Pollutants of Concern (POC) Loads Monitoring Data, Water Year (WY) 2011

Final Report

Prepared for The Regional Monitoring Program for Water Quality in San Francisco Bay (RMP) Small Tributaries Loading Strategy (STLS)

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Abstract

This study characterized water quality during wet weather conditions in watersheds of varying size and land use to support management decisions on where to sample for loads determinations. Candidate watersheds were selected to include multiple representatives of the most common small to medium sized watershed types in the Bay Area, approximately equal distribution throughout the four counties where loads monitoring is required by the MRP, smaller catchments with heavy urban and industrial land uses, watersheds where stormwater programs are planning enhanced management actions to reduce PCB and mercury discharges, and watersheds with historic mercury or PCB occurrences or related management concerns. A set of 30 candidates was narrowed to 17 based upon balancing these criteria and financial resources. At each site, a minimum of four samples for analysis of turbidity, SSC, POC, PBCs and Hg were collected from the water column over the rising, peak, and falling stages of the hydrograph using clean hands techniques. At fewer targeted sites, samples were also analyzed for PBDEs, PAHs, and Se.

Maximum concentrations of PCBs (468 ng/L) were observed in the Santa Fe watershed in Richmond, a watershed with a long industrial history. Maximum concentrations of mercury (1665 ng/L) were observed in the Zone 5 Line M, a residential watershed with moderate imperviousness and a small amount of industrial land uses. Given the influence of variable sediment erosion, particle normalized concentrations of mercury and PCBs were used to rank the watersheds from most polluted (potentially the highest leverage in relation to treatment efficiency) to least polluted (likely lower leverage). The same six watersheds that ranked highest for PCBs also ranked highest in terms of mercury particle ratios (San Leandro Creek, Glen Echo Creek, Pulgas Creek (north and south), Santa Fe Channel, and Ettie Street Pump Station watersheds) although the order of ranking is slightly different. Ranking on the basis of methylmercury particle ratios, Santa Fe Channel, San Leandro Creek, Ettie St. Pump Station, Pulgas Creek – North, and Glen Echo Creek also ranked in the top six. Thus overall, the mercury data collected during this study appears to corroborate the PCBs findings; management efforts in these watersheds may be more cost-effective than managing any sources in some of the lower ranked watersheds.

To continue to build our understanding of the relationship between land uses and source areas, Pearson correlation, principal components analysis, and linear regression were carried out on particle ratio data transformed to achieve approximate normal distributions. Seventeen geographic variables were extracted from our GIS database for the 17 watersheds investigated in this study and an additional five watersheds that had been previously studied (Coyote Creek at Hwy 237: Z4LA, Hayward; Gellert Park, Daly City; Guadalupe River; North Richmond Pump Station). All 17 spatial variables required transformation to achieve approximate normal distribution, with the majority (10) log-normally distributed. Imperviousness was positively correlated to 1954 urban and 1954 industrial, commercial high density, and transportation; and negatively correlated to open infiltrative. Area was positively correlated to residential low density. The first four principal components explained 64% of the variation in the 17 watershed spatial attributes. The first principal component (i.e., PC1) explained 29% of the variance in the data and was largely associated with indicators of historic industrial development, including imperviousness and the historical measures of urbanization and industrialization. PC2 explained 16% of additional variance and was associated with watershed area and low density residential. PC1, PC2, and PC3 in combination explained 54% of variation in land use and were associated with land use attributes that could be hypothesized to explain pollutant concentrations. Concentrations were positively correlated across pollutants; the strongest correlations were between mercury (total and methyl) and both PBDEs and PAHs. Total Hg and PCBs were positively related to PC1, indicating an association with imperviousness and historic

industrialization. For HgT, the variability explained by PC1 was low. MeHg was not significantly related to PC1, PC2, or PC3. PBDEs and TOC were negatively related to PC3, and thus positively related to industrial land use. PAHs were negatively related to PC2, and thus positively related to watershed area. The PBDE relationship was driven by the watershed with the lowest PBDE concentration and the highest PC2 score (Borel Creek; 30 pg/mg). PC1, PC2, and PC3 were most strongly associated with imperviousness, watershed area, and industrial land use, respectively, so each pollutant was individually regressed against these three land use traits. In bivariate linear regression models, HgT, PCBs, and PAHs were most related to imperviousness, PBDEs was only related to industrial land use, and MeHg was not related to any of the three land use traits. Thus, the statistical analysis using PCA and correlation matrix identified two predominant gradients within Bay Area watersheds; one of urbanization and industrialization characterized by a variety of correlated land categories (historic urban, historic and current industrial, imperviousness, high density commercial) and the second related to watershed area and low density residential.

In the five watersheds (Lower Marsh Creek, Walnut Creek, Ettie Street Pump Station, San Lorenzo, and Sunnyvale East Channel) with historical empirical flow data available, loads calculations were made by multiplying a climatically averaged runoff volume by the particle weighted mean concentration (PWMC) of each contaminant. Maximum yields (mass per unit watershed area) were calculated for Ettie Street Pump Station watershed (82 µg PCB/m^2/y; 79 µg Hg/m^2/y). In contrast, Lower Marsh Creek was estimated to have the lowest yield for PCBs and the second lowest yield for mercury. Yield was found to generally increase with increasing imperviousness for HgT, PCBs and PAHs. Suspended sediment (SS) loads normalized to watershed area generally decreased with increasing imperviousness. Our first order HgT yield estimate in Lower Marsh Creek is relatively low despite a former mercury mine in the upper watershed, though sampling in this creek occurred only on the rise due to logistical constraints. The results presented here support previous studies which identified Ettie Street Pump Station as a high leverage watershed for PCBs. First order suspended sediment loads estimated using these methods for Walnut Creek were in general agreement with previous estimates.

Data and interpretations from this study provided support for selection of two additional loads monitoring sites (North Richmond Pump Station watershed and Pulgas Creek Pump Station watershed). In addition, the data are being used to support both the input side and for calibration and verification of the Regional Watershed Spreadsheet Model being developed by the STLS to improve estimates of regional loads. In the future, this dataset will likely have utility for measuring trends in relation to management actions over the next 5 to 10 years. Although the dataset collected is extremely valuable, should a similar effort be repeated in the future for further identification of high leverage watersheds, to support development of the RWSM, or to further explore cause-and-effect statistical relationships, some minor cost-effective improvements in sampling design were identified.

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Introduction

The ecological, economic and cultural values of estuaries and the land surrounding them typically attract dense human settlements (e.g. Chesapeake Bay, New York Bay, Wouri Estuary), and runoff from these urban landscapes is often contaminated with organic compounds (Marsalek and Ng, 1989; Walker et al., 1999; Foster et al., 2000; Curren et al., 2011) and mercury (Mason and Sullivan, 1998; Lawson and Mason, 2001) that are known to be toxic to fish and invertebrates (Davis et al., 2003; 2007). In an effort to try to reduce potential or realized impacts to coastal water resources, there have been large efforts to better define problems and causes through quantification of loads in wet and dry weather flow (Mason and Sullivan, 1998; Lawson et al., 2001; McPherson et al., 2002; Foster et al., 2003; Stein and Tiefenthaler, 2005), determine pollutant sources and source areas (Bannerman et al., 1993; Van Metre and Mahler, 2003; Stein and Tiefenthaler, 2005; Stein et al., 2006), and remove polluted sediment or treat runoff (Pitt et al 1995; Greb et al., 2000; Kang et al., 2009; Aryal et al., 2010; Park et al., 2010). Urban areas or watersheds with greater pollution sources per unit area are described as "high leverage" and are expected to be more cost effective to manage through treatment or abatement (Stenstrom et al., 1984; Park and Stenstrom, 2007; Park et al., 2009).

San Francisco Bay is listed on the Clean Water Act regulatory 303(d) list as impaired for multiple pollutants (Table 1); among these, mercury and polychlorinated biphenyls (PCBs) are the highest priority pollutants found in the Bay. Mercury has legacy sources dating back to 19th-century gold and mercury mining, as well as current use sources such as ongoing electronic uses and aerial deposition. PCBs, banned from production in the US in 1979, are a predominantly legacy contaminant from high usage during mid-20th century industrial and commercial applications (Erickson and Kaley II, 2011), though a significant source remains in enclosed transformers (USEPA, 2011) and caulking material in buildings. Mercury and PCBs are persistent contaminants that bioaccumulate in food webs and many Bay sportfish species are above regulatory thresholds for these contaminants. The San Francisco Bay Regional Water Quality Control Board has developed cleanup action plans (Total Maximum Daily Load plans, or TMDLs) to reduce contaminant loads entering the Bay from local tributaries. Mercury and PCBs TMDLs have been completed and load allocations established. In the northern embayments of San Francisco Bay, which receive stormwater flow from selenium-rich watersheds, a TMDL for selenium is in development. Polycyclic aromatic hydrocarbons (PAHs) have been placed on the 303(d) list, though a TMDL has not been developed. Polybrominated diphenyl ethers (PBDEs) remain under examination with a focused interest on the response of watersheds to the phase out of the penta- and octa-formulations, and proposed phase-out of the deca-formulation by 2013 (USEPA, 2012).

The Small Tributaries Loading Strategy (STLS) team, a subgroup of the San Francisco Bay Regional Monitoring Program for Water Quality, is focused on developing the knowledge-base necessary to inform management actions directed at reducing loads and impacts of pollutants of concern (POC) entering the Bay from small tributaries. The STLS oversees a suite of related projects aimed at answering one or more of the

Table 1. Summary of pollutants listed on the regulatory 303(d) list for San Francisco Bay and status of TMDLs.

Pollutant	**Water Body**	**TMDL Status**
Mercury	San Francisco Bay	Completed
PCB	San Francisco Bay	Completed
PBDE		Not Listed
PAH	Central San Francisco Bay	Listed
Selenium	North San Francisco Bay	In Development

following four questions: 1) Impairment: *Which are the "high-leverage[1]" small tributaries that contribute or potentially contribute most to Bay impairment by pollutants of concern?*, 2) Loads: *What are the loads or concentrations of pollutants of concern from small tributaries to the Bay?*, 3) Trends: *How are loads or concentrations of pollutants of concern from small tributaries changing on a decadal scale?*, and 4) Support for Management Actions: *What are the projected impacts of management actions on loads or concentrations of pollutants of concern from the high-leverage small tributaries and where should management actions be implemented in the region to have the greatest impact?* (SFEI, 2009).

These questions were developed in parallel with the Municipal Regional Stormwater NPDES Permit (MRP) (Water Board, 2009). As such, the same questions are also found in Provision C.8.e. of the MRP which requires Permittees to monitor a number of watersheds to generate loads information over a minimum of three wet seasons and use this data to estimate both long term watershed specific average loads (Question 1) and to make estimates of regional scale loads (Question 2). However, even though there was available local knowledge about old industrial areas (areas that were industrial during the peak use period of Hg and PCBs (1950-1990) and likely pollutant sources supported by a GIS based statistical correlation analysis (Greenfield et al., 2010), the STLS was unable to reach a consensus on which six watersheds to select for a long term loads monitoring program out of a preliminary list of 30.

The primary goal of this study was to characterize water quality during wet weather conditions in a subset of watersheds of varying size and land use to support management

[1] A concept developed by Michael Stenstrom in 1984 to describe areas of a watershed where there is a greater portion of the load coming from smaller areas. Mathematically it is calculated as the percentage of pollutant loading divided by the percentage of the area of each land use – high leverage areas are indicated by a numerical outcome >1 where a leverage of 2-5 is typical and desirable for areas of management focus (Stenstrom et al., 1984; Park and Stenstrom, 2007; Park et al., 2009). Also coopted in the Bay Area RMP mercury strategy to include habitats with high rates of net methylation (due to the combined effects of methylation and demethylation) and food web characteristics that allow for substantial and efficient uptake into the food web. Because this study was a characterization study that does not attempt to estimate loads for most watersheds sampled, we determined "high leverage" watersheds by ranking them according to their particle ratios (term defined on page 13 of this report).

decisions on where to sample for loads determinations. The key objectives were three-fold:

1. Rank the watershed outflows from greatest to least pollution[2],
2. Evaluate the strength of relationships between land uses and pollutant concentrations,
3. Develop some hypothesis level loading estimates for selected watersheds to compare with existing data.

Methods

Reconnaissance and site selection

The San Francisco Bay estuary is surrounded by nine highly urbanized counties with a total population greater than seven million people (US Census Bureau, 2010). Although urban runoff from approximately 500 small tributaries flowing from the adjacent landscape represents only about 6% of the total freshwater input to the San Francisco Bay, this input has broadly been identified as a significant source of pollutants of concern (POCs) to the estuary (Davis et al., 2007; Oram et al., 2008; Davis et al., in press; Gilbreath et al., 2012). The question remains which of these broad set of tributaries might be considered more typical of a general category of urban watershed and which might be higher leverage?

In a further effort to support management, a preliminary list of 30 candidate sampling locations for reconnaissance was developed by the STLS. The candidate watersheds were selected to include multiple representatives of the most common small to medium sized watershed types in the Bay Area, approximately equal distribution throughout the four counties where loads monitoring is required by the MRP, smaller catchments with heavy urban and industrial land uses, watersheds where stormwater programs are planning enhanced management actions to reduce PCB and mercury discharges, and watersheds with historic mercury or PCB occurrences or related management concerns.

Site visits were then conducted in each of the 30 candidate watersheds to identify the most downstream sampling location for each watershed that was upstream from tidal influence and had safe suitable overhead structure for sampling from during storm flow. Geomorphic, logistical and safety attributes were recorded as well as photo documentation at each potential sampling location. Six watersheds were removed from consideration mainly due to logistical constraints. Of the 24 remaining watersheds, the STLS selected 17 locations (Figure 1; Table 2) based upon balancing the criteria above and the financial resources available (Note, Appendix B includes a table of the watersheds not selected).

[2] In this report, we use the contaminant particle ratio (defined on page 13) to rank and determine higher leverage watersheds.

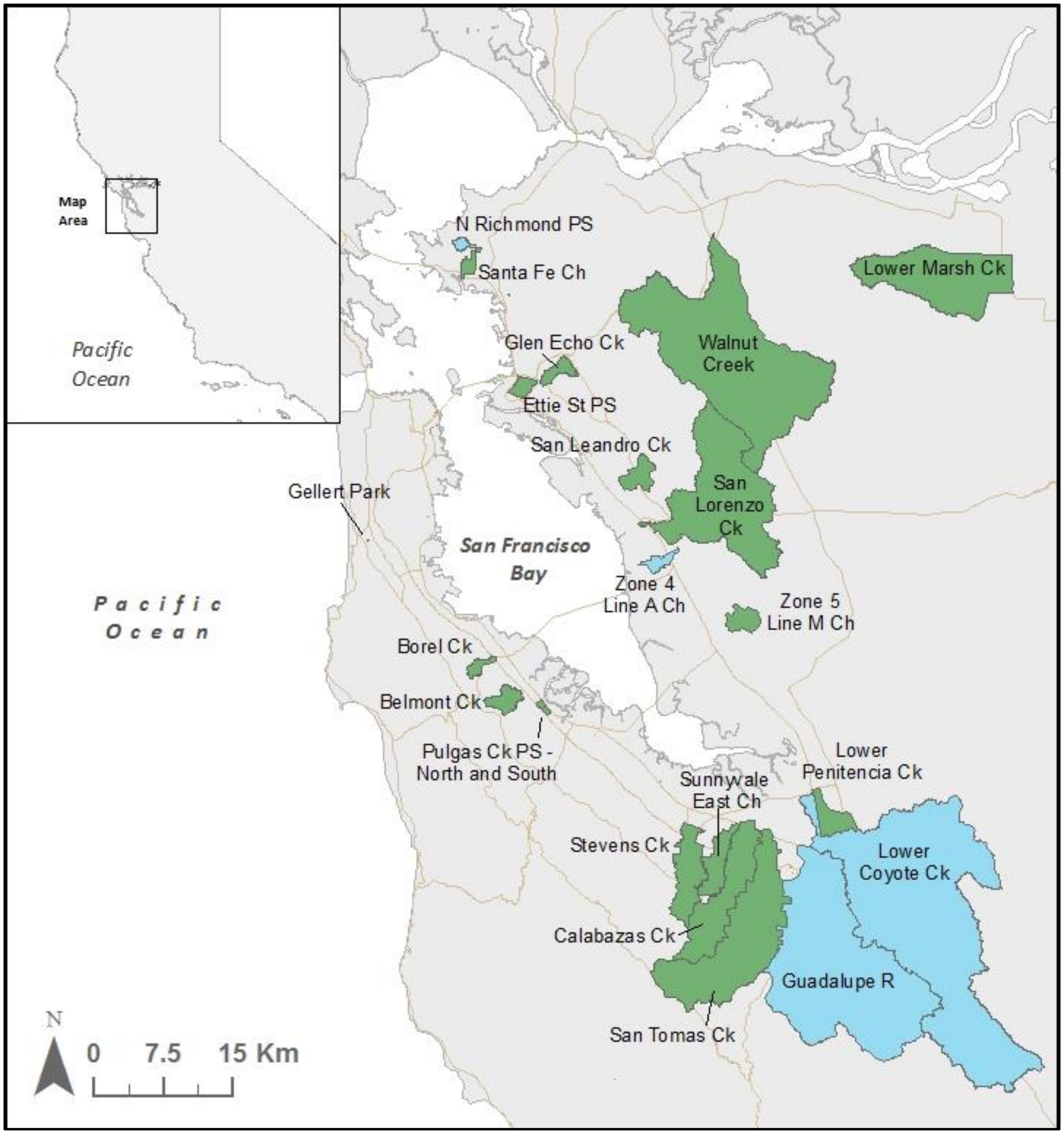

Figure 1. Map of watersheds selected for WY 2011 Characterization Study (in green). Watersheds also included in land use statistical analysis in blue (Gellert Park is blue). Channel (Ch), Creek (Ck), Pump Station (PS), River (R).

Table 2. Drainage area and land use characteristics of each watershed. The last five watersheds are included in the land use statistical analysis but were not sampled in this study.

Watershed Name	Drainage Area (km^2)	Imperviousness (%)	Old Urban (%)	Old Industrial (%)	Agriculture (%)	Open (%)	Residential (%)	Commercial (%)	Industrial (%)	Transportation (%)
Belmont Creek	7.2	27	55	0	0	10	53	15	0	23
Borel Creek	3.2	31	26	0	0	18	34	28	0	20
Calabazas Creek	50.1	44	5	0	1	14	46	13	8	18
Ettie St. Pump Station	4.0	75	100	13	0	7	28	19	13	33
Glen Echo Creek	5.4	39	100	0	0	30	39	10	0	21
Lower Marsh Creek	83.6	10	2	0	4	73	14	3	0	6
Lower Penitencia Creek	11.4	65	9	0	0	8	27	24	14	26
Pulgas Creek Pump Station - North	0.6	84	100	32	0	6	9	31	32	22
Pulgas Creek Pump Station - South	0.6	87	98	22	0	3	2	57	23	16
San Leandro Creek	8.9	38	50	0	0	27	48	6	0	19
San Lorenzo Creek	124.8	13	21	0	0	65	25	4	0	5
San Tomas Creek	108.2	33	15	0	0	28	45	10	2	16
Santa Fe Channel	3.3	69	97	2	0	4	41	18	3	34
Stevens Creek	26.0	38	23	0	0	17	52	8	1	22
Sunnyvale East Channel	14.8	59	29	2	0	3	51	19	3	23
Walnut Creek	231.8	15	15	0	0	45	37	8	0	9
Zone 5 Line M Channel	8.1	33	10	1	0	36	31	15	7	11
Gellert Park	0.02	90	98	0	0	10	0	0	0	90
Guadalupe River	232.7	39	20	1	1	26	39	11	3	19
Lower Coyote Creek	319.5	21	4	0	6	55	21	6	2	10
North Richmond Pump Station	2.0	62	68	18	0	6	27	8	33	25
Zone 4 Line A Channel	4.2	68	33	6	0	2	27	35	18	18

[1]Old industrial and urban areas are defined as areas that were in either industrial or urban land use during the peak use period of Hg and PCBs (1950-1990).

Field methods of data collection

Prior to sampling, a statistical analysis was performed to determine the number of samples needed to adequately describe pollutant concentrations for determination of loads and trends of specified accuracy, precision and power (Melwani et al., 2010). Data sets for Hg, PCBs and suspended sediment concentration (SSC) collected in two Bay Area watersheds over three wet seasons were resampled following a range of typical urban stormwater sampling designs. This analysis showed that ideally between eight and 16 samples collected during each winter season would be suitable for determining loads and trends and in some instances as few as four samples could be suitable (Melwani et al., 2010). Therefore, in part due to available financial resources and the lesser objectives here to use the data only for ranking purposes rather than loads computations, four to seven discrete samples were collected at each of the 17 sampling locations.

Samples for analysis of multiple analytes were collected from the water column over the rising, peak, and falling stages of the hydrograph. When necessary, two storms were sampled at a site in order to collect the minimum of four samples. Staff aimed to collect samples over a range of turbidity from moderately low to very high. Manual sampling of stormwater at each location was carried out either from a bridge over the water body or by dipping into a stormdrain. A Federal Interagency Sediment Program (FISP) US D-95 depth integrating water quality sampler, boom truck, crane and b-reel winch mechanism was used to lower bottles into the creek when sampling from a bridge, and a handheld FISP US DH-81 was held in the storm flow when sampling in a stormdrain.

For all sampling events, a single 1 L Teflon bottle was used as the collection apparatus and trace clean sampling protocols were used for the collection of all analytes. The 1 L Teflon collection bottle was first rinsed with site water prior to the collection of the first sample set, and prior to the collection of each subsequent sample set. The 1 L bottle was filled, inverted three times to ensure full mixing, and decanted into appropriately cleaned sample bottles. Total mercury (HgT), total methyl Hg (MeHgT), suspended sediment concentration (SSC), and turbidity were all collected from the same 1 L dip. Total and dissolved selenium (SeT and SeD) were both collected from the same 1 L dip. Polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and polybrominated diphenyl ethers (PBDEs) all were collected into 2.5 L amber bottles and thus required decanting multiple 1 L dips to fill the bottles. Total organic carbon was collected last. Turbidity was sampled intermittently throughout each sample collection and measured with a Hach 2100P portable turbidimeter.

Field duplicates were collected using the same methods and in sequential dips rather than from the same dip as the original field sample. Thus, field duplicates reflect environmental variability over a short period of time (minutes) as well as other issues of sample integrity such as inconsistences in preservation, shipping, storage, and handling prior to analysis. Field blanks were collected by first rinsing the 1 L collection bottle with approximately 100 mL of high purity water (HPW) provided by a laboratory, then filling the collection bottle with HPW and decanting into the appropriate analyte bottle.

All samples were labeled, placed on ice, transferred back to San Francisco Estuary Institute (SFEI), and refrigerated at 4 °C until transport to the laboratory for analysis. Mercury samples were preserved with hydrochloric acid (HCL) and selenium samples were preserved with nitric acid (HNO_3) at SFEI within 48 hours of sample collection.

Laboratory methods

Water samples for analysis of turbidity were collected multiple times during fieldwork before and after samples collected for pollutant analysis in an effort to better understand the correlation between SSC (which correlates strongly with turbidity) and the pollutant of interest. Samples were analyzed in the field using a portable Hach 2100 P instrument that was calibrated at the beginning of each day of fieldwork using Formazin standards provided by the manufacturer. Samples that exceeded the instrument range (0-999 NTU) were measured at SFEI's field preparation laboratory after a 5 to 1 dilution.

Water samples for analysis of SSC were collected at all 17 locations and analyzed by Moss Landing Marine Laboratory (Moss Landing, California) using a modified version of ASTM D 3977: Standard Test Methods for Determining Sediment Concentration in Water Samples, in which the entire volume of sample collected is filtered (see Gilbreath et al. 2012 for more detailed description of all laboratory methods cited in this paper). The detection limit defined by replicate blanks, was sufficiently low that no samples were reported as non-detects. Batch blank averages were all below the method detection limit. Lab replicates were not possible for SSC as the entire volume was consumed for each analysis, however field sample replicates were collected and resulted in <10% relative standard deviation (RSD).

Total organic carbon (TOC) samples were analyzed by Columbia Analytical Services using Standard Methods 5310C, 20^{th} edition (APHA, 1998). The method was sufficiently sensitive to report quantitative results for all samples. Blank samples were all non-detect. The RSD for lab, field and matrix spike (MS) replicates were all <5%. Recoveries of spiked blank samples averaged within target error range (<5%).

Water samples for PCB analysis were collected at all 17 locations. Trace organic contaminants were analyzed by AXYS Analytical Services Limited (Sidney, British Columbia, Canada). Samples were analyzed for 40 individual and co-eluting PCB congeners using AXYS Method MLA-010, including lab-specific modifications to EPA Method 1668 Revision A. Detection limits were sufficiently low that none of the congeners had extensive non-detects. Approximately 40% of the congeners reported were detected in lab blanks, and 31% of the congeners were detected in field blanks, though most were <1/3 the concentrations found in field samples. Nine PCB congeners, in some samples, were censored (not reported) due to blank contamination in laboratory blanks. Blank contamination did not affect ability to calculate a sum of PCB congeners (all samples had <50% congeners censored). There are no certified reference materials (CRMs) for PCBs in water, so recoveries were evaluated on blank spikes, with average errors within the 35% target for all analytes. Average precision (RSDs) from lab replicate analyses were generally good (<35% RSD), except for one of the congeners. PCB 052 was flagged but not censored. Field replicates were not used to assess precision because the variability in the rapidly changing watersheds prevents adequate characterization of field replicability.

Eighteen PBDE samples were collected in five of the 17 watersheds spanning a diverse range of land uses and impervious cover (31-69%). Samples were analyzed for PBDEs by AXYS Method MLA-033, a lab-specific implementation of EPA Method 1614. Sample specific method detection limits were generally sufficient for the analyses with 29% (14 of 49) of the PBDE congeners (PBDE 010, PBDE 012, PBDE 030, PBDE 032, PBDE 035, PBDE 077, PBDE 079, PBDE 105, PBDE 116, PBDE 126, PBDE 128, PBDE 181, PBDE 190, and PBDE 205) showing >=50% non-detects. Approximately 39% of the PBDE congeners reported had some contamination in the lab blanks, and three PBDE congeners had sample results censored because field sample results were <3x the blank results. No congener had more than 50% of sample results censored and results were not blank corrected. Precision on lab replicate samples was generally good, with only a few showing variation outside of the target range (35% RSD). Accuracy was evaluated by recovery in blank spike samples, as there are no CRMs available for PBDEs in water. Recoveries were within the target of 35% average error for all congeners.

Water samples for analysis of PAHs were collected in a subset of six of the 17 locations. PAHs were analyzed by AXYS Method MLA-021, a variant of EPA Methods 1624 and 8270. Total PAH concentrations reported represent the sum of 25 individual congeners routinely reported by the RMP. Method sensitivity was sufficient for most analytes with only two (5%) of the PAHs (Dibenzothiophene and Fluorene) having >=50% non-detects. Both lab blanks and field blanks had 54% of the PAHs measured for detected in one or more batches, but concentrations were usually <1/3 those in field samples. Results were not blank corrected. Field replicates were not collected given the dynamic flows and suspended sediment transport processes in the monitoring watersheds. Average precision from blank spikes was good (<35% RSD) for all analytes. Accuracy was also assessed based on blank spike results and was generally good with only one PAH (Tetramethylnaphthalene, 1,4,6,7-) having >35% error.

Water samples for total mercury and methylmercury analysis were collected at all 17 locations. Total Hg and MeHgT were also measured by Moss Landing Marine Laboratory (Moss Landing, California) using lab-specific variants of EPA Method 1631 Revision E and 1630, respectively. Detection limits for mercury (0.2 ng/L) and methylmercury (0.02 ng/L) were sufficient to report concentrations in all Hg field samples and 91% for MeHgT samples (9% non-detects). Total mercury results were blank corrected, with all blank samples except one below detection limits (however, the two other blanks in this batch were not detected, which calculated an average blank <MDL so no qualifiers were needed). Methylmercury results were not blank corrected. All MeHgT blanks were below detection limits except for one, but the average blank concentration for this batch was <MDL. No field results were flagged for blank contamination. Recoveries on certified reference materials (CRMs) and matrix spikes

showed average errors of 12% or better, well within the target average 35% error. Replicates were fairly consistent for both field (n = 4) and lab (n = 10) replicates, averaging 11% RSD or better.

Selenium was analyzed in water samples collected at three sites in Contra Costa County: Lower Marsh Creek, Walnut Creek, and Santa Fe Channel. Samples were analyzed for SeT and SeD by Brooks Rand Labs (Seattle, Washington) following Brooks Rand method BR-0060, a lab specific implementation of EPA Method 1638. Detection limits for total and dissolved Se analyses (both species ~0.03 µg/L) were sufficient with all results reported (no non-detects). Field samples were all reported blank corrected, and though Se was found in blank samples, no flags were applied for blank contamination. Precision on lab and field replicate analyses were all <5% RSD, well within the target (<35% RSD). The average error on CRM and MS recoveries were <10%, also all well within the target average error of <35%.

Analysis methods

Particle normalized concentrations

In order to rank the watersheds from most polluted to least polluted, confounding factors such as sediment erosion characteristics need to be controlled. This is particularly necessary in the Bay Area because watersheds here produce a wide range of sediment loads relative to watershed area (Lewicki and McKee, 2010). This leads to the possibility of variation of pollutant concentrations caused by erosional properties of the landscape in addition to variation associated with release of pollutants from various sources. Concentrations of pollutants of concern were normalized by the corresponding suspended sediment concentration to derive an estimate of particle concentration (mass of POC per mass of suspended sediment concentration either directly measured or estimated using turbidity). The resulting ratio, for example the ratio of total PCBs concentration to suspended sediment concentration (pg PCB: mg SSC) is an estimate of the particle concentration if we assume all PCBs are transported in a particle form. Since this is likely not true, we preferred to use the term "particle ratio" rather than particle concentration. These normalized concentrations were used for the correlation statistical analysis of pollutants in relation to land uses and for ranking the watersheds from most polluted to least polluted as a means of identifying likely high leverage watersheds.

Statistical correlation analysis between pollutants of concern and land use

To continue to build our understanding of the relationship between land uses and source areas and the release of contaminants to stormwater conveyances and ultimately to the Bay, the field data collected in this project was investigated for land use relationships. All statistical analyses were performed in R version 2.15.0 (R Development Core Team 2012). Prior to analysis, POC particle ratio data were transformed to achieve approximate normal distributions (Kirchner, 2001) and multivariate linear relationships. Each parameter was then centered and scaled by subtracting the mean and dividing by the standard deviation[3]. Transformations included members of the Box-Cox transformation series (Draper and Smith 1998): log transformation, square root transformation and 1/(square root) transformation, with some variables being arcsine (square root) transformed, and one variable being cube root transformed[4].

The correlation structure of the geographic variables was examined using a correlation matrix based on Pearson correlation coefficients on the transformed data. Because land use is described by a large number of correlated variables, Principal Components Analysis (PCA) was then performed on the correlation matrix for the 17 geographic variables (Everitt and Hothorn 2011). PCA condenses the majority of variation in the land use variables into a smaller number of principal components. The number of components retained was selected based on the cumulative proportion of variance explained and the component beyond which the variance on a scree diagram declined linearly (Figure 2). PCA was performed with a dataset comprising 206 watersheds (based on SFEI 2010), in order to be representative of spatial variability in all Bay Area watersheds[5].

In addition to the 17 locations described in this study, a further five locations (Coyote Creek at Hwy 237: SFEI unpublished data; Z4LA, Hayward: Gilbreath et al., 2012; Gellert Park: David et al., 2011; Guadalupe River: McKee et al., 2006; North Richmond Pump Station: Hunt et al., 2012) were previously monitored and included in this analysis. Therefore, pollutant and TOC concentrations in 22 watershed locations were available. The sampling locations were generally upstream from the "pour point" of the full watershed, to refrain from sampling in the tidal portion of the drainage. Nevertheless, for 17 of the 22 sampling watersheds, the land use abundance and types corresponded approximately or exactly to the full watershed in the GIS database. However, five of the field-monitored watersheds were very small in comparison to the full watersheds defined in the GIS database (ranging from 0.04% to 8% of the size of the full watershed). These five sampling "subwatersheds" (Zone 5 Line M, Gellert Park, Pulgas Creek South, Pulgas Creek North, and North Richmond Pump Station) were added to the 201 GIS watersheds data set prior to performing the PCA, resulting in 206 data points for the PCA.

[3] These treatments are needed to perform principal components analysis. Transformation also improves linear regression in two ways: 1. Transformation achieves a linear relationship between predictor and response variables that is needed for regression parameter estimates to be valid; 2. Transformation achieves residual normality and variance homoskedasticity (homogeneity), which are needed for valid estimates of statistical significance.

[4] The transformation selected for each variable was based on achieving approximate normality of the result. This judgment was based on: comparing the mean and quartiles for symmetry; skewness and standard error; kurtosis and standard error; coefficient of variation; examination of histogram and normal scores plots; and the Shapiro-Wilk test for normality. These are the standard approaches used to decide upon appropriate transformation prior to linear modeling (Draper and Smith 1998, Crawley 2007, Hair et al. 2009).

[5] This sample size is sufficient to achieve high confidence in PCA results (Hair et al. 2009).

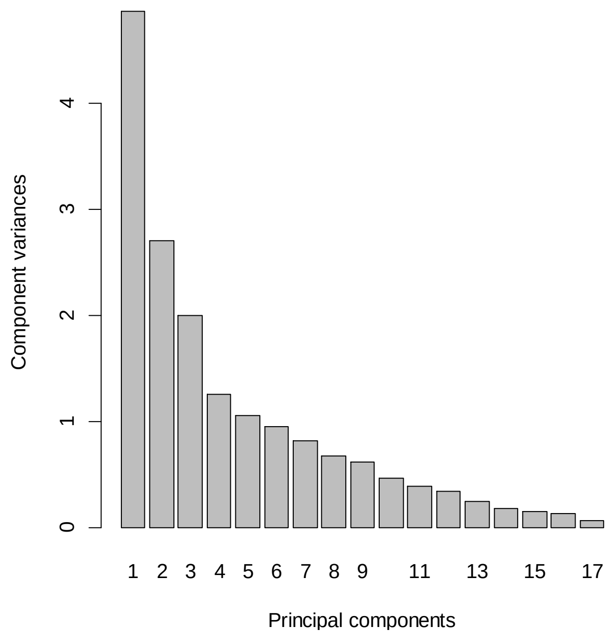

Figure 2. Scree plot of PCA results for spatial attributes in 206 watersheds.

Linear regression analysis was performed to evaluate the association between land use attributes and pollutant concentrations. For each pollutant (Table 3), particle normalized average watershed concentrations were regressed against the PCs that explained the most variability in land use attributes (Everitt and Hothorn 2011)[6]. All linear combinations of the PCs were evaluated, and those with $p < 0.05$ were retained in the final model. Pollutant concentrations were then regressed against the individual land use attributes (Table 3) that were most associated with the significant PCs. For all regression models, variables were transformed (Table 3). Model residuals were checked for normality, variance homoskedasticity, curvature, and influential outliers (Kirchner, 2001).

All 17 spatial variables required transformation to achieve approximate normal distribution, with the majority (10) log-normally distributed (Table 3). Most watersheds exhibited less than 10% abundance of any single land use attribute types (median < 10%), with only a few watersheds relatively high (maximum > 50%). However, results were generally higher for imperviousness, 1954 urban, and 1974 urban, indicating the highly developed and urbanized nature of Bay Area watersheds. Greater than 30% of the watersheds had zero values for agriculture, water, 1954 industrial, and null (Table 3).

[6] Regressing the pollutants versus the land use Principal Components is beneficial because 17 land use attributes is too many to examine as meaningful predictor variables. A small number of principal components that explain most of the variation in land use attributes serve as "master variables" that can be tested to see if they also describe pollution.

Table 3. Summary statistics and chosen transformation for all study variables.

Parameter	Abbreviation	N	Median	Maximum	CV (%)	Zero values (%)	Transform [a]
Area (km^2)	area	206	4.1	960.2	392	0	Log
Imperviousness (%)[b]	imperv	206	42	90	57	0	Arcsine(Sqrt)
1954 Urban (%)[c]	oldurb	206	31	100	91	17	Sqrt
1954 Industrial (%)[c]	oldind	206	0.3	37	208	33	Log
1974 Urban (%)[c]	oldurb74	206	86	100	54	7	Arcsine(Sqrt)
1974 Industrial (%)[c]	oldind74	206	2	67	177	22	Log
Agriculture (%)	ag	206	0	18	510	80	Log
Commercial high density (%)	cohigh	206	4	95	174	18	Log
Commercial low density (%)	colow	206	4	100	216	18	Log
Industrial (%)	oldind	206	3	80	165	15	Log
Null (%)	null	206	0.02	40	311	34	1/Sqrt
Open infiltrative (%)	openinf	206	5	100	147	18	Cube Root
Open impervious (%)	opencomp	206	6	100	135	7	Log
Residential high density (%)	reshigh	206	18	68	91	18	Arcsine(Sqrt)
Residential low density (%)	reslow	206	1	53	192	29	Log
Transportation (%)	trans	206	15	98	89	4	Sqrt
Water (%)	water	206	0.1	55	319	37	Log
Total mercury (ng/mg)		22	0.33	4.5	145	0	1/Sqrt
Methylmercury (ng/mg)		18	0.0019	0.008	88	0	Log
PCBs (pg/mg)		22	61	1,403	166	0	Log
PBDEs (pg/mg)		8	192	453	62	0	Sqrt
PAHs (pg/mg)		8	3,953	130,241	179	0	Log
TOC (mg/mg)		19	0.037	0.17	82	0	Log

[a.] Sqrt = $X^{1/2}$. Cube root = $X^{1/3}$. 1/Sqrt = $X^{-1/2}$. Log = Ln(X)

b The National Land Cover Dataset (2001) (Homer et al., 2004) spatial datalayer was used to estimate percent impervious for each watershed.

c. 1954 and 1974 urban and industrial is classified for each watershed using the USGS spatial datalayers for the Dynamic Mapping of Urban Regions for the San Francisco Bay Area project (http://landcover.usgs.gov/urban/umap/pubs/urisa_cb.php) developed by the USGS Land Cover Institute. Industrial areas are specifically called out in the 1974 layer. To derive 1954 industrial, we took the intersection of 1954 urban areas and current industrial areas, making the following assumptions: 1) industrial areas in 1954 are mapped as "urban" in that layer, 2) current industrial landscapes that were urban in 1954 were also industrial in 1954, and 3) industrial areas in 1954 have not been redeveloped into other types of landscape.

Loads estimation methods

In the five watersheds (Lower Marsh Creek, Walnut Creek, Ettie Street Pump Station, San Lorenzo Creek, and Sunnyvale East Channel) with historical empirical flow data available, loads calculations were made by multiplying a climatically averaged runoff volume by the particle weighted mean concentration (PWMC) of each contaminant. Because the five watersheds have varying periods of runoff record and the San Francisco Bay Area experiences extreme climatic variability, a climatically averaged volume (CAV) was developed for all of the watersheds in order to improve the validity of comparisons between them and with other existing loads data. CAV was estimated by scaling local rainfall records to the watershed and then using the empirical rainfall versus flow volume regression to estimate the CAV based on watershed average annual rainfall for the period 1971 to 2000 (PRISM rainfall data). Two rainfall-runoff regressions were used for the Sunnyvale East Channel, where flow data published by Santa Clara Valley Water District are known to have some anomalies[7]. Based on rainfall runoff relations, land use and imperviousness in the watershed and the results of our spreadsheet model (Lent and McKee, 2011; Lent et al., 2012), we suggest that the higher runoff volume estimate is likely more accurate given the imperviousness of the watershed, but we cannot be certain without further field verification and therefore we report a higher and lower estimate. Given the majority of loads are transported in Bay Area rivers and urban creeks during peak flow when SSC is also high, and given flow was not measured during water sampling, a weighted mean concentration for each contaminant in each watershed was estimated using SSC as a surrogate for flow computed by:

$$\text{PWMC} = \frac{\sum_{i=1}^{n} SSC * POC}{\sum_{i=1}^{n} SSC}$$

Where:
SSC and POC represent the analytical pairs of field samples for suspended sediment concentration and any pollutants of concern.

The particle weighted mean concentration (PWMC), thus derived, is probably a slight under estimate of the true flow weighted mean concentration since the variation in flow during a run-off event is likely to be three orders of magnitude whereas the variation of suspended sediment concentration was less than one order of magnitude in some of our impervious urban systems. With this caveat in mind, the derived PWMC for each contaminant was then applied to the climatically averaged annual runoff volume to make first order estimates of total average annual loads. Results are considered coarse, though

[7] Santa Clara Valley Water District is aware of problems with the gauge they operate on Sunnyvale East channel and are planning to upgrade and re-rate discharge measurements collected as part of their ongoing gauging maintenance program.

within believable ranges for the San Francisco Bay Area. Furthermore, first order suspended sediment loads estimated using these methods for Walnut Creek were also in general agreement (<5% RPD) with average annual sediment loads previously reported based on five years of data (WYs 1966-70) for Walnut Creek (110,600 US short tons; 100,300 metric t: Porterfield, 1972).

Results and interpretation of concentration data

Variation of pollutants of concern between watersheds

Total organic carbon (TOC)

Concentrations of TOC ranged from 2.11 mg/L (Santa Fe Channel) to 13.2 mg/L (Walnut Creek), with <2.5-fold variation within sites (Table 4). The correlation between TOC concentration and SSC concentration (r^2=0.27) was poor likely due to the lack of concurrence of the sample peers that were on average collected about 17 minutes apart and up to 45 minutes apart. Turbidity and TOC samples that were collected simultaneously provided an improved correlation, though still not strong (r^2=0.36). The average particle ratio of TOC to SSC, for each site, ranged between 0.0091 mg TOC: mg SSC and 0.17 mg/mg with an average of 0.043 mg/mg and a median of 0.031 mg/mg. TOC was somewhat correlated with pollutants measured. In decreasing order, TOC was correlated with HgT, PCBs, PAHs, MeHgT, and PBDEs (r^2 ranged from 0.46 down to 0.04, respectively).

Table 4. Concentration range, mean and sample count (separated by a semicolon) for organics contaminants and TOC in the 17 watersheds.

	PCBs (ng/L)			PAH (ng/L)			PBDEs (ng/L)			TOC (mg/L)		
Site	Range	Mean	N	Range	Mean	N	Range	Mean	N	Range	Mean	N
Belmont Creek	2.83-4.91;	3.60;	3							6.20-10.4;	8.05;	4
Borel Creek	3.41-8.67;	6.13;	3	437-1044;	842;	3	9.03-19.9;	13.6;	3	7.06-8.43;	7.72;	5
Calabazas Creek	5.11-24.8;	11.5;	5							4.54-9.70;	6.92;	5
Ettie Street Pump Station	35.8-69.0;	59.0;	4							2.62-5.16;	3.67;	4
Glen Echo Creek	5.64-85.8;	30.0;	4							5.32-8.00;	6.54;	4
Lower Marsh Creek	0.70-4.14;	2.15;	6	76.8-212;	140;	5				4.50-8.86;	6.69;	6
Lower Penitencia Creek	1.14-1.85;	1.48;	4	135-456;	248;	5	12.8-21.5;	17.9;	4	2.98-4.55;	3.73;	4
Pulgas Creek South	19.1-53.9;	31.5;	4							5.00-6.98;	5.76;	4
Pulgas Creek North	43.3-84.5;	60.3;	4							4.00-4.48;	3.96;	4
Santa Fe Channel	25.4-468;	198	5	667-4684;	2117;	3	24.1-30.5;	27.3;	2	2.11-4.59;	3.29;	5
San Leandro Creek	4.59-31.3;	12.4;	7							5.42-7.95;	6.14;	7
San Lorenzo Creek	5.70-20.4;	12.9;	5							4.87-7.08;	5.90;	6
Stevens Creek	3.17-17.6;	7.53;	6							5.73-7.23;	6.14;	6
San Tomas Creek	1.62-4.37;	2.83;	5							5.44-8.99;	7.18;	5
Sunnyvale Channel	9.41-67.5;	39.2;	5	1203-2790;	2148;	4	12.3-99.6;	56.2;	5	2.80-3.42;	3.07;	4
Walnut Creek	3.69-24.4;	9.00;	6							6.07-13.2;	9.70;	6
Zone 5 Line M	16.7-26.3;	20.8;	4	709-1413;	1061;	2	34.0-128;	75.0;	4	5.04-7.86;	5.94;	4

Polychlorinated biphenyls (PCBs)

Total PCB concentrations in water ranged over three orders of magnitude between sites, whereas variation within sites was less extreme (all sites <1 order of magnitude) (Table 4). We measured maximum PCB concentrations in Santa Fe Channel (468 ng/L) that were greater than measured in any other Bay Area watersheds (McKee et al., 2006; Gilbreath et al., 2012). The most contaminated watersheds by concentration were also low in SSC, creating a large divide between them and the majority of the watersheds sampled when normalized to SSC (Figure 3). These watersheds may be thought of as high leverage with regards to PCB contamination and loads; the evidence generated here would suggest that management efforts in these watersheds may be more cost-effective for reducing PCB loading to the Bay than looking for and managing any sources in some of the lower ranked watersheds. Lower Marsh Creek was ranked the lowest in relation to PCB particle ratios. Management of PCB mass in Lower Marsh Creek would likely be the least cost effective per unit mass of PCBs treated or removed.

Changes in total PCB concentrations were well explained by changes in SSC for some of the watersheds (35% watersheds having least squares regression correlation coefficients (r^2) > 0.8). Variation in other watersheds was poorly correlated with SSC (30% of the watersheds with $r^2 < 0.3$). PCB congeners 110, 138 and 153 proportionally contributed the greatest amount of PCBs (7, 9 and 7% respectively) to the total based on the median for all samples combined. Indicators of Aroclors 1242 (PCBs 18, 28, 31 and 33), 1248 (PCBs 44, 49, 66, 70 and 74), 1254 (PCBs 87, 95, 99, 101, 110 and 118), and 1260 (PCBs 149, 170, 180 and 187) on average summed to 4, 7, 26 and 19%, respectively, showing a predominance of the more highly chlorinated Aroclors (Table 5).

Polybrominated diphenyl ethers (PBDEs)

Concentrations in the sample set ranged two orders of magnitude, and mean concentrations differed between sites five-fold (Table 4). Normalized to SSC, Borel Creek stands out as being lower than the others; at this time we cannot speculate on the reason. As also noted in the three other studies that report on whole water concentrations of PBDEs for urbanized river or storm drain systems (Guan et al. 2007; Oram et al. 2008; Gilbreath et al. 2012), the sum of PBDEs for each sample was dominated by BDE 209, comprising between 41-77% of the sum (avg. 66%). BDE 99 was the next highest average contributor to the sum (9%), followed by BDE 207 (7%), BDE 47 (6%), and BDEs 206 and 208 (each 5%). All other congeners summed together for each sample comprised less than 14% of the total sum, and on average only 8%.

Generally, correlations between PBDEs and SSC were challenged at all five sampling locations due to the small variation in SSC between samples. Only Sunnyvale Channel, which had the best r^2 correlation (0.92), had SSC variation greater than four-fold, while other watershed SSC samples ranged by less than two-fold. Despite these limitations, SSC explained changes in total PBDEs concentration well at three sites (Sunnyvale Channel, Lower Penitencia, Zone 5 Line M, r^2>0.7) and poorly at Borel Creek (r^2>0.3). Correlation between SSC and PBDE concentration was not possible for Santa Fe Channel due to the small sample size.

Polyaromatic hydrocarbons (PAHs)

At all sites, PAH concentrations ranged three orders of magnitude (Table 4) with the lowest concentration sampled equaling three times the average water column PAH concentration in the San Francisco Bay (26 ng/L; Greenfield and Davis, 2004). Similar to findings in other studies reporting PAHs in urban runoff (Stein et al., 2006; Hwang and Foster, 2006), PAH composition was dominated by high molecular weight PAHs (HPAHs), averaging 84% of the sum.

The explanatory strength of SSC for total PAH concentrations appeared to be influenced by imperviousness. Of the six watersheds sampled for PAHs, three have relatively high imperviousness (59-69%), one has moderate imperviousness (31%), and one has relatively low imperviousness (11%) (the 6th site has moderate imperviousness (33%) but only two samples were collected and could not be regressed with turbidity). The highly impervious watersheds all had strong correlations (r^2>0.76), while the moderate and low impervious watersheds had lower correlations (r^2=0.64 and 0.0014, respectively).

As might be expected for urban watersheds, the ultimate origin of the PAH loading appears to be mixed. A chi-square test was used to help indicate if any particular PAH source dominated at any of the sites. Average percent contributions of 12 common PAHs at each site were compared to common source profiles (see Van Metre and Mahler, 2010 for chi-square test procedures and source profile references). No single source resulted in a chi-square value less than 0.25. A first flush effect was noted in all four of the watersheds in which three or more PAH samples were collected in the same storm event; total PAH particle ratios were generally higher in the early part of the storm and steadily decreased as the storm continued.

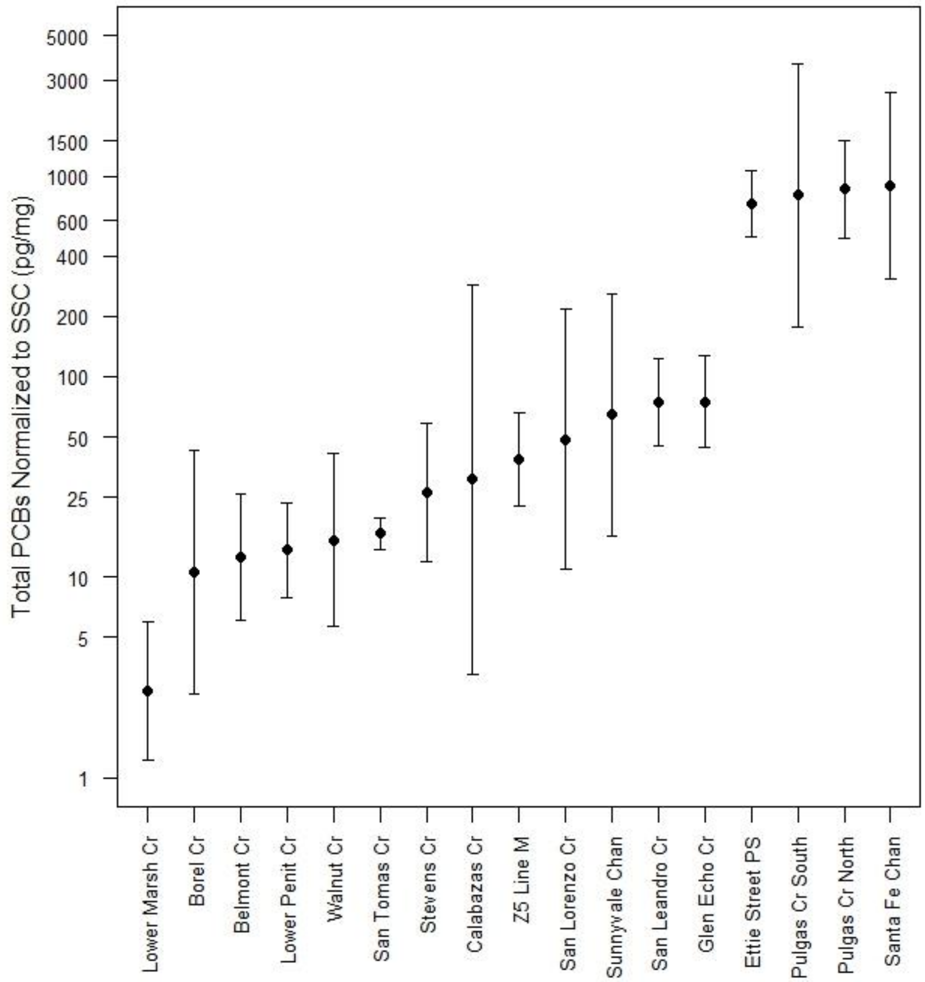

Figure 3. Comparative watershed ranking based on SSC normalized PCB concentrations (pg/mg).

Table 5. Common uses of PCB Aroclors in relation to the average percentage of Aroclor indicators in each of the sampled watersheds. Use information adapted from Erickson and Kaley II, 2011.

	Aroclor and primary use			
	1242	1248	1254	1260
Watershed	Capacitors; transformers; transmission turbine lubricants; heat transfer; carbonless copy paper	Heat transfer; vacuum pumps	Transformers; heat transfer; vacuum pumps; cutting oils; inks; adhesives; caulk and joint sealants; insulation and other building materials; rubber products; wire and cable coatings	Transformers; hydraulic fluids; immersion oils for microscopes; wire and cable coatings
Belmont Creek	1	3	14	31
Borel Creek	1	3	18	28
Calabazas Creek	2	8	36	14
Ettie Street Pump Station	13	12	22	15
Glen Echo Creek	2	8	40	11
Lower Marsh Creek	7	14	25	16
Lower Penitencia Creek	3	6	25	21
Pulgas Creek North	2	4	27	18
Pulgas Creek South	8	6	10	28
San Leandro Creek	2	7	31	15
San Lorenzo Creek	6	9	29	16
San Tomas Creek	4	8	28	18
Santa Fe Channel	1	6	45	11
Stevens Creek	3	7	31	17
Sunnyvale East Channel	1	2	15	30
Walnut Creek	5	8	27	17
Zone 5 Line M	1	5	26	21
Median	2	7	27	17
90th	7	10	38	29
Maximum	13	14	45	31

Suspended sediment concentration (SSC)

SSC concentrations ranged from 31.0 to 4,139 mg/L between all sites. These results are within a similar range as reported for an urban stormdrain (Zone 4 Line A) in Hayward (range 1.4-2,700 mg/L, Gilbreath et al., 2012) and a moderately urban river in San Jose (Guadalupe River, range 6-1,100 mg/L, McKee et al., 2006), both well sampled watersheds in the Bay Area. Except at Santa Fe Channel, which was sampled during a more intense storm, the watersheds with the greatest percentage imperviousness (Ettie St. Pump Station, Lower Penitencia Creek, Pulgas Creek North and South) all had relatively low SSC (<200 mg/L), and within a narrow range (<5-fold). Conversely, the largest and least impervious watersheds (Walnut Creek, Lower Marsh Creek, San Lorenzo Creek) all had much greater SSC (maximum in all watersheds >2,000 mg/L) with much greater variability between samples (>18-fold).

Selenium

Dissolved selenium concentrations ranged from 0.06-0.56 µg/L. Maximum concentrations were found at Lower Marsh Creek (Table 7). Total selenium concentrations ranged from 0.2-6 µg/L, with maximum concentrations also found at Lower Marsh Creek. Particulate Se, in the less urban watersheds, generally increased with increasing SSC while SeD remained fairly constant over varying SSC. Percent SeD ranged from 4-74% of SeT, and generally decreased with increasing SSC indicating particulate selenium dominated during higher storm flows. There was a strong relationship between SSC and SeT concentrations at all three stations (Table 7).

Mercury

Total Hg concentrations in water ranged over 200 times between sites, whereas variation within sites was less extreme (all sites <24 times between the highest and lowest observed concentration) (Table 6). Changes in HgT concentrations were well explained by changes in SSC for some of the watersheds (35% watersheds having least squares regression correlation coefficients (r^2) > 0.84). Others exhibited variation that was poorly correlated with SSC (30% of the watersheds with $r^2 < 0.36$). Methylmercury concentrations observed during the study varied from 0.029 ng/L to 2.9 ng/L, a variation of 100-fold between watersheds. Methylmercury as a percentage of HgT varied considerably between samples from 0.05-2.7% and between watersheds (the mean for each watershed ranges from 0.1-1.4%). There appears to be no pattern in the data with respect to imperviousness proportion of methylated mercury. For example, Walnut Creek with low percentage imperviousness also had a low percentage of mercury in methylated forms. San Lorenzo Creek, also with low imperviousness, had a high portion of mercury in methylated forms. Santa Fe channel, with a relatively high imperviousness, had a relatively low percentage of mercury in methylated forms whereas Pulgas Creek, also with high watershed imperviousness, had a relatively high percentage of mercury in methylated forms.

Table 6. Concentration range, mean and sample count (separated by a semicolon) for SSC and mercury in the 17 watersheds.

	SSC (mg/l)			HgT (ng/l)			MeHgT (ng/l)			% MeHgT		
Site	**Range**	**Mean**	**N**	**Range**	**Mean**	**N**	**Range**	**Mean**	**N**	**Range**	**Mean**	**N**
Belmont Creek	148-382;	240;	4	47.4-58.9;	52.7;	4	0.222-0.244;	0.233;	2	0.38-0.45;	0.41;	2
Borel Creek	238-612;	362;	5	47.0-73.6;	57.9;	5	0.174-0.338;	0.256;	2	0.37-0.67;	0.52;	2
Calabazas Creek	193-585;	393;	5	35.0-88.8;	58.8;	5			0			
Ettie Street Pump Station	45.4-141;	79.5;	4	44.4-72.5;	53.7;	4	0.371-0.447;	0.409;	2	0.62-0.84;	0.73;	2
Glen Echo Creek	38.9-1068;	341;	4	26.4-179;	72.8;	4	0.295-1.94;	1.12;	2	1.08-1.19;	1.1;	2
Lower Marsh Creek	123-4139;	1368;	6	35.7-167;	73.7;	6	0.311-0.603;	0.410;	3	0.62-0.88;	0.77;	3
Lower Penitencia Creek	38.5-172;	97.8;	4	8.17-18.9;	13.9;	4	0.159-0.241;	0.192;	3	0.94-1.49;	1.2;	3
Pulgas Creek North	31.0-87.5;	60.3;	4	21.4-27.2;	24.2;	4		0.029;	1	1.73;	1.7;	1
Pulgas Creek South	17.4-60.1;	38.4;	4	18.2-28.4;	24.3;	4		0.370;	1	0.16;	0.16;	1
Santa Fe Channel	49.3-469;	152;	5	34.6-217;	86.0;	5	0.271-0.711;	0.491;	2	0.33;	0.33;	2
San Leandro Creek	49.1-965;	270;	7	21.5-477;	193;	7	0.558-0.855;	0.657;	4	0.14-2.7;	0.84;	4
San Lorenzo Creek	85.2-2084;	484;	8	18.4-77.1;	30.8;	6	0.254-0.596;	0.398;	3	1.38-1.52;	1.4;	3
Stevens Creek	48.1-751;	357;	6	21.3-112;	76.3;	6	0.587-0.635;	0.611;	2	0.57-0.97;	0.77;	2
San Tomas Creek	99.3-293;	206;	5	23.5-129;	58.6;	5	0.0380-0.0990;	0.0677;	3	0.05-0.26;	0.16;	3
Sunnyvale Channel	56.9-1050;	372;	4	27.9-151;	77.7;	4	0.304-0.588;	0.446;	2	0.4-0.65;	0.52;	2
Walnut Creek	128-2392;	1301;	6	27.3-181;	92.8;	5	0.0340-0.142;	0.0843;	3	0.07-0.13;	0.10;	3
Zone 5 Line M	250-2174;	874;	4	70.8-1665;	503;	4	0.639-2.90;	1.77;	2	0.17-0.9;	0.53;	2

Table 7. Concentration range, mean and sample count (separated by a semicolon) for the three watersheds in which selenium was monitored, as well as linear regression statistics for the relationship between total selenium and SSC in those watersheds.

	SeT (µg/l)			SeD (µg/l)			SeT and SSC linear regression statistics			
Site	Range	Mean	N	Range	Mean	N	Slope	Intercept	p value	r^2
Marsh Creek	0.626-6.00;	2.06;	14	0.276-0.555;	0.384;	14	0.0011	0.51	1.2E-12	0.99
Santa Fe Channel	0.195-0.416;	0.277;	7	0.0580-0.171;	0.119;	7	0.00060	0.16	0.016	0.72
Walnut Creek	0.518-4.36;	2.67;	7	0.114-0.533;	0.311;	7	0.0015	0.95	0.0082	0.86

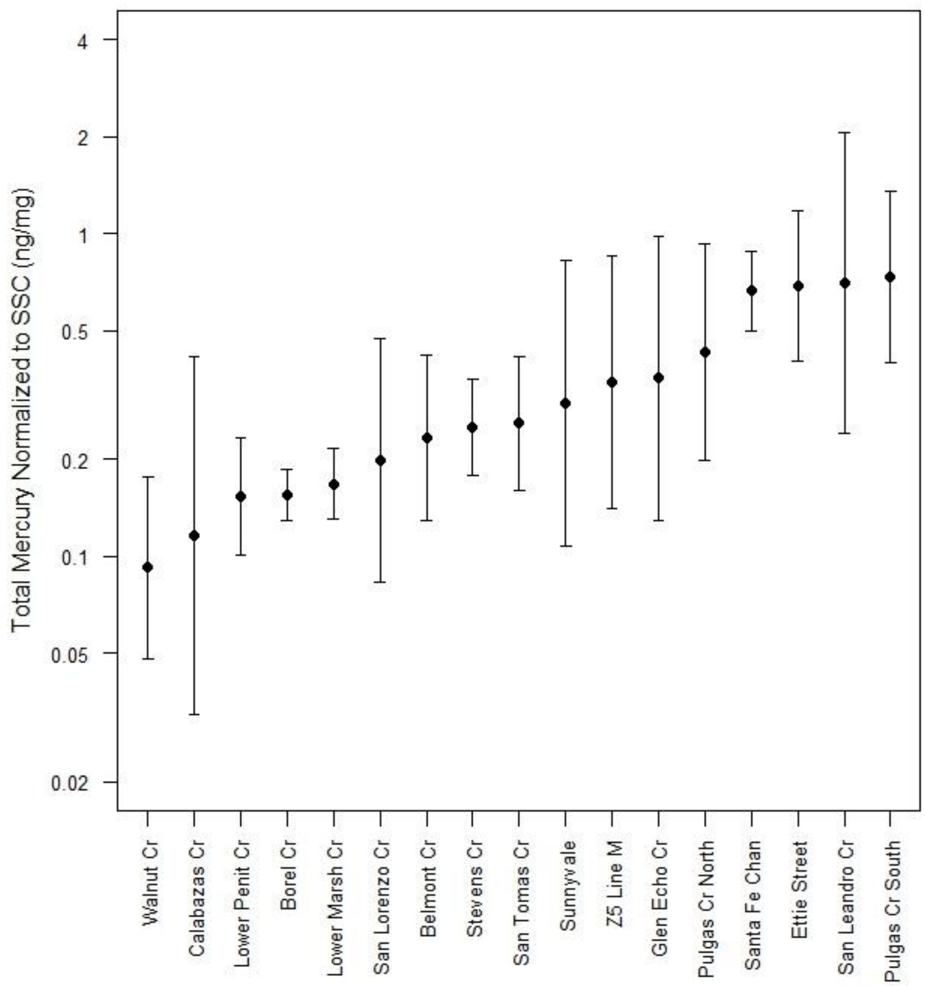

Figure 4. Comparative watershed ranking based on SSC normalized total Hg concentrations (ng/mg).

Some of the most mercury contaminated watersheds were also the watersheds that produced the greatest suspended sediment loads, thus, even more dramatically than for PCBs, the choice of ranking method had a large impact on the outcome. For example, Walnut Creek would rank third highest if ranked by mean concentration, whereas it ranks the lowest amongst all 17 using particle ratios (Figure 4). In contrast, Ettie Street Pump Station watershed is ranked third highest on the basis of particle ratios due to a relatively low sediment production in the watershed and San Leandro Creek ranks high using either method due to relatively high mercury concentrations and moderate suspended sediment production. Most of the six watersheds that ranked highest for PCBs also rank highest for mercury in terms of particle ratios (Glen Echo Creek, Pulgas Creek (north and south), Santa Fe Channel, San Leandro Creek and Ettie Street Pump Station watersheds) although the order of ranking is slightly different (Table 8). If we were to rank on the basis of methylmercury particle ratios, Santa Fe Channel, San Leandro Creek, Ettie St. Pump Station, Pulgas Creek – North, and Glen Echo Creek also rank in the top six. Thus overall, the mercury data collected during this study appears to corroborate the PCBs findings. Sunnyvale East Channel, Glen Echo Creek, Pulgas Creek (North and South), Santa Fe Channel, Ettie Street Pump Station watershed and San Leandro Creek appear to be high leverage; management efforts in these watersheds may be more cost-effective than managing sources in some of the lower ranked watersheds. Lower Marsh Creek were ranked 17th and 13th, and Walnut Creek ranked 13th and 17th for PCB and mercury particle ratios, respectively (Table 8). Management of PCBs or mercury in either of these watersheds would likely be the least cost effective per unit mass removed and would unlikely result in any measurable downward trend in either particle ratios or loads at a lower watershed sampling site.

Table 8. Ranking of watershed based on PCB and Hg particle ratios organized by the PCB:SSC ratio.

Watershed	PCB/SSC Avg Ratio (pg/mg)	THg/SSC Avg Ratio (ng/mg)	PCB Rank	Hg Rank	Rank Sum
Santa Fe Channel	1403	0.68	1	4	5
Pulgas Creek North	1050	0.47	2	5	7
Pulgas Creek South	906	0.83	3	1	4
Ettie Street Pump Station	745	0.78	4	3	7
Glen Echo Creek	86	0.41	5	6	11
San Leandro Creek	85	0.80	6	2	8
Sunnyvale Channel	79	0.34	7	8	15
San Lorenzo Creek	75	0.28	8	9	17
Zone 5 Line M	48	0.40	9	7	16
Calabazas Creek	41	0.16	10	16	26
Stevens Creek	34	0.26	11	11	22
San Tomas Creek	20	0.27	12	10	22
Walnut Creek	19	0.10	13	17	30
Lower Penitencia Creek	17	0.16	14	15	29
Belmont Creek	14	0.24	15	12	27
Borel Creek	13	0.17	16	14	30
Lower Marsh Creek	3	0.20	17	13	30

Relationships between land use attributes and pollutants of concern

Codependence in land use attributes

For the 206 watersheds, Pearson product moment correlations were moderate among some spatial attributes. In particular, imperviousness was positively correlated to 1954 urban and 1954 industrial, commercial high density, and transportation; and negatively correlated to open infiltrative. Area was correlated to residential low density (Table 9). The first four principal components explained 64% of the variation in the 17 watershed spatial attributes (Table 10), with substantially lower variation explained by the remaining 13 principal components. The first principal component (i.e., PC1) explained 29% of the variance in the data and was largely associated with indicators of historic industrial development, including imperviousness and the historical measures of urbanization and industrialization (Table 10, Figure 5). PC2 explained 16% of additional variance and was associated with watershed area and low density residential (Table10, Figure 5). PC3 explained 12% of additional variance, and was negatively related to the industrial categories but positively associated with residential, 1974 urban, and transportation (Table 10). Thus, PC3 appeared to indicate the difference between industrial versus other urban land use. PC4 explained about 7% of additional variance and was negatively associated with open impervious, water, and the null category (Table 10). PC1, PC2, and PC3 in combination explained 54% of variation in land use and were associated with land use attributes that could be hypothesized to explain pollutant concentrations; therefore, these three components were regressed against pollutant concentrations.

The 22 monitored watersheds were not representative of the full range of Bay Area watersheds considered in the PCA. Within the bivariate space represented by PC1 and PC2, the monitored watersheds tended to exhibit higher PC2 scores and lower PC1 scores than the majority of Bay Area watersheds (O versus + symbols in Figure 5). This can be explained by the selection of watersheds that tended to have higher urbanization or greater size than the majority of Bay Area watersheds (Figure 6). This targeted selection of watersheds resulted in a much stronger negative correlation between watershed area and urbanization attributes for the study monitored watersheds than the full range of Bay Area watersheds. For example, the Pearson correlation coefficient between watershed area (log transformed) and imperviousness (arcsine(sqrt) transformed) was $r = -0.86$ for the 22 monitored watersheds but only $r = -0.23$ for the 206 study watersheds. One of the monitoring stations, the Gellert Park storm drain, had the highest imperviousness and lowest area of all 206 watersheds, and the lowest PC2 score (Figure 5, bottom O symbol), and is thus an influential observation.

Table 9. Matrix of Pearson product-moment correlation coefficients for Bay Area watersheds (N = 206). Correlations (r) greater than 0.4 are indicated in boldface.

Parameter	Area	Imperviousness	1954 Urban	1954 Industrial	Agriculture	Commercial high density	Industrial	Open infiltrative	Residential high density	Residential low density
Imperviousness	-0.23									
1954 Urban	0	**0.53**								
1954 Industrial	0.06	**0.43**	**0.59**							
Agriculture	0.38	-0.22	-0.27	-0.18						
Commercial high density	0.15	**0.59**	0.29	0.33	-0.02					
Industrial	0.14	0.29	0.11	**0.63**	-0.06	0.35				
Open infiltrative	0.36	**-0.72**	**-0.46**	-0.2	0.32	-0.39	-0.01			
Residential high density	0.39	0.2	0.31	0.17	0.01	0.3	0.06	-0.24		
Residential low density	**0.58**	-0.12	0.06	-0.05	0.24	0.27	-0.06	0.1	**0.54**	
Transportation	0	**0.58**	**0.4**	0.28	-0.11	0.31	0.08	**-0.47**	0.26	0.15

Table 10. Loadings and proportion of variance explained for the first four principal components. To aid in visualization, loadings below 0.1 are excluded and loadings above 0.35 are boldfaced.

	Principal components			
Parameter Loading	1	2	3	4
Area		**0.52**		
Imperviousness	**0.38**	-0.16		
1954 Urban	0.33			
1954 Industrial	0.30		**-0.37**	
1974 Urban	**0.38**		0.23	
1974 Industrial	0.31		**-0.45**	
Agriculture	-0.13	0.30		
Commercial high density	0.30	0.13		0.18
Commercial low density	0.18	0.33		
Industrial	0.20		**-0.58**	
Null		-0.17		**-0.61**
Open infiltrative	-0.32	0.21	-0.29	
Open impervious		0.20		**-0.45**
Residential high density	0.21	**0.35**	0.19	-0.14
Residential low density		**0.49**	0.18	
Transportation	0.28		0.19	
Water			-0.21	**-0.60**
Variance explained (%)	28.6	16.0	11.8	7.4
Cumulative variance (%)	29	45	56	64

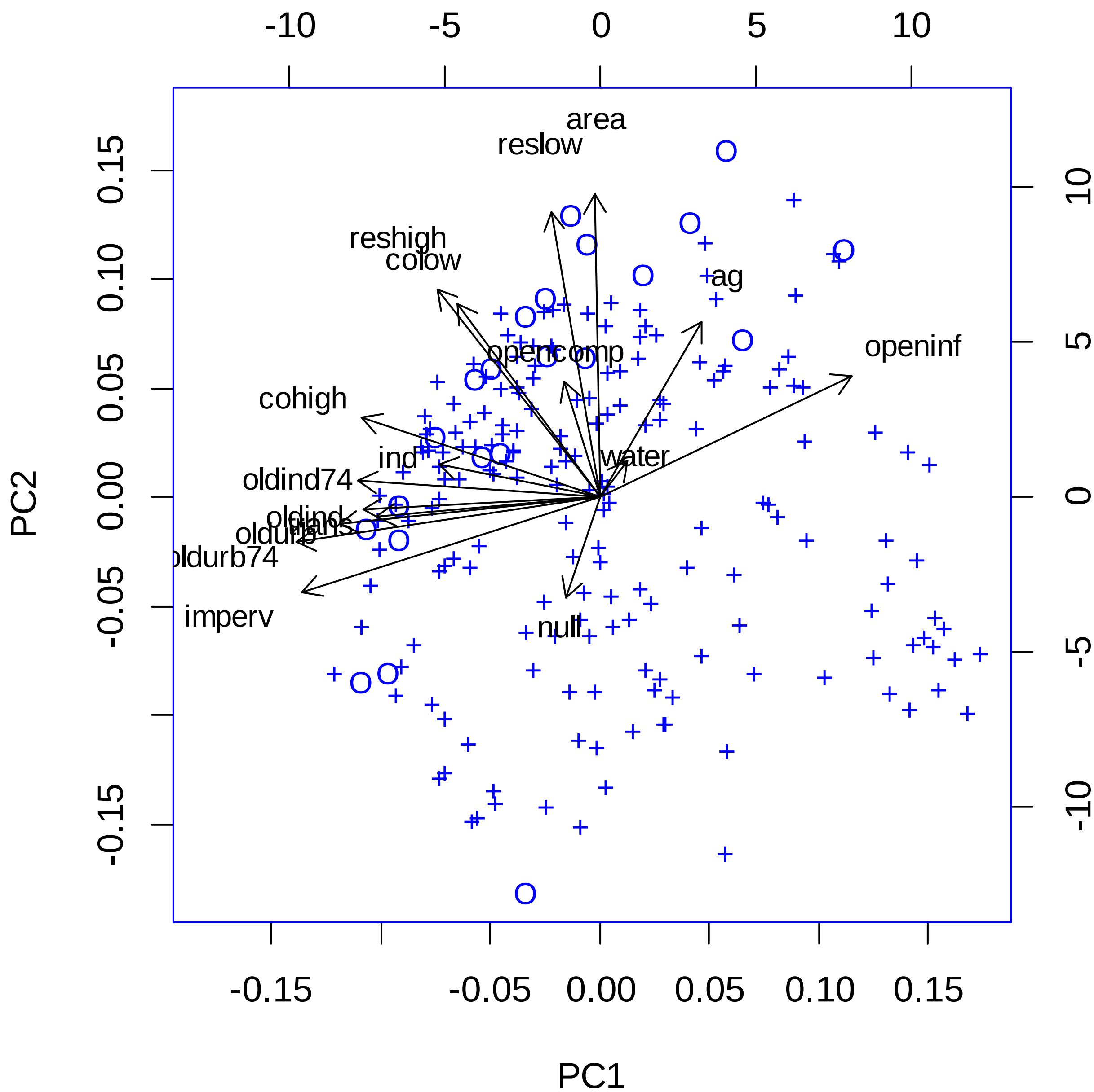

Figure 5. Biplot of principal components 1 and 2. Arrows indicate relative parameter loading. O symbols indicate monitored watersheds and + symbols indicate non-monitored watersheds. Table 1 lists parameter abbreviations.

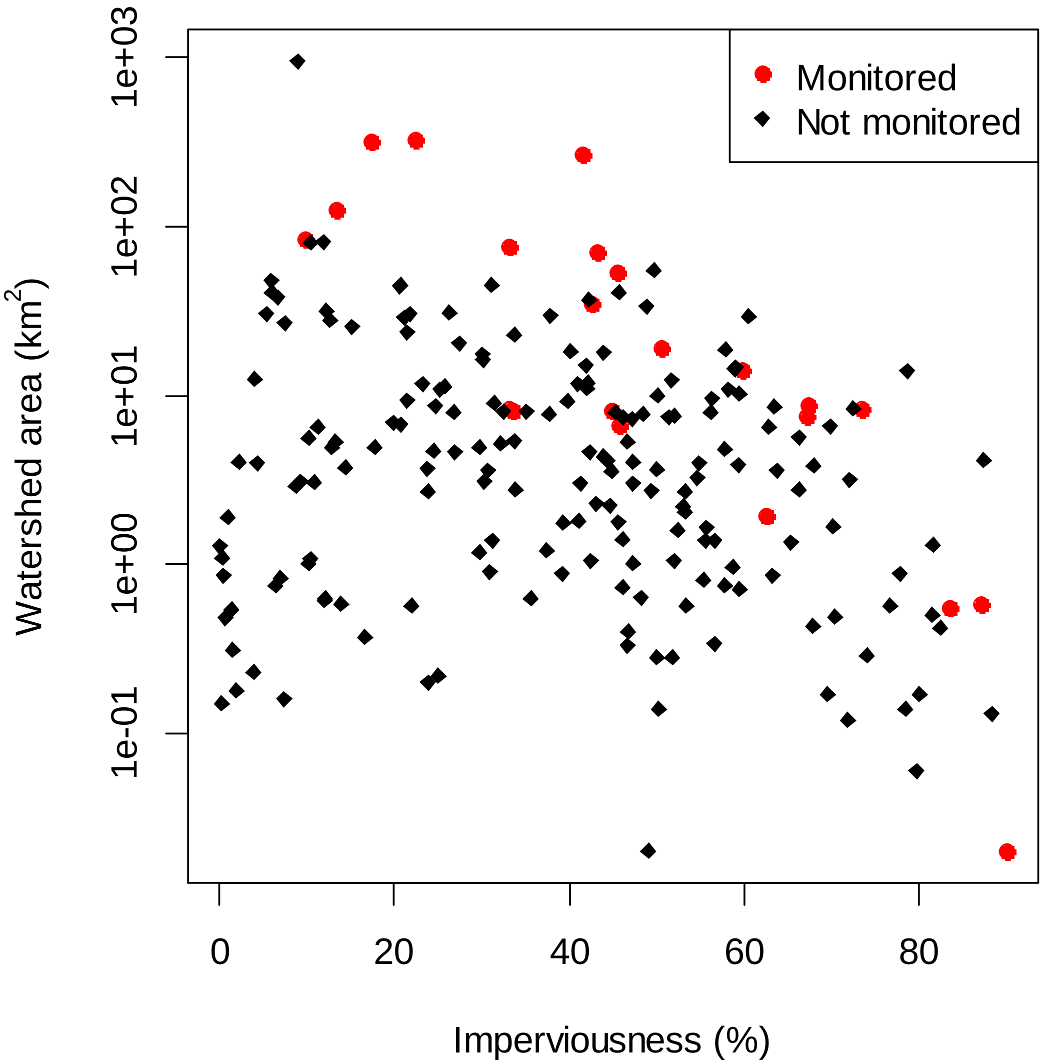

Figure 6. Watershed area and imperviousness for the 22 watersheds monitored in the study and 184 additional Bay Area watersheds not monitored. The Gellert Park site is the lower right most point. Note log scale y-axis.

Relationship between land use attributes and pollutant particle concentrations

Concentrations were positively correlated across pollutants; the strongest correlations were between mercury (total and methyl) and both PBDEs and PAHs (Table 11). Total Hg and PCBs were positively related to PC1, indicating an association with imperviousness and historic industrialization. For HgT, the variability explained by PC1 was low (Table 12). MeHg was not significantly related to PC1, PC2, or PC3. PBDEs and TOC were negatively related to PC3, and thus positively related to industrial land use. PAHs were negatively related to PC2, and thus positively related to watershed area. The PBDE relationship was driven by the watershed with the lowest PBDE concentration and the highest PC2 score (Borel Creek; 30 pg/mg; Figure 7).

Table 11. Pearson product-moment correlation coefficients for pollutants measured in Bay Area watersheds (N in parentheses). Correlations (r) greater than 0.6 are indicated in boldface.

	Total mercury	Methylmercury	PCBs	PBDEs	PAHs
Methylmercury	**0.68** (18)				
PCBs	**0.65** (22)	0.48 (18)			
PBDEs	**0.72** (8)	**0.90** (7)	0.54 (8)		
PAHs	**0.70** (8)	**0.86** (7)	0.54 (8)	**0.65** (6)	
TOC	0.35 (19)	0.27 (18)	0.45 (19)	0.57 (7)	**0.63** (7)

Table 12. Linear regression model results for pollutant concentrations versus land use attributes. PCA model and R^2: which of the three principal components (PC) were significantly related to the pollutant, and the direction of the relationship. Remaining columns indicate R^2 for individual parameters most associated with the PCs. Not significant (NS): $p > 0.05$.

Pollutant	**PCA model**	**PCA R^2**	**Imperviousness R^2**	**Area R^2**	**Industrial R^2**
HgT	+PC1	0.32	0.37	0.24	NS
MeHg	NS	NS	NS	NS	NS
PCBs	+PC1	0.57	0.43	0.18	0.35
PBDEs	-PC3	0.64	NS	NS	0.58
PAHs	-PC2	0.75	0.90	0.68	NS
TOC	-PC3	0.26	0.28	NS	0.20

PC1, PC2, and PC3 were most strongly associated with imperviousness, watershed area, and industrial land use, respectively, so each pollutant was individually regressed against these three land use traits. In bivariate linear regression models, HgT, PCBs, and PAHs were most related to imperviousness (Figure 7, Table 12), PBDEs was only related to industrial land use, and MeHg was not related to any of the three land use traits (Table 12). The regression R^2 was generally similar for the strongest PC versus the corresponding individual land use trait. For example, HgT and PCBs were related to imperviousness which corresponded to PC1, while PBDEs was only related to industrial land use, corresponding to PC3, and were not related to imperviousness. An exception was PAH being more related to imperviousness than watershed area, which did not correspond to the PAH association with PC2. However, the relative importance of area versus imperviousness was driven by Gellert Park, which had the highest PAH concentration and lowest watershed area. Removing Gellert Park from the analysis resulted in a stronger association with PC1 (i.e., imperviousness). Because the three land use attributes were correlated for the monitored sites (e.g., Figure 5), no models including two land use attributes were significant for both variables.

First order estimates of average annual loads

The five watersheds with loadings estimates ranged two orders of magnitude in size, seven-fold in imperviousness, and varied in mean annual rainfall by 43% RPD. Area normalized loads are reported in Table 13, with watersheds listed in increasing order of imperviousness. Yields generally increase with increasing imperviousness for HgT, PCBs and PAHs. In contrast, suspended sediment (SS) loads normalized to watershed area generally decrease with increasing imperviousness. Our first order HgT yield estimate in Lower Marsh Creek is relative low despite a former mercury mine in the upper watershed, though sampling in this creek occurred only on the rise due to logistical constraints. It is possible that more mercury contaminated runoff occurs on the falling limb but at this time we have no evidence to support that hypothesis. Since the BASMAA sediment studies (Salop et al., 2002a; 2002b), Ettie Street Pump Station has been considered a high leverage watershed for PCBs; the first order loads estimates presented here provide further support for that hypothesis. In addition, the Ettie Street Pump Station watershed also appears to be a high leverage watershed for HgT and MeHgT. Urban landscapes are not typically described as methylation areas for mercury, though the yields shown below suggest otherwise, especially in a highly impervious watershed such as Ettie St. Pump Station.

Discussion

Relative concentrations and rankings

PCBs were used in a wide variety of products and applications between the post WW II period of the 1950s and the late 70s when they were phased out for all new applications. The largest use was as an electrical dielectric fluid but other uses included heat transfer fluids, vacuum pumps, cutting oils, inks, adhesives, caulk and joint sealants, insulation and other building materials, rubber products, wire and cable coatings, and carbonless copy paper (Erickson and Kaley II, 2011). Conceptually, evidence collected to-date in the Bay Area supports a hypothesis that watersheds containing older denser urban areas, old industrial areas and higher imperviousness are likely to be higher leverage for PCBs (SFEI, 2010; Lent and McKee, 2011). A similar pattern is expected for mercury although the magnitude of variation between land use or source area classes is likely to be less for mercury due to the use history being relatively more dispersed as well as the atmospheric component of the mercury cycle being more dominant leading to wider mercury dispersal (SFEI, 2010; Lent and McKee, 2011).

Relative concentrations across the watersheds generally fit our conceptual models for which San Francisco Bay watersheds might be higher leverage (Table 14). BASMAA sediment studies for the region (Salop et al., 2002a; 2002b; KLI, 2002) and a more recent study by SFEI (Yee and McKee, 2010) had previously indicated relatively high PCB concentrations in soils and stormwater sediments in the five of the six highest ranked watersheds (Glen Echo Creek, Pulgas Creek (North and South), Santa Fe Channel, and Ettie Street Pump Station watershed) (Table 14). The watersheds selected as representative of more typical urban residential areas in the Bay Area (Belmont, Borel, Calabazas, Lower Penitencia, San Tomas) have moderate to lower

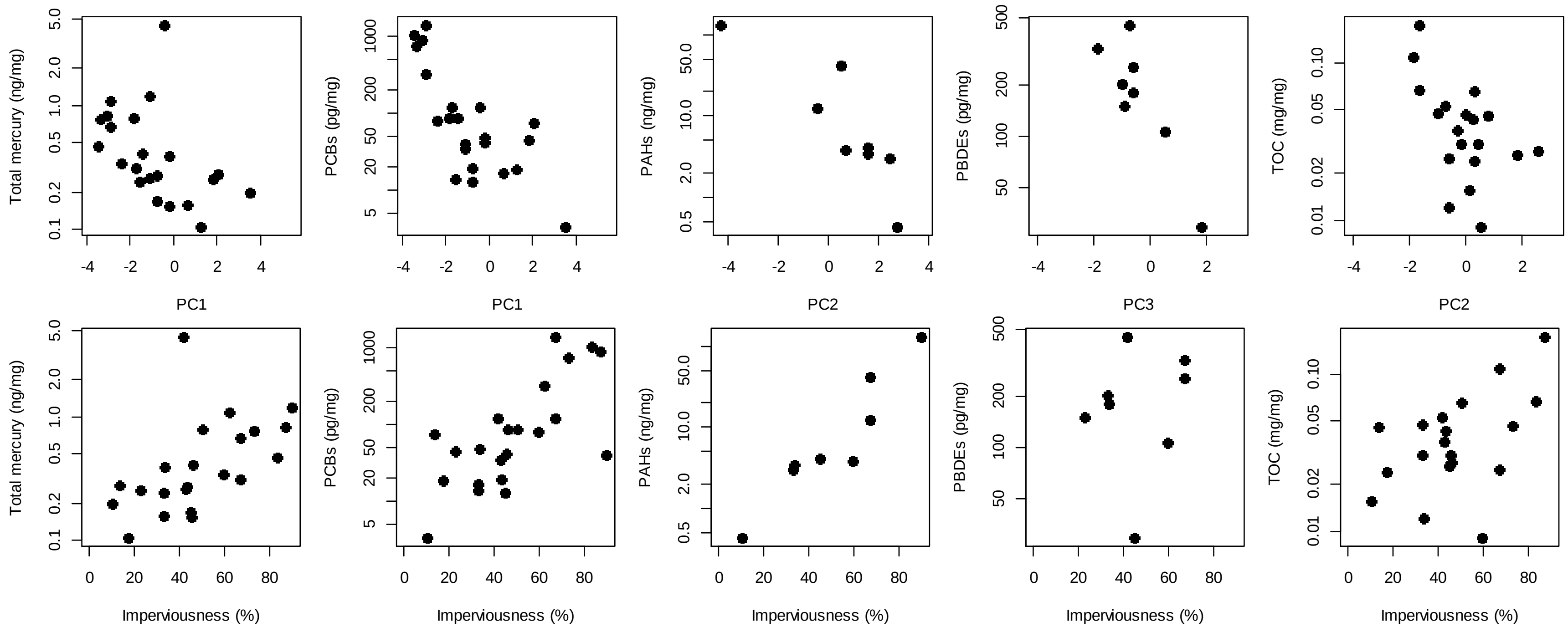

Figure 7. Relationship between particle normalized average watershed pollutant concentrations and the strongest associated principal components (PC; top row) or percent imperviousness (bottom row). Note log scale y-axis and varying scales among pollutants.

Table 13. Average annual yields (mass per unit watershed area).

Watershed	Area (km^2)	Impervious (%)	Mean Annual Rainfall (m)	Mean Annual Volume (10^6 CM)	HgT (ug/m^2)	MeHgT (ug/m^2)	SeT (ug/m^2)	SeD (ug/m^2)	TOC (t/km^2)	PCBs (ug/m^2)	PAHs (ug/m^2)	PBDEs (ug/m^2)	SS (t/km^2)
Lower Marsh Ck	84	10	0.42	8.6	9	0.05	356	39	0.7	0.3	12		278
San Lorenzo Ck	125	13	0.65	19.9	8	0.07			1.0	2.6			221
Walnut Ck	232	15	0.58	50.2	29	0.02	846	86	2.7	2.0			433
Sunnyvale East Channel (High)	15	59	0.43	2.7	23	0.10			0.6	8.8	438	13	147
Sunnyvale East Channel (Low)	15	59	0.43	1.5	13	0.05			0.3	4.8	240	7	80
Ettie St. Pump Station	4	75	0.54	5.7	79	0.57			4.9	82			133

concentrations and are ranked lower (Table 14). However, there were some surprises. Sunnyvale East Channel watershed had not previously been identified as high leverage for PCBs although there were some manufacturing plants that used PCBs in the watershed historically. It was perhaps surprising to see San Leandro Creek ranking moderately high given the sampling location is upstream of the majority of industrial land uses. It was also surprising that San Leandro Creek ranked the second highest of the 17 watersheds for mercury; at this time we provide no hypothesis of the source. Stevens Creek watershed is influenced by mercury deposition from the only currently operating cement plant in the Bay Area (Rothenberg et al., 2010a; 2010b). Despite this known atmospheric loading source, Stevens Creek watershed ranked in the middle relative to the other 17 watersheds. Lower Marsh Creek, with a history of mercury mining, appears to be relatively low leverage for both mercury and PCBs. This was predicted for PCBs but was not expected for mercury; further sampling during more intense rainstorms and carefully sampling on the falling stage to try to capture any mercury signal from the mines will likely confirm or reject this hypothesis.

The range of PCB concentrations we observed during the study generally coincide with those reported in the literature for other urban areas (ND-34 ng/L, Curren et al., 2011; 2.0-28.9 ng/L, Foster et al., 2000; 27-179 ng/L, Marsalek and Ng, 1989; 26.9-1,120 ng/L, Walker et al., 1999). Except for the Ettie St. Pump Station watershed, a known high leverage area for PCBs, yields reported for the other four watersheds were within similar ranges reported for other SF Bay local watersheds (3.0-5.0 $\mu g/m^2/y$, Davis et al., 2000; 3.1 $\mu g/m^2/y$, Gilbreath et al., 2012).

Much less data are available regionally and internationally for stormwater concentrations of PBDEs to guide our hypotheses for expected concentrations relative to watershed attributes. A conceptual model for PBDE pollution in relation to land uses and source areas will be developed through the STLS in the near future. However, concentrations observed during the study were within the lower half of the range reported for Z4LA, a small, 100% urban watershed in Hayward (Gilbreath et al., 2012). On average, concentrations in the 17 watersheds were mostly higher than reported by Guan et al. (2009) for eight urban watersheds in the Pearl River Delta, a global electronic-waste hotspot. However samples in Guan's study were collected on a monthly time interval rather than focusing on stormwater collection when PBDE concentrations are known to be elevated (McKee et al 2006; Gilbreath et al 2012).

Concentrations of PAHs observed appear to fall in the lower end range of those reported for other urban watersheds (13,400 ng/L, Crunkilton and DeVita, 1997; 1,510-12,500 ng/L, Hwang and Foster, 2006; 5,393 ng/L, Ngabe et al., 2000; 676-2,351 ng/L for EMCs and 4,061 to 30,020 for rising limb concentrations, Shinya et al., 2000; 817-5,821 ng/L for EMCs, Stein et al., 2006). Lower Penitencia Creek appeared to have anomalously lower PAH concentrations than might have been expected given the level of development and imperviousness found in the watershed (though the storms sampled at Lower Penitencia were of generally lower intensity and may not have been able to mobilize sediments from some of the more developed areas). Likewise, Lower Penitencia is

Table 14. Summary of PCB and mercury concentrations determined during this study and ranking from highest (1) to lowest (17) estimated leverage based on particle ratios (mass of pollutant in water in relation to mass of suspended sediment in water) of PCBs. NA = no samples collected.

Watershed	Old industrial	Old urban	Impervious	Total Mercury (ng/L)	Total Mercury particle ratio (ng/mg)	Total Methylmercury (ng/L)	Total Methylmercury particle ratio (pg/mg)	PCB (ng/L)	PCB particle ratio (ng/ mg)	Rank based on PCB particle ratio
Santa Fe Channel	2%	97%	69%	86	0.68	0.49	2.1	198	1.4	1
Pulgas Creek - North	32%	100%	84%	24	0.47	0.37	4.2	60	1.1	2
Pulgas Creek - South	22%	98%	87%	24	0.83	0.03	0.55	31	0.91	3
Ettie St. Pump Station	13%	100%	75%	54	0.78	0.41	3.9	59	0.75	4
Glen Echo Creek	0%	100%	39%	73	0.41	1.12	5.2	30	0.09	5
San Leandro Creek	0%	50%	38%	193	0.80	0.66	3.7	12	0.085	6
Sunnyvale East Channel	2%	29%	59%	78	0.34	0.45	1.0	39	0.079	7
San Lorenzo Creek	0%	21%	13%	31	0.28	0.40	2.1	13	0.075	8
Zone 5 Line M	1%	10%	33%	503	0.40	1.8	1.9	21	0.048	9
Calabazas Creek	0%	5%	44%	59	0.16	NA	NA	11	0.041	10
Stevens Creek	0%	23%	38%	76	0.26	0.61	1.6	7.5	0.034	11
San Tomas Creek	0%	15%	33%	59	0.27	0.07	0.33	2.8	0.020	12
Walnut Creek	0%	15%	15%	77	0.10	0.08	0.07	9.0	0.019	13
Lower Penitencia Creek	0%	9%	65%	14	0.16	0.19	1.8	1.5	0.017	14
Belmont Creek	0%	55%	27%	53	0.24	0.23	0.78	3.6	0.014	15
Borel Creek	0%	26%	31%	58	0.17	0.26	0.91	6.1	0.013	16
Lower Marsh Ck	0%	2%	10%	74	0.20	0.41	0.54	2.1	0.0033	17

notably low for HgT and PCBs. On a somewhat more surprising note, the concentrations sampled in the six watersheds of this study were all lower than most sample concentrations measured in the Z4LA urban watershed in Hayward (up to 22,000 ng/L, Gilbreath et al., 2012). It was noted by Gilbreath et al. that congener profiles from the Zone 4 Line A samples may indicate usage of coal tar sealcoat which can greatly amplify PAH concentrations in stormwater (Van Metre and Mahler, 2010).

Stormwater is one of the primary pathways of moving selenium into North San Francisco Bay (Tetra Tech, 2008). Previous local stormwater studies have shown total selenium concentrations in the range of 0.3-0.8 µg/L (Sacramento River/San Joaquin River), 0.053-2.86 µg/L (Zone 4 Line A, Hayward, CA), and 1.3-2 µg/L (North Bay tributaries) (David et al., 2011; Gilbreath et al., 2012; Tetra Tech, 2008). Findings from this study, showed total selenium concentrations up to 6 µg/L (Lower Marsh Creek). Estimated total selenium yields, where flow data were available, were 356 $\mu g/m^2$ (Lower Marsh Creek) and 846 $\mu g/m^2$ (Walnut Creek). Estimated yields from other local studies were 27 $\mu g/m^2$ (Zone 4 Line A) and 106 $\mu g/m^2$ (Sacramento/San Joaquin River).

Correlation between land use attributes and PCB and mercury particle ratios

The statistical analysis using PCA and correlation matrix identified two predominant gradients within Bay Area watersheds. The first was a gradient of urbanization and industrialization characterized by a variety of correlated land categories (historic urban, historic and current industrial, imperviousness, high density commercial). The second was related to watershed area and low density residential. These results are generally consistent with findings in Greenfield et al. (2010). In addition, they are also consistent with Hatt et al. (2004) who used regression analysis to demonstrate the impact of imperviousness and drainage connection on concentrations and loads of nutrients in urbanization. Future sampling of watersheds, conceptual model development, and mapping could take into consideration these two gradients. A general correlation was observed between pollutant concentrations, historic and urban land cover, and imperviousness. This supports the conceptual model of land cover explaining a portion of variation in a range of local pollutants of concern. MeHg exhibited no correlation to land use attributes. Possible reasons could include within-creek/stormdrain processes, confounding of nutrient loading with other attributes (Driscoll et al. 2012), or other factors. The usual caution is warranted that controlling HgT may not control MeHg. The Gellert Park watershed was an influential watershed for PAHs given its small size, high imperviousness, and having the maximum PAH concentrations in the watersheds dataset. Omitting Gellert Park reduced the strength of the relationship between PAH and watershed area. This is probably largely a sample size issue. Overall, the statistical analysis presented does provide confidence that as our concentration data in Bay Area watersheds continues to expand in number and in quality, and our knowledge about the spatial distribution of pollution sources also continues to improve, we are likely to continue to improve our understanding of the potential influences of land uses and source areas on pollutants such as mercury and PCBs.

Potential future uses of the data set?

In addition to identifying watersheds or urban areas that are higher leverage (STLS question 1), the small tributaries loading strategy and MRP calls for development of a regional loading estimate for mercury and PCBs (provision C.8.e.) and for PBDEs, OC pesticides and selenium (provision C.14.). A regional watershed spreadsheet model (RWSM) is being developed to assist Permittees to develop improved regional loading estimates (Lent and McKee 2011; Lent et al., 2012). In addition to information on the land use, rainfall, run-off coefficients, soils, slope, and imperviousness, input data are needed on pollutant concentrations, either in water (referred to as event mean concentrations or EMCs) or on suspended sediment particles (referred to as particle concentrations), in relation to each land use or source area identified. Additionally, data are needed to verify the output of the regional model in the form of averaged concentrations (referred to as flow weighted mean concentrations or FWMCs) or annual average loads (mass). The observations made in the 17 watersheds of this study and in previous RMP SPLWG loading studies can be used for either of these two purposes providing the input data set is chosen as a subset and therefore independent of the calibration and verification datasets. During Year 3 of the RWSM development, these uses are being explored. In addition, should stakeholders be interested, future versions of the RWSM could include modules to explore the potential loads reductions associated with various kinds of management actions (STLS question 4). Although only 4 to 7 data points have so far been collected in these 17 watersheds during storm events, the number of data points might still allow the possibility of observing trends over the next five or 10 years as management actions are implemented (Melwani et al., 2010).

Sampling design improvements

This report presents data from 17 watersheds across four counties in the Bay Area. The number of samples collected per watershed was based on a statistical analysis (Melwani et al., 2010). In some instances only four or five samples were collected due to budget constraints; this number of samples is at the low end of the recommendation by Melwani et al. This made developing statistically significant regression relationships between either turbidity or SSC challenging. In some cases (for example some watersheds for PBDEs), the dataset spanned only a small range of turbidity also making statistical correlation challenging. In addition, due to budget constraints, only one sample for SSC analysis was collected with each pollutant sample set. This made pairing the samples for particle normalization challenging, an issue partially rectified by using turbidity as a surrogate. Stage height data were collected but inconsistently between sites due to logistical constraints, negating the possibility of making consistent estimates of flow weighted mean concentrations for each watershed; an issue that was partially rectified by averaging based on suspended sediment concentration (method referred to as PWMC). An improvement for future reconnaissance studies, where so few samples are being collected, is to do so in watersheds that we know will vary in turbidity; fully urban watersheds that have less variation may need a larger number of samples to better characterize them. The methodology developed for the study could be implemented again in a future reconnaissance to identify high leverage watersheds, or for supporting either the input side or the calibration and verification side of the RWSM. Should this occur, some recommendations to consider include:

- sampling design should be improved with a minimum of six samples per site for each pollutant
- the collection of an additional water sample for suspended sediment analysis for each sample set
- continued vigilance in the field for collecting samples over a wide range of turbidity
- and at minimum, collect a manual record of stage during sampled storm events to couple with a cross section of the sampling location for estimating flow using Manning' s formula.

In relation to statistical analysis for land use relationships, a future question that could be considered is how PCB, PBDE, and PAH congener profiles vary across watershed type or attributes? Multivariate analysis of congener profiles may indicate different kinds of source categories or depuration processes (Rodenburg et al. 2011, Greenfield and Allen 2012). This could help in verifying source categories in relation to management actions or improvement of the underlying structure of the RWSM. A current weakness in the dataset is the inherent biases associated with targeted sampling of certain kinds of watersheds. A wide range of urbanization conditions (i.e., PC1) were sampled, and pollution concentrations seemed to be related to PC1 (land cover) rather than PC2 (watershed area), suggesting that the most important gradient (imperviousness and urbanization) was successfully sampled. However a disadvantage in the currently available dataset is that sampling to-date is not fully representative of all Bay Area watersheds. Therefore, the results presented here should not be extrapolated to all watershed types in the Bay Area. This is consistent with Cuevas et al. (2006) who argued against the spatial generality and predictive power of land cover-stream property models. In particular, small and moderate sized watersheds with low imperviousness were not sampled at all so far in Bay Area studies. Should there be further interest in exploring land use relationships with pollutant particle concentrations, a more statistically valid technique for choosing watershed sampling locations should be employed. This would be particularly true if there was interest in developing a predictive statistical model. The number of watersheds selected in the current study, as well as future studies, is likely to be limited by budget resources. Therefore, an unbiased sample draw based on the previous STLS characterization study (Greenfield et al 2010) will likely remain difficult to achieve.

Conclusions

Data and interpretations from our reconnaissance of 17 watersheds in the Bay Area have provided information to support management. Based partially on the preliminary information from this study, two additional loads monitoring sites were selected (North Richmond Pump Station watershed and Pulgas Creek Pump Station watershed (which receives water from Pulgas Creek North and South subwatersheds reported in this study). Using the data collected during the study, it was possible to rank watersheds from most polluted to least polluted for PCBs and mercury, effectively providing managers with assurance that management effort in a few more polluted watersheds would likely be more cost-effective towards reducing loads. In relation to continuing to develop regional load estimates using RWSM, the data collected has utility on both the input side and for calibration and verification. The data collected could be used as a statistical baseline for temporal trend comparisons in relation to management actions. The statistical analysis using PCA and correlation matrix identified a gradient within Bay Area watersheds in relation to urbanization and industrialization and a second gradient related to watershed area and low density residential. These gradients are consistent with and continue to provide support for our hypothesis that both imperviousness and land use age and intensity impact contaminant loads. The dataset collected is extremely valuable. However, should a similar effort be repeated in the future (for further identification of high leverage watersheds, to support development of the RWSM, or to further explore cause-and-effect statistical relationships) some minor cost-effective improvements in sampling design were identified.

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