Mercury Concentrations Bordering The Hamilton Army Air Field Remediation Site: February, 2003

Wet Season - Dry Season Contrast

Report to USACE District,
San Francisco

Prepared by
USACE Engineer Research and Development Center
Waterways Experiment Station
Vicksburg, MS

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Authors

The following staff scientists at the U.S. Army Engineer Research and Development Center, Environmental Laboratory, Ecosystem Processes and Engineering Division\(^1\) and Environmental Chemistry Division\(^2\), were responsible for the conduct of the research and preparation of this report:

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Background

The re-establishment of wetlands in the San Francisco Bay/Delta System using dredged material from bay channels has the potential for mobilizing mercury (Hg) present in the sediments. The primary route of entry of mercury from sediments into the Bay ecosystem is through the formation of the highly mobile and bioavailable form, methylmercury (monomethylmercury, CH$_3$Hg$^+$, MeHg). Methylmercury formation is favored under certain conditions characteristic of wetland environments. In the summer of 2001 (dry season) at the request of USACE District, San Francisco, the USACE Engineer Research and Development Center, Waterways Experiment Station (ERDC-WES) conducted extensive soil and sediment sampling and analysis for total mercury (THg) and MeHg at selected stations bordering the former Hamilton Army Airfield (HAAF) wetlands restoration site located on the western edge of San Pablo Bay, California. This work was continued in FY03 with wet season sampling and analysis at the same stations, and the results are here reported and contrasted with the dry season concentrations.

This report references:


Wet Season Sampling

Descriptions of the sampling locations, sampling and analysis methods, personnel involved, theoretical rationale for the study, and dry season results are contained in reference 2 and the appendices therein. The present study follows the proposed outline described in the scope-of-work (Reference 1). Briefly, in the dry season study seven locations were selected bordering the HAAF wetlands remediation site and one reference location (China Camp Marsh) at which three to twelve sample sites were located and identified by GPS coordinates. Five replicate soil surficial samples were taken at each site within an approximate 0.75 m radius. The original sample sites were located and re-sampled for the wet season study. Clean techniques were used in the field (gloves, SS

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trowel, acid-washed glass sample jars with teflon-lined lids) and samples were chilled on site and kept at 4°C until analysis. The sampling interval was chosen to occur during the wettest time of the year (Jan-Feb) and all samples were collected and shipped between 3 and 6 February, 2003. The dry season collection (Reference 2) occurred following the driest months, on 10-14 September, 2001. All analyses were performed by the ERDC Environmental Chemistry Branch, CEERD-EP-C, Omaha, Nebraska.

Statistical Methods

Statistical analyses for comparisons within and between locations were performed using SAS procedures (SAS Institute, Inc. 2001). Total mercury and MeHg concentrations in soil, and MeHg as percent of THg were tested statistically for differences between wet and dry season at each of the eight locations, and at each site within each location. Parametric comparison test assumptions were checked for violations using the Shapiro-Wilk’s test for normality of residuals, and a modification of Hartley’s Fmax test for equality of variances (Shoemaker 2003). Site data failing the normality assumption were compared using the nonparametric Kruskal-Wallis test. Site data passing the normality test were compared using t-tests for equal or unequal variances.

For comparisons of season at each location, all sites combined, two-way analysis of variance (ANOVA) was performed with site as a blocking variable. Ranks were used in the ANOVA instead of raw data when the normality assumption was violated. A significance level of 0.05 was used for all statistical tests. Non-detects were substituted as one-half detection limit prior to analysis.

Results and Discussion

Historical and current rainfall measurements were not found for the HAAF Remediation Site. However, the city of San Rafael is located in the vicinity of HAAF and rainfall records there are available as a surrogate (Western Regional Climate Center 2003). The average monthly rainfall and temperatures over the past 55 years at San Rafael are shown in Figure 1. Rainfall in the month preceding the wet season collection totaled 4.39 inches at San Rafael, and was about half the historical average for January. Virtually no rainfall occurs during the average summer, and none was reported in the north SF Bay area during the month preceding the dry season collection (NOAA NWS 2001). Temperatures were somewhat cooler than historical averages during both dry and wet season collection intervals. Historical September temperatures average 81°F/54°F day/night, and during the dry season collection the averages were 74°F/54°F day/night. In February the historical averages are 61°F/44°F day/night, and during the wet season collection they were 56°F/36°F day/night.

Overall summary statistics calculated for all soil/sediment samples taken bordering the HAAF Remediation Site and the China Camp Reference Marsh are given in Table 1. Units of THg and MeHg concentration in Table 1 and throughout this document are expressed as ng g⁻¹ (parts-per-billion, ppb) on a dry weight basis. This
**Figure 1.** San Rafael Civic Center, San Rafael California (047880). Monthly Climate Summary, Period of Record: 7/1/1948 to 3/31/2003. Western Regional Climate Center 2003. **W:** Total rainfall for month preceding wet season collection, 4.39 inches. **D:** Total rainfall for month preceding dry season collection, 0.00 inches.

**Table 1.** Overall HAAF soil/sediment mercury summary statistics, ng g⁻¹.

<table>
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<tr>
<th>Analyte/Season</th>
<th>Mean</th>
<th>n</th>
<th>SD</th>
<th>Std. Error of Mean</th>
<th>95% Confidence Limits of Mean</th>
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<td>292</td>
<td>305</td>
<td>103</td>
<td>5.93</td>
<td>280, 304</td>
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<tr>
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<td>301^1</td>
<td>305</td>
<td>110</td>
<td>6.23</td>
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<td>MeHg dry</td>
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<td>305</td>
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<td>3.58, 5.20</td>
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<td>305</td>
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<td>0.126</td>
<td>1.24, 1.74</td>
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<th>5%ile</th>
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<td>0.073</td>
<td>6.14</td>
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</table>

^1Based on statistical comparisons of wet and dry seasons, bold indicates significantly greater mean (P < 0.05)
convention is used in order to clearly show relationships between the mercury species. Wet season means were statistically higher than dry season means for all Hg metrics. Although a statistical difference was found for mean THg between seasons the difference was small, amounting to only a three percent higher concentration in the wet season. It appears unlikely that the higher value is anything more than a statistical artifact; the data were significantly non-normally distributed, and wet and dry season medians were essentially the same. Location differences for THg are notable in the case of the HAAF Bay Edge sites and the Bel Marin Creek sites (Figure 2), although not much difference can be seen among the other six locations. The overall THg concentration in the intertidal and mid- to high marsh soils bordering HAAF during both seasons (~ 0.3 ppm) is the same value measured in upper near-shore San Pablo Bay sediments by Hornberger et al. (1999), and within the range (0.166-0.515 ppm) for North Bay wetland soils reported by Lee et al. (2000). Although THg was essentially constant over seasons, the overall wet season MeHg concentrations averaged threefold higher than were measured during the dry season, whether mean or median was taken as the central tendency. This amounted to a 2.4 to 2.8-fold increase in the percentage of MeHg in surficial soil THg. This effect could be the result of a shallower anoxic or suboxic zone producing MeHg.

Results of seasonal means comparisons at the eight locations and at sample sites within locations are given in Table 2. For each mercury metric (THg, MeHg, and %MeHg), a significantly greater difference was observed for wet season samples than for dry season at about one-third to one-half of all the individual sample sites. Mean wet season concentrations significantly exceeded mean dry season concentrations in 26 percent of THg and 40 percent of MeHg soil analyses. Percent MeHg significant wet season increases paralleled MeHg at 43 percent. Mean dry season concentrations were greater than mean wet season concentrations in 15 percent of all sites for THg only. All sites within the Mid Marsh and Bel Marin Creek exhibited significant seasonal differences in concentration for at least one mercury metric. Mercury methylation was significantly increased most often at the Mid Marsh location (six out of eight sample sites) but the highest levels of MeHg were found at the High Marsh sites, reaching as much as 23.5 ng g$^{-1}$ at SM-20, a 30-fold increase at that site. The increase in MeHg averaged 12-fold across all sites at the High Marsh location.

For all sites combined within a location, MeHg (and %MeHg) were significantly greater during the wet season than during the dry season in six of the eight locations. In decreasing order, these were: High Marsh > Mid Marsh > Reference Marsh > BM Seasonal > Bay Edge = Antenna Field. The fraction of MeHg in THg reported in coastal estuarine sediments and soils is most typically in the range of a few tenths to about one percent, with highest levels reported being on the order of six percent (Bartlett and Craig 1981, Kannan et al. 1998). High soil/sediment THg did not correlate with high mercury methylation. Locations with the highest wet season MeHg were Mid- and High Marsh which ranged to over 20 ng g$^{-1}$, and 9 percent MeHg in THg. The Bay Edge samples where the highest THg concentrations were found were all less than 1% MeHg. This observation is consistent with other temperate estuaries.
The observed increase in methylation in surficial soils/sediments is consistent with expectations. In the sediments and soils of a moderately high mercury content, climatically temperate, estuary such as the San Francisco Bay system we hypothesize that rates of mercury methylation will tend to increase in the wet season. Processes that favor increased methylation include increased atmospheric deposition of mercury-bound particulates due to rainfall, a dampening of volatilization of mercury with decreased temperatures and shorter photoperiod, decreased salinity and increased transport and deposition of organic material with increased freshwater runoff and river discharge volume, seasonal dormancy of plant rhizosphere oxidative processes and wider areal redox shift toward reducing conditions in wetted soils and sediments. These conditions and the primary processes dependent on them may be sufficient to dominate counter-processes that sum to reduce mercury methylation rates, e.g., lowered sulfate-reducing microbial activity with lowered seasonal temperatures, or a reduction in photo-oxidation of MeHg with shorter photoperiods. The complexity of the geochemical, microbial, and physical interactions that favor or reduce the rate of net methylation of inorganic mercury
<table>
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<th>Location</th>
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<th>Percent MeHg</th>
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<td></td>
<td>Dry</td>
<td>Wet</td>
<td>Dry</td>
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over demethylation, and differences in higher organism exposure, make it impossible to predict the extent of seasonal variations in mobilization and uptake of mercury without creating a body of region- and ecosystem-specific data. Increases and decreases in mercury tissue levels of invertebrates and fish have both been reported during the same season. For example, killifish, *Fundulus heteroclitus*, monitored in Berry's Creek, New Jersey, a highly mercury-contaminated saltwater marsh, showed a five-fold increase in mercury levels during the summer (Weis et al. 1986). In contrast, bay mussels, *Mytilus edulis*, monitored over a two-year period in the estuary and Gulf of St. Lawrence showed a marked decrease in tissue mercury content during the summer months (Cossa and Rondeau 1985).
Conclusions

Total mercury concentrations in surficial soils and sediments at seven locations bordering the HAAF Wetlands Restoration Site and at a near-by reference saltmarsh (China Camp) in San Pablo Bay, CA, were mostly similar during the dry season of 2001 and the wet season of 2003. Methylmercury in the same samples increased an average of three-fold during the wet season. Highest THg was consistently found in samples at the intertidal zone (Bay Edge) but MeHg was increased less than two-fold in the same samples during the wet season. Highest methylation occurred in samples taken closer to the levee and less influenced by tidal fluctuation. In the Mid Marsh and High Marsh samples, MeHg in THg averaged up to nine percent, and MeHg concentrations were as high as 23.5 ng g⁻¹ in soil, suggesting that meteorological or surface water play a role in Hg methylation of the higher marsh areas.

References


Western Regional Climate Center. (2003). Historical Climate Information. Desert Research Institute, 2215 Raggio Parkway, Reno, Nevada 89512. 
http://www.wrcc.dri.edu/
Appendix A. Analytical Results

Department of the Army
Engineer Research and Development Center
Environmental Chemistry Branch - Omaha
420 South 18th Street Omaha, NE 68102
(402) 444-4300

Sample Results for Mercury and Methylmercury
Project Name: San Francisco Bay System Wetland Testing
Project No.: 6836

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<th>Client Sample ID</th>
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<th>Hg Digestion Date</th>
<th>Hg Analysis Date</th>
<th>Hg Result (ng/g)</th>
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**Total Mercury QC**

**Project Name:** San Francisco Bay System Wetland Testing  
**Project No.:** 6836

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