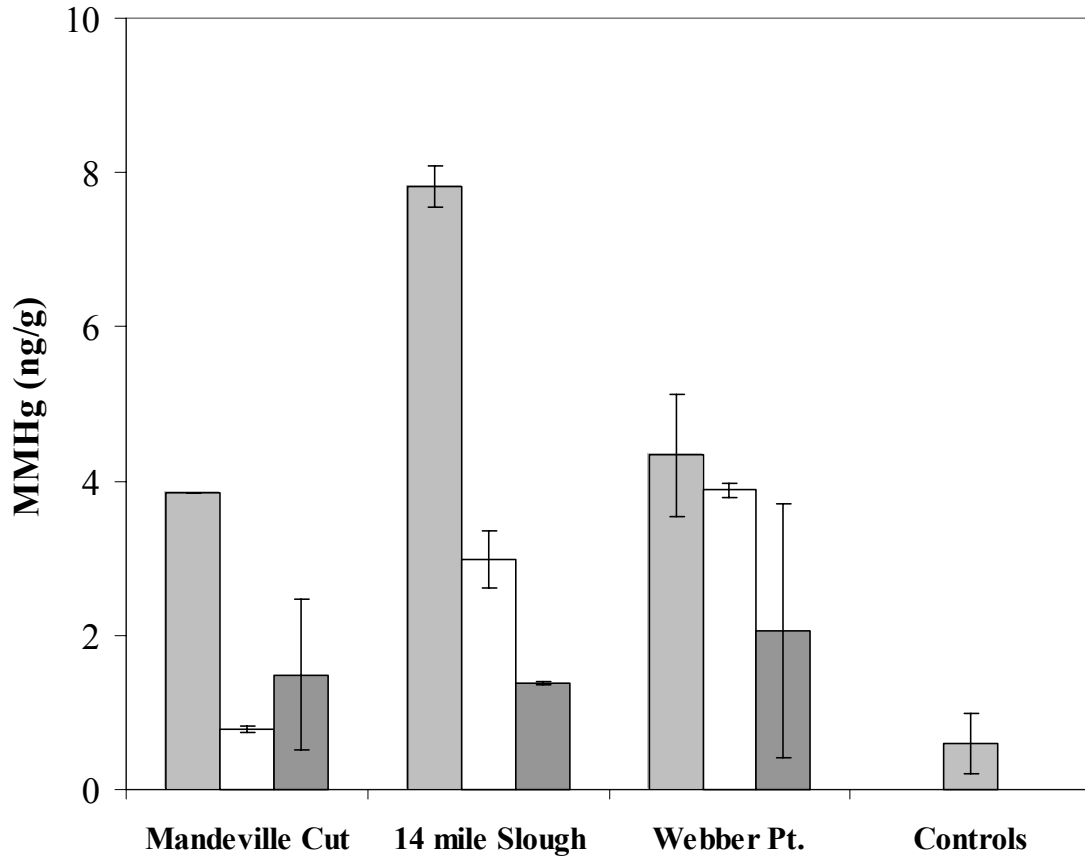
**Figure 5. Methyl/Total Mercury Concentrations in Coastal and Sierra Nevada Ranges**



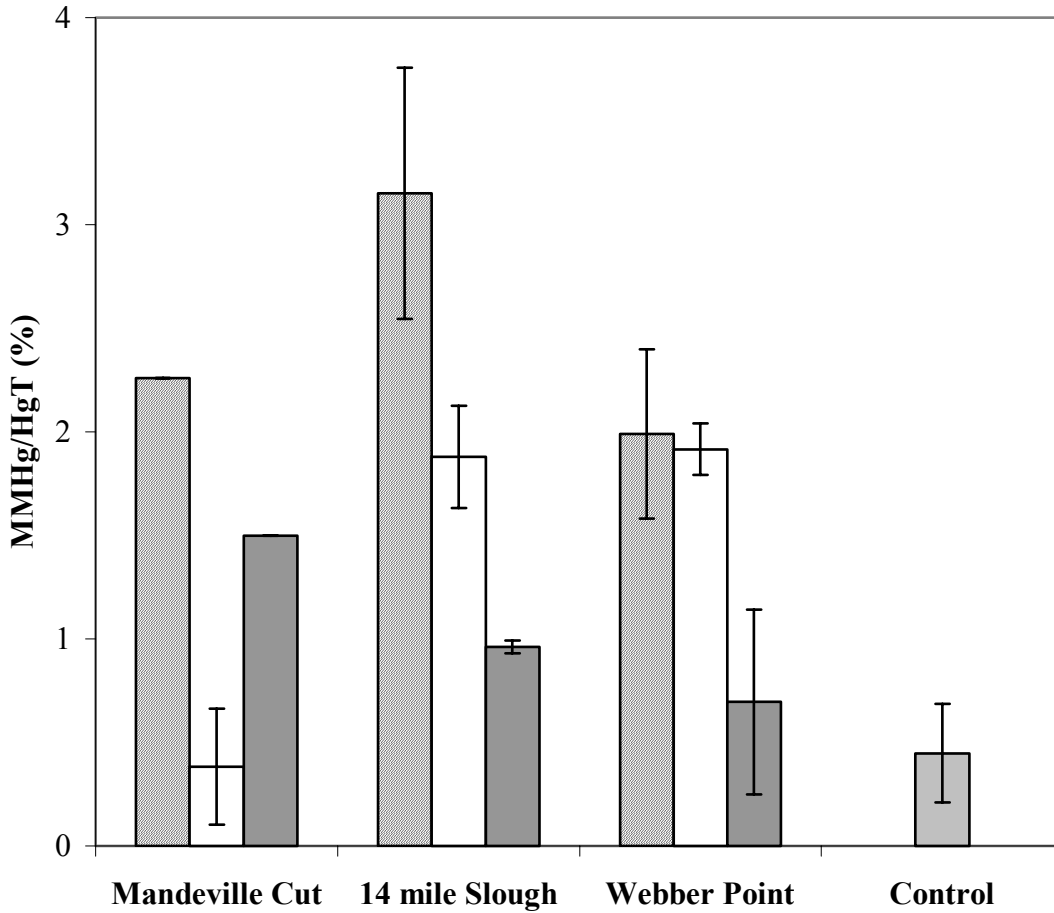
**Figure 6. Seasonal changes in methyl mercury ( $\blacktriangle$ ), total mercury ( $\blacksquare$ ), and the methyl mercury to total mercury ratio (as a percent)( $\blacklozenge$ ) in surficial sediments (0-0.5 cm) of the Bay-Delta. Total mercury is shown on the second y-axis as a log scale. Error bars represent analytical uncertainty rather than field duplication. The following locations were sampled between May 2000 and November 2001: Prospect Slough (panel a), Sherman Island (panel b), Franks Tract (panel c), Connection Slough (panel d), White Slough (panel e), and Cosumnes River (panel f).**



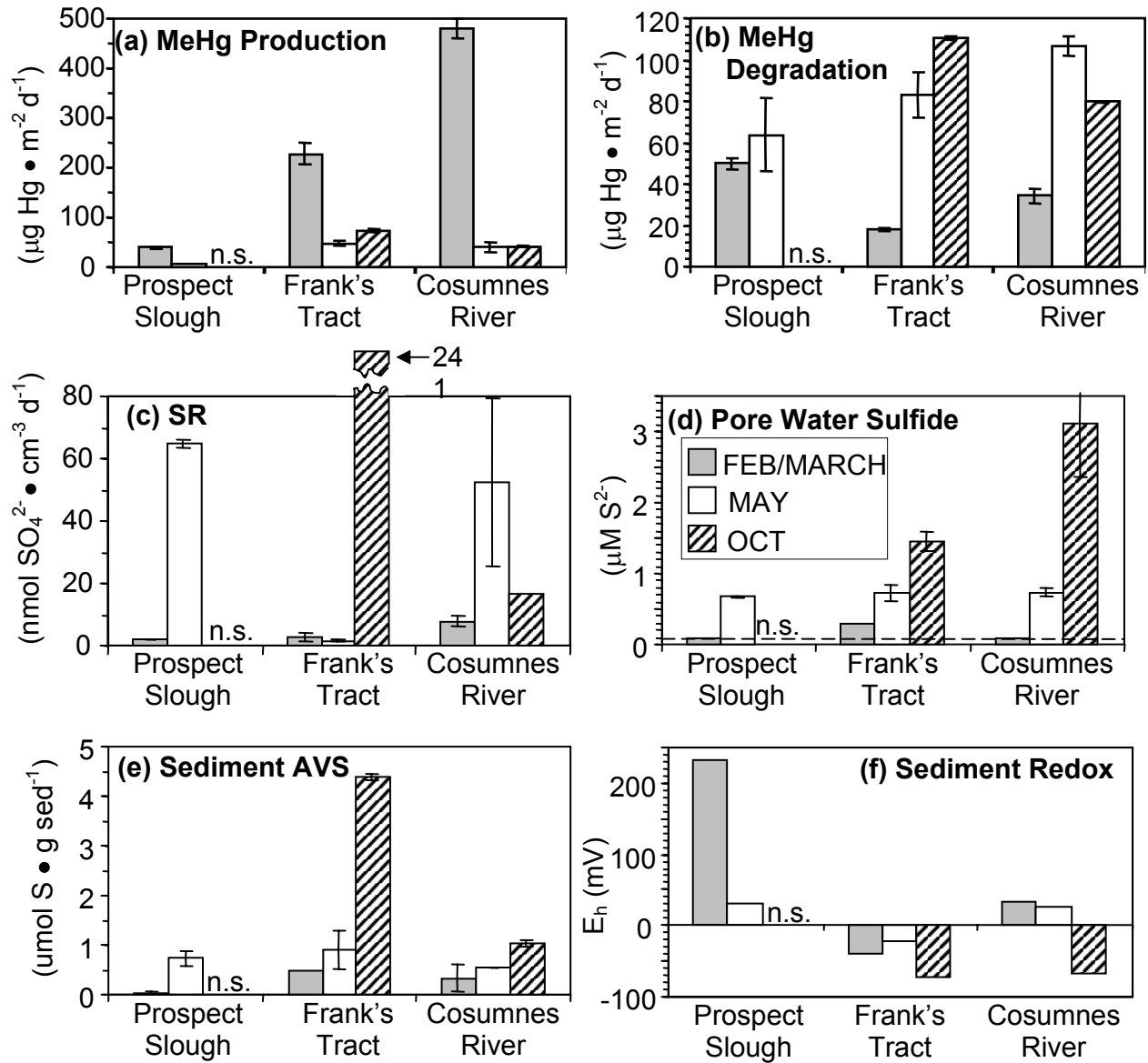
**Figure 7. MMHg in sediment samples in three wetland areas. Diagonal hatching = innermost station, no hatching = middle station, solid = station nearest edge of wetland. Control was taken a few miles away in the San Joaquin River. Error bars = ranges.**



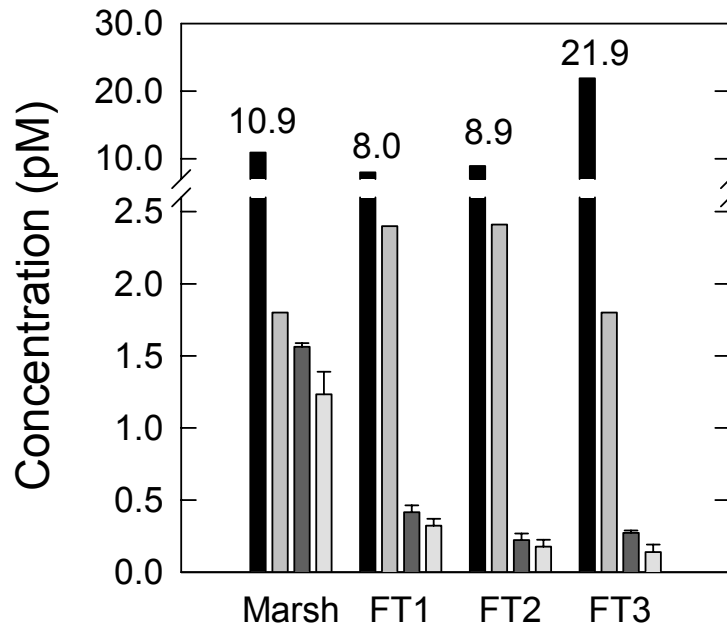
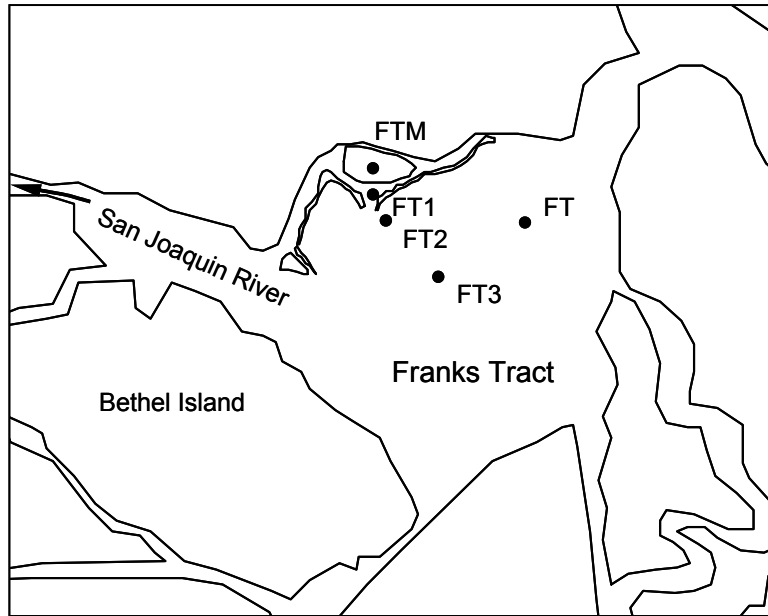
**Figure 8. MMHg/Total Hg in sediment samples in three wetland areas. Diagonal hatching = innermost station, no hatching = middle station, solid = station nearest edge of wetland. Control was taken a few miles away in the San Joaquin River. Error bars = ranges.**



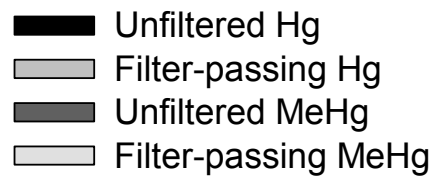
**Figure 9. Mercury methylation and de-methylation rate potentials determined using radioisotopic assays along with selected parameters which can influence these processes. Figure Provided by Dr. Mark Marvin (USGS)**



**Figure 10. Surface Water Transect Studies in Franks Tract**



**Franks Tract Transect**



**Figure 11. Time Series Measurements of Hg and MeHg at the Franks Tract Marsh site in May 2001.**

