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The Contribution of Rice Agriculture to Methylmercury in Surface Waters: A Review of Data from the Sacramento Valley, California

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Abstract

Methylmercury (MeHg) is a bioaccumulative pollutant produced in and exported from flooded soils, including those used for rice (*Oriza sativa* L.) production. Using unfiltered aqueous MeHg data from MeHg monitoring programs in the Sacramento River watershed from 1996 to 2007, we assessed the MeHg contribution from rice systems to the Sacramento River. Using a mixed-effects regression analysis, we compared MeHg concentrations in agricultural drainage water from rice-dominated regions (AgDrain) to MeHg concentrations in the Sacramento and Feather Rivers, both upstream and downstream of AgDrain inputs. We also calculated MeHg loads from AgDrains and the Sacramento and Feather Rivers. Seasonally, MeHg concentrations were higher during November through May than during June through October, but the differences varied by location. Relative to upstream, November through May AgDrain least-squares mean MeHg concentration (0.18 ng L^{-1}, range 0.15–0.23 ng L^{-1}) was 2.3-fold higher, while June through October AgDrain mean concentration (0.097 ng L^{-1}, range 0.6–1.6 ng L^{-1}) was not significantly different from upstream. June through October AgDrain MeHg loads contributed 10.7 to 14.8% of the total Sacramento River MeHg load. Missing flow data prevented calculation of the percent contribution of AgDrains in November through May. At sites where calculation was possible, November through May loads made up 70 to 90% of the total annual load. Elevated flow and MeHg concentration in November through May both contribute to the majority of the AgDrain MeHg load occurring during this period. Methylmercury reduction efforts should target elevated November through May MeHg concentrations in AgDrains. However, our findings suggest that the contribution and environmental impact of rice is an order of magnitude lower than previous studies in the California Yolo Bypass.

Core Ideas

- We studied the impact of rice production on MeHg at the watershed scale.
- MeHg concentration was elevated in agricultural drainage water during November through May.
- Watershed-scale MeHg loads were lower than expected based on field studies.

In anoxic soils, a portion of the inorganic mercury (Hg) pool can be methylated, predominantly by sulfate- and iron-reducing bacteria (Compeau and Bartha, 1985; Gilmour et al., 1992; Fleming et al., 2006; Kerin et al., 2006), forming methylmercury (MeHg). Methylmercury binds strongly to thiols in proteins (Ballatori, 2002) and is extremely toxic to organisms, causing neurological problems and decreased reproductive success (Crump and Trudeau, 2009). Environmental levels of MeHg as low as 0.1 ng L^{-1} in freshwater ecosystems can have negative effects on high trophic-level organisms via bioaccumulation and biomagnification (Rudd, 1995; Watras et al., 1998; Chan et al., 2003). Humans are exposed to MeHg primarily through the consumption of fish and other wildlife from Hg-contaminated environments, resulting in negative health effects (Chan et al., 2003). Mercury and MeHg contamination of surface water is widespread. In the United States, fish consumption advisories have been issued in all 50 states and one US territory, including 1.8 million km of river and 6.6 million ha of lake (USEPA, 2011).

Wetlands provide anoxic soil conditions in which Hg-methylating microbes thrive, resulting in elevated MeHg production and bioaccumulation in a variety of wetland types (Marvin-DiPasquale et al., 2003; Hall et al., 2008). Unlike many crops, rice (*Oriza sativa* L.) is grown in flooded fields that are effectively agricultural wetlands. Rice is grown on approximately 150 million ha globally (Czech and Parsons, 2002), comprising a substantial portion of the world's estimated 1.2 billion ha of wetlands (Finlayson et al., 1999) and serving as important wildlife habitat (Czech and Parsons, 2002). Seasonal wet–dry cycles and inputs of labile organic carbon from root exudates and rice straw promote MeHg production and bioaccumulation in rice fields (Windham-Myers et al., 2009, 2014a; Ackerman and Eagles-Smith, 2010; Ackerman et al., 2010; Rothenberg and Feng, 2012). Many studies have found that MeHg can accumulate in rice grain and that rice can be the primary route of MeHg exposure for people living in Hg-contaminated inland rice-growing

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Abbreviations: AgDrain, agricultural drainage; AgDrain-E, drainage from agricultural areas east of the Sacramento River, also known as Sacramento Slough; AgDrain-W, drainage from agricultural areas west of the Sacramento River, also known as the Colusa Basin Drain; AgDrain-$W_{outfall}$, flows from AgDrain-W that enter the Sacramento River via the Knight's Landing Outfall Gates; AgDrain-$W_{diversion}$, flows diverted from AgDrain-W to the Yolo Bypass via Ridge Cut Slough; LSM, least-squares mean; MeHg, methylmercury.

areas (e.g., Feng et al., 2008; Meng et al., 2010, 2011; Zhu et al., 2015). In contrast, few studies have investigated export of MeHg in rice-field drainage water and its impact on downstream ecosystems, with the exception of studies in the California Yolo Bypass (Windham-Myers et al., 2014a). While it is clear that rice fields produce and export MeHg (Alpers et al., 2014), their effect has not been evaluated at a watershed scale.

MeHg concentration in stream channels and exposure to biota in those channels result from the interaction of sources, sinks, and transport processes in the watershed (Bradley et al., 2011). Though floodplains may also produce MeHg when temporarily flooded (Singer et al., 2016), at a watershed scale, the best predictor of MeHg concentration in a water body is the percent wetland cover upstream (Krabbenhoft et al., 1999). Methylmercury production may be decoupled from export (Bachand et al., 2014), and processes such as photodemethylation in open water (Seller et al., 1996) and particle settling can remove MeHg from the water column. As a result, MeHg concentrations can be lower than would be predicted from upstream MeHg inputs (Bradley et al., 2011).

The Sacramento River watershed (1.7 million ha) in California provides a good opportunity to study how rice contributes to watershed-scale MeHg concentrations and loads. Rice is grown on more than 240,000 ha and is the main crop in the low-lying central drainage area of the valley (USDA–NASS, 2016). Naturally enriched Hg conditions, plus a legacy of Hg and gold mining (which used elemental mercury to amalgamate the gold) in the mountains surrounding the valley, have resulted in elevated Hg concentrations in river sediments, water, and fish (Domagalski, 1998, 2001; Davis et al., 2008; Springborn et al., 2011; Singer et al., 2013; Domagalski et al., 2016; Donovan et al., 2016a, 2016b). Elevated MeHg concentrations in fish have led to fish consumption advisories and enactment of a Total Maximum Daily Load to reduce MeHg loads into the Sacramento–San Joaquin Delta (Delta Mercury Control Program, 2010).

The overall objective of this study was to evaluate the contribution from rice-producing areas to MeHg loads in the lower Sacramento River by compiling and analyzing historic surface water MeHg concentration and flow data. We evaluated MeHg concentrations and loads in major agricultural drains from rice-dominated areas and at mainstem sampling points in the Sacramento and Feather Rivers, both upstream and downstream of agricultural drain inputs. Furthermore, we evaluated seasonal trends to identify the time of year in which MeHg loads exported from rice fields may be of greatest concern, and long-term trends to see if changes in postharvest rice straw management practices have influenced MeHg concentrations.

Materials and Methods

The Sacramento Valley covers 1.7 million ha and is situated in the low-lying area between the Coast Ranges and Sierra Nevada of California. It is 240 km long from north to south and ranges from 32 km wide in the north to 72 km wide in the south (Bennett et al., 2011). The Sacramento Valley has a Mediterranean climate, with typically hot, dry conditions during April to September and a cool, rainy season from October through March

The Sacramento River watershed has both natural and anthropogenic Hg sources. Mineral springs (Youngs, 1994) and erodible surficial soils (US Bureau of Mines, 1965) in the Coast Range to the west and Cascade Mountains to the northeast are naturally Hg enriched. Mercury mining occurred in the Coast Range to the west (Jasinksi, 1995). Between 1846 and 1981, an estimated 34.5 million kg of Hg was released into the environment, partially as Hg vapor during ore processing (Churchill, 2000). Most of the Hg mining occurred in the Cache Creek watershed and farther south; thus, runoff from mine wastes in these areas enters the Yolo Bypass downstream of the current study area (Domagalski, 1998). Beginning in 1848, elemental Hg was used by the gold mining industry in the Klamath and Sierra Nevada mountain ranges, drained by the upper Sacramento River and Feather River, respectively (Domagalski, 1998; Churchill, 2000). Combined Hg losses from gold mining totaled 5.8 million kg, with 97% of losses occurring before 1935 and 80 to 90% of losses occurring in the Sierra Nevada (Churchill, 2000). While Hg losses were substantial, it is unclear to what degree rice-growing areas in the central Sacramento Valley have been contaminated with Hg. Reservoirs limit downstream transport of MeHg and total Hg (Slotton et al., 1995). Domagalski (1998) measured total Hg in riverbed sediments: sites in the central Sacramento River had 40 to 70 ng Hg g^{-1}, while sites within and downstream of the Feather River had 140 to 370 ng Hg g^{-1}. To our knowledge, there are no published Hg concentration data for Sacramento Valley rice soils. Atmospheric deposition has not been measured directly for the Sacramento Valley. However, the San Francisco Estuary to the south received 19 μg Hg m^{-2} yr^{-1} as dry deposition and 4.2 μg Hg m^{-2} yr^{-1} as wet deposition in 1999 to 2000 (Tsai and Hoenicke, 2001).

Within the Sacramento Valley, there are two main waterways: The Sacramento River on the western side of the valley and the Feather River on the east. Additional watershed inflows occur along the eastern and western edge of the valley, including the Yuba and Bear rivers. Both of the main rivers flow from north to south and converge near the southern end of the Valley's primary rice-growing region, north of the city of Sacramento (Fig. 1). After confluence with the Feather River, the Sacramento is joined by the American River and continues south to the Sacramento–San Joaquin Delta. Surface water hydrology is highly manipulated, with a number of constructed waterways used to direct irrigation water (supply and drainage) and to mitigate flooding.

Drainage from rice agriculture (hereafter, "rice"), and to a lesser extent nonagricultural wetlands, makes up the majority of water in agricultural drainage canals. Rice is planted during late April through May. Most fields are water-seeded, whereby pregerminated rice seed is dropped from an airplane onto flooded fields (Linquist et al., 2015). Water management during the first month of the growing season is highly variable, with the water level and outflow adjusted for seedling establishment and herbicide application. Later in the growing season, maintenance flow is established where irrigation water is applied to rice in excess of evapotranspiration demand in part to limit salinity build up (Grattan et al., 2002; Scardaci et al., 2002). Excess water is exported from rice fields as drainage water. Fields are drained in August or September, 3 to 4 wk before harvest. Irrigation of other crops in the watershed is managed to match evapotranspiration demand closely so that little or no drainage water is produced. Drainage water from managed wetlands is also present; however, wetland cover within the drainage area is approximately one fourth that of rice (Fig. 1) (US Fish and Wildlife Service, 2016). Water-export data from wetlands managed for wildlife is limited, but rates of outflow

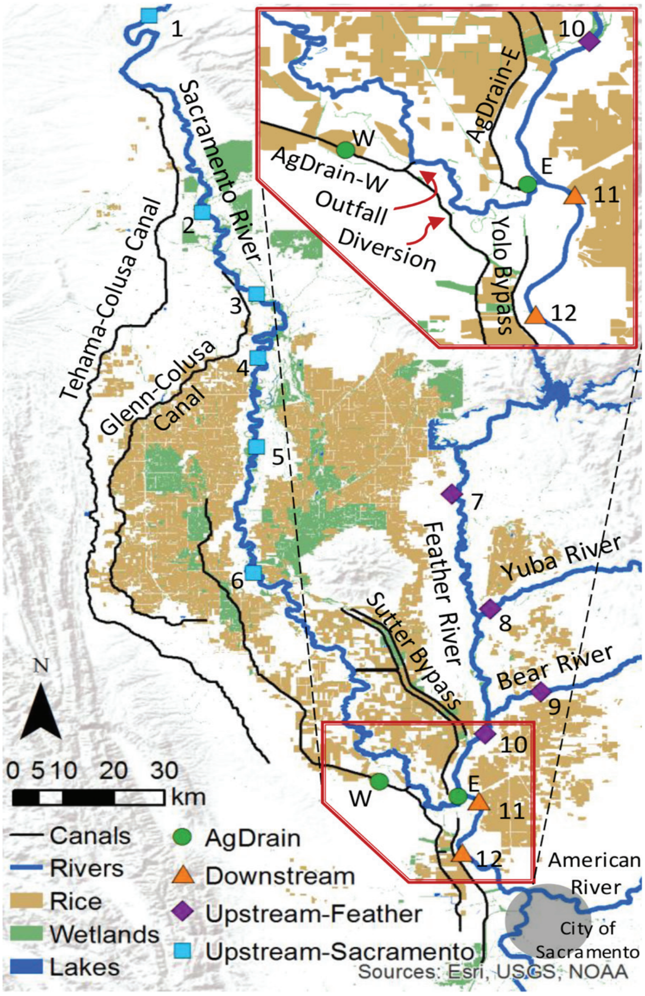

Fig. 1. Map of the study area. Site labels correspond to site identification field in Table 1. Rice area was obtained from the California Department of Water Resources (2013). Wetland area is from the National Wetland Inventory (US Fish and Wildlife Service, 2016). Water ways are depicted according to the National Hydrography Dataset (USDA–NRCS et al., 2013). AgDrain, agricultural drainage.

are also managed to prevent salinity build up (this management objective is similar to those of rice growers). Periods of high water export from managed wetlands are the drawdown of seasonal wetlands (March–May) and areas flooded during the summer (July–August) (S. Emmons, personal communication, 2016).

Drainage water from agricultural fields and other wetlands is collected by two major agricultural drains, hereafter referred to as "AgDrain-E" and "AgDrain-W." AgDrain-E collects drainage water from the eastern valley area (e.g., Sutter Bypass) between the Sacramento and Feather Rivers and discharges into the Sacramento River immediately upstream of the confluence of the Sacramento and Feather Rivers (Fig. 1). Low flows during summer in AgDrain-E are made up primarily of rice drainage water; however, during high winter flows, large volumes of floodwater from the Sutter Bypass floodplain are mixed with rice drainage water. AgDrain-W collects drainage water from the area west of the Sacramento River. Flows from AgDrain-W are divided immediately downstream of the sampling location, with some water directed into the Sacramento River (AgDrain-W$_{\text{outfall}}$) and the remainder diverted to the Yolo Bypass floodplain (AgDrain-W$_{\text{diversion}}$) (Fig. 1). Drainage from AgDrain-E and the Feather River enter the Sacramento River 25 km downstream of AgDrain-W$_{\text{outfall}}$.

We categorized sampling sites into "site groups" based on their location with respect to the two main AgDrains (Fig. 1): Sampling sites located at the mouths of AgDrain-E and AgDrain-W are referred to as "AgDrain" sites. "Upstream-Sacramento" and "Upstream-Feather" sites were located on the Sacramento and Feather Rivers, respectively, upstream of where AgDrains empty into the Sacramento River. Upstream sites serve as control sites because they represent river water that has been minimally influenced by rice. All Upstream-Sacramento sites were upstream of rice drainage water inputs, while Upstream-Feather site 5 may have been influenced by rice drainage water from rice grown east of the Feather River and north of the Bear River. Upstream sites also represent irrigation source water: sites 1 and 3 on the Sacramento River were located close to the Tehama–Colusa and the Glenn–Colusa Canals, respectively, which are major agricultural diversions that provide irrigation water to the western side of the Sacramento Valley. "Downstream" sites were located downstream of the confluence of the Sacramento River, Feather Riverm and AgDrains, but upstream of the Sacramento urban area and the confluence with the American River (Fig. 1). Downstream sites represent Sacramento River water that has been influenced by Sacramento Valley rice drainage.

Data Sources

We used data from programs that monitored aqueous, unfiltered MeHg concentrations in the Sacramento River watershed from 1996 to 2007. Programs include the USGS National Water Quality Assessment Program (Domagalski et al., 2000) dataset from 1996 to 1998 (I); three programs conducted by the Sacramento River Watershed Program (2005, 2008), including data from 2000 to 2003 (II), 2004 (IV), and 2006 to 2007 (V); and one dataset from the CALFED Bay-Delta Program (Foe et al., 2008), including data from 2003 to 2006 (III) (Supplemental Table S1). Samples were collected either as grab samples or as depth-integrated samples at the midpoint of the channel cross-section. Detection limits ranged from 0.0114 to 0.0234 ng L^{-1} (Supplemental Table S1). Methylmercury concentrations were below the detection limit in 1.6% of samples. Nondetects were equally distributed among Upstream-Sacramento, Upstream-Feather, AgDrain, and downstream samples (n = 4, 2, 3, and 2, respectively). If the lab value was reported, it was used in the analysis; otherwise undetected MeHg concentrations were treated as 0.5 × detection limit in data analysis. Sampling sites varied among programs, and sampling frequency ranged from monthly to quarterly. A number of sites were omitted from the analysis due to lack of relevance to the study questions or lack of data. These sites included small, seasonal creeks, sites on the Sacramento River distantly upstream or downstream of rice, and sites with fewer than 10 samples. For sites included in this analysis, the number of samples available by site and program is shown in Table 1.

Both precipitation and burned rice area were tested as possible explanatory variables. Precipitation from storm events may influence MeHg concentration by dilution or flushing MeHg from areas where it is produced (Balogh et al., 2006). Precipitation data were obtained from the California Irrigation Management Information System (California Department of Water Resources, 2014b). Data from two weather stations that represented the study area within the watershed were averaged for analysis (station identifications 12 and 3). Precipitation totals from 3, 5, and

Table 1. The number of methylmercury (MeHg) samples collected by each sampling program at sites used in this analysis. AgDrain, agricultural drainage water from rice-dominated regions.

Site	Site ID†	Samples in each program‡					Total number of samples
		I	II	III	IV	V	
Upstream-Sacramento							
Sacramento River above Bend Bridge	1	–	17	30	4	18	69
Sacramento River at Woodson Bridge	2	–	–	31	–	–	31
Sacramento River at Hamilton City	3	–	17	31	4	18	70
Sacramento River at Ord Ferry Bridge	4	–	–	22	–	–	22
Sacramento River at Butte City	5	–	–	30	–	–	30
Sacramento River at Colusa	6	29	17	30	4	18	98
Upstream-Feather							
Feather River at Gridley	7	–	–	31	–	–	31
Yuba River at Marysville	8	–	18	31	4	18	71
Bear River below Wheatland	9	–	–	30	–	–	30
Feather River at Nicolaus	10	–	18	31	4	18	71
AgDrains							
Colusa Basin Drain at Knights Landing	W	25	16	31	4	18	94
Sacramento Slough at Karnack	E	23	16	28	4	18	89
Downstream							
Sacramento River at Verona	11	27	–	—	–	–	27
Sacramento River at Veteran's Bridge	12	–	–	—	–	17	17

† Site locations are denoted in Fig. 1 using site identification (Site ID).

‡ See Supplemental Table S1 for more information about the sampling programs.

7 d before MeHg sample collection were tested as predictors of MeHg concentration. Burning of rice straw removes carbon from fields, possibly influencing microbial activity (Windham-Myers et al., 2014b; Zhu et al., 2015). Burned rice area data were obtained from the California Rice Commission (2016).

Flow data for load estimations were obtained from the California Data Exchange Center, the California Water Data Library (California Department of Water Resources, 2014a, 2016), and the National Water Information System (USGS, 2016) databases (Supplemental Table S2). Mean daily flow was used when available; otherwise, hourly or quarter-hourly flow measurements were averaged to obtain mean daily flow. Missing flow data prevented load calculation at key sites, including AgDrain-$W_{diversion}$, AgDrain-$W_{outfall}$ during the 2005 water year (October 2004–September 2005), and AgDrain-E during winter.

Data Analysis

Season was included in the analysis as a categorical variable because seasonal variation occurs in rice management as well as climate. We wanted to identify times of the year when concentrations were elevated to inform future studies. We plotted MeHg concentration data from all years by day and month and defined two seasons based on periods with relatively higher or lower MeHg concentrations.

Linear mixed-effects regression analysis was used to assess differences in MeHg concentration, allowing us to account for the fact that sites were measured repeatedly but not always concurrently. Site and year were used as random effects in the model. Fixed effects tested included season (June–October or November–May), site group (Upstream-Sacramento, Upstream-Feather, AgDrain, or downstream), time (as a continuous variable), fraction of rice area burned, and precipitation, as well as site group × season, site group × time, and site group × fraction of rice area burned interaction terms. Models were fitted using the lme4 package (Bates et al., 2015) in R (R Core Team, 2014). We selected the final model using backward stepwise regression. Beginning with the fullest model, first random effects, then fixed effects, were dropped stepwise if nonsignificant ($p > 0.05$). The p-values for random effects were calculated using likelihood ratio tests, whereas significance of fixed effects was determined using F tests with denominator degrees of freedom, calculated by Satterthwaite's approximation. Post hoc Tukey-corrected differences of least-squares means (LSM) were used to identify significant differences among categorical fixed effects; differences significantly different from zero ($p < 0.05$) were considered significant. Model selection and tests for differences between fixed effects were done using the lmerTest package (Kuznetsova et al., 2016). Assumptions of normalcy and homogeneity of variance were assessed using standard diagnostic plots. A natural log transformation was used on MeHg, but not on other variables. This transformation successfully corrected for heteroscedasticity and normalized the residuals of the model.

Beale's ratio estimator was used to calculate MeHg loads (Beale, 1962; Richards, 1998) because it was found to be unbiased and accurate in a number of studies comparing load calculation methods (Dolan et al., 1981; Beirman et al., 1988). The mean daily load (concentration × flow) for days on which concentration was measured was multiplied by the ratio of average flow during the period of interest to the average flow on days when concentration was measured, then multiplied by a bias correction factor (Supplemental Eq. S1 and S2).

At each site, loads were calculated separately for monitoring programs I, II, and V. Programs III and IV were combined because samples were collected concurrently and program IV did not have enough data for a separate load calculation. Additionally, loads were calculated using data from all programs combined. Separate calculations were done for the November through May and June through October seasons.

Upstream-Sacramento and Upstream-Feather loads were calculated at sites 6 and 10, respectively. These sites were located immediately upstream of AgDrain inputs but downstream of other known MeHg inputs, including the Bear and Yuba Rivers (Fig. 1). Additionally, these sites had the most complete datasets (Table 1).

Results and Discussion

A total of 681 unfiltered water MeHg concentration measurements were compiled from the various programs and used in this study. Only three sites were sampled by all five programs, two of which were the main agricultural drainage canals that discharge directly into the Sacramento River (AgDrain-E and AgDrain-W), while the third was Upstream-Sacramento site 6.

Methylmercury concentrations ranged from below the detection limits (0.02–0.0114) to 1.97 ng L^{-1}, and only three samples in the dataset were above 1 ng L^{-1} (Fig. 2). Waterways with MeHg concentrations above 0.1 ng L^{-1} are potentially negatively impacted (Rudd, 1995), and 57% of samples in this study had concentrations below this level. Twenty percent of samples were below 0.06 ng L^{-1}, the regulatory standard for the Sacramento–San Joaquin Delta.

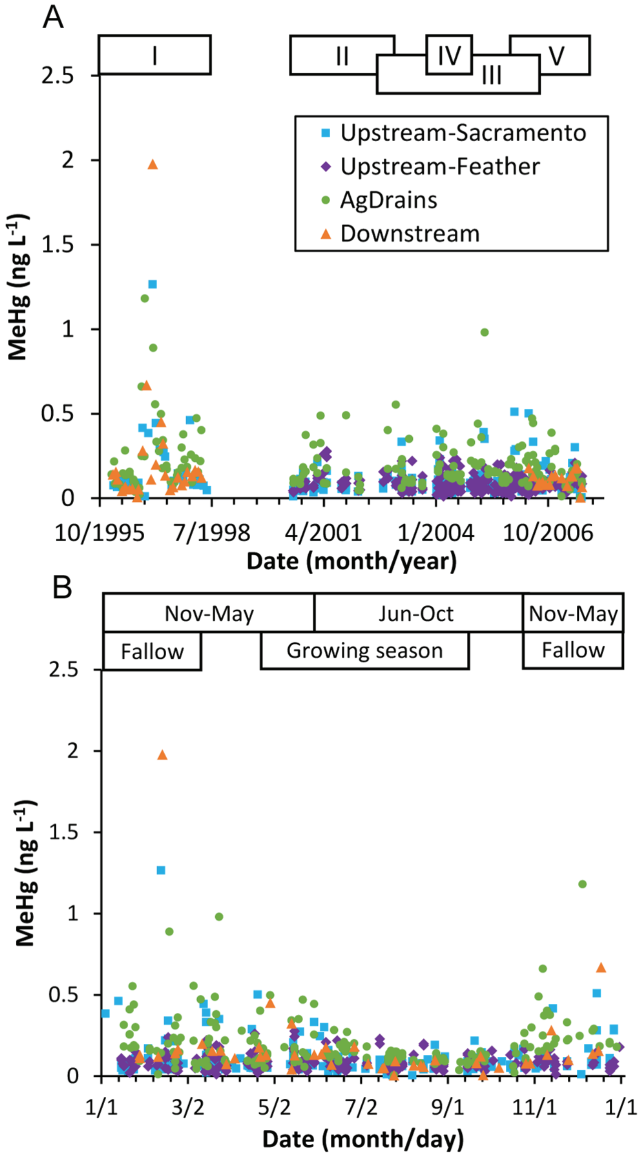

Fig. 2. (A) Time series of methylmercury (MeHg) concentrations throughout the study period. Bars at the top indicate when each sampling program occurred (Supplemental Table S1). (B) MeHg concentrations plotted by day of year. The figure shows data from the whole study period. Bars at top show the rice-growing season and winter fallow, and June to October and November to May seasons as defined in this study.

Methylmercury Concentrations

Despite changes in rice straw management during the study period, mixed-effects regression analysis did not detect a significant change in MeHg concentration over time (Fig. 2A), and there was no interaction between time and location (The results of variable selection for the mixed-effects model are shown in Supplemental Table S3). During the study period, straw removal practices changed as a result of the Connelly–Areias–Chandler Rice Straw Burning Reduction Act of 1991 (California Environmental Protection Agency, 1991), which mandated that rice straw burning in the Sacramento Valley be phased down starting in 1992 and be allowed only under specified conditions for disease control by 2001. The practice of burning rice straw was replaced by incorporating rice straw into soil during winter, followed by flooding to facilitate its decomposition (Linquist et al., 2006). The percentage of area where rice straw was incorporated during winter increased from <15% in 1992 to >80% by 2001 (California Air Resources Board, 2003). Rice straw is a source of labile organic carbon that may increase Hg methylation in rice fields (Windham-Myers et al., 2014a; Zhu et al., 2015); thus, an increase in AgDrain MeHg concentration over time was expected due to this shift from burning to incorporation and flooding during the study period. However, the fraction of rice area burned did not significantly affect MeHg concentration and did not interact with site group. This result is consistent with a recent controlled, replicated experiment testing the effect of straw removal from rice fields (Eagles-Smith et al., 2014). Other sources of variation in MeHg concentration may obscure any effect of rice straw management on MeHg concentration at the valley scale.

We did not detect a significant effect of precipitation on MeHg concentrations. Elevated MeHg concentrations were found in early 1997, 1 mo after a rain on snow event that caused major flooding throughout the region (samples were not collected during the event) (Fig. 2A). Although Balogh et al. (2006) reported elevated MeHg concentrations during high flows, concentration changes in response to storm events can be complex, depending on the transported material, as well as watershed characteristics (Richards and Holloway, 1987). The sampling frequency in this dataset may be too low to detect any MeHg concentration changes in response to precipitation.

Plotting all data by month and day revealed a seasonal pattern in which MeHg concentrations were consistently lower from June to October (range: less than detection limit to 0.3 ng L^{-1}), while concentrations from November through May were higher and more variable (less than detection limit to 1.98 ng L^{-1}) (Fig. 2B). We hereafter refer to these seasons as June to October (153 d) and November to May (212 d), respectively. Mixed effects regression revealed a strong seasonal effect, with November to May concentrations 65% higher than June to October concentrations ($F_{1,644} = 62.3, p < 0.001$); however, there was an interaction between site group and season ($F_{3,634} = 11.8, p < 0.001$; Fig. 3). The final model includes these significant fixed effects, as well as site group because it was part of a significant interaction (Supplemental Table S3). Significant random effects for both site and year were also included.

With respect to water management and inputs into these systems, June to October includes runoff from rice fields in the form of maintenance flow and the final drain in preparation for harvest (August). November to May corresponds to the flooding of rice and managed wetlands (October and November) and subsequent runoff and final drains from rice (February) and natural wetlands (April and May). For the years studied, 92 ± 6% of the annual rainfall occurred in November to May. During high rainfall years or large storm events, regional flooding occurs, resulting in runoff from other agricultural fields (and urban areas), and river water may be diverted into bypasses, both resulting in comingling of water sources, particularly for AgDrain-E.

Methylmercury concentrations did not differ between Upstream-Sacramento and Upstream-Feather in November to May (LSM = 0.079 and 0.077 ng L^{-1}, respectively; $p = 0.3$) or June to October (0.060 and 0.075 ng L^{-1}, respectively; $p = 0.9$) (Fig. 3), indicating that rice on the east and west sides of the Valley receive similar MeHg inputs in irrigation water. Upstream-Feather MeHg concentrations did not differ between seasons ($p = 0.8$) (Fig. 3), but Upstream-Sacramento had slightly but significantly higher MeHg concentrations during November to May ($p = 0.002$).

During June to October, AgDrain MeHg concentrations (LSM = 0.097 ng L^{-1}) appeared elevated compared with other site groups; however, there were no significant differences ($p > 0.05$) in MeHg concentrations among site groups. In contrast, November to May AgDrain MeHg concentrations (LSM = 0.18 ng L^{-1}; $p < 0.05$) were significantly higher than November to May Upstream-Sacramento and Upstream-Feather concentrations. Previous studies also reported elevated MeHg concentrations in rice drainage water relative to irrigation water (Alpers et al., 2014; Zhao et al., 2016).

Similar to AgDrains, downstream MeHg concentrations were significantly higher than upstream during November to May, and there was no significant difference between AgDrain and downstream MeHg concentrations during either June to October or November to May. However, the flows and loads of AgDrain, upstream, and downstream sites must be considered when determining the degree to which AgDrains were influencing downstream MeHg concentrations.

In this study, Ag Drains exhibited a much stronger seasonal pattern than upstream sites, suggesting that rice and wetlands influence concentrations. Studies in the Yolo Bypass reported a similar seasonal pattern (Bachand et al., 2014; Marvin-DiPasquale et al., 2014; Windham-Myers et al., 2014b). While MeHg is produced in rice fields throughout the year (Marvin-DiPasquale et al., 2014), transpiration during the growing season results in the downward movement of surface water, causing MeHg to be transported into and stored in the rootzone (Bachand et al., 2014). The absence of transpiration during the fallow season allows MeHg to be released into surface water via diffusion (Bachand et al., 2014). Additionally, rice plants can store a significant amount of MeHg during the growing season (Windham-Myers et al., 2014b). Other studies have reported that MeHg stored in dry sediment may be quickly mobilized into the water column on flooding (Kelly et al., 1997; Rumbold and Fink, 2006; Alpers et al., 2014) or proposed that methylation may occur shortly after inundation (Singer et al., 2016). Thus, the early growing season drainage events, in addition to the fallow season, may be periods of MeHg export from rice. (e.g., Fig. 2B).

Methylmercury Loads

Using all available data, June to October MeHg loads for AgDrain-$W_{outfall}$, AgDrain-E, Upstream-Sacramento, and Upstream-Feather were (mean ± SD) 0.14 ± 0.03, 0.23 ± 0.03, 2.2 ± 0.3, and 0.88 ± 0.3 g d^{-1}, respectively (Fig. 4). The downstream MeHg load is expected to be the sum of all upstream

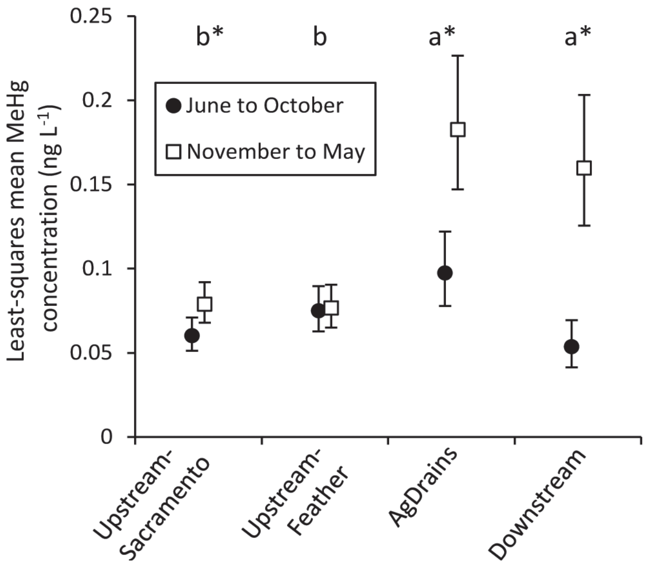

Fig. 3. Least-squares mean ± SE of methylmercury (MeHg) concentrations based on mixed-effects modeling for site groups and seasons. There is a significant interaction between site group and season. Site groups that were not significantly different during November to May have the same letter. Site groups did not differ significantly during June to October. Asterisks (*) indicate a significant difference between seasons at each site group. See Supplemental Table S4 for *p*-values of pairwise comparisons. AgDrain, agricultural drainage.

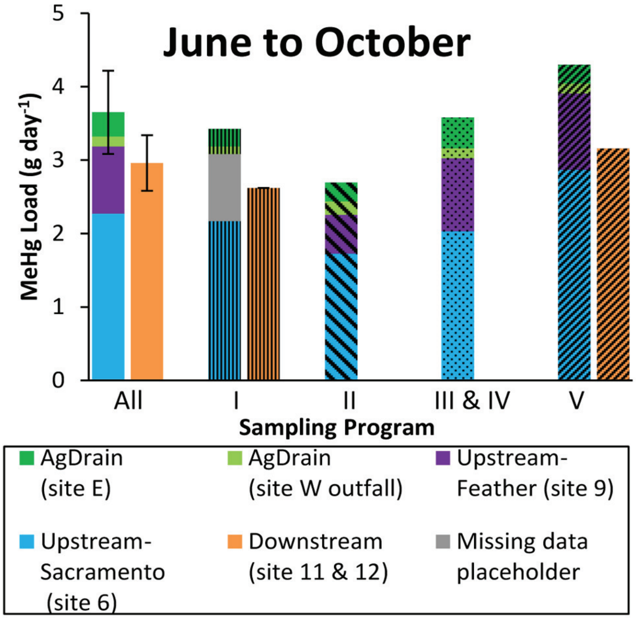

Fig. 4. June to October methylmercury (MeHg) loads by sampling program (indicated by *x*-axis groups and shading) and site (indicated by color in legend). Loads from tributary sources are shown as a stacked bar and are expected to equal downstream load. "All" represents loads calculated using data from all sampling programs. Error bars represent the standard deviation of sampling program estimates. Data was not available for Upstream-Feather, so a placeholder (gray) was used with the value from "All." Color and shading is consistent with Fig. 5. AgDrain, agricultural drainage.

tributary loads: Upstream-Sacramento, Upstream-Feather, AgDrain-W$_{\text{outfall}}$, and AgDrain-E. The downstream load (2.5 ± 0.6 g d^{-1}) was 73% of the total June to October tributary load (3.4 ± 0.4 g d^{-1}). Although the downstream load was within the range of variability of the tributary load, the downstream load was less than the tributary load in both programs where it was measured (I and V), possibly suggesting MeHg loss. June to October load estimates differed among datasets by less than a factor of two (Fig. 4). AgDrain loads accounted for 10.7% of the total June to October upstream tributary load, or 14.8% of the downstream load.

November to May loads made up 70 to 90% of the total annual load (within sites), and estimates differed among programs by approximately a factor of four (Fig. 5). November to May MeHg loads for AgDrain-W$_{\text{outfall}}$, Upstream-Sacramento, Upstream-Feather, and downstream were 0.22 ± 0.1, 7.3 ± 4, 1.7 ± 0.8, and 24 ± 17 g d^{-1}, respectively. November to May loads for AgDrain-E were not possible to estimate due to gaps in flow data and floodwater being diverted into and out of the river at a number of locations.

Sampling frequency in this study was lower than recommended for accurate load calculation (Dolan et al., 1981). Low sampling frequency is problematic because it reduces the likelihood of accurately capturing loads during storm events. This is particularly evident for November to May downstream loads, where removal of one elevated sample concentration (1.98 ng L^{-1}, 13 Feb. 1997, see Fig. 2A) decreases the downstream load estimate by 40%. This sample was taken more than a month after a major flood event, during an extended period of high flow, and elevated MeHg concentrations were also observed at other sites (quality control data did not suggest this was a result of contamination [Domagalski, 1998]). It is possible that higher concentrations occurred during the peak of this or other storm events, but samples were not collected then. Therefore, uncertainty surrounding storm event loads limits our confidence in November to May loads. There are typically no storm events and MeHg concentrations are less variable during June to October (Fig. 2B), so the variation in MeHg concentration can be captured using fewer samples; thus, the sampling frequency is likely adequate for June to October loads.

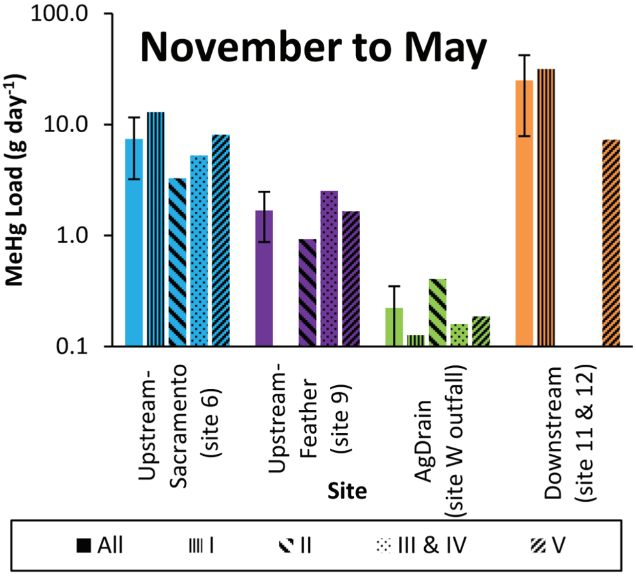

Fig. 5. November to May methylmercury (MeHg) loads by site (indicated by color and *x*-axis groups) and sampling program (indicated by shading in legend). Note the log scale *y*-axis. "All" represents loads calculated using data from all sampling programs. Error bars represent standard deviation of sampling program estimates. Color and shading is consistent with Fig. 4. AgDrain, agricultural drainage.

The true MeHg load at a site should be expected to show considerable interannual variation, with larger loads occurring in years with high flow (Hill, 1986). It is important to consider whether the years measured in this study are representative of historical flow regimes. Hill (1986) recommends using data from 6 to 7 yr to obtain robust estimates of average annual loads. With the exception of downstream, loads presented here were based on 7 to 9 yr of data, suggesting that this study adequately captures the interannual load variation at these sites. Flow during the study period was greater than averages of historical average flows at all sites except AgDrain-W in June to October (Supplemental Table S5 and Supplemental Fig. S1), suggesting that loads during this study may have been higher than historical average loads.

Full accounting of rice field loads requires the total MeHg load from AgDrain-W, which is the sum of AgDrain-W$_{\text{outfall}}$ (reported above) and AgDrain-W$_{\text{diversion}}$ (not calculated because of missing flow data). However, AgDrain-W$_{\text{diversion}}$ flow data from 2007 and 2012 shows that AgDrain-W$_{\text{outfall}}$ carried 36 ± 9% of November to May flows and 73 ± 7% of June to October flows from AgDrain-W. Through discussions with irrigation district managers, we determined that management of AgDrain-W flows have not changed since the beginning of the study period (Bair, personal communication, 2016). Based on the fraction of flows carried by AgDrain-W$_{\text{outfall}}$, the total AgDrain-W load was 0.61 ± 0.17 g d^{-1} in November to May and 0.19 ± 0.02 g d^{-1} in June to October.

Windham-Myers et al. (2014a) reported MeHg loads exported from rice fields in the Yolo Bypass. If these loads are extrapolated to other rice-growing regions and multiplied by the total area of rice in the Sacramento Valley (240,000 ha), the predicted MeHg loads are 12± 7 and 26 ± 24 g d^{-1} in June to October and November to May, respectively—an order of magnitude higher than the results of this study. This substantial difference may result from two potential sources. First, there are known site differences between the field-scale study site in the Yolo Bypass and typical Sacramento Valley rice-growing areas. For example, MeHg concentrations in irrigation source water were an order of magnitude higher in the Yolo Bypass than in irrigation source water (Upstream-Sacramento and Upstream-Feather) in this study. The Yolo Bypass is known to accumulate Hg-laden sediment during storm events (Springborn et al., 2011), while much of the rice land is not in the path of storm flows that would deliver Hg-laden sediment. Secondly, differences may result from scale-dependent factors. Bradley et al. (2011) found that MeHg loads exported from wetland sources overestimated the watershed-scale load because MeHg was lost through sink processes (including photo- and microbial demethylation and particle settling) during transport. Windham-Myers et al. (2014a) showed that rice fields are net MeHg sources; however, aqueous MeHg exported from rice fields in Sacramento Valley must be transported through a network of canals before it reaches the Sacramento River. Future research should seek to determine the degree to which MeHg is lost during canal transport and to quantify MeHg budgets for Sacramento Valley rice fields.

Rice Field Drainage Contribution

Elevated November to May downstream concentrations and the large November to May downstream load relative to the upstream tributary loads suggest that the uncalculated November to May AgDrain-E was considerably larger than AgDrain-W. However, AgDrain-E carries a mixture of floodwater and rice drainage water, as well as drainage from wetlands. Determining the MeHg load from rice requires further research on the effect of floodplains on MeHg and the contribution of managed wetlands.

It is misleading to consider only MeHg loads exported from rice because fields receive MeHg in irrigation water and atmospheric sources. Irrigation source water (upstream) had a MeHg concentration of 60 to 70% of that of AgDrain water (Fig. 3). Due to evapotranspiration and percolation losses, the volume of drainage water is $\leq$40% of irrigation water applied to a rice field during the growing season (Linquist et al., 2015). Thus, growing season increases in MeHg concentrations between irrigation and drainage water during the growing season were of a similar magnitude to that expected from evapoconcentration alone. The June to October AgDrain MeHg load contributed to the Sacramento River may be similar to the MeHg load diverted from the river for irrigation. During November to May (the fallow season), upstream MeHg concentrations were 43% of AgDrain, while drainage water exports are expected to be a larger fraction of irrigation water applied (no transpiration is expected in fallow rice fields), resulting in increased MeHg exports relative to imports. This is consistent with Bachand et al. (2014), who found that rice fields might store MeHg during the growing season but release it during the winter fallow. Wet and dry atmospheric deposition may be important sources of MeHg and total Hg (Munthe et al., 1995, Conaway et al., 2010) and have been shown to influence runoff fluxes from catchments (Hultberg and Munthe, 1995). However, atmospheric deposition data is not available for the Sacramento Valley. Deposition studies would help quantify the degree to which rice fields are MeHg sources or sinks.

Elevated November to May AgDrain MeHg concentrations and November to May AgDrain-W loads being fourfold higher than June to October loads both indicate that November to May is the period of higher concern for MeHg export from rice fields. Without knowledge of the full annual cycle of MeHg production in and export from a system, studies risk missing important periods of MeHg export. If the environmental management objective is to reduce annual loads of MeHg, control efforts should focus on the November to May period, when MeHg concentrations in AgDrains are elevated. Finally, this study indicates that care should be taken when extrapolating the impact of rice production on MeHg in surface water at the field to valley scale. While the seasonal patterns of MeHg concentrations and exports were similar to those observed at the field scale, there were substantial differences in the magnitudes of concentrations and loads.

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