# Mercury and Methylmercury in the San Francisco Bay area: land-use impact and indicators

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**May 16th 2008**

**California Water Symposium 2008**

Thank you for the comments. I appreciated your responds and I tried to do my best to reflect on my paper.

First, I wanted to compare the mercury and methylmercury sediment concentrations with the draining point of each watershed, but I failed to obtain relevant data. I also tried to collect data about point sources, for example gasoline refinery and wastewater treatment plant, but also I did not find available data. If I had collected those data, the results would have been more clear and stronger.

In this study, I just tried to highlight the relationship between methylmercury and environmental factors including land use and the fact that historical mining activities are the main sources of mercury is clear I did not mentioned much of that. However, to draw the conclusion that mercury and methylmercury should have different control strategies, the sources of them have to be claimed. Therefore, I explained the sources of mercury and the importance of them in Conclusion section.

There were many researched about mercury in the San Francisco Bay area, but mostly studies were simply about analysis or interpretation of monitoring data. However, the interpretation should take account for environmental factors, especially in the case of methylmercury because there are lots of factors relate to methylation. In my study, I took a look at the relationship between environmental factors- water quality indicators and land use- and mercury species. My conclusion showed good agreement with previous study and I examined these facts with new method- correlation coefficient analysis.

I would like to thank you for the opportunity to explore my research interests, best lectures and interesting filed trips.

# Mercury and methylmercury in the San Francisco Bay area: land-use impacts and indicators

Hyojin Kim*

Abstract

In this study, I analyzed the impact of land-use on mercury and methylmercury in the San Francisco Bay area and I explored correlations of mercury and methylmercury with various water quality indicators using water and sediment quality data from the Regional Monitoring Program. To understand the relationships of land-use and water and sediment quality with mercury and methylmercury concentrations, I conducted a correlation coefficient analysis using Microsoft EXCEL 2007. In the San Joaquin Delta watershed and the Suisun Bay watershed, heavy metals showed strong relationship with methylmercury. Developed land uses such as industrial, commercial services and urban built-up had a strong relationship with methylmercury, while agricultural land uses generally had a negative relationship with methylmercury. Mercury and methylmercury had a strong positive relationship with clay, silt, and fine sand. Mercury had significant negative correlation with pH and significant positive correlation with silver. Methylmercury was strongly related to temperature and total nitrogen. Although underestimated in this study, strip mines had a fairly strong correlation with mercury, indicating that they may be a major source of mercury to the San Francisco Bay. Restoration efforts should target areas with developed land-use, high clay, silt, fine sand and heavy metals.

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Keywords

Mercury, Methylmercury, Water Quality Indicators, Land-use, TMDL

1. Introduction

Mercury is a rising concern worldwide, especially in the San Francisco bay area, where mercury issues have a long history. In the 19th century, during the Gold Rush Era, tremendous amounts of mercury were introduced into water bodies and accumulated in stream beds. At present, more than 70% of total mercury loading into the San Francisco bay area is of gold mining legacy, i.e., bed erosion in the central valley watershed (Clean Estuary Partnership, 2004).

Most properties such as toxicity and bioavailability are strongly dependent on mercury species. Among mercury species, methylmercury is the most toxic and highly bioavailable. Methylmercury is produced by biotic and abiotic methylation processes and unfortunately the methylation process is not still well understood. Seven major factors determining the rate of mercury methylation are 1) microbiology; 2) temperature; 3) pH; 4) organic matter; 5) redox conditions; 6) sulfide and 7) salinity (Ullrich et al., 2001). However, due to the complexity of the process it is hard to predict the rate of methylation.

Given the concern about mercury and methylmercury, the California Environmental Protection Agency decided to adopt a Total Maximum Daily Load (TMDL) for the San Francisco Bay Area and the surrounding watersheds (State Water Control Resource Board, http://www.waterboards.ca.gov/water_issues/programs/tmdl/). The mercury TMDL is proposed 1) to reduce total mercury and 2) to minimize mercury methylation which produces methylmercury, more toxic and highly bioavailable. However, the TMDL is a water quality regulation for a single pollutant and the rate of mercury methylation process is determined by various environmental factors.    Therefore, the additional regulation for relevant chemical species and land-use will be needed to reduce the rate of methylation.

To meet the objectives of the mercury TMDL will require further study of the water quality indicators and land-uses that relate to mercury methylation in the bay area. Understanding these relationships will make TMDL implementation more successful and effective.    Therefore, the goals of this study are 1) to identify water quality indicators associated with mercury methylation and 2) to assess the relationship between land use and methylmercury.

## 2. Method

### *Data Collection*

The study area was determined according to USGS hydrologic units, which divided by watershed are Suisun Bay (18050001), San Pablo Bay (18050002), Coyote (18050003), San Francisco Bay (18050004), San Joaquin Delta (18040003) and Lower Sacramento (18020109) (Fig 1, USGS, http://water.usgs.gov/GIS/huc_name.html#Region18).    Basic GIS data including hydrologic unit boundary, stream, and land use was collected using BASINS 4, a tool that is applicable to various purposes of environmental modeling (EPA, http://www.epa.gov/waterscience/basins/).    This tool allows collection of nation-wide watershed data.

Water and sediment quality data were collected from the Regional Monitoring Program (RMP) in San Francisco Estuary institute (San Francisco Estuary Institute, http://www.sfei.org/index.html).    Since 1993, the RMP has been monitoring water and sediment quality and conducting bioassay throughout bay area in 25 locations with more than 180 sampling points (Fig. 2). Sampling points were divided into 5 groups based on hydrologic units. Methylmercury data has only been available since July and August of 2000. In this paper, all statistical analysis on methylation takes into account data from 2000 through 2006.

*Statistic Analysis*

To identify the relationship between mercury/methylmercury and land-use and water and sediment quality, I performed, a multiple correlation coefficient analysis using the EXCEL 2007 Data Analysis Toolbox. I calculated areas of land use types for each watershed using ArcGIS 9.2. For comparison purposes, the analysis considers, for each watershed, land use types both the whole watershed, as well as within a 100km buffer zone from the estuary.

3. Results and Discussion

*Land use*

Large portions of the total area (1,696k km$^2$) are used as agriculture and forests (Table 1). Coyote watershed, which includes the Guadalupe River watershed that is speculated to introduce 7% of total mercury load into San Francisco Bay area, was the southern most part of San Francisco Bay area (Clean Estuary Institute, 2004). Residential (20 %), evergreen forest land (16%) and herbaceous rangeland (15%) are the three largest land uses in total watershed area. However, within a 100km zone from the estuary, residential (27%) and industrial (24%) land-use were dominant. The percentages of industrial and commercial and commercial and services were relatively high compared to the other sectors, 4% and 11%, respectively.

Lower Sacramento watershed, as the largest hydrological units, was 455 km$^2$, 28% of the total area. In the Lower Sacramento watershed, the largest land use type was cropland and pasture in both the total area and the 100km buffer zone.

The San Francisco bay watershed had various land use throughout, including herbaceous rangeland (21%), mixed forest land (20%), residential (15%) and evergreen forest (13%). However, in the 100km buffer zone, the main land use was residential areas (38%) and developed land use type -commercial and services and industrial- was the second largest portion, 23%. In San Joaquin Delta watershed, same as Lower Sacramento watershed, the largest land use type was cropland and pasture in both total area and 100km buffer zone.

San Pablo watershed was the second largest hydrologic unit in study area. In total watershed area, evergreen forest (29%), cropland and pasture (15%), herbaceous rangeland (13%) and residential (12%) were the major land use. However, nearby the estuary, residential area was the largest land use type.

Suisun bay watershed was the smallest region and most land uses were herbaceous rangeland (20%), cropland and pasture (15%) and non-forested wetland (15%). However, within 100km from estuary was more than 30% of land was non-forested wetland.

*Mercury and methylmercury*

Seasonal changes could not be assessed from interstitial water and sediment quality data obtained from RMP website because available methylmercury data were only from July and August, except for one February, 2002. Differences between daily sampling dates of water and sediment were negligible, so the average values of each month were taken.

The concentration of mercury varied year to year. However, in Coyote watershed and San Pablo watershed, mercury concentrations were higher than average concentration throughout the year (Fig 3). Methylmercury concentrations in Coyote watershed and San Francisco Bay watershed were significantly high. Even though mercury concentration in Coyote watershed and San Francisco Bay watershed were steady since 2001, methylmercury concentration slightly increased (Fig 3). The ratio of methylmercury/mercury generally increased over time.

*Correlation coefficient analysis*

Although correlation coefficient analysis does not indicate a cause and effect relationship, it shows the strength and direction of a linear relationship, which will be adequate for this study to show the relationships between land-use and water quality indicators and land-use and mercury.

Water and Sediment quality

In the Coyote watershed, Silicates had the highest positive correlation coefficient with Hg but temperature showed fairly strong negative relationship to Hg. In the case of methylmercury, it was difficult to see strong correlation but Dissolved Oxygen (DO, 0.40), heavy metals including Ni (0.45), Zn (0.41), and Cu (0.40), and temperature (0.40) were all slightly related to methylmercury (Fig.11).

Hg in the San Francisco Bay watershed also had high positive coefficient with heavy metals such as Cu, Pb, and Al. However, both SSC (-0.544) and Total Suspended Solid (TSS, -0.62) showed negative correlation with Hg. Methylmercury in the San Francisco Bay watershed did not have significant positive correlation with any parameters, but rather had a negative relationship with nitrite, silicates and phaeophytin (Fig 11). Negative correlation between nitrite and methylmercury might be the anoxic condition that nitrite exists. The rate of methylation is the highest in an oxic/anoxic interface so this condition might reduce the rate of methylation (Ullrich et al., 2001).

In the Coyote watershed and San Francisco Watershed, methylmercury showed no significant correlation to most of the parameters. Therefore, the main sources of methylmercury in these areas might be external inputs, especially industrial wastewater discharge. Conaway et al (2003), in their study, observed similar trends of methylmercury in the southern Bay area and they suggested further studies on methylmercury loading from out of the system.

In the Lower Sacramento watershed, Ag (0.96) and Suspended Solid Concentration (SSC, 0.82) were strongly correlated with Hg (Fig 12). This high value indicates that the main sources of Hg in central valley are historic mining legacy and bed erosion. These results show a good agreement with the prediction of Clean Estuary Institute (Clean Estuary Institute 2004). The pH in Lower Sacramento showed strong negative correlation with metals because the solubility of most metals decrease as pH went up. Methylmercury concentration in Lower Sacramento watershed had strong positive relationship to various parameters, such as Total Organic Carbon (TOC, 0.94); Fine (0.88); total nitrogen (0.86); silt (0.85); conductivity (0.855); salinity (0.82); clay (0.80); and temperature (0.69).

In the San Joaquin Delta, heavy metals showed strong positive relationship with methylmercury: Zn (0.846), Cd(0.816), As (0.724), Pb (0.675) and Cu (0.637). This relationship also showed up in the results for the Suisun Bay area: Cu (0.57), Cd (0.54), Pb (0.53), Zn (0.518) and Fe (0.50). The common factor between these two watersheds was that the largest land use types in both watersheds were agriculture (Fig 13).

In general, agricultural areas rarely change in land use and the usage of fertilizers or pesticide is high in these areas. Furthermore, agricultural areas are vulnerable to contaminants accumulation. Mercury along with other heavy metals, seem to be accumulated in the soil and is later on introduced into water body. Especially, the Suisun watershed had large proportions of non-forested wetland, which made this watershed more vulnerable to contaminant accumulation.

In these three regions, Lower Sacramento Watershed, San Joaquin Delta Watershed and Suisun Bay watershed, where the main land uses were agriculture and forest, analysis results showed strong correlation to TOC, Total Nitrogen, temperature and solids phase. Based on these results, it can be inferred that the main sources of methylmercury might be the product of on–site methylation process. These indicators – total organic carbon, temperature and total nitrogen, are closely linked to the methylation process (Ullrich et al., 2001). Table 1 and table 2 show summary of correlation coefficient analysis results.

Land use

Table 3 shows results of correlation coefficient analysis with total area land use. Hg had fairly high positive relationship with evergreen forest, industrial, mixed rangeland and reservoirs but with agricultural land uses, had a strong negative relationship (Table 3). With strip mines, the results showed a significantly low negative coefficient. In the case of MeHg, generally developed land use types - commercial and services, industrial and commercial complex, industrial, residential showed strong positive values. Methylmercury had high negative coefficient with agricultural land uses as well. The MeHg/Hg had generally similar correlation patterns with MeHg. From these results, one can deduce that methylmercury has a strong relationship with developed areas such as industrial and urban areas.

Table 4 shows the results of correlation coefficient analysis for the 100km buffer zone, and they were mostly similar to the results obtained in the total area analysis. However, the relationship between strip mines and Hg showed strong differences: the results of total area and 100km buffer zone are -0.86 and 0.53, respectively. This significant disagreement might be caused by the simple analysis method, and the fact that the strip mine land use data for correlation coefficient analysis was only based on a very small portion of the total area.

## 4. Conclusion

Mercury and methylmercury should have different strategies to control loading and meet the TMDL goals, because between mercury and methylmercury, there were no significant relationships over all the study areas.

The main source of mercury in the San Francisco Bay area was historic mining legacy. In the Lower Sacramento Watershed, where most of the mines existed, Ag showed strong positive correlation to mercury. Therefore, to reduce the concentration of mercury in this area, there should be more efforts on the restoration of abandoned mines. Clay, silt and fine sand also showed strong positive correlation to mercury, so monitoring should be more focused on sediment loading.

Methylmercury concentration is positively related to total nitrogen concentration, temperature and TOC in the San Joaquin Delta watershed and the Suisun Bay watershed. In these areas, methylmercury concentration increased along with other heavy metals as well. On site methylation might have played a key role in methylmercury loading.

In developed areas such as the Coyote Watershed and San Francisco Bay Watershed, there should be more researches on the relationship between methylmercury and industrial wastewater discharge.

Methylmercury and methylmercury/mercury was strongly linked to developed land uses including industrial, commercial and urban built-up, illustrating the need for further studies between industrial or municipal wastewater discharges and methylmercury. In conclusion, if restoration is to be effective in the San Francisco bay area, it is important to know the places to look for mercury contamination and the indicators to look for

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Table 1 Land use type of San Francisco Bay area

| Land use type | AREA(km2) | (%) |
|---|---|---|
| CROPLAND AND PASTURE | 5822.04 | 34.31 |
| HERBACEOUS RANGELAND | 2473.23 | 14.58 |
| EVERGREEN FOREST LAND | 1887.17 | 11.12 |
| MIXED FOREST LAND | 1586.26 | 9.35 |
| RESIDENTIAL | 1540.00 | 9.08 |
| ORCH,GROV,VNYRD,NURS,ORN | 623.16 | 3.67 |
| SHRUB & BRUSH RANGELAND | 545.54 | 3.22 |
| COMMERCIAL AND SERVICES | 451.07 | 2.66 |
| NONFORESTED WETLAND | 439.09 | 2.59 |
| INDUSTRIAL | 337.89 | 1.99 |
| OTHER URBAN OR BUILT-UP | 269.31 | 1.59 |
| MIXED RANGELAND | 243.67 | 1.44 |
| TRANS, COMM, UTIL | 193.31 | 1.14 |
| STREAMS AND CANALS | 178.92 | 1.05 |
| TRANSITIONAL AREAS | 94.38 | 0.56 |
| RESERVOIRS | 69.01 | 0.41 |
| STRIP MINES | 55.92 | 0.33 |
| MXD URBAN OR BUILT-UP | 39.08 | 0.23 |
| INDUST & COMMERC CMPLXS | 36.14 | 0.21 |
| LAKES | 27.11 | 0.16 |
| OTHER AGRICULTURAL LAND | 25.35 | 0.15 |
| CONFINED FEEDING OPS | 13.52 | 0.08 |
| FORESTED WETLAND | 5.74 | 0.03 |
| SANDY AREA (NON-BEACH) | 4.45 | 0.03 |
| DECIDUOUS FOREST LAND | 2.62 | 0.02 |
| BARE EXPOSED ROCK | 0.72 | 0 |
| MIXED BARREN LAND | 0.69 | 0 |
| BEACHES | 0.38 | 0 |
| TOTAL | 16967 | 100 |

Table 2. Physicochemical properties of sediment in San Francisco Bay area

| Year | 2000-07 | 2001-02 | 2001-08 | 2002-07 | 2003-08 | 2004-07 | 2005-08 | 2006-08 |
|---|---|---|---|---|---|---|---|---|
| pH | 7.34 | 7.31 | 7.21 | 7.32 | 6.90 | 6.96 | 7.03 | 7.07 |
| Clay (%) | 46.25 | 51.02 | 42.63 | 52.75 | 49.07 | 44.48 | 48.26 | 44.83 |
| Fine (%) | 70.92 | 78.02 | 64.42 | 76.81 | 76.77 | 75.81 | 78.75 | 74.54 |
| Granule + Pebble (%) | 0.98 | 0.37 | 0.73 | 2.61 | 2.49 | 2.90 | 2.37 | 2.34 |
| Sand (%) | 28.11 | 21.61 | 34.85 | 20.60 | 20.80 | 21.30 | 18.88 | 23.14 |
| Silt (%) | 24.65 | 27.00 | 21.79 | 24.06 | 27.71 | 31.33 | 30.49 | 29.71 |
| TOC (mg/L) | 1.12 | 1.06 | 1.10 | 1.65 | 1.05 | 1.15 | 1.08 | 1.10 |
| Total Nitrogen (mg/L) | 0.15 | 0.12 | 0.13 | 0.13 | 0.13 | 0.13 | 0.10 | 0.11 |
| Ag (mg/L) | 0.24 | 0.21 | 0.28 | 0.22 | 0.16 | 0.14 | 0.22 | 0.26 |
| Al (mg/L) | 41785.19 | 38207.42 | 38099.08 | 35363.17 | | 24906.54 | 36621.91 | 31974.33 |
| As (mg/L) | 8.34 | 12.08 | 7.99 | 8.78 | 6.11 | 6.90 | 9.53 | 7.78 |
| Cd (mg/L) | 0.32 | 0.25 | 0.25 | 0.26 | 0.24 | 0.29 | 0.26 | 0.18 |
| Cu (mg/L) | 42.14 | 49.13 | 39.98 | 37.79 | 38.43 | 43.77 | 40.50 | 40.12 |
| Fe (mg/L) | 38290.03 | 43653.11 | 37278.69 | 39542.75 | 37624.39 | 38240.15 | | 40792.67 |
| Hg (mg/L) | 0.21 | 0.16 | 0.29 | 0.26 | 0.23 | 0.23 | 0.18 | 0.23 |
| MeHg (µg/L) | 0.71 | 0.08 | 0.30 | 0.52 | 0.45 | 0.51 | 0.63 | 0.72 |
| Mn | 695.42 | 604.66 | 992.97 | 772.89 | 689.18 | 669.29 | 711.43 | 806.34 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| (mg/L) | | | | | | | | |
| Ni (mg/L) | 84.68 | 87.54 | 71.29 | 76.66 | 79.09 | 85.55 | 78.35 | 85.19 |
| Pb (mg/L) | 23.12 | 22.91 | 19.63 | 18.83 | 17.90 | 20.48 | 18.64 | 18.25 |
| Se (mg/L) | 0.31 | 0.22 | 0.29 | 0.29 | 0.26 | 0.23 | 0.30 | 0.14 |
| Zn (mg/L) | 117.05 | 125.60 | 103.06 | 107.17 | 110.64 | 118.23 | 116.41 | 109.68 |

Table 3 Significant correlation coefficient with mercury in San Francisco Bay area

| | | | | | |
|---|---|---|---|---|---|
| Ag | | | Se | | |
| | Lower Sacramento | 0.958 | | Lower Sacramento | 0.589 |
| | San Francisco | 0.494 | | San Joaquin Delta | 0.495 |
| | San Joaquin Delta | 0.409 | | Suisun Bay | 0.548 |
| | Suisun Bay | 0.469 | Silt 0.0039 to <0.0625 mm | | |
| Clay <0.0039 mm | | | | Lower Sacramento | 0.484 |
| | Lower Sacramento | 0.467 | | San Joaquin Delta | 0.658 |
| | San Francisco | 0.465 | | San pablo | 0.411 |
| | San pablo | 0.463 | | Suisun Bay | 0.640 |
| | Suisun Bay | 0.675 | Fe | | |
| Cu | | | | Lower Sacramento | 0.628 |
| | Lower Sacramento | 0.482 | | San Francisco | 0.469 |
| | San Francisco | 0.617 | | San Joaquin Delta | 0.739 |
| | San pablo | 0.542 | | Suisun Bay | 0.771 |
| | Suisun Bay | 0.659 | Fine <0.0625 mm | | |
| pH | | | | Lower Sacramento | 0.479 |
| | Lower Sacramento | -0.815 | | San Joaquin Delta | 0.624 |
| | San Joaquin Delta | -0.832 | | San pablo | 0.487 |
| Sand 0.0625 to <2.0 mm | | | | Suisun Bay | 0.704 |
| | Lower Sacramento | -0.473 | Total Nitrogen | | |
| | San Joaquin Delta | -0.576 | | Lower Sacramento | 0.453 |
| | San pablo | -0.483 | | San Francisco | 0.452 |
| | Suisun Bay | -0.700 | | San Joaquin Delta | -0.806 |
| | | | | San pablo | 0.564 |
| | | | | Suisun Bay | 0.506 |

Table 4 Significant correlation coefficient with methylmercury in San Francisco Bay area

| | | | |
|---|---|---|---|
| Clay <0.0039 mm | | Temperature | |
| Lower Sacramento | 0.805 | Coyote | 0.401 |
| Suisun Bay | 0.612 | Lower Sacramento | 0.697 |
| Fine <0.0625 mm | | San Joaquin Delta | 0.500 |
| Lower Sacramento | 0.878 | San pablo | 0.407 |
| Suisun Bay | 0.638 | Suisun Bay | 0.602 |
| Sand 0.0625 to <2.0 mm | | Total Nitrogen | |
| Lower Sacramento | -0.883 | Lower Sacramento | 0.866 |
| San Francisco | -0.463 | San Joaquin Delta | 0.417 |
| Suisun Bay | -0.634 | Suisun Bay | 0.498 |
| Silt 0.0039 to <0.0625 mm | | Zn | |
| Lower Sacramento | 0.853 | Coyote | 0.412 |
| San Francisco | 0.429 | San Joaquin Delta | 0.846 |
| Suisun Bay | 0.589 | Suisun Bay | 0.518 |
| SSC | | SSC | |
| Lower Sacramento | 0.445 | Suisun Bay | 0.857 |
| San pablo | 0.463 | | |

Table 5 Total area correlation coefficient of land use and mercury

| | Rate | Hg | MeHg |
|---|---|---|---|
| COMMERCIAL AND SERVICES | **0.765** | **0.622** | **0.809** |
| CONFINED FEEDING OPS | **-0.555** | -0.095 | -0.420 |
| CROPLAND AND PASTURE | **-0.588** | **-0.827** | **-0.775** |
| EVERGREEN FOREST LAND | 0.157 | **0.752** | 0.456 |
| INDUST & COMMERC CMPLXS | **0.841** | 0.514 | **0.821** |
| INDUSTRIAL | **0.824** | **0.701** | **0.898** |
| MIXED FOREST LAND | **0.908** | 0.381 | **0.817** |
| MIXED RANGELAND | 0.458 | **0.772** | 0.695 |
| ORCH,GROV,VNYRD,NURS,ORN | **-0.845** | -0.265 | **-0.715** |
| OTHER AGRICULTURAL LAND | -0.510 | **-0.971** | **-0.801** |
| OTHER URBAN OR BUILT-UP | **0.709** | 0.509 | **0.721** |
| RESERVOIRS | 0.535 | **0.785** | **0.756** |
| RESIDENTIAL | **0.734** | **0.699** | **0.821** |
| SANDY AREA (NON-BEACH) | -0.224 | **-0.895** | -0.545 |
| SHRUB & BRUSH RANGELAND | **0.811** | **0.672** | **0.880** |
| STREAMS AND CANALS | -0.564 | -0.304 | -0.526 |
| STRIP MINES | -0.146 | **-0.865** | -0.503 |
| TRANS, COMM, UTIL | **0.662** | 0.010 | 0.485 |
| Rate | 1.000 | 0.439 | **0.915** |

Table 6 100km buffer zone correlation coefficient of land use and mercury

| | Rate | Hg | MeHg |
|---|---|---|---|
| COMMERCIAL AND SERVICES | **0.868247** | **0.736415** | **0.954838** |
| CONFINED FEEDING OPS | **-0.54696** | **-0.61586** | **-0.65267** |
| CROPLAND AND PASTURE | -0.49478 | **-0.90685** | -0.74713 |
| EVERGREEN FOREST LAND | -0.19497 | **0.500427** | 0.094462 |
| HERBACEOUS RANGELAND | -0.57413 | 0.248781 | -0.32954 |
| INDUST & COMMERC CMPLXS | **0.780737** | **0.503681** | **0.773997** |
| INDUSTRIAL | **0.82161** | **0.660029** | **0.876926** |
| MIXED FOREST LAND | -0.03608 | **0.502348** | 0.213626 |
| MIXED RANGELAND | 0.450543 | **0.611109** | 0.618759 |
| MXD URBAN OR BUILT-UP | **0.540208** | **0.595989** | 0.640966 |
| OTHER AGRICULTURAL LAND | **-0.75537** | **-0.80627** | **-0.90697** |
| OTHER URBAN OR BUILT-UP | **0.815875** | **0.773016** | **0.934949** |
| RESERVOIRS | 0.424217 | **0.547067** | 0.562754 |
| RESIDENTIAL | **0.770878** | **0.793667** | **0.917608** |
| SANDY AREA (NON-BEACH) | -0.22413 | **-0.89478** | -0.54529 |
| SHRUB & BRUSH RANGELAND | **0.594948** | 0.311803 | 0.585275 |
| STREAMS AND CANALS | -0.46713 | **-0.78887** | **-0.66801** |
| STRIP MINES | 0.005181 | 0.531961 | 0.208595 |
| TRANS, COMM, UTIL | **0.777846** | 0.504889 | **0.788663** |
| TRANSITIONAL AREAS | -0.44215 | **-0.65139** | **-0.63009** |

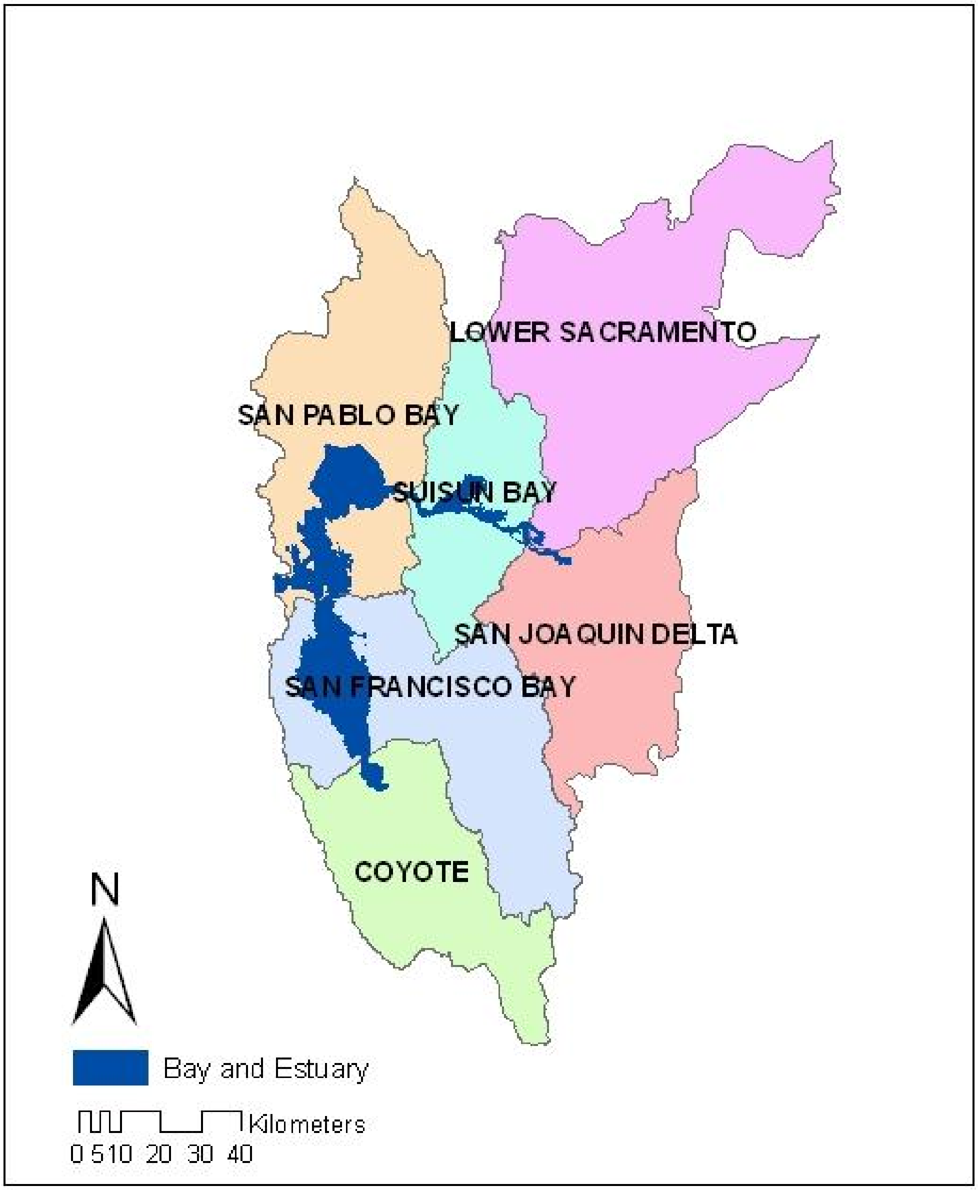



Fig. 1 Hydrological units in the study site, San Francisco Bay area

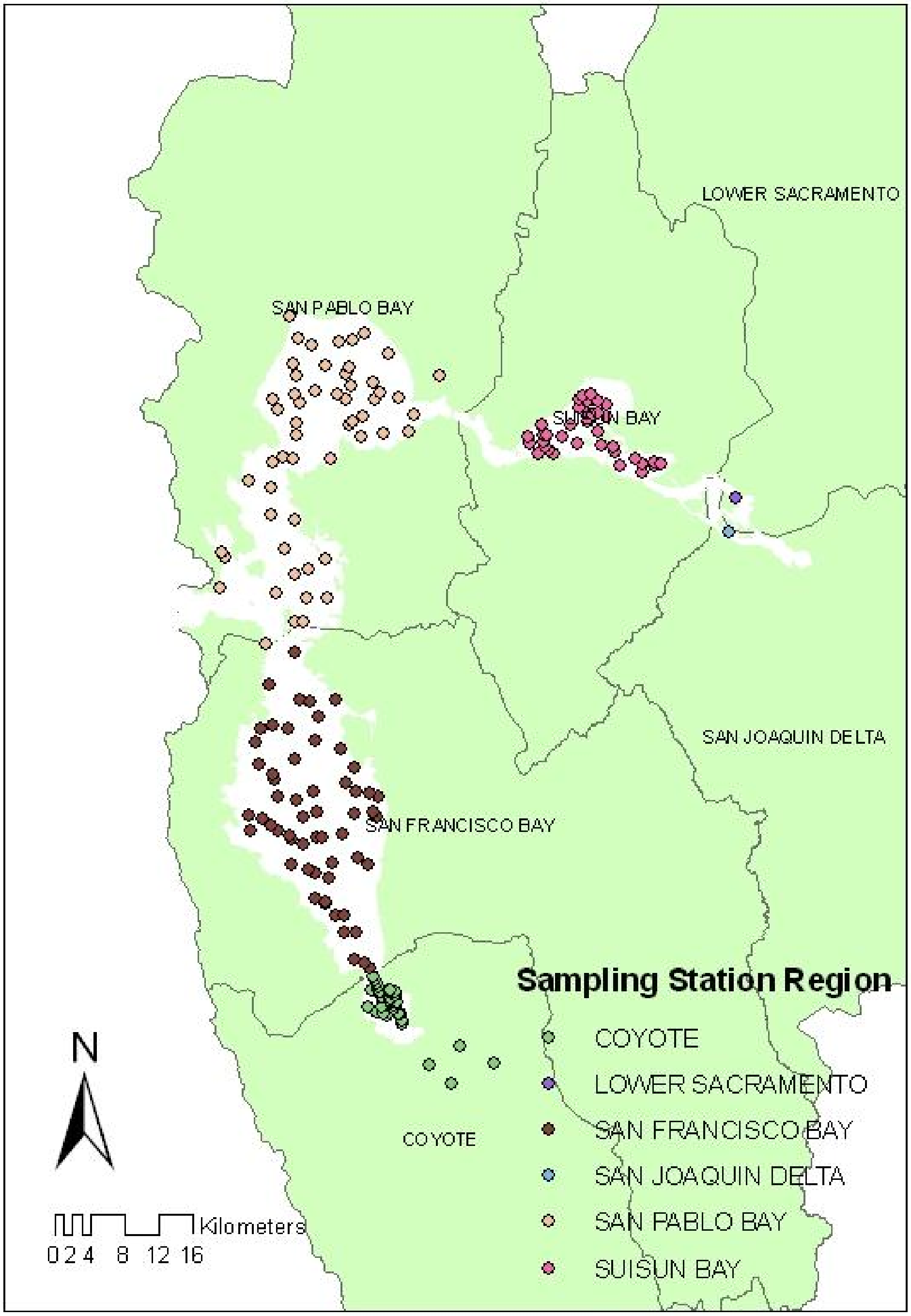



Fig. 2 Sampling points of Regional Monitoring Program (RMP)

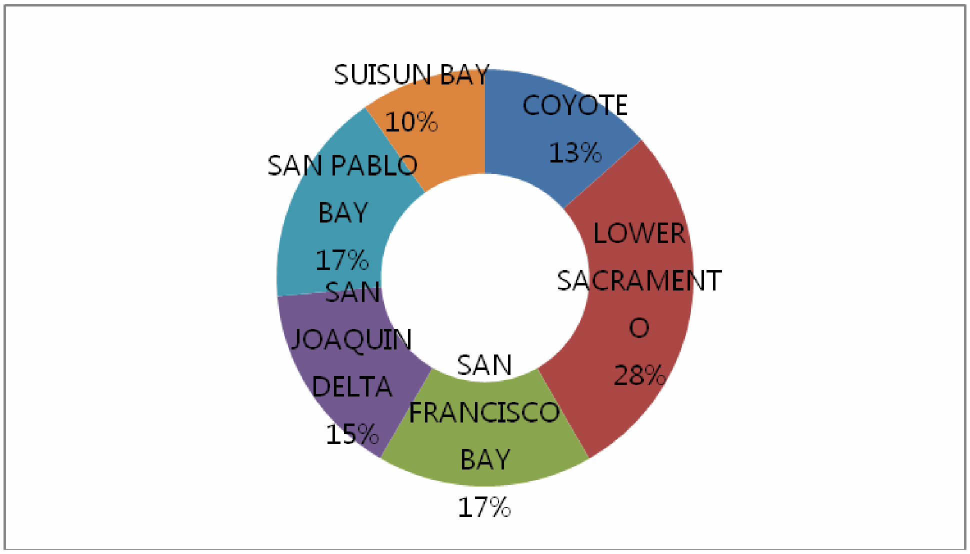
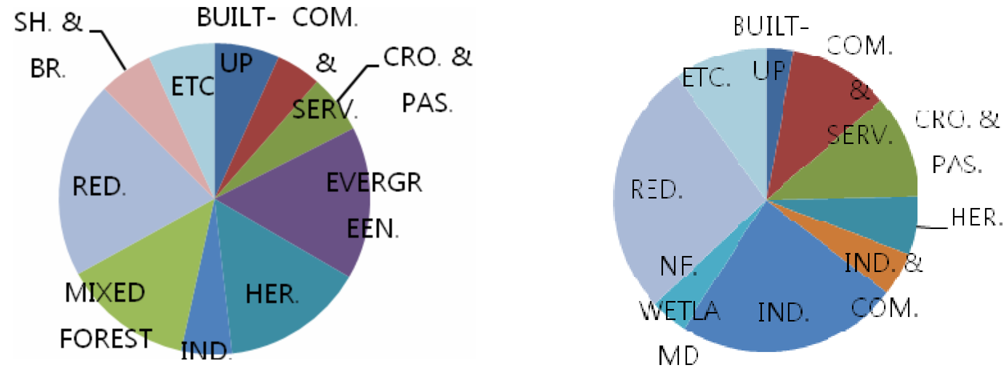



Fig. 3 Percents of total area of each watershed within the San Francisco Bay area

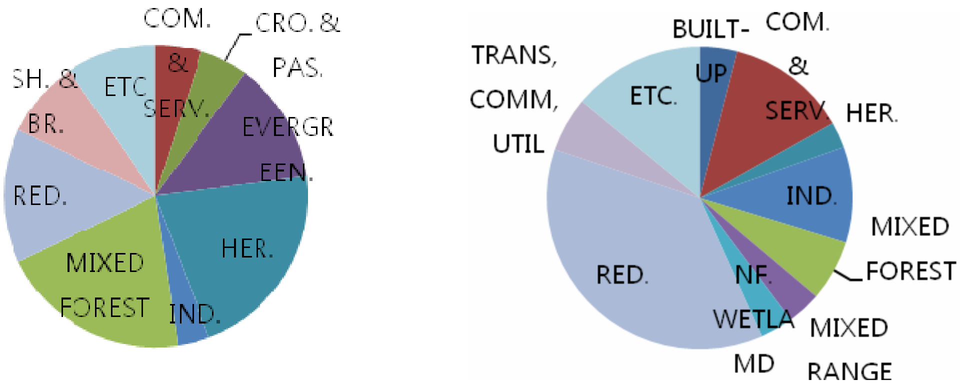



(a) (b)

Fig. 4 Land use type of total area (a) and 100 km buffer zone (b) in Coyote Watershed

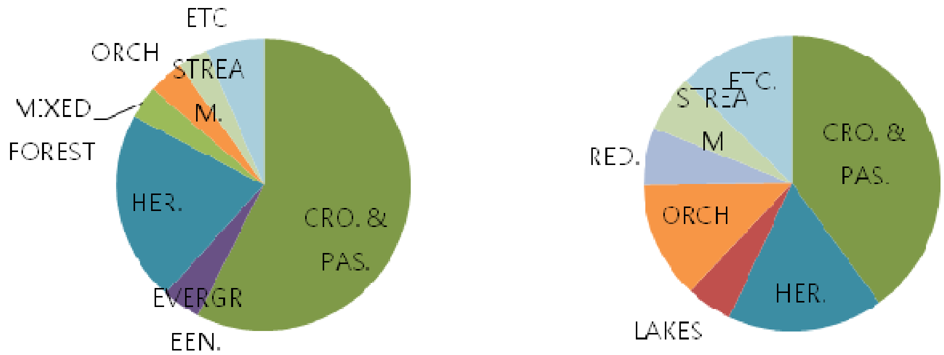

(a) (b)

Fig. 5 Land use type of total area (a) and 100 km buffer zone (b) in Lower Sacramento watershed



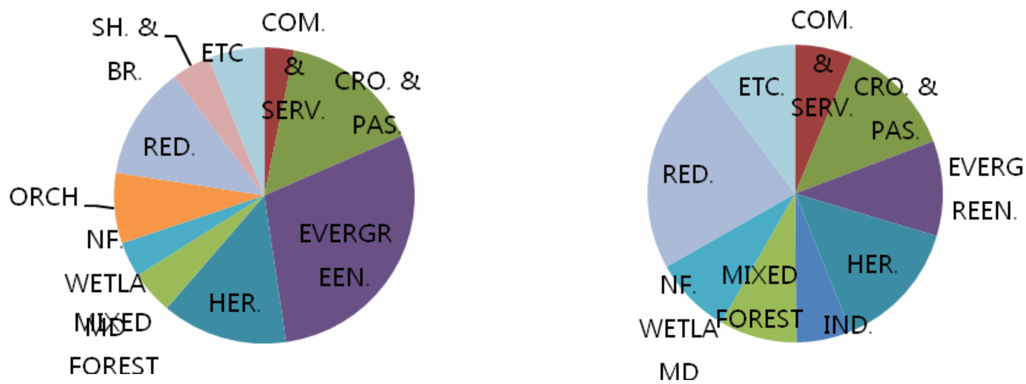

(a) (b)

Fig. 6 Land use type of total area (a) and 100 km buffer zone (b) in San Francisco Bay watershed



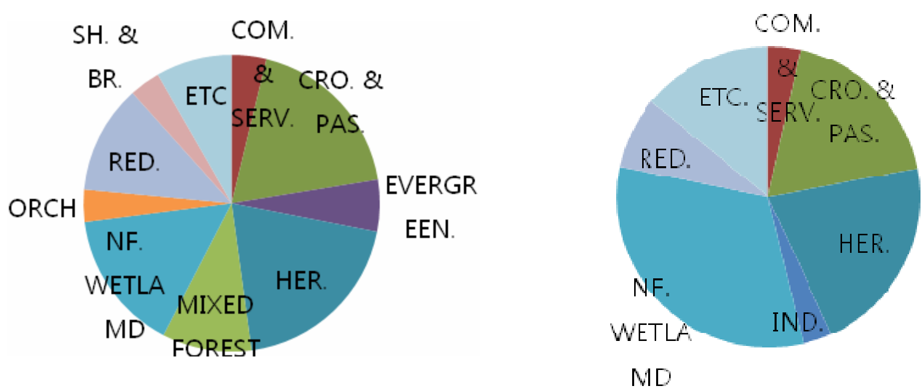

(a) (b)

Fig. 7 Land use type of total area (a) and 100 km buffer zone (b) in San Joaquin Delta Watershed




(a) (b)

Fig. 8 Land use type of total area (a) and 100 km buffer zone (b) in San Pablo Watershed




(a) (b)

Fig. 9 Land use type of total area (a) and 100 km buffer zone (b) in Suisun Watershed

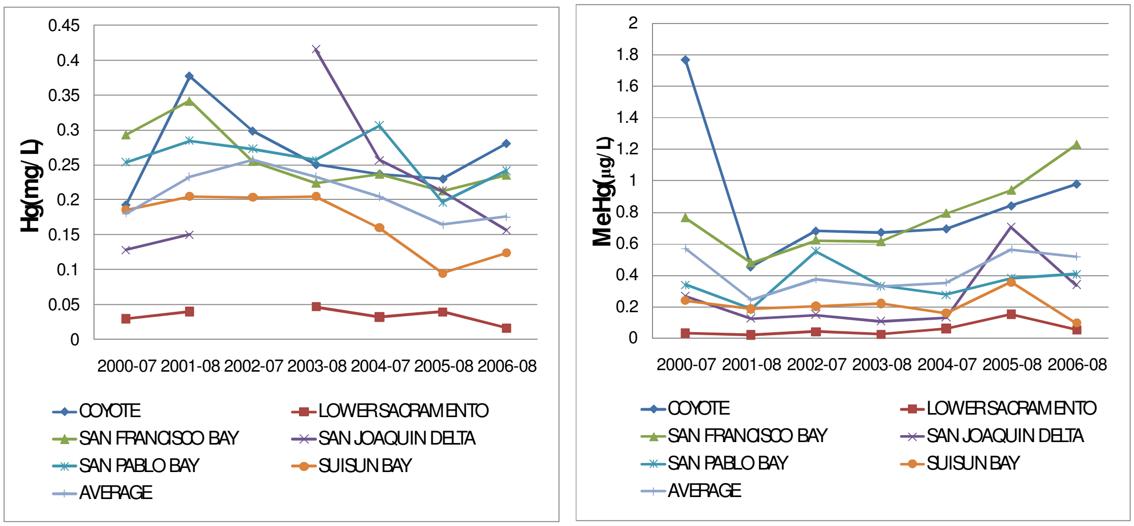
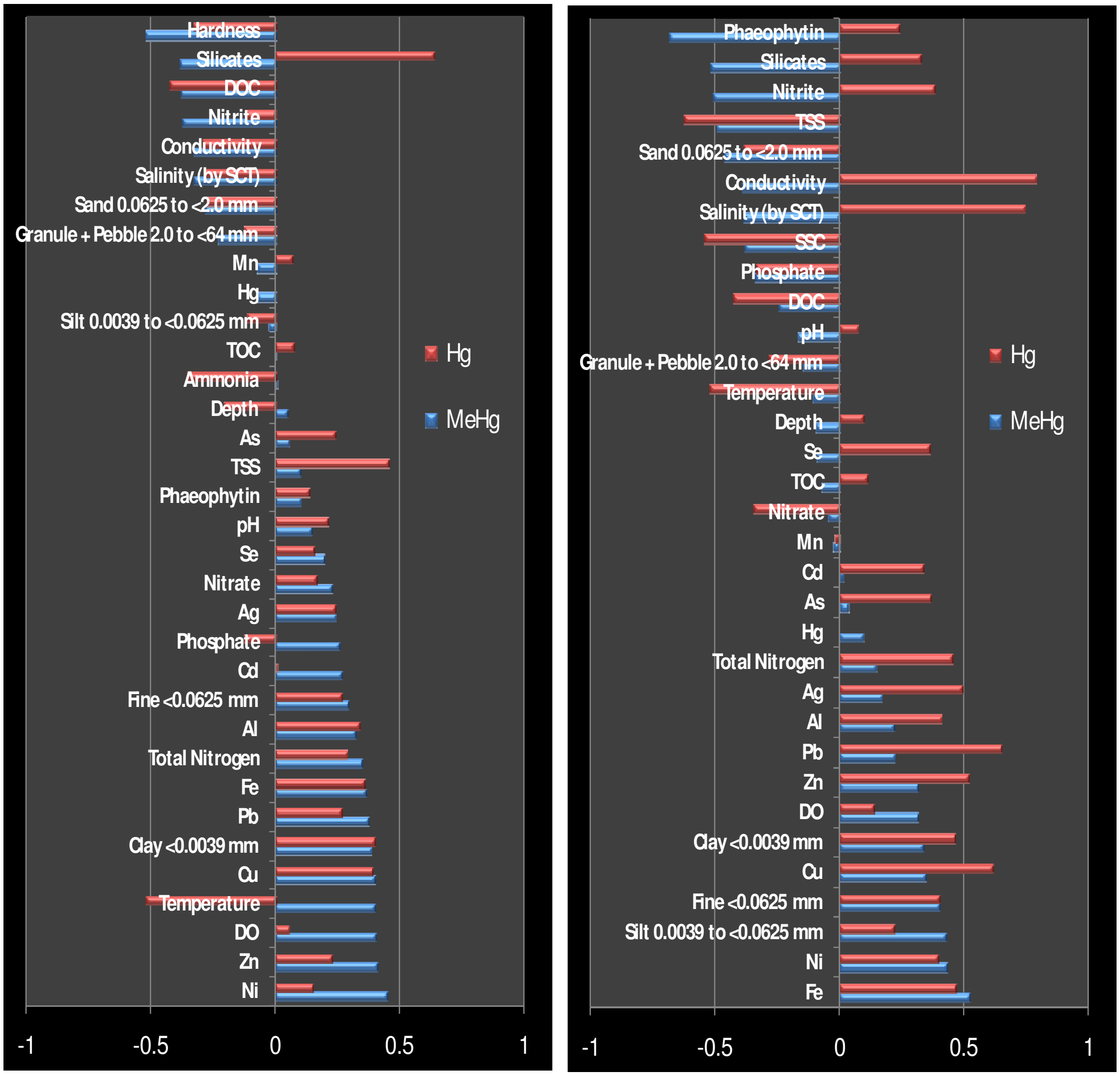
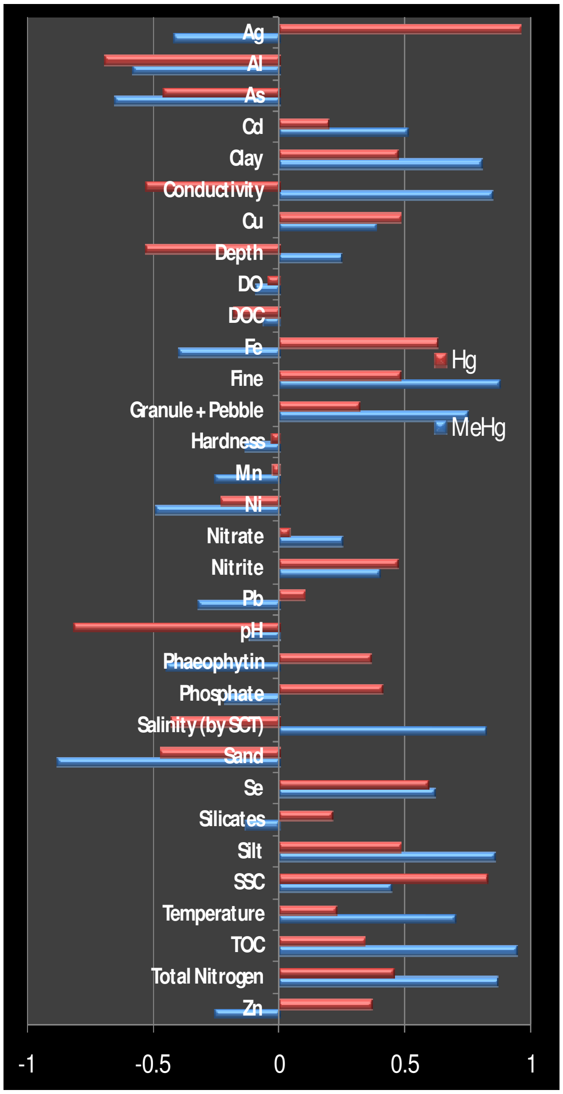



Fig. 10 Annual average of mercury and methylmercury concentration

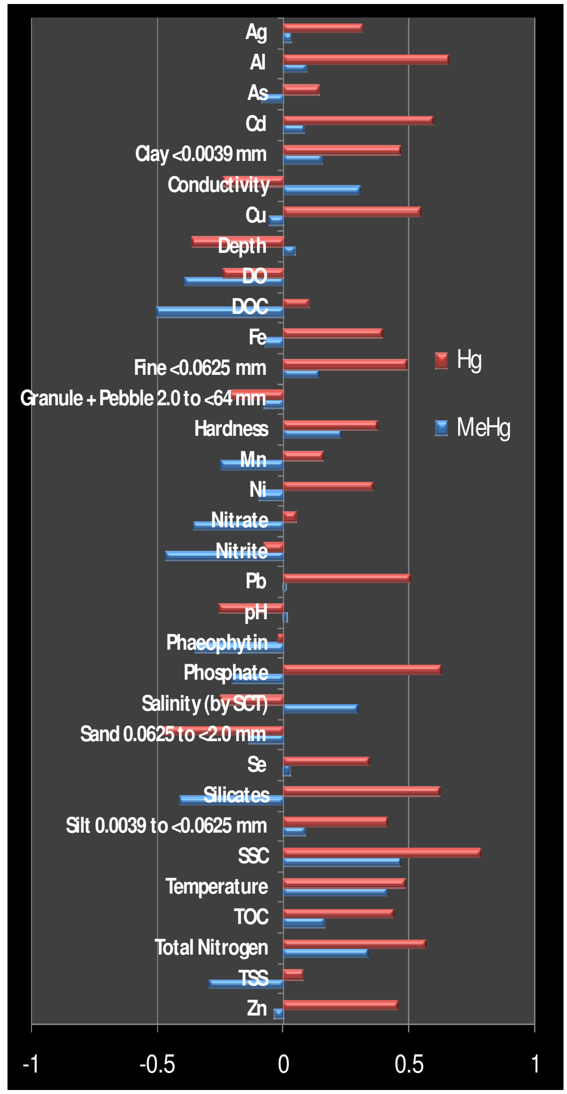



(a) (b)

Fig. 11 Results of correlation coefficient analysis in Coyote watershed (a) and San Francisco Bay watershed (b)

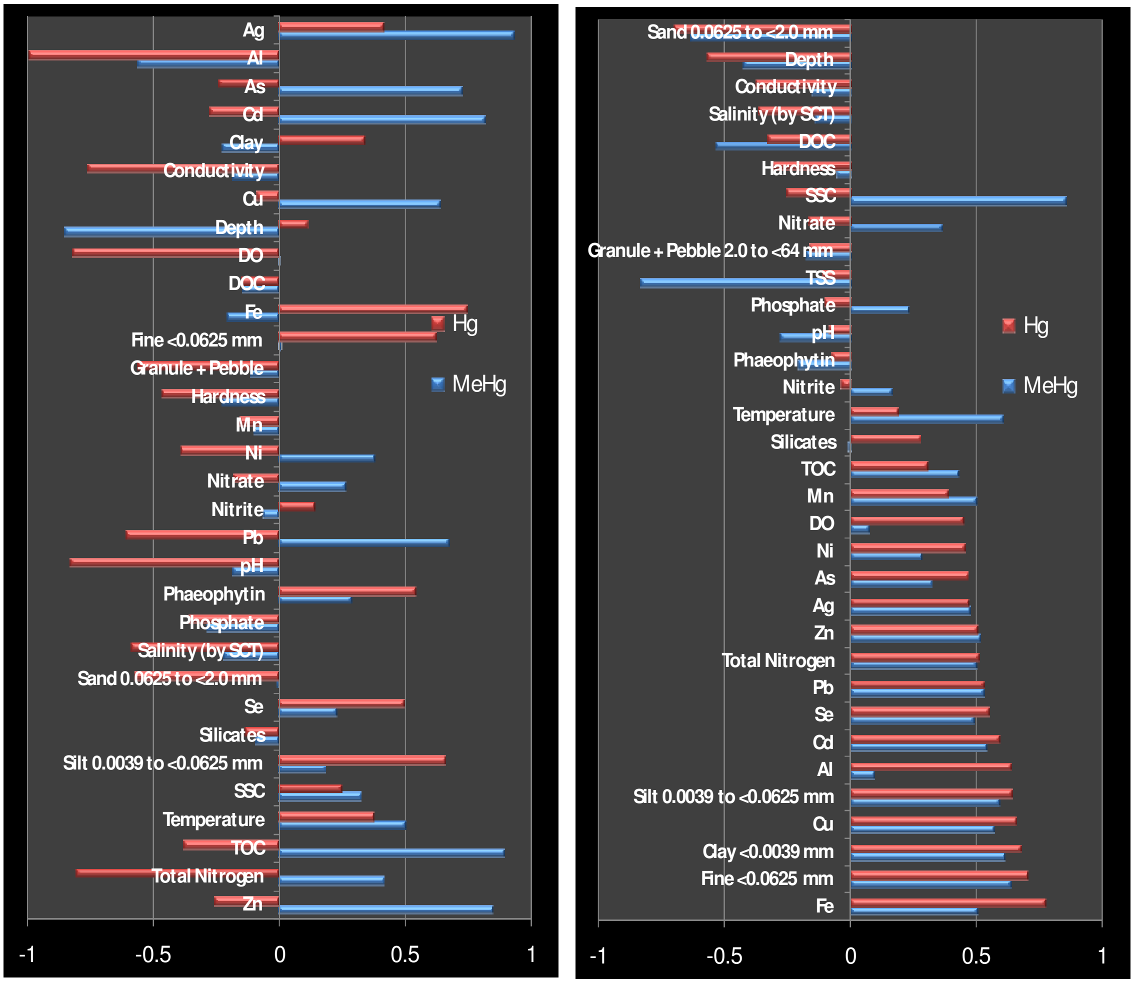



(a)




(b)

Fig. 12 Results of correlation coefficient analysis in Lower Sacramento Watershed (a) and San Pablo watershed (b)




(a) (b)

Fig. 13 Results of correlation coefficient analysis in San Joaquin Delta Watershed (a) and Suisun Bay watershed (b)