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Sediment-adsorbed total mercury flux through Yolo Bypass, the primary floodway and wetland in the Sacramento Valley, California

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A B S T R A C T

The fate and transport of mercury are of critical concern in lowland floodplains and wetlands worldwide, especially those with a history of upstream mining that increases the mobility of both dissolved and sediment-bound Hg in watersheds. A mass budget of total mercury (*THg*) quantifies sources and storage for particular areas — knowledge that is required for understanding of management options in lowland floodplains. In order to assess contaminant risk in the largest flood-control bypass, prime wetland, and restoration target in the Sacramento River basin, we estimated empirical relationships between *THg*, suspended sediment concentration (*SSC*), and streamflow (*Q*) for each of the major inputs and outputs using data from various publicly available sources. These relationships were improved by incorporating statistical representations of the dynamics of seasonal and intra-flood exhaustion (hysteresis) of sediment and mercury. Using continuous records of *Q* to estimate *SSC* suspended sediment flux and *SSC* to estimate *THg* flux, we computed the net transfer of sediment-adsorbed mercury through the Yolo Bypass over a decade, 1993–2003. Flood control weirs spilling Sacramento River floodwaters into the bypass deliver ~75% of the water and ~50% of the river's suspended sediment load, while one Coast Range tributary of the bypass, Cache Creek, contributes twice the *THg* load of the mainstem Sacramento. Although estimated sediment flux entering Yolo Bypass is balanced by efflux to the Sacramento/San Francisco Bay-Delta, there is much evidence of deposition and remobilization of sediment in Yolo Bypass during flooding. These factors point to the importance of the bypass as sedimentary reservoir and as an evolving substrate for biogeochemical processing of heavy metals. The estimates of mercury flux suggest net deposition of ~500 kg in the 24,000 ha floodway over a decade, dominated by two large floods, representing a storage reservoir for this important contaminant.

1. Introduction

1.1. Statement of the problem

Much attention has been paid to mercury sources and sinks in the Sacramento River basin of California, due to the legacy of hydraulic gold mining in the Sierra Nevada and Hg mining in the Coast Ranges, which has created persistent contamination of lowland sediments and food webs (Bouse et al., 2010; Conaway et al., 2007; David et al., 2009; Davis et al., 2008; Domagalski, 2001; Eagles-Smith et al., 2009; Gehrke et al., 2011a, 2009; Greenfield et al., 2005; Greenfield and Jahn, 2010; Marvin-DiPasquale et al., 2009, 2003; Roth et al., 2001; Rytuba, 2000). However, there is still great uncertainty in the mass balance of total mercury (*THg*) delivered to and stored in lowland floodplain environments from different source areas. Such estimates are particularly important given that lowland floodplains integrate basinwide delivery of chemical constituents and engender conditions favorable for mercury methylation (Compeau and Bartha, 1985; Gilmour et al., 1992; Marvin-DiPasquale et al., 2003). In this paper, we develop empirical relationships from hydrology, sediment, and mercury data collected by various regional and state programs to evaluate the delivery of *THg* to a large, engineered floodplain in the Lower Sacramento River basin and its storage over a decade. The work is relevant to contamination of food webs within a biologically productive lowland floodplain ecosystem (Sommer et al., 2001b), as well as to past and future data collection efforts in this region. These will enable better understanding of prevailing and potential contamination risks to Yolo Bypass and Sacramento/San Francisco Bay-Delta food webs. We intend the results of this analysis to augment current understanding of sediment and mercury storage in the Bypass, to clarify the relative importance of the contributing watercourses, and to inform the targeting of efforts to stabilize mercury sources.

Although the Sacramento River watershed contributes an estimated 80% of the *THg* moving through the Sacramento-San Joaquin Bay-

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Delta (Foe, 2003), a region now classified as impaired under the Clean Water Act for excessive levels of mercury and other contaminants (Larry Walker & Associates, 2002), Hg concentrations in water and suspended sediments have been assessed inconsistently in space and time. Data collection campaigns have generally been conducted within particular regions of the basin over short periods. Consequently, generalizations from such data to broader spatial and time scales are ill-defined.

1.2. Previous mercury studies

Most previous studies report on a single set of event and/or annual sample data from a particular monitoring program. Estimates from Larry Walker & Associates (LWA) (2002) took advantage of several monitoring programs spanning the years 1992–2000. Mercury concentration estimates from that study were derived from univariate regressions with discharge data, where available. Where no discharge data were available, they used daily values or monthly means from the nearest discharge gauging stations to estimate regressions. There are several potential problems with such an approach. First, *THg* concentrations can vary significantly for a given flow rate (see LWA (2002), Fig. 2-2b). Second, since the majority of mercury tends to be adsorbed to fine sediment particles (e.g. (Domagalski, 2001; Maurice-Bourgoin et al., 2002)), a direct relationship between flow and mercury would tend to overestimate concentrations in water of the falling limb of the hydrograph if sediment hysteresis occurs (illustrated in Fig. 2-2a from LWA (2002)). Third, monthly mean discharge data dampen flood peaks that are generally responsible for the majority of the sediment/mercury flux (Singer and Aalto, 2009; Singer and Dunne, 2001).

The LWA study (and other shorter term studies) did not estimate Hg concentrations in flow overtopping Fremont Weir. This is an important omission because it is the largest source of flow, sediment, and *THg* derived Sacramento Basin (Domagalski, 2001; Singer and Aalto, 2009). Overall, there is a lack of long-term analyses on mercury mass balance within the Yolo Bypass, where higher rates of methylation are expected because of the organic carbon-rich, stagnant wetland environments that occur there (Rudd, 1995; Zilloux et al., 1993).

Here we will quantify the relative contributions of each major *THg* source to the bypass and flux out to the Bay-Delta. The study is intended to build on existing analyses that have assessed mercury in and around Yolo Bypass by providing a longer view based on mass fluxes of water, sediment and *THg*.

2. Study Area and Historical Data

2.1. Study area

The Yolo Bypass, 66 km long, is a 24,000 ha conduit for flood flow from Sacramento River, Feather River, Sutter Bypass, Knights Landing Ridge Cut (KLRC), Cache Creek, and Putah Creek before reconnecting with the Sacramento River upstream of the Sacramento/San Francisco Bay-Delta head at Rio Vista (Fig. 1). It has a flow capacity of 14,160 m^3 s^{-1}, or 4.5 times that of the lower Sacramento River channel; this capacity has been approximately reached twice in its ~80-year history, in February of 1986 and January of 1997. Fremont Weir is the primary source of Yolo Bypass inundation; the smaller tributary inputs are perennial and their storm flows more frequent (Schemel et al., 2002).

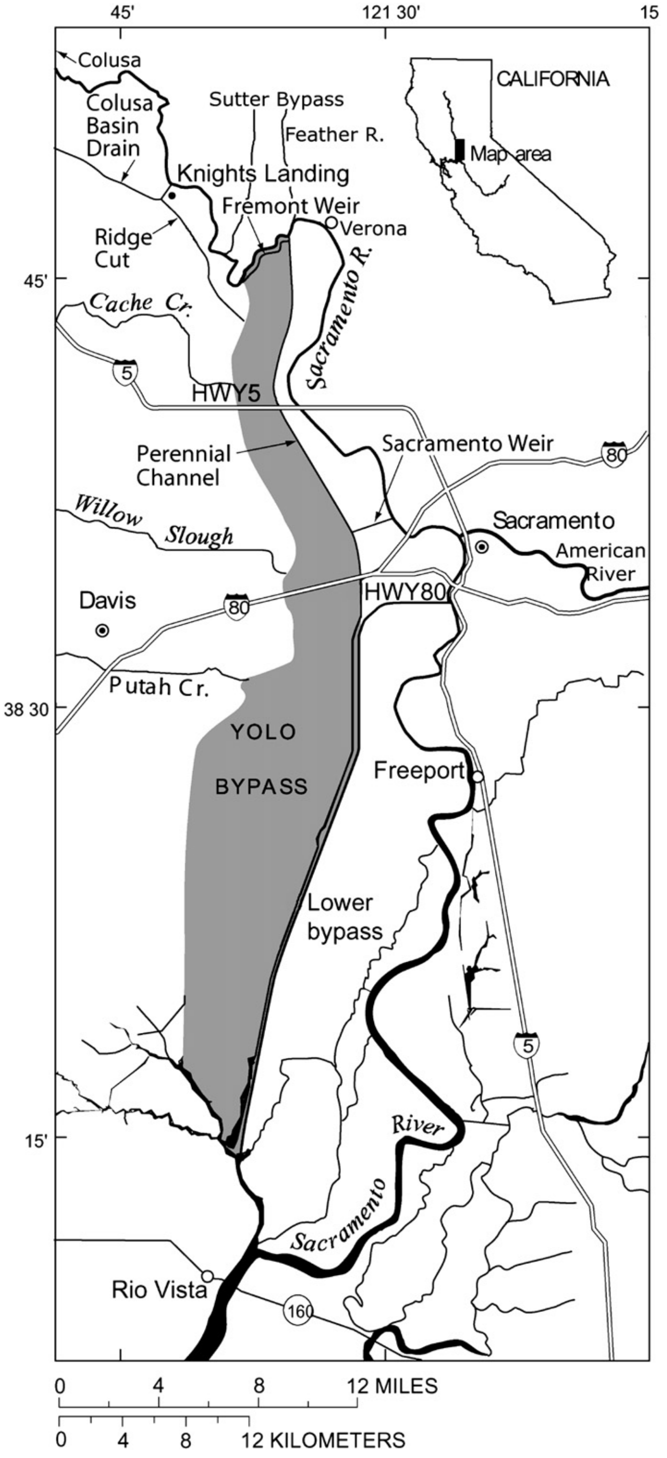

Fig. 1. Map of study area (adapted from (Schemel et al., 2002)).

Yolo Bypass is fundamentally important for various purposes, including flood control and biological habitat (Sommer et al., 2001a), and plays an important role in the fate and transport of fine sediment and adsorbed mercury in the basin. Much of the Bypass is farmed during the growing season, and parts of it are used for wildlife habitat. The engineering of this lowland floodway was completed by the 1930s, allowing for the safe diversion of up to 80% of basin flow in floods which previously had inundated most of the valley floor (Kelley, 1998). At that time, the lower Sacramento Valley was still recovering rom extensive hydraulic mining in the Sierra foothills, which delivered huge volumes of sediment to what was then Yolo Basin and other lowland flood basins in the region (Gilbert, 1917). This deposition occurred at repeatedly reoccupied crevasses and natural overflow loci within natural levees, such as at the entrance to Yolo Bypass, which is now controlled by a passive overflow weir, but which still allows sediment to overpass (Singer and Aalto, 2009; Singer et al., 2008). The deposit that has built up along this margin is being gradually dissected by headward erosion (Singer and Aalto,

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2009), which releases stored mercury to wetland areas near the Bay-Delta.

In the Sacramento basin, the leading source of Hg contamination is not atmospheric deposition (as is the case for most of the United States), but geological sources and mining activity (Domagalski, 1998; Larry Walker & Associates, 2002). There are 52 registered, productive (past or present) mercury mines in the Sacramento Basin, all but three of which lie in the Putah Creek and Cache Creek watersheds (Fig. 1). Processed mercury sulfide (HgS, or cinnabar), mined from these Coast Range drainages (on the western boundary of the basin), was used throughout the hydraulic and subsequent mining periods in the Sierra foothills to separate gold from lighter materials, a process which resulted in mercury losses of up to 30% (Bowie, 1905; Domagalski, 1998; Schemel et al., 2002). In total, between 1.4 and 4.5×10^6 kg of Hg are estimated to have been lost to the rivers draining the western slope of the Sierra Nevada (Alpers and Hunerlach, 2000; Churchill, 2000). This Hg has led to contamination of Sierra-draining rivers in their piedmont courses that is registered in elevated bioaccumulation in the tissues of local aquatic species (Davis et al., 2008; Gehrke et al., 2011b; Hunerlach et al., 1999; Slotton et al., 1997; Slotton et al., 2003) and waterfowl (Eagles-Smith et al., 2009; Greenfield et al., 2005; Greenfield and Jahn, 2010), as well as in sediments (Alpers et al., 2005; Choe et al., 2004; Conaway et al., 2007; Domagalski, 2001; Domagalski et al., 2004; Gehrke et al., 2011a; James et al., 2009; Roth et al., 2001) at locations downstream of both gold and mercury mining sites in the basin.

The US Geological Survey (USGS) National Water-Quality Assessment (NAWQA) program has investigated mercury concentrations in bed sediments, suspended load, dissolved load, and in tissues of aquatic organisms at various locations. These studies indicate high *THg* levels in bed sediments in Sierra Nevada gold mining streams such as Bear and Yuba Rivers (tributary to Feather River), and in Cache Creek and Yolo Bypass (Domagalski, 1998, 2001; Domagalski et al., 2004; Maccoy and Domagalski, 1999) and high concentrations of *THg* in suspended sediments within Yolo Bypass (Domagalski, 2001; Roth et al., 2001). Cache Creek is the largest tributary source of sediment and mercury, and enters Yolo Bypass over a passive weir after depositing 60% of its sediment load and 39% of its Hg in the Cache Creek Settling Basin (Cooke et al., 2004). Although *THg* in mining sediments appear to be hot-spot dominated (Ashley et al., 2002), mercury has been documented on sediments in high concentrations on the periphery of the delta (Choe et al., 2003; Rytuba, 2000; Slotton et al., 2002), indicating recent flood-based inputs from the major rivers and Yolo Bypass. The Central Valley Regional Water Quality Control Board conducted regional water sampling that indicated Yolo Bypass constitutes a substantial mercury loading source to the Bay-Delta in years when it is flooded (Domagalski, 2001; Foe, 2003).

Up to ~5×10^6 tonnes of sediment can be delivered to Yolo Bypass during floods, much of which is stored near its northern entrance (Singer and Aalto, 2009). Upon deposition in floodplains, sediment-adsorbed Hg may be methylated by sulfate-reducing bacteria under suitable chemical and physical conditions (Compeau and Bartha, 1985; Gilmour et al., 1992), making it available for bioaccumulation through the food chain. Indeed, filtered sampling of Yolo Bypass waters in conjunction with a Cache Creek watershed mercury loading study found that the bypass environment promotes the production of methylmercury (Domagalski et al., 2004). Several other studies are ongoing.

Chemical studies of streamflow, sediments, and tissues of Sacramento Valley aquatic organisms have revealed high levels of mercury and other trace metals used in gold extraction (Domagalski, 1998, 2001; Foe, 2003; Heim et al., 2003; Roth et al., 2001). Likewise, studies in the Delta channels found elevated levels of mercury bioaccumulation in North Delta regions exposed to inflows from the Yolo Bypass (Slotton et al., 2002). And recent work has istopically linked Hg in forage fish tissues to that found in basin surface sediments (Gehrke et al., 2011b). Storage of contaminants like Hg poses a risk to biota in Central Valley floodplains, many of which serve as the last regional remnants of productive lowland aquatic habitat (Sommer et al., 2001a,b). Bioaccumulation of mercury in fish poses a serious health risk to humans (Bloom, 1992; Maurice-Bourgoin et al., 2002; White et al., 1995). These factors have led the CALFED Bay-Delta Authority to develop a Mercury Strategy, which includes "comprehensive quantitative assessments of residual mercury…in the alluvial deposits in the Central Valley upstream of the Bay-Delta" as its first core component (Weiner et al., 2003).

2.2. Data sources

Data compiled for this study include daily mean flow rates *(Q)*, mean, depth-integrated suspended sediment concentration *(SSC)*, and concentration of *THg* in unfiltered surface grab samples from various studies. Tables 1 and 2 provide of a summary of the observations and data sources for the three direct inputs (Cache Creek, Putah Creek, and KLRC) and for the source waters providing flow over the Fremont and Sacramento Weirs. While flow rates over the weirs are available, sufficient records of *SSC* and *THg* at the weirs are not,

Table 1
Sediment and Hg data.

		SSC		THg		Source
	Gaging Site	n	Date Range	n	Date Range	
Direct Inputs	Knights Landing Ridge Cut	56	2/7/96–4/13/03	4	3/6/96–4/13/03	1,5
	Cache Creek (low flow)	7	2/23/96–6/11/97	15	12/23/96–2/22/98	3,6
	Cache Creek (high flow)	8	1/6/97–2/22/98			
	Putah Creek	18	3/28/00–10/1/01	18	3/28/00–10/1/01	3
Sources for Fremont Weir	Sacramento River @ Colusa	76	3/10/95–4/13/03	46	3/10/95–4/13/03	1,5,6
	Sacramento Slough (low flow)	66	4/22/96–9/23/03	38	4/22/96–4/13/03	4
	Sutter Byapss (high flow)	9	2/12/96–1/22/03	8	2/12/96–1/22/03	1,5
	Feather River near Nicolaus	51	2/23/96–4/13/03	48	2/23/96–4/13/03	1,5
Sources for Sacramento Weir	Sacramento River @ Verona	26	2/2//96–5/20/98	26	2/2//96–5/20/98	1
	American River @ Sacramento	119	1/15/96–8/5/03	106	1/15/96–8/5/03	1,2
Bypass outlet	Yolo Bypass (outlet)	41	1/12/95–10/1/01	43	1/10/95–10/1/01	3
	Sacramento River @ Freeport	305	1/10/90–9/23/03	140	2/15/94–8/6/03	1,2

1 US Geological Survey National Water-Quality Assessment Program (http://ca.water.usgs.gov/sac_nawqa/waterindex.html).
2 Sacramento Coordinated Water Quality Monitoring Program (http://www.sfei.org/rmp/1997/c0802.htm).
3 Central Valley Regional Water Quality Control Board (http://bdat.ca.gov).
4 California Department of Water Resources (http://cdec.water.ca.gov).
5 Sacramento River Watershed Program (http://www.sacriver.org).
6 US Geological Survey (http://waterdata.usgs.gov/nwis).

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Table 2
Flow Data.

Gaging site	Site #	Source
Sacramento River below Wilkins Slough	11390500	1
Sacramento River at Colusa	11389500	1
Sacramento River at Verona	11425500	1
Sacramento River at Freeport	11447650	1
American River at Fair Oaks	11446500	1
Cache Creek at Yolo	11442500	1
Colusa Drain at Knights Landing	A02945	2
Colusa Drain at Hwy 20	A02976	2
Putah Creek	PUT	2
Sacramento Slough near Karnak	A02925	2
Fremont Weir Spill to Yolo Bypass	A02930	2
Feather River near Nicolaus	11425000	2,3
Yolo Bypass outlet	DAYFLOW	4

1 US Geological Survey (http://waterdata.usgs.gov/nwis).
2 California Department of Water Resources (http://cdec.water.ca.gov/).
3 US Geological Survey National Water-Quality Assessment Program (http://ca.water.usgs.gov/sac_nawqa/waterindex.html).
4 Interagency Ecological Program (http://iep.water.ca.gov/dayflow/).

necessitating the analysis of source water concentrations as a proxy. The final two sites at the bottom of Table 1 are for the output from the Yolo Bypass and for the Sacramento River at Freeport, a site representing mainstem input to the Bay-Delta (allowing for the estimation of *THg* efflux and comparisons to previous studies). For this study, Prospect Slough represents the downstream boundary of Yolo Bypass.

In order to characterize Hg flux over numerous locations and over several years incorporating many storm seasons, it was necessary to synthesize data from many different studies and agencies. In some settings the advent of ultra-clean sampling methods have led to reductions in measured *THG* by over two orders of magnitude (Bloom, 1995). While all of the sampling programs accessed for this study follow stringent sampling standards, inter-laboratory variance is likely, but cannot be quantified. Since it has been shown elsewhere that errors in individual sediment concentration measurements are between 5% and 20% (Topping et al., 2000), we expect similar errors for sediment-adsorbed *THg*.

2.3. Sediment and THg

Table 1 presents the number of sample observations of *SSC* and *THg*, as well as the date range of their collection, to provide a sense of temporal representation. *SSC* and *THg* observations are most limited temporally for Putah Creek and the Sacramento River at Verona. Other limitations in the dataset include only nine observations for the Sutter Bypass in flood and 15 total observations for Cache Creek downstream of the Cache Creek Settling Basin. We identified sampling sites as close to Yolo Bypass as possible for all inputs (see below).

2.4. Flow

All flow data (Table 2) were either obtained as mean daily *Q* or converted to this form from hourly values. For Putah Creek (Fig. 1) we used *Q* from a station 35 km upstream of the Bypass. Cache Creek *Q* values were available at Yolo, ~17 km upstream of the Bypass, and considering the limited water storage capacity in the Settling Basin, this gauge provides an accurate estimate of flow over the Cache Creek Weir. While flow rates for the Feather River near Nicolaus, 17 km upstream of the Sacramento confluence, are only available for the period 1995–1998, stage data for the period of study (1993–2003) are available. However, because an updated stage-discharge rating curve could not be located, we developed one with a limited number of discharge measurements to calculate *Q* for years other than 1995–1998.

Due to lack of data, it was necessary to estimate the flow rate over the entire temporal domain for three cases: Sutter Bypass in flood, Sacramento River above Fremont Weir, and the KLRC. In low flow, Sutter Bypass flow is generally constrained to the Sacramento Slough, which spills into the Sacramento River just downstream of the Fremont Weir. Flood flows from the Sutter Bypass were calculated as the sum of Fremont and Verona flows minus the sum of the Sacramento River upstream of Fremont Weir and the Feather River at Nicolaus. Flow upstream of Fremont was taken as the sum of the Sacramento River at the Wilkins Slough gauge (71 km upstream from Verona) and the Colusa gauge.

The nearest gauge site that captures flow rate for the Sacramento River above Fremont Weir is below Wilkins Slough. In between this point and the weir, *Q* from the Colusa Drain at Knights Landing spills into the Sacramento when the outfall gates are opened. Thus, the water contribution of the Sacramento above Fremont Weir is set equal to the Wilkins Slough gauge value combined with that of the Colusa Drain. Further details can be found in the Supplementary Data.

Estimates of *Q* at the downstream outlet of Yolo Bypass (Prospect Slough) were extracted from the Interagency Ecological Program's Dayflow calculator, which combines flow in the Yolo Bypass at Woodland with spill over the Sacramento Weir (entering the Bypass downstream of Woodland) and discharge from Putah Creek (as calculated in this study). This proxy is necessary because a significant tidal influence from the Delta on flows at this site affect direct measurements. While a digital filter may be used on direct measurements to remove the tidal signal, such data are not historically available. The Dayflow calculation is believed to underestimate *Q* when values are in the low range and when the floodplain is draining because the estimates may depend on gauged inflows (http://www.water.ca.gov/dayflow/).

3. Methodology and statistical model

Our estimation approach involved a two-stage regression procedure for estimating *SSC* and *THg* at each site. In the first stage, we utilized daily *Q* records and event-based samples of *SSC* to estimate a *Q-SSC* relationship. In the second stage, we modeled *THg* levels as a function of both *Q* and *SSC*. In both stages, estimates at some gauges were improved by using additional hydrologic variables intended to capture hysteresis effects. Using the relationships characterized in both stages, we translated the *Q* record into daily *SSC* predictions, which were subsequently used to calculate daily *THg* estimates. From *Q* and *SSC* we calculated the daily mass flux of suspended sediment and *THg* flux for each site. Thus our estimates of *THg* are relevant to the sediment-bound Hg only. Because samples of *SSC* and *THg* are not widely available for spill over Fremont Weir and Sacramento Weir, we estimated their concentrations in the source waters and developed two simple models of how these waters might mix in the Sacramento River before the weir is overtopped. Finally we develop a bootstrapping method for characterizing the uncertainty of our predictions, which also allows for hypothesis testing at any level of temporal aggregation, from days to the entire 10-year period.

3.1. Estimating SSC

In the first stage, we use linear regression to estimate a relationship between log transformed *Q* and *SSC* for any time *t*:

$$\ln SSC_t = \alpha + \beta \ln Q_t + \varepsilon_t \tag{1}$$

where α is the intercept, β is regression slope, and ε_t is normally-distributed error term. An example of the positive correlation between *SSC* and *Q* is shown in Fig. 2.

Some aspects of antecedent hydrology might affect the *Q-SSC* relationship, including the timing and size of previous floods, hydrograph shape, antecedent soil conditions and the relative

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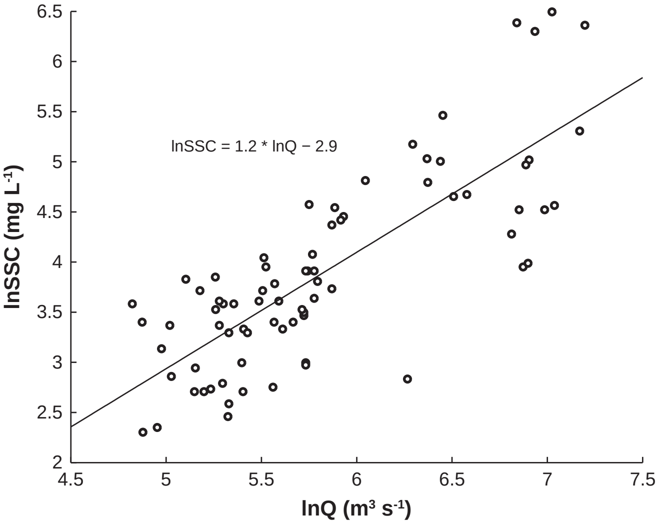

Fig. 2. A scatterplot of log-*SSC* versus log-*Q* with a linear best-fit line, from samples taken in the Sacramento River at Colusa.

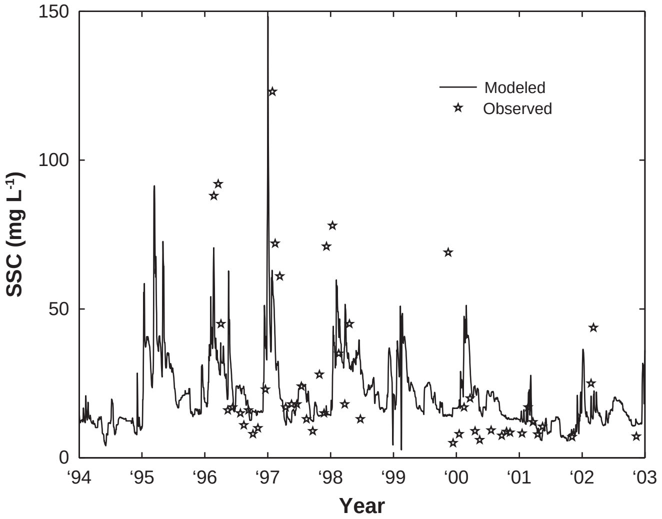

Fig. 3. Example of the first estimation stage prediction, estimated daily *SSC* levels (continuous line) and actual *SSC* samples (stars) for the Feather River at Nicolaus.

amounts of rainfall and snowmelt runoff. These effects might contribute hysteresis to the relationship if the readily available supply of sediment declines during a period of high runoff (Heidel, 1956). In our first-stage model, we explored a small set of functions of previous values of Q that might capture sediment hysteresis dynamics. The simplest such function was Q from the previous day ($lagQ_{1day}$). To capture hysteresis at work over a longer time frame, we included the sum of Q over the previous week ($sumQ_{1week}$). To account for the effect of sustained extreme flows, we used the sum of Q over the previous two weeks ($sumQ_{2weeks}$). For each site we first assess whether the specification in [1] is appropriate and then attempt to improve on the predictive power of the model by including a hysteresis variable in a multiple regression, which is a simpler, yet more transferable method than previously developed (c.f., (Singer and Dunne, 2001)).

Table 3 summarizes the regression specifications used for each of the three direct tributary inputs, six weir-spilled water sources, the Bypass output and the Sacramento River gauge at Freeport. All variables were log-transformed for the regression and only significant ones ($p<0.1$) were used as the criterion for inclusion in the regression equation (Table 3). Flow (*Q*) is a significant variable in explaining the variation in *SSC* at each site except for Putah Creek and during lower flows at Cache Creek. At these sites sediment supply is likely to be spatially and temporally variable and discharge is affected by flood control structures, so we use simple averages of *SSC* for all available data for these two cases.

Accounting for exhaustion by inclusion of variables that represent antecedent conditions improves estimates for several sites. Hysteresis variables had a significant impact on *SSC* for the Sacramento at Colusa and Verona ($lagQ_{1day}$), for the Sutter Bypass in flood (*sumQ1wk*), and for the Yolo Bypass output (*sumQ2weeks*). Predicted daily *SSC* v. observed *SSC* for the Feather River at Nicolaus is shown in Fig. 3.

3.2. Estimating THg

Log transformed *THg* was regressed against log transformed *SSC* for time *t* as

$$\ln THg_t = \beta_0 + \beta_1 \ln SSC_t + \mu_t, \tag{2}$$

where β_0 is regression intercept, β_1 is regression slope, and μ_t is a normally distributed error term. For several sites, log-transformed *Q* was also significant and therefore was included as a second stage variable in the multiple regression. Table 3 summarizes regression models for *SSC* and *THg*. The hysteresis variable $lagQ_{1\mathrm{day}}$ was also significant for the Sacramento River at Colusa, the Feather River at Nicolaus and the Bypass outflow.

3.3. Mixing models

There are three major inputs to the spill over Fremont Weir and two major inputs to the spill over Sacramento Weir (Table 1) that

Table 3
Regression Models.

			SSC		THg			
	Gauging Site	Const.	Q	site-specific	Const.	CSS	Q	lagQ
Direct Inputs	Knights Landing Ridge Cut	3.05	0.27	−0.52 wet season	−2.14	0.90		
	Cache Creek (low flow)	4.94			−1.10	1.03		
	Cache Creek (high flow)	−7.70	1.53		−1.10	1.03		
	Putah Creek	3.37			−0.07	0.69		
Sources for Fremont Weir	Sacramento River @ Colusa	5.76	2.44	−1.42 lagQ1day	−5.52	0.46	1.60	−1.03
	Sacramento Slough (low flow)	4.11		−0.44 sumQ1week	−0.60	0.73		
	Sutter Byapss (high flow)	3.58	0.57		0.91	0.25		
	Feather River near Nicolaus	−1.44	0.50		−1.70	0.44	1.81	−1.57
Sources for Sacramento Weir	Sacramento River @ Verona	−0.68	3.36	−2.90 lagQ1day	−2.65	1.15		
	American River @ Sacramento	−3.81	0.66		−3.45	0.23	0.45	
Bypass outlet	Yolo Bypass (outlet)	3.50	0.22	−0.07 sumQ2weeks	−0.94	0.94	0.27	−0.31
	Sacramento River @ Freeport	−4.15	1.89		−3.05	0.25	0.41	

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affect the sources of flow, sediment, and Hg to Yolo Bypass. Since observations of *SSC* and *THg* are not available at the weirs, we estimated the concentrations of these constituents in the spill water by mixing estimates from the various sources under two different scenarios: perfect mixing (PM) and hierarchical allocation (HA). In the PM scenario we assume that source waters mix perfectly before overtopping the weir. Therefore, *SSC* and *THg* are computed as a simple additive function. Under the HA approach, discharge from the first source to reach the weir each day is completely exhausted before the next source contributes. This continues until the measured discharge over the weir is reached. For the Fremont Weir the order of contribution, based on upstream distances, is (1) Sacramento River, (2) Sacramento Slough/Sutter Bypass, and (3) Feather River. It is therefore possible under HA that the Feather River will not contribute sediment and mercury to Fremont Weir spillage during certain flooding periods. For the Sacramento Weir the order of contribution is (1) Sacramento River and (2) American River. An alternative approach could have been to estimate the degree of lateral mixing of sediment as function of distance upstream of each weir, but since the difference in the two outcomes considered here is not large, we did not add the extra complication.

In Fig. 4, we present an example of the proportion of *Q* from each source for the Sacramento Weir under the two assumptions. For this particular date range, the figure shows how the contribution of the Feather River (the last to contribute under the HA scenario) to Fremont Weir spillage ranges from ~20% to zero depending on the mixing model used. In Fig. 5, the proportions from Fig. 4 are translated via [1] and [2] into Hg flux over the weir by source.

3.4. Estimating confidence intervals

A bootstrap approach is used to calculate confidence intervals for predictions of concentration and flux for both *SSC* and *THg* at each site over the decadal time domain. For each site, we began by assuming that residuals from the first stage are normally distributed and directly calculated the variance from them. The validity of this assumption was evaluated by plotting residuals and analyzing their statistics. We then drew an error term from this distribution for each day and added it to the *lnSSC* prediction in [1]. This process was repeated, creating 500 bootstrapped samples of daily *lnSSC* predictions. We then performed the inverse transformation (correcting for bias as instructed by Duan (1983)), sorted the bootstrapped samples, and selected values within the 90% confidence interval from the resulting empirical distribution (i.e., between the 26th and 475th highest values).

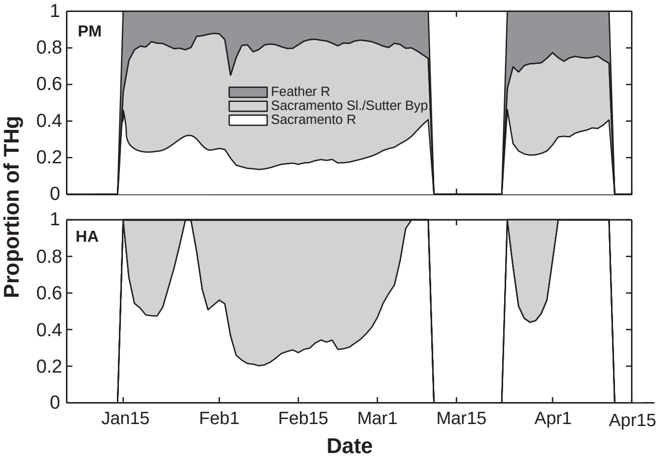

Fig. 4. Relative contributions of *THg* in flow from three sources over Fremont Weir under PM (top) and HA (bottom) for a flooding period in 1998.

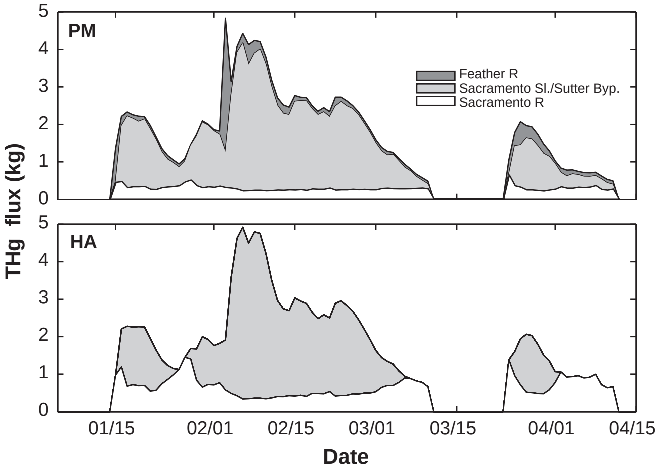

Fig. 5. Relative contributions of Hg flux from three sources over Fremont Weir under PM (top) and HA (bottom) for a flooding period in 1998.

Next, the 500 bootstrapped first-stage predictions of daily *lnSSC* were used to create 500 second-stage predictions of daily *lnTHg* according to [2]. We pursued the same 90% confidence interval strategy as described for *lnSSC*. Fig. 6 shows daily *THg* predictions and the estimated 90% confidence interval for a selected period on the Sacramento River at Colusa site. To calculate confidence intervals for flux estimates we converted the unsorted bootstrapped samples of concentration values (*SSC* or *THg*) into flux estimates using *Q* and summed them over the desired time period (e.g. one year) before sorting to create the empirical distribution.

4. Results

While our central goal is quantifying the supply or mobilization of Hg within the Bypass over the ten-year period, our approach allows for the estimation of the mass flux of water, sediment and mercury from each of the five sources and through the single outlet over any time scale at or greater than one day. For each water year and for the period as a whole we present mass flux estimates of sediment and mercury flux and their 90% confidence intervals. Mass fluxes for the Sacramento River at Freeport are also calculated and combined with Bypass fluxes to characterize total Sacramento River basin contributions to the Bay-Delta, which begins at Rio Vista (Fig. 1). The mixing of source waters has a bearing on the results, so we

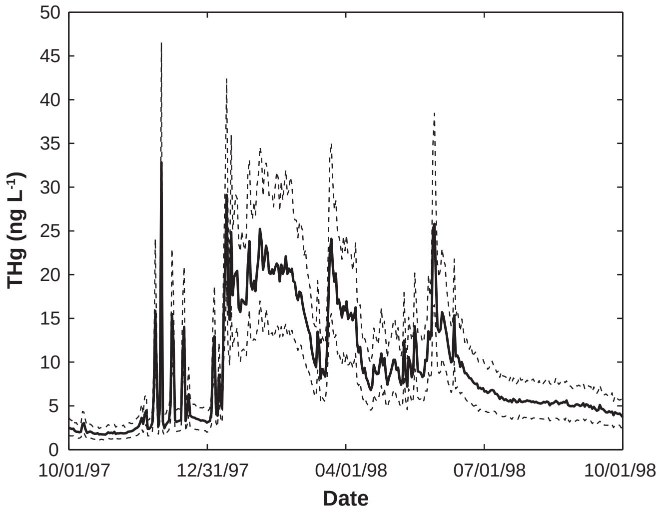

Fig. 6. Example of the second stage prediction of daily mercury concentration (bold line) and 90% confidence interval, Sacramento River at Colusa, water year 1998.

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summarize the impact of the two scenarios on sediment and Hg loading below.

4.1. Mixing

Estimates of Hg and suspended sediment loading over both weirs under the alternative assumptions of PM and HA appear in Tables 4 and 5. In every year with nonzero loading, suspended sediment fluxes over the weirs are greater under HA than PM, though only significantly so for 50% of years. For both weirs, 10-year suspended sediment totals under HA are approximately 25% greater than under PM, a difference which is significant at the 90% level. This suggests that sources last to contribute in the HA scenario, the Feather and American Rivers, generally contribute less sediment per unit of water than the Sacramento River, which is the first source to contribute in both cases. One caveat on this conclusion is that *SSC* for Feather River is poorly defined for high flows (Singer and Aalto, 2009), so we may underestimate sediment flux from this basin. Indeed this may lead to underestimation of high *SSC* values (and thus *THg*) during floods (Fig. 3). Thus, our estimates of mercury loading to Yolo Bypass are likely to be conservatively low.

Given the significant differences for sediment flux, the results for mercury flux are quite surprising—there is no significant difference in mercury flux under PM and HA for any year or in the 10-year total (see Table 5). This outcome is not driven by more generous confidence intervals for Hg flux. In fact, the percent deviation from the yearly flux estimate of the lower and upper bounds is *smaller* for mercury than it is for suspended sediment. A potential explanation is that sediment eroded from the Feather River basin contains higher concentrations of Hg, due to its legacy of hydraulic mining (primarily in its tributary basins, Yuba and Bear). This lower-in-sediment, higher-in-mercury mixture from the Feather basin becomes diluted by the relatively higher Sacramento River sediment mass under HA.

Table 4
SSC flux results.

	Fremont Weir		Sacramento Weir	
Water year	PM	HA	PM	HA
1994	0	0	0	0
1995	1198	1520	45	55
	(1103,1312)	(1397,1684)	(40,49)	(49,61)
1996	313	423	2	2
	(280,350)	(375,481)	(2,2)	(2,3)
1997	1182	1322	116	152
	(1018,1392)	(1167,1504)	(98,138)	(129,173)
1998	1103	1419	27	32
	(1019,1183)	(1312,1536)	(24,31)	(28,36)
1999	207	310	0	0
	(184,231)	(267,360)		
2000	464	602	0	0
	(418,517)	(531,681)		
2001	0	0	0	0
2002	28	36	0	0
	(21,37)	(24,53)		
2003	33	47	0	0
	(26,41)	(33,66)		
Total	4528	5679	190	241
	(4300,4776)	(5432,5958)	(170,212)	(218,263)

Table 5
THg Flux Results.

	Fremont Weir		Sacramento Weir	
Water Year	PM	HA	PM	HA
1994	0	0	0	0
1995	195	197	7	8
	(184,207)	(186,210)	(6,7)	(7,9)
1996	48	50	0	0
	(45,51)	(47,54)		
1997	219	198	20	22
	(198,245)	(182,218)	(17,22)	(18,25)
1998	166	177	4	4
	(157,173)	(168,186)	(3,5)	(4,5)
1999	37	34	0	0
	(34,40)	(32,36)		
2000	68	74	0	0
	(64,73)	(70,79)		
2001	0	0	0	0
2002	4	4	0	0
	(3,4)	(3,4)		
2003	4	5	0	0
	(4,5)	(4,5)		
Total	741	739	31	34
	(713,770)	(718,765)	(26,33)	(30,38)

4.2. Sediment and mercury deposition in Yolo Bypass

Total annual inputs, output, and storage estimates for Yolo Bypass under the PM model are presented in Table 6. On average, ~1000 kilotonnes of suspended sediment move through the Bypass in one year. While the net suspended sediment flux (storage) for the 10-year period is negative, suggesting mobilization of sediment from the Bypass, this value is not significantly different from zero. However, under the HA model net total flux is significantly positive at ~1000 kilotonnes (90% confidence interval, [148, 2129]). While a rough compromise between the PM and HA assumptions would lead to an estimate of net deposition on the order of 500 kilotonnes, the width of the confidence intervals under both scenarios (over 1800 kilotonnes) suggests that this would not be significantly different from zero at the 90% confidence level. Singer and Aalto (2009) modeled fractional (by grain size) concentration profiles based on weighted mixing of sediment from the various sources to compute silt-clay flux over Fremont Weir during the large flood of 1964. They computed a flux of ~5000 kilotonnes over more than a month of weir spillage. There are several possible explanations for the large disparity between the flux computed here for water year 1997 (~1000 kilotonnes), which contained a large flood, and the five times higher value computed in Singer and Aalto (2009) for the 1964 flood, which had a peak discharge ~10% larger than the 1997 event. First, the prior study employed regressions between instantaneous flow and instantaneous sediment concentration measurements that include higher peak values than are usually present in historical mean daily values. Second, the prior study developed such relationships using records from the late-1970's at the Feather River at Nicholaus, which were poorly defined for higher flows. As such, the computed a regression slope 42% higher than that estimated herein (c.f., Table 3 with Table 2 from Singer and Aalto (2009), and thus higher absolute values of sediment flux past Nicholaus. That prior study also estimated deposition in Yolo Bypass (~6000 kilotonnes), but that analysis was limited to the area between Fremont Weir and Hwy 5 (Fig. 1), so this indicates net event-based deposition during major floods could be much larger than estimated here. It should also be noted that regressions for the Bypass outlet (Yolo Bypass at Woodland) in that prior study were quite poor and therefore limit the certainty of the storage estimate (Singer and Aalto, 2009).

4.3. Annual variation

For simplicity of presentation and because Hg flux results were not sensitive to the alternative mixing scenarios, only the PM results will be discussed here. Table 6 indicates that ~500 kg of mercury was deposited in the Yolo Bypass during the decade with a small number of high flow years dominating the overall result. Providing a snapshot of relative yearly magnitudes, Fig. 7 displays total inputs (a), output (b) and net flux (c) of water, suspended sediment, and mercury.

Table 6
SSC and THg flux and storage results.

		SSC			THg	
Water year	Input	Output	In-Out	Input	Output	In-Out
1994	19	4	15	3	2	1
	(18,21)	(3,5)	(13,17)	(3,4)	(1,2)	(1,2)
1995	2650	2814	−164	712	551	161
	(2319,3104)	(2445,3239)	(−572,417)	(573,907)	(485,632)	(1,372)
1996	659	784	−124	152	157	−5
	(607,734)	(678,904)	(−246,3)	(134,177)	(137,179)	(−32,29)
1997	1974	2470	−496	486	467	18
	(1755,2259)	(2066,2927)	(−1037,22)	(415,573)	(397,550)	(−99,137)
1998	2962	2109	853	791	404	387
	(2636,3376)	(1856,2358)	(426,1336)	(647,969)	(360,449)	(242,570)
1999	367	394	−28	88	83	4
	(339,396)	(349,451)	(−90,27)	(81,96)	(74,95)	(−9,17)
2000	574	646	−73	92	137	−45
	(523,627)	(552,758)	(−189,37)	(86,98)	(116,167)	(−75,24)
2001	51	79	−28	10	18	−8
	(45,58)	(71,88)	(−40,-18)	(8,12)	(16,20)	(−11,-5)
2002	139	171	−32	27	38	−10
	(123,160)	(144,211)	(−75,1)	(23,33)	32,46)	(−20,-2)
2003	235	319	−84	58	68	−10
	(207,275)	(286,358)	(136,-29)	(47,74)	(62,76)	(−25,8)
Total	9630	9790	−161	2419	1925	493
	(9089,10358)	(9147,10448)	(−1067,862)	(2197,2734)	(1810,2041)	(224,821)

Eighty-four percent of the net flux of Hg occurred in 1995 and 1998, both years resulting in Yolo Bypass deposition. Including the year 2000, 91% of the mercury flux occurs during three water years. Not surprisingly then, floods play the most significant role in Hg delivery to and transport through Yolo Bypass.

A closer examination of the water balance provides reason to suspect that water outflow from the Bypass may be underestimated for the 1998 water year. Excluding 1998, the average annual difference between total water input and output is 7.4×10^8 m^3 with a range of -1.2×10^8 m^3 to 17×10^8 m^3. While some amount of loss may be expected from evaporation and pumping, the annual difference for 1998 is 4.3×10^9 m^3. While most of the discharge inputs to Yolo Bypass were at their maxima in 1998, outflow was only at the third highest value. This suggests that estimated outflow from the Bypass may be too low for 1998 (Fig. 7).

To estimate a possible lower bound for the 1998 outflow from the Bypass, it is useful to consider water year 1995, which had Bypass inflows similar to 1998 (~1.4 v. ~1.5×10^{10} m^3, respectively). Assuming

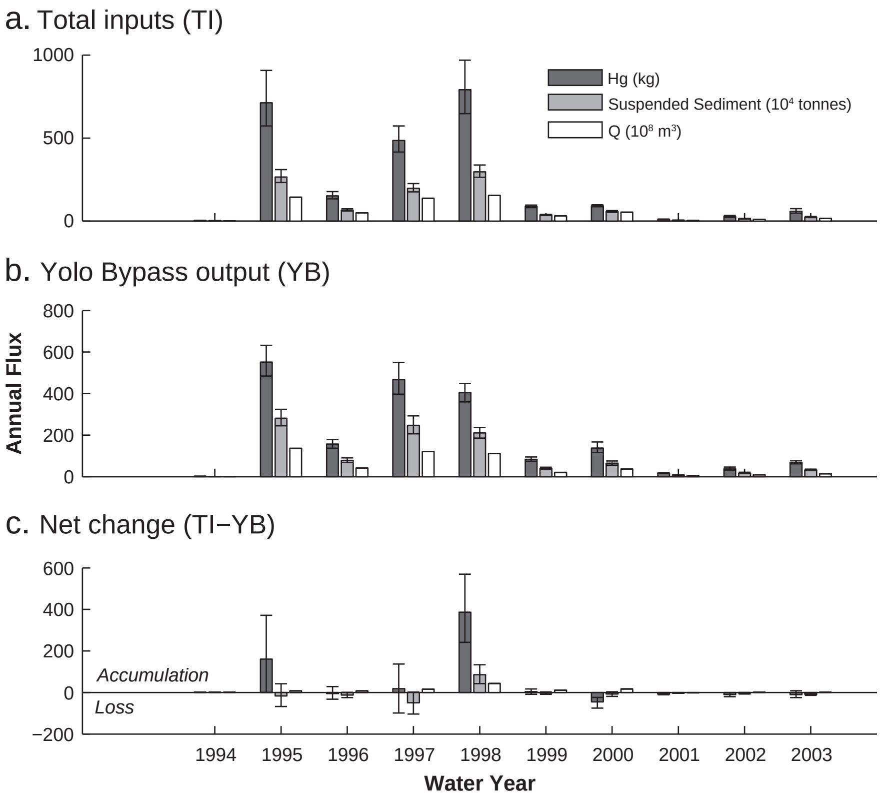

Fig. 7. Annual mass flux and storage of mercury, suspended sediment, and water through the Yolo Bypass.

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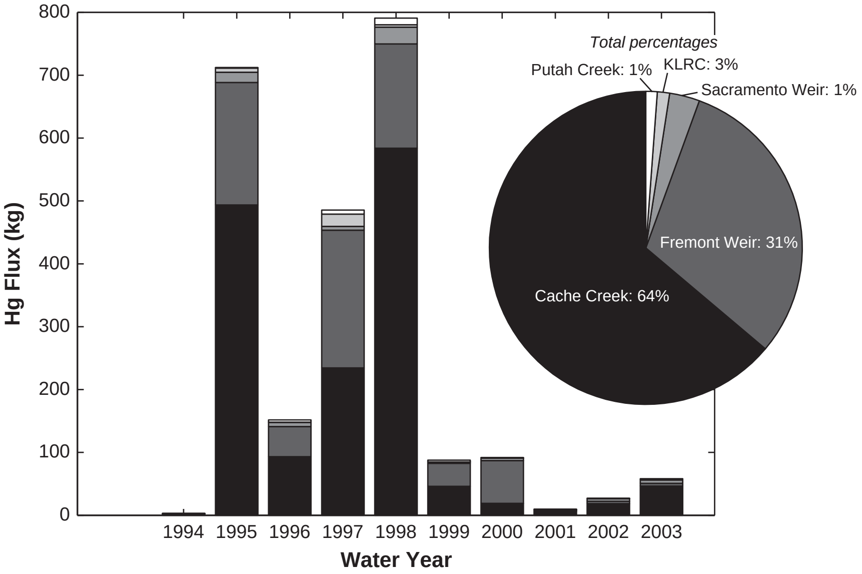

Fig. 8. Annual mercury contribution to Yolo Bypass from major inputs, 1994–2003.

the same outflow value as 1995 (~1.4×10^{10} m^3), a conservative estimate, would reduce the water storage in 1998 to 1.9×10^9 m^3)—still high, but much closer to that of other years (Fig. 7). This outflow adjustment would reduce net mercury deposition in Yolo Bypass by ~146 kg, or 38% less for 1998 and a 30% reduction in mercury storage for the entire period of study.

While Cache Creek and Fremont Weir trade off the role of largest sediment source from year to year, the Hg load from Cache Creek is greater in all water years except for 2000. Fig. 8 shows yearly mercury contributions by source. Contributing only ~11% of the water influx to the Bypass, Cache Creek contributes 38% of the sediment load and 64% of the mercury load. By contrast, the Fremont Weir is the leading source of water (71%) and sediment (47%) but delivers only 31% of the mercury mass, according to this analysis. The remaining Hg inputs are far smaller. KLRC is the next leading source (3%) while the roles of the two smallest sources, Sacramento Weir and Putah Creek are statistically indistinguishable (1%).

4.4. Comparison of mercury loading to Bay-Delta

Fig. 9a displays contributions of water, suspended sediment, and mercury from the Sacramento River at Freeport, 53 km upstream from the Yolo Bypass outlet near Rio Vista (Fig. 1). Fig. 8b is a reproduction of Yolo Bypass output from Fig. 5b. Total loading to the Delta and Yolo Bypass shares are presented in Fig. 8c and d, respectively.

Over the 10-year period, the Yolo Bypass supplied 17% of the water, 38% of the suspended sediment and 46% of the Hg contributed

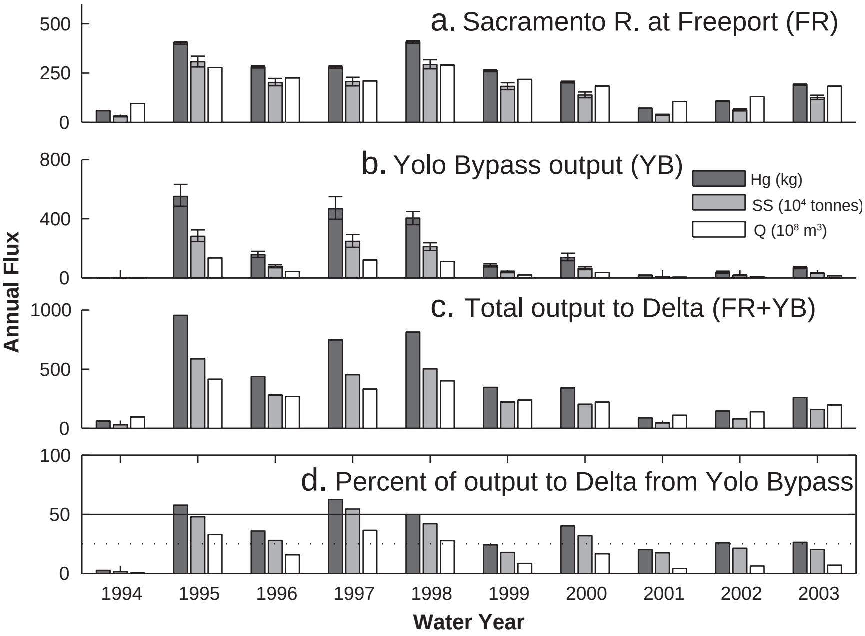

Fig. 9. Annual mass flux of mercury, suspended sediment, and water for the Sacramento River at Freeport versus output from the Yolo Bypass. 90% confidence intervals are included for suspended sediment (SS) and mercury (HG) flux estimates.

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from the Sacramento River basin to the Bay-Delta. The share of watershed Hg delivered to the Bay-Delta from Yolo Bypass shows high interannual variation, ranging from under 3% in 1994 to over 62% in 1997, indicating it plays a large role during major floods.

5. Discussion

Mass flux results from previous studies at our sampled locations are presented in Table 6. For five different previous studies, Table 6 presents the location and time frame of the existing result and compares each reported value to our own. The final column describes how the time frame and location of the original study differs from our own. Various approaches, as described in the introduction, were used in these calculations. In each case, our estimate is at least 16% larger. This result is inconclusive when the time frames compared are not well-matched (Domagalski, 2001; Foe and Croyle, 1998; Larry Walker & Associates, 2002).

Our larger estimates may also be driven by our use of a nonstationary and nonlinear relationships between *Q*, *SSC* and *THg*, while other approaches, where reported, relied on simpler models and estimates. For example, Domagalski et al. (2004) collected sample Hg concentration data in the Cache Creek watershed upstream of Yolo Bypass. Finding a poor linear relationship between *Q* and *THg*, they estimated annual loading using a dry season and a wet season average Hg concentration. For Cache Creek *above* the settling basin they find mercury loadings of approximately 12 kg in 2000 and 4 kg in 2001. These are significantly lower than our estimates of 19 kg and 7 kg for the same years for flux *below* the settling basin (where some portion of the mercury mass load should be deposited). The difference probably results from the fact that 2000–2001 were low-flow water years for Cache Creek, and accounted for less than 2% of the Hg flux in our 10-year total.

LWA (2002) used a single-stage regression framework with a nonlinear relationship between *Q* and *THg*. Instead of daily flow data, the median value of *Q* for the month was employed in the regression estimation. For the Sacramento River at Verona, the only site which was an exact match, our estimate of flux was 17% greater than the *LWA* estimate, as would be expected from our use of higher-frequency flow data (Walling and Webb, 1987). While the 35% lower *LWA* estimate of Hg for Yolo Bypass at Woodland does not include Putah Creek or Sacramento Weir inputs included in this study's estimate at the Bypass outlet, this effect should be minor since, on average, the combined Hg supply of these downstream inputs is ~3 kg per year, according to our study. Comparisons between our estimates and others can be found in the Supplemental Material.

A few other points are worth mentioning. Estimates of Hg flux and storage presented herein are based on surface grab samples of unfiltered water. It is well known for cases where suspended sediment is eroded from a bed surface that its concentration declines exponentially with increasing distance from the bed, so that the surface waters have the lowest concentrations of sediment (Rouse, 1937). It is also well appreciated that Hg tends to adsorb to fine sediment particles (Maurice-Bourgoin et al., 2002) at concentrations far higher than is found in water solution. Given these factors, it is likely that our estimates of Hg flux and storage for the Yolo Bypass are underestimates of actual values.

The estimated value of sediment storage in Table 6 is not significantly different from zero, suggesting a mass balance of suspended sediment in Yolo Bypass. However, Singer and Aalto (2009) documented extensive event-based deposition (several decimeters in depth) along the entrance to Yolo Bypass that is removed every few years by the managing agency to maintain flood conveyance. So it seems initially surprising that sediment flux entering the Bypass is balanced by flux out of the Bypass. But the aforementioned study also pointed out gully-like erosion of sediment deposits within Yolo Bypass (Singer and Aalto, 2009). It is possible that deposition along the bypass entrance and other parts of the Bypass is balanced by erosion of former flood-borne deposits. To follow this line of reasoning, net Hg storage results presented here suggest that newly arrived sediment at the Bypass entrances (dominated by Cache Creek and Fremont Weir spillage) is richer in Hg than sediments leaving the Bypass at its downstream end. In other words, there is local enrichment of sediments in Yolo Bypass with mercury. In essence, cleaner or processed Hg-laden sediments are being remobilized to the Bay-Delta and replaced with relatively contaminated ones. Instead, we suggest either that remobilized sediment within and leaving the Yolo Bypass is being mobilized from non-depositional (low Hg) areas or that remobilized sediment has already undergone some processing by sulfate-reducing bacteria, so that the lower Hg content of sediments exported from the Bypass indicate a net loss of Hg into the Yolo Bypass ecosystem. The latter explanation is more likely but further interdisciplinary work would be required to test this hypothesis.

6. Conclusions

We estimate a mass balance of Hg for Yolo Bypass that suggests this lowland floodway is an important compartment for the storage and processing of Hg. Since it is inundated for up to several months a year, Hg stored in this zone may be methylated during anoxic conditions generated by long floods or wet seasons when soil drainage is slow. Mass balance estimates presented here and substantial reworking of deposited sediment-adsorbed Hg identified in prior work (Singer and Aalto, 2009), indicate a source of bioavailable Hg to the Bay-Delta.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at doi:10.1016/j.scitotenv.2011.10.004.

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