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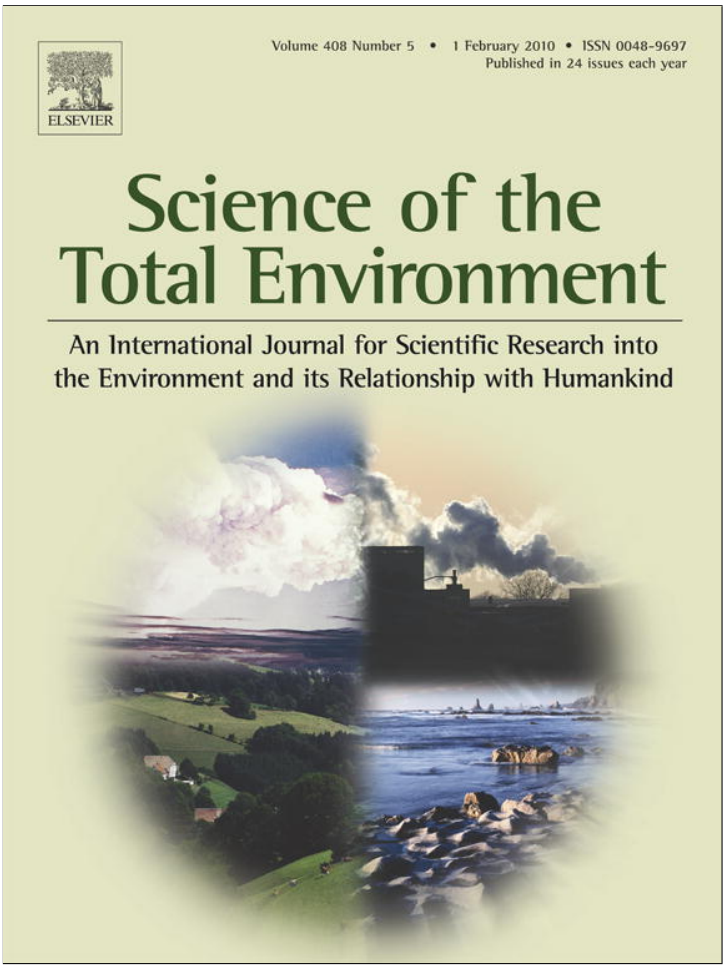

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Temporal and spatial distributions of sediment mercury at salt pond wetland restoration sites, San Francisco Bay, CA, USA

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ABSTRACT

Decommissioned agricultural salt ponds within south San Francisco Bay, California, are in the process of being converted to habitat for the benefit of wildlife as well as water management needs and recreation. Little is known of baseline levels of contaminants in these ponds, particularly mercury (Hg), which has a well established legacy in the Bay. In this study we described spatial and short-term temporal variations in sediment Hg species concentrations within and among the Alviso and Eden Landing salt ponds in the southern region of San Francisco Bay. We determined total Hg (Hg_t) and methylmercury (MeHg) in the top 5 cm of sediment of most ponds in order to establish baseline conditions prior to restoration, sediment Hg_t concentrations in a subset of these ponds after commencement of restoration, and variation in MeHg concentrations relative to sediment Hg_t, pH, and total Fe concentrations and water depth and salinity in the subset of Alviso ponds. Inter-pond differences were greatest within the Alviso pond complex, where sediment Hg_t concentrations averaged (arithmetic mean) 0.74 μg/g pre and 1.03 μg/g post-restoration activity compared to 0.11 μg/g pre and post at Eden Landing ponds. Sediment Hg_t levels at Alviso were fairly stable temporally and spatially, whereas MeHg levels were variable relative to restoration activities across time and space. Mean (arithmetic) sediment MeHg concentrations increased (2.58 to 3.03 ng/g) in Alviso and decreased (2.20 to 1.03 ng/g) in Eden Landing restoration ponds during the study. Differences in MeHg levels were related to water depth and pH, but these relationships were not consistent between years or among ponds and were viewed with caution. Factors affecting MeHg levels in these ponds (and in general) are highly complex and require in-depth study to understand.

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1. Introduction

Restoration of human altered landscapes to historical habitat has been a major goal of resource and conservation agencies over the past several decades. Two of the largest wetland restoration projects in the United States are progressing in the Florida Everglades and more recently in the San Francisco Bay (Bay), California. More than 10,000 ha of decommissioned commercial salt ponds throughout the Bay have been identified for conversion from salt production to habitat that not only benefits wildlife but also water quality, flood control, and recreation (Takekawa et al., 2006). Natural resources associated with the pond complex in the southernmost Bay (hereafter, the Alviso ponds) are managed largely by the U.S. Fish & Wildlife Service (USFWS) while those located in south central Bay (hereafter, the Eden Landing ponds) are managed by the California Department of Fish and Game (CDFG) (Fig. 1). These agencies must address habitat and water quality issues that include reduction of salinity in these ponds, accomplished by direct or indirect dilution with waters from the Bay or its tributaries.

Sediment can sequester contaminants and therefore serves as a pathway for exposure to food webs. The Bay estuary in general is renowned for mercury (Hg) contamination particularly from historic Sierra Nevada gold mining in the North Bay and specifically Hg mining in the Guadalupe River watershed in the South Bay (Hornberger et al., 1999; Thomas et al., 2002; Conaway et al., 2004). The restoration of salt ponds to tidal marsh or seasonal mud flat habitat may result in mobilization of Hg due to, for example, wetting and drying or slough scour and sediment disturbance, and thus increase its availability and methylation (Marvin-DiPasquale and Cox, 2007).

Initial restoration activities at the Alviso and Eden Landing salt ponds involved increased water exchange to certain ponds via control gates or breached levees that opened directly to adjacent tidal tributaries or increased water flow between ponds; hereafter, restoration. Obvious changes in water or sediment qualities (e.g. salinity, pH) will result from water management changes. Changes in these or other qualities (e.g., sediment iron [Fe]) and Hg could influence MeHg production, fueled by effects on sulfur or iron-reducing bacteria (Compeau and Bartha, 1985; Fleming et al., 2006; Liu et al., 2009). The potential long-term effect of restoration actions

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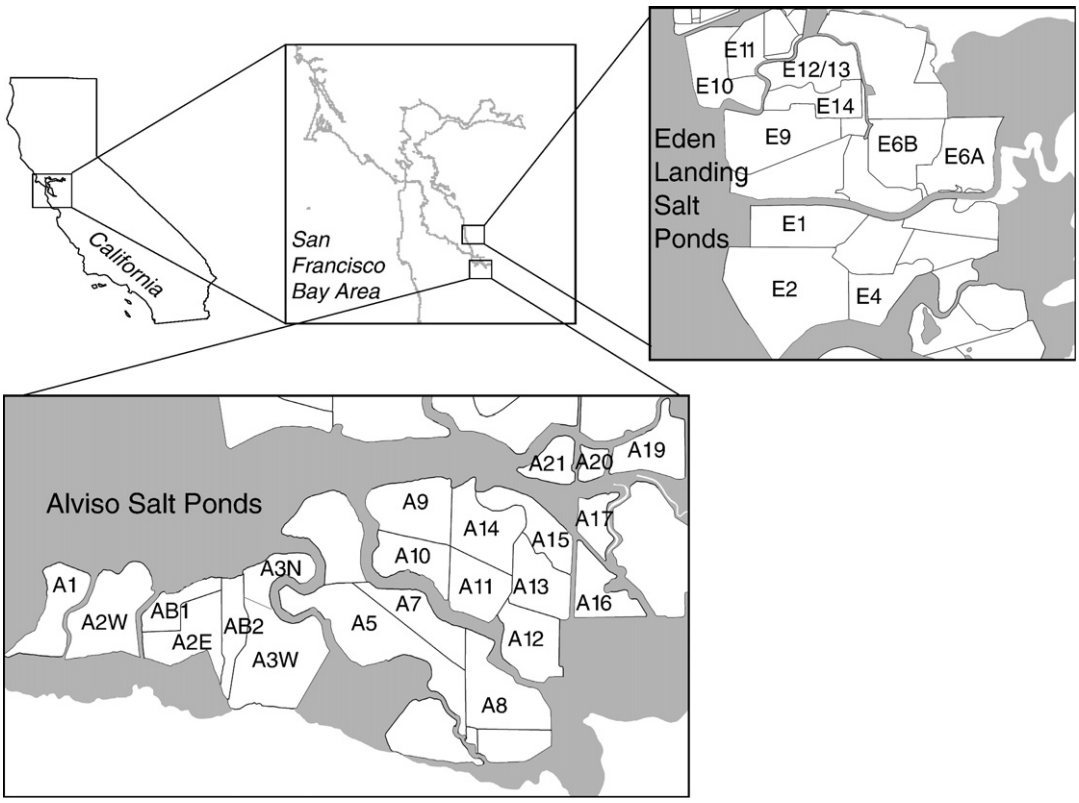

Fig. 1. Location of former commercial salt ponds at San Francisco (Bay), California, identified for restoration to natural or semi-natural conditions. Sediment samples were collected for mercury analyses from numbered ponds (note that in text, Eden Landing pond identification numbers are preceded by an "E" and Alviso ponds are preceded by an "A") periodically from summer or winter 2003–2007. Gray shading indicates Bay or tributaries.

such as lowering salinity in high salinity ponds, agitation of sediment, or tidal wetting and drying of sediment may be increased MeHg production (Compeau and Bartha, 1987; Gilmour et al., 2004; Gustin et al., 2006). Monitoring is important to understand changes in water and sediment chemistry that ensue from proposed or enacted water management plans and subsequent effects on Hg.

Levels of Hg in sediment and related concentrations in biota are not well understood but modeling of empirical data has been used to assess this relationship (Chapman, 1989; Long et al., 1995). When compared to other media (e.g., tissue concentrations), sediment can be a relatively stable medium for understanding differences in ambient, elemental contamination over space and time and can also provide a means of assessing remediation for improving environmental quality (Miles and Tome, 1997; Chapman, 1989). In a review of San Francisco Bay data from the 1970s and 1980s, Long et al. (1988) suggested a grand (arithmetic) mean of 0.50 μg/g dry weight (dw) Hg in sediment, in which 93% of these samples had concentrations ≤1.0 μg/g.

In this study we describe spatial and short-term temporal variations in sediment Hg species concentrations within and among the Alviso and the Eden Landing salt ponds. Our objectives were to determine: a) total Hg (Hg_t) and methylmercury (MeHg) in surficial (top 5 cm) sediment in order to establish baseline conditions prior to restoration, b) sediment Hg_t concentrations in a subset of Alviso and Eden Landing ponds after commencement of restoration, and c) variation in Hg species concentrations relative to discrete measures of sediment pH and total Fe (Fe_t) concentrations and water depth and salinity in a subset of ponds. Monitoring is important to understand how changes in water and sediment chemistry that ensue from any salt pond restoration activities subsequently affect Hg cycling. The information derived from this study is important to resource agencies that adaptively manage restoration at the ponds to water quality and natural resource goals.

2. Methods

2.1. Sediment sample collection

We collected sediment samples using a 2.0 cm diameter × 1.5 m corer made of PVC. The surficial layer of the sediment cores was used for all Hg analyses. We established in pilot work at the salt ponds that MeHg concentrations were higher in surficial sediment compared to deeper interval (15–20 cm) sediment, but Hg_t did not differ between surface and deeper sediment (unpublished data); Marvin-DiPasquale et al. (2003) and Hammerschmidt et al. (2004) reported similar findings. Sediment samples were placed in acid-rinsed glass jars, stored on ice, and transferred to a −20 °C freezer within several hours of collection. The corer was scrubbed with pond water and rinsed with deionized water between each point. At each site within a pond, 3 sub-samples of sediment were collected within a 10 m radius and composited as one sample per site (Elder et al., 1980). All sites were georeferenced with a Global Positioning System.

2.2. Sampling design

We conducted large-scale sampling in ponds that comprised the Alviso and Eden Landing salt pond complexes (Fig. 1) during August–October (summer) or January–February (winter) 2003–2007 to provide baseline Hg concentrations during initial restoration activities. All ponds were sampled at least once before and after restoration, which occurred approximately between summer 2004 and winter 2006 depending on the pond. Alviso ponds A3N, A8, A10, A15, and A2E were not subjected to restoration during the study (Table 1). Depending on funding constraints, samples collected within ponds were either composited as 1 sample per pond (Gilbert, 1987) or analyzed individually. A minimum of 3 sites located in proximity of inflow and outflow structures and mid-pond were sampled in each pond.

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Table 1
Geometric mean (and range) concentrations of total mercury (μg/g dw) in surficial sediment from Alviso Salt Ponds, San Francisco Bay, California.

Pond	Sampling dates						
	Summer – 2003	Summer – 2004	Winter – 2005	Summer – 2005	Winter – 2006	Summer – 2006	Winter – 2007
A3N	0.38 (0.22–0.77)	–	0.26 (0.11–0.39)	0.29 (0.08–0.709)	0.26 (0.13–0.63)	0.23 (0.05–0.55)	0.13 (0.05–0.46)
A7	0.81 (0.56–1.24)	0.31	0.77 (0.30–1.69)	0.95 (0.54–2.52)	1.38 (0.43–4.77)	1.22 (0.677–2.52)	1.04 (0.72–2.56)
A9	–	–	0.56 (0.51–0.66)	0.63 (0.48–0.76)	0.72 (0.62–0.79)	0.68 (0.584–0.90)	0.68 (0.62–0.73)
A12	1.70 (0.90–4.49)	–	0.95 (0.27–4.23)	2.17 (0.93–3.92)	2.79 (1.73–6.74)	1.75 (0.93–3.58)	1.98 (1.03–4.93)
A16	0.41 (0.25–0.63)	–	–	0.67 (0.55–0.824)	0.68 (0.53–0.82)	0.73 (0.614–0.86)	0.60 (0.34–0.956)
A1	–	0.30	–	–	–	0.29	–
A5	–	0.44	0.41 (0.29–0.51)	–	–	0.66	–
A8	0.70 (0.13–4.37)	–	–	–	–	0.54	–
A10	0.72 (0.69–0.79)	–	–	–	–	0.83	–
A11	0.65 (0.51–1.05)	–	–	–	–	0.27	–
A13	1.07 (0.62–3.11)	–	0.83 (0.61–1.53)	–	–	1.25	–
A14	0.28 (0.19–0.50)	–	–	–	–	0.10	–
A15	–	–	0.49 (0.27–0.72)	–	–	0.71	–
A17	–	–	0.20 (0.16–0.27)	–	–	0.38	–
A19	–	–	0.14	–	–	0.18	0.14
A20	–	–	0.25	–		0.27	0.39
A21	–	–	0.11	–		0.23	0.15
A2E	0.49 (0.45–0.51)	0.18	–	–		0.46	–
A2W	–	0.74	–	–		0.53	–
A3W	–	0.55	–	–		0.15	–
AB1	–	0.39	0.38 (0.34–0.42)	–		0.55	–
AB2	–	0.39	–	–		0.46	–

Values without ranges represent a single composite sample. Box = approximate time of implementation of a change in water management strategy for that pond. Dash = not sampled.

To evaluate effects of restoration on sediment Hg in ponds of different water management schemes, a repeated measures time series design was implemented in summer 2005 whereby 25 randomly selected fixed sites in Alviso ponds A3N, A7, A9, A12, and A16 were sampled twice annually until winter 2007. Pond A3N (4 sites) was a seasonal pond, A9 (4 sites) was a deep water pond with an inlet from Alviso Slough, A12 (4 sites) was the deepest pond with an outlet to Alviso Slough, A16 (4 sites) was targeted for reduction in water salinity, and A7 (9 sites) had a pronounced depth gradient subjected to variable wetting and drying. A similar design was implemented at Eden Landing whereby 4 sub-pond complexes interconnected by breached levees were sampled during summer 2005 and winter 2006. We sampled 5 randomly selected sites in sub-complexes E6A/6B, E10/11, and E9/12/13/14, and 6 sites in sub-complex E1/2/4. We measured sediment pH and Fe_t concentrations and water depth and salinity, which may influence or interact with MeHg levels, at each point within Alviso ponds. We measured water depth to the nearest cm with a 2-m ruler and water salinity with a refractometer.

2.3. Chemistry analyses

Frontier Geosciences, Inc. (Seattle, WA) and Battelle Marine Sciences (Sequim, WA) analyzed samples as follows: for Hg_t, sediment was completely digested using aqua regia and released gaseous mercury measured by cold vapor atomic florescence (Method 1631 Appendix A, EPA, 1996). Samples for MeHg were digested using $KBr/H_2SO_4/CuSO_2$ and then extracted into CH_2Cl_2. An ethylating agent was added to the extract to form a volatile methyl–ethylmercury derivative, and then purged onto graphitized carbon traps as a means of preconcentration and interference removal (Bloom et al., 1997). Samples were then isothermally chromatographed, pyrolitically broken down to elemental mercury, and detected using a cold vapor fluorescence detector (Bloom, 1989). Volatilized Hg compounds were then separated via isothermal gas chromatography, pyrolitically reduced to elemental Hg, and quantified via a cold vapor fluorescence detector (Bloom, 1989). Standard reference materials (SRMs) used were from the International Atomic Energy Agency (IAEA-405; estuarine soils) for Hg_t (certified value = 0.81 ± 0.04 mg/g) and MeHg (certified value = 5.49 ± 0.53 ng/g), and the National Institute of Standards and Technology (NIST 2709; San Joaquin soils, certified value = 1.44 ± 0.07 mg/g) and National Research Council Canada (NRCC MESS-3; marine sediments, certified value = 0.09 ± 0.01 mg/g) for Hg_t. Quality control and assurance included calculating relative percent difference (RPD) for laboratory duplicates and matrix spike recoveries on 5% of all samples. For Hg_t, RPD averaged 5% (standard deviation [SD] = 4%), matrix spike recoveries averaged 97% (SD = 9%), and SRM recoveries averaged 100% (SD = 5%). For MeHg, RPD averaged 7% (SD = 9%), matrix spike recoveries averaged 94% (SD = 11%), and SRM recoveries averaged 88% (SD = 7%). Minimum detection limits averaged 0.004 μg/g (SD = 0.001) for Hg_t and 0.01 ng/g (SD = 0.005) for MeHg. Total (Hg_t) is reported as μg/g and MeHg as ng/g, dry weight.

Sediment was analyzed for Fe_t and pH by the Agricultural and Natural Resources Analytical Laboratory, University of California, Davis (UCD). Sediment Fe_t was determined using a nitric acid/hydrogen

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peroxide closed vessel microwave digestion and then atomic absorption spectrometry (Sah and Miller, 1992). Standard reference materials included an in-house standard (UCD 187) and NIST 2709. Sediment pH was determined from a saturated paste extract with a pH meter (USDA, 1954). This method measures the disassociated H-ions affecting the sensing electrode and is reproducible within 0.2 pH units.

2.4. Statistical analyses

We used geometric means and ranges or report values from single composite samples to describe general spatial and temporal (where appropriate) patterns of sediment Hg_t and MeHg concentrations within salt ponds. We calculated percent changes in mean Hg relative to the timing of restoration, or between early and late sampling for ponds without restoration activity.

2.4.1. Alviso and Eden Landing time series

For the Alviso time series ponds, we constructed a set of repeated measures ANOVA models to examine spatial and temporal variations in sediment Hg_t. Pond (spatial, 5 levels), year, and season (temporal, 2 levels each) were treated as categorical fixed effects. The year effect consisted of early (samples collected in summer 2005 and winter 2006) and later interval samplings (collected in summer 2006, winter 2007), herein, Year 1 and Year 2. For MeHg, we first constructed the same set of spatial and temporal models (set 1). We then constructed a second model set to examine the influence of covariates (i.e. sediment Hg_t, Fe_t, and pH and water depth and salinity) on MeHg singularly and in combination with the best spatial–temporal factor(s) determined from set 1. All models within each set were constructed using SAS (v8.12) PROC Mixed with Maximum Likelihood Estimation, whereby the repeated measures sampling point ($n=25$) was treated as a random effect.

We used Akaike's Information Criteria (AIC) (Burnham and Anderson, 2002) to assess which of the above model(s) best explained variation in sediment Hg when contrasted with all models within set 1 or 2. The AIC approach is better suited than traditional testing for statistical significance because the null hypothesis is often not reasonable for observational data. Factors related to Hg distribution would be exceedingly complicated in a single model, and AIC provides a strategy for identifying the model that provides the best balance of complexity (i.e. explaining large patterns of variation in the data) and parsimony (i.e. avoiding overly complex models). The AIC process is a relative measure of model fit, related to the deviance between the data and the model. We used the second-order AIC (AIC_c) corrected for sample size bias ($AIC_c = -2(\text{log-likelihood}) + 2KN/(N-K-1)$, where K = number of estimable parameters and N = sample size). The model with minimum AIC_c is construed as the best model in the set, and the relative fit of all other models was scaled to the best model in the form of delta (Δ) AIC_c. Models with a ΔAIC_c value <2 (i.e. difference between the AIC_c value for $model_j$ and the top $model_i$) were evaluated as competing models: Akaike weights (i.e. $w=(-\exp$

Table 2
Geometric mean (and range) concentrations of methylmercury (ng/g dw) in surficial sediment from Alviso Salt Ponds, San Francisco Bay, California.

Pond	Sampling dates						
	Summer – 2003	Summer – 2004	Winter – 2005	Summer – 2005	Winter – 2006	Summer – 2006	Winter – 2007
A3N	5.77 (2.62–11.22)	–	3.03 (1.78–4.87)	2.65 (1.00–9.51)	3.14 (2.36–5.29)	4.24 (2.69–8.03)	3.35 (2.78–4.66)
A7	2.83 (1.13–11.57)	2.15 –	2.04 (0.41–5.61)	4.68 (0.88–15.50)	4.31 (2.16–13.50)	1.55 (0.53–8.19)	1.62 (0.36–5.14)
A9	–	–	2.59 (1.23–5.21)	1.93 (1.23–2.83)	1.61 (1.10–2.18)	1.03 (0.85–1.36)	1.50 (0.94–2.19)
A12	3.34 (1.46–6.00)	–	3.82 (1.77–8.97)	2.68 (0.98–5.05)	4.26 (1.92–8.69)	4.04 (2.45–7.24)	3.42 (2.04–9.51)
A16	1.21 (0.76–2.42)	–	–	1.66 (0.92–4.12)	0.67 (0.25–2.38)	0.65 (0.40–1.21)	1.29 (0.32–6.53)
A1	–	0.32		–	–	0.11	–
A5	–	0.23	2.12 (1.16–3.04)	–	–	0.31	–
A8	1.45 (0.76–6.13)	–	–	–	–	2.30	–
A10	1.43 (1.10–1.75)	–	–	–	–	3.10	–
A11	2.31 (0.96–4.95)	–	–	–	–	4.95	–
A13	3.03 (1.67–4.68)	–	3.08 (2.87–3.47)	–	–	1.83	–
A14	1.44 (0.90–1.92)	–	–	–	–	1.55	–
A15	–	–	2.22 (1.83–2.51)	–	–	1.00	–
A17	–	–	1.94 (0.95–3.46)	–	–	0.38	–
A19	–	–	0.68	–	–	3.77	2.83
A20	–	–	1.69	–	–	8.60	6.55
A21	–	–	1.22	–	–	1.57	0.49
A2E	0.72 (0.40–1.08)	1.19	–	–	–	0.17	–
A2W	–	2.54	–	–	–	0.33	–
A3W	–	0.27		–	–	0.64	
AB1	–	1.91	0.99 (0.43–1.97)	–	–	0.75	–
AB2	–	0.73		–	–	0.89	–

Values without ranges represent a single composite sample. Box = approximate time of implementation of a change in water management strategy for that pond. Dash = not sampled.

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$(-\Delta AIC/2)))$ were used as a measure for model likelihood. Interpreting these model weights as the probability of the model being the best in the set is heuristic. Evidence ratios of AIC_c weights (i.e. w_i/w_j, where w_i is the weight of the best ΔAIC_c model and w_j the weight of a model of interest in the set) were used to further evaluate the likelihood of the competing models. Increasing evidence ratios indicated more support for model$_i$. These ratios can be viewed as a type of odds ratio for identifying the relative likelihoods of different models. Finally, model set 1 was balanced, i.e., each main effect or interaction was represented the same number of times within the set. This enabled direct comparison of factor weights (i.e. AIC_c weights summed across all models where a particular factor occurred) to determine the importance of that factor relative to other factors (Burnham and Anderson, 2002, p. 169).

Similarly, we used nested repeated measures ANOVA fitted in PROC Mixed and interpreted with the AIC approach to determine whether sediment Hg_t and MeHg concentrations differed between Alviso and Eden Landing pond complexes and seasons during interval Year 1 when data for both pond complexes were collected. Ponds nested within complexes were treated as an additional random effect. Only the first model set was applied to Alviso and Eden Landing pond comparisons because we lacked covariate data for all sites at Eden Landing.

All variables except sediment pH were $\log_e$ transformed prior to analysis for all ANOVA/ANCOVA models. Back transformed least squares mean concentrations (i.e., geometric mean) of Hg_t and MeHg controlled for model effects are reported unless stated otherwise.

Table 4
Akaike's Information Criterion (AIC) used to determine the best approximating model(s) to explain spatial (Ponds A3N, A7, A9, A12, and A16) and temporal (Year[a], season = summer, winter) variations in total mercury in sediment at the Alviso Salt Ponds, San Francisco Bay, CA.

Model	K[b]	$(-\text{Log } \mathcal{L})$[c]	AIC_c	ΔAIC_c	(w_i)[d]
Pond + Year + Season + Year*Season	10	121.9	144.4	0	0.38
Pond + Year	8	127.7	145.2	0.8	0.25
Pond	7	130.6	145.8	1.4	0.19
Pond + Year + Season	9	127.6	147.6	3.2	0.08
Pond + Season	8	130.6	148.2	3.8	0.06
Pond + Year + Pond*Year	12	121.9	149.5	5.1	0.03
Pond + Year + Season + Pond*Year	13	121.9	152.1	7.7	0.01
Pond + Year + Season + Pond*Season	13	121.9	152.2	7.8	0.01
Pond + Season + Pond*Season	12	125.1	152.7	8.3	0.01
Year + Season + Year*Season	6	153.3	166.2	21.8	0
Year + Season + Year*Season	4	159	167.4	23	0
Intercept	3	162	168.2	23.8	0
Year + Season	5	159	169.6	25.2	0
Season	4	162	170.4	26	0

$N = 100$. See methods for explanation of AIC values.
[a] Year = Year 1 = sampling interval summer 2005, winter 2006; Year 2 = summer 2006, winter 2007.
[b] K = parameters.
[c] $(\mathcal{L})$ = likelihood.
[d] w_i = Akaike weight.

Table 3
Geometric mean concentrations of total mercury (Hg_t, μg/g, dry weight) and methylmercury (MeHg ng/g, dry weight) in surficial sediment from Eden Landing salt ponds, San Francisco Bay, California.

Pond ID	Pre-conversion Hg_t; MeHg		
	Summer 2003	Summer 2004	Winter 2005
E1	–	0.14; 1.71	0.13; 2.16
E2	0.13; 0.65[a]	0.14; 0.62	0.12; 1.24
E4	–	0.10; 2.17	0.12; 1.48
E5	–	–	0.09; 0.82
E6	–	–	0.07; 0.48
E6A	0.07; 0.12[b]		0.08; 1.60
E6B	–	–	0.09; 2.33
E7	–	0.08; 0.47	0.09; 0.26
E8	–	–	0.07; 0.66
E8A	–	–	0.13; 0.98
E9	–	–	0.09; 2.24
E10	–	–	0.09/0.99[c]
E11	0.16; 10.7[d]		0.13; 3.07[e]
E12	0.06; 2.07[f]		0.12; 2.76
E14	–	–	0.09; 1.68
E1C	–	–	0.16; 1.79
E2C	–	–	0.07; 0.54
E4C	–	–	0.05; 0.48
E5C	–	–	0.12; 0.41
E6C	–	–	0.07; 0.32

Pond complex	Post-conversion			
	Hg_t		MeHg	
	Summer 2005	Winter 2006	Summer 2005	Winter 2006
E1/2/4	0.15 (0.10–0.30)	0.16 (0.13–0.20)	0.62 (0.07–1.21)	0.78 (0.41–3.12)
E6A/6B	0.07 (0.05–0.09)	0.08 (0.05–0.11)	0.81 (0.38–1.75)	1.08 (0.47–2.15)
E10/11	0.11 (0.07–0.16)	0.17 (0.14–0.24)	0.93 (0.40–2.16)	1.68 (1.08–3.43)
E9/12	0.03 (0.01–0.05)	0.06 (0.03–0.09)	0.62 (0.39–1.55)	0.42 (0.35–0.57)

Values without ranges footnoted or in parentheses represent three site samples within pond composited into one sample. Dash indicates not sampled.
[a] Range Hg = 0.12–0.16, MeHg = 0.29–1.02.
[b] Range Hg = 0.07–0.07, MeHg = 0.09–0.20.
[c] Range Hg = 0.09–0.09.
[d] Range Hg = 0.15–0.16, MeHg = 10.5–10.9.
[e] Range Hg = 0.13–0.13.
[f] Range Hg = 0.05–0.09, MeHg = 1.88–2.29.

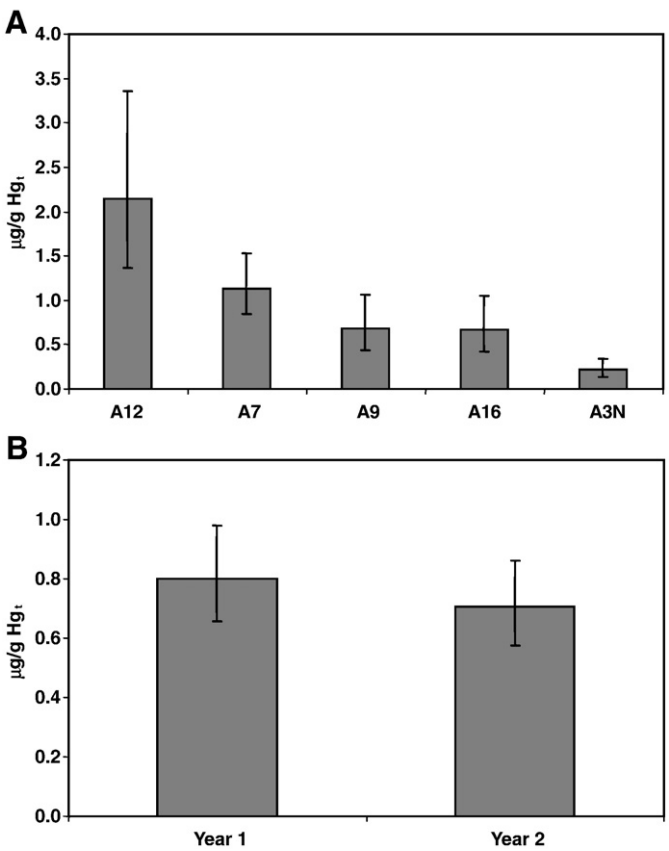

Fig. 2. Back transformed least squares mean concentrations and 95% confidence intervals of Hg_t determined from the most parsimonious AIC model Pond (A) + Year (B) for Alviso time series ponds; Year 1 = sampling interval summer 2005, winter 2006 and Year 2 = summer 2006, winter 2007.

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3. Results

3.1. Overview — pattern of sediment Hg levels

Mean sediment Hg_t concentrations within the Alviso ponds were relatively stable over time. Less than a 2-fold difference in Hg_t concentrations occurred in 8 of the 12 ponds sampled ≥3 time intervals, and only 1 measure of Hg_t differed 3–4-fold from those at other sampling intervals in the remaining 4 ponds (A3N, A7, A12, and A2E) (Table 1). Sediment Hg_t concentrations were also stable between intervals in 7 of 10 ponds sampled once early and late study; 4 of these ponds (A1, A2W, AB2, and A17) had restoration activity whereas the other 3 (A8, A10, and A15) had none. Sediment Hg_t concentrations declined 2–4-fold in the remaining 3 (of 10) ponds (A11, A14, and A3W) that had restoration activity.

Patterns of change in MeHg were more pronounced than those in Hg_t (Table 2). For example, dramatic changes in sediment MeHg concentrations were illustrated by an 8-fold decline in A2W and a >5-fold increase in Ponds A19 and A20 following implementation of restoration activities. Sediment MeHg concentrations increased markedly in Pond A5 (9-fold) and to a lesser extent in Pond A7 (2-fold) following restoration activities, but then declined during subsequent sampling events. In contrast, sediment MeHg concentrations increased >2-fold in Ponds A3W and A11, declined >2-fold in Ponds A1, A9, A13, and A2W, and remained relatively stable in Ponds A12, A16, AB2, and A14 following restoration activities. Among ponds without restoration activities, sediment MeHg concentrations decreased over time in A2E and A15, increased in A8 and A10, and were variable in A3N. The highest MeHg concentration (8.6 ng/g) was detected in A20 in summer 2006.

Sediment Hg_t and MeHg concentrations at the Eden Landing ponds were consistently low (overall geometric mean $Hg_t = 0.09$ μg/g, MeHg = 0.98 ng/g) relative to the Alviso Ponds ($Hg_t = 0.63$ μg/g MeHg = 1.94 ng/g). Sediment MeHg concentrations were approximately stable temporally within Eden Landing ponds with some exceptions

Table 5
Akaike's Information Criterion (AIC) used to determine the best approximating model(s) to explain spatial and temporal variations in sediment methylmercury (MeHg; Set 1), Alviso Salt Ponds, San Francisco Bay, CA.

Model set	Model	K[a]	$(-\text{Log}\ \mathcal{L})$[b]	AIC_c	ΔAIC_c	(w_i)[c]
1	Pond + Year + Pond*Year	12	191.6	219.2	0.0	0.7802
	Pond + Year + Season + Pond*Year	13	191.6	221.8	2.6	0.2126
	Pond + Year	8	211.9	229.5	10.3	0.0045
	Pond + Year + Season	9	211.9	231.9	12.7	0.0014
	Pond + Year + Season + Year*Season	10	211.1	233.5	14.3	0.0006
	Year	4	226.5	235.0	15.8	0.0003
	Pond	7	221.5	236.7	17.5	0.0001
	Year + Season	6	226.5	237.2	18.0	0.0001
	Year + Season + Year*Season	7	225.7	238.6	19.4	0.0000
	Pond + Season	8	221.5	239.1	19.9	0.0000
	Intercept	3	236.1	241.1	21.9	0.0000
	Pond + Year + Season + Pond*Season	13	211.4	241.7	22.5	0.0000
	Season	4	236.1	244.5	25.3	0.0000
	Pond + Season + Pond*Season	12	221.0	248.6	29.4	0.0000
2 (Year 1)	Pond + pH	8	97.5	117.0	0.0	0.3053
	Pond + Depth	8	98.5	118.0	1.0	0.1852
	Pond	7	101.7	118.3	1.3	0.1594
	Pond + Hg_t	8	99.3	118.8	1.8	0.1241
	Pond + Salinity	8	99.5	119.0	2.0	0.1123
	Pond + Total Fe (Fe_t)	8	100.8	120.3	3.3	0.0586
	Pond + Fe_t + Pond*Fe_t	12	93.9	122.9	5.9	0.0160
	Salinity	4	114.9	123.8	6.8	0.0102
	pH	4	115.3	124.1	7.1	0.0088
	Pond + Depth + Pond*Depth	12	92.1	124.5	7.5	0.0072
	Hg_t	4	115.7	124.6	7.6	0.0068
	Pond + Hg_t + Pond*Hg_t	12	94.7	127.1	10.1	0.0020
	Intercept	3	121.1	127.6	10.6	0.0015
	Pond + pH + Pond*pH	12	96.8	129.2	12.2	0.0007
	Depth	4	120.9	129.8	12.8	0.0005
	Fe_t	4	121.0	129.0	12.9	0.0005
2 (Year 2)	Pond + Depth	8	86.9	106.4	0.0	0.4823
	Pond + pH	8	87.5	107.0	0.6	0.3573
	Pond + Salinity	8	91.1	110.6	4.2	0.0591
	pH	4	101.9	110.8	4.4	0.0534
	Pond	7	96.3	113.0	6.6	0.0178
	Depth	4	106.2	115.1	8.7	0.0062
	Pond + Hg_t	8	96.2	115.7	9.3	0.0046
	Intercept	3	109.3	115.8	9.4	0.0044
	Pond + Fe_t	8	96.3	115.8	9.4	0.0044
	Pond + Depth + Pond*Depth	12	83.8	116.2	9.8	0.0036
	Salinity	4	108.0	116.9	10.5	0.0025
	Fe_t	4	109.1	118.0	11.6	0.0015
	Hg_t	4	109.2	118.1	11.7	0.0014
	Pond + pH + Pond*pH	12	85.7	118.2	11.8	0.0013
	Pond + Hg_t + Pond*Hg_t	12	90.7	123.1	16.7	0.0001
	Pond + Salinity + Pond*Salinity	12	90.7	123.1	16.7	0.0001
	Pond + Fe + Pond*Fe_t	12	92.7	125.2	18.8	0.0000

We then examined covariate and total mercury (Hg_t) effects on MeHg by Year (Year = Year 1 = sampling interval summer 2005, winter 2006; Year 2 = summer 2006, winter 2007) (Set 2). $N = 100$. See methods for explanation of AIC values.

[a] K = parameters.
[b] $(\mathcal{L})$ = Likelihood.
[c] w_i = Akaike weight.

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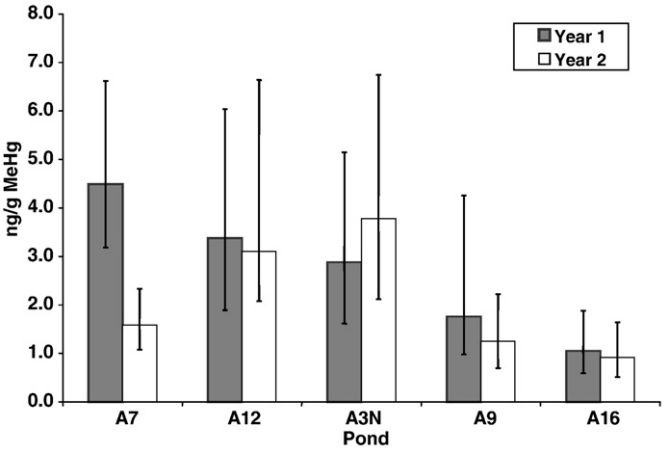

Fig. 3. Back transformed least squares mean concentrations and 95% confidence intervals of MeHg levels determined from AIC model of Pond + Year + Pond*Year for Alviso time series ponds; Year 1 = sampling interval summer 2005, winter 2006 and Year 2 = summer 2006, winter 2007.

(Table 3). Most noteworthy was a high average MeHg sediment concentration (10.7 ng/g) detected in Pond E11 in summer 2003. However, this concentration was >3-fold lower when sampled in winter 2005, indicating that we probably detected an isolated hotspot previously. Sediment MeHg concentrations declined after summer 2005 in all pond complexes (E1/2/4, E6A/6B, and E9/12/14) when compared to those detected in specific ponds pre-restoration (Table 3).

3.2. Alviso time series ponds: AIC–Hg_t

The model containing Pond + Year + Season + Year*Season had the lowest AIC_c value (Table 4). However, simpler competing models within <2 ΔAIC_c units included the models containing Pond and Year, and Year only. Evidence ratios indicated that the top model (containing 2–3 more parameters) was only ~2 times better than either of these models. Factor weights indicated the strongest relative influence of Pond (0.99), then Year (0.75), and then Season (0.53). Therefore, we chose the simpler Pond + Year model. Least squares means indicated that Hg_t concentrations were highest in A12 (2.1 μg/g), intermediate in A7 (1.1 μg/g), and lowest in A3N (0.2 μg/g) (Fig. 2A). Conversely, Hg_t changed little between Year 1 and Year 2, which further indicated that Hg_t varied more strongly spatially than temporally (Fig. 2B).

3.3. Alviso time series ponds: AIC–MeHg

Spatial and temporal variations in MeHg were best explained by the model Pond + Year + Pond*Year ($\Delta AIC_c = 0.0$; $w_i = 0.78$); no other model had $\Delta AIC_c \leq 2$ or $w_i > 0.22$ (Table 5 [set 1]). Factor weights indicated that Pond (0.99) and Year (0.99) had the strongest influence, while Season had little relative influence (0.30). Concentrations of

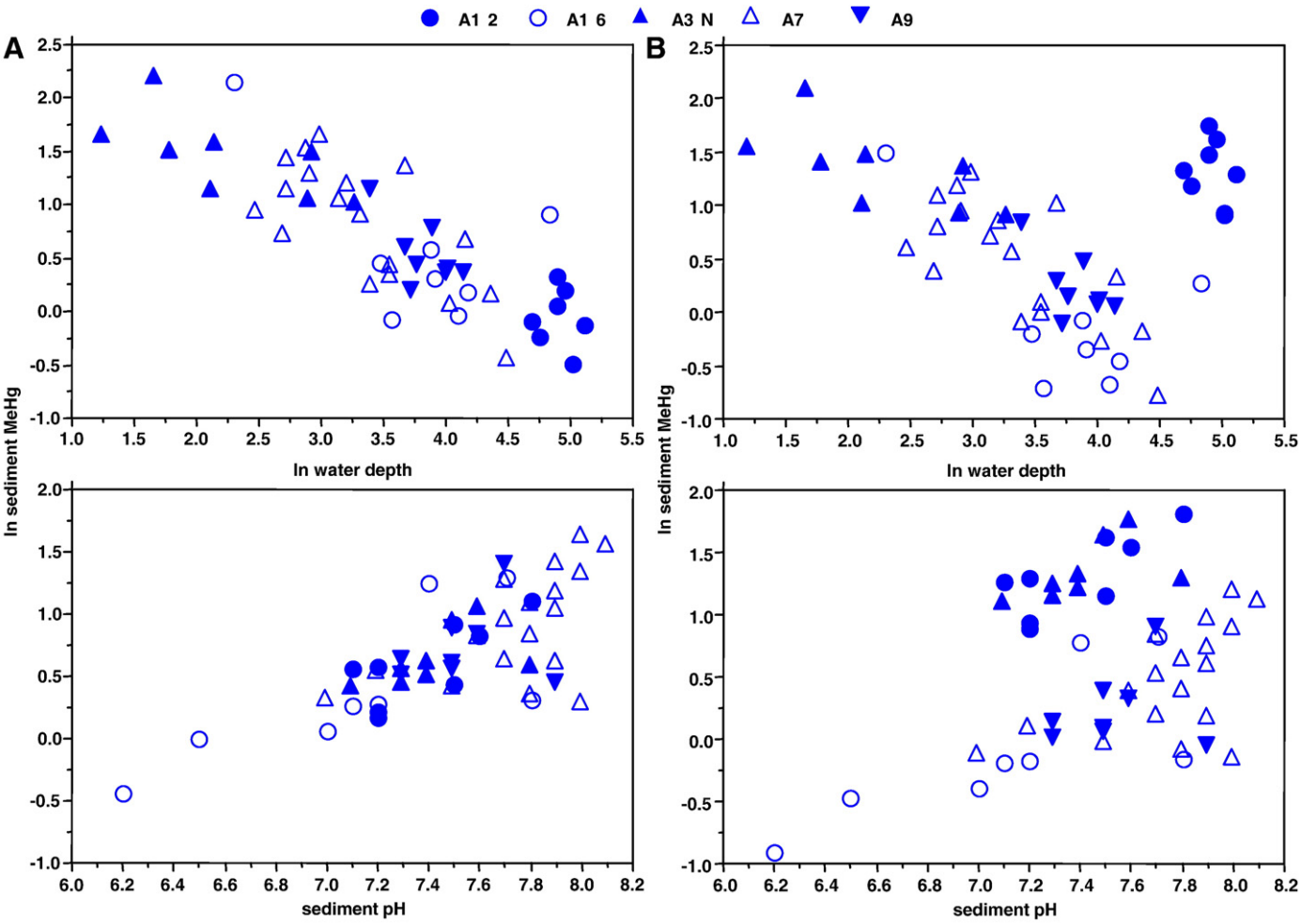

Fig. 4. Relationships between MeHg and sediment pH or water depth in sampling interval Year 2 (summer 2006, winter 2007) for Alviso time series ponds. MeHg (*y*-axes) concentrations are adjusted for variation between sampling sites with (A) and without (B) controlling for Pond effects.

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MeHg were highest in Pond A7, intermediate in A12 and A3N, and lowest in A9 and A16 in Year 1. However, MeHg declined about 2-fold in A7 in Year 2, but concentrations for all other ponds remained similar between Year 1 and Year 2 as indicated by overlapping confidence intervals (Fig. 3).

We examined covariates separately by Year 1 and Year 2 because the effect of Pond on sediment MeHg concentrations was confounded by time. Although Pond + pH comprised the top model in Year 1, four other competing models had $\Delta AIC_c \leq 2$ (Table 5 [set 2, Year 1]). Evidence ratios provided insight into the contribution of factors within these competing models: the top model was only ≤2.7 times better than competing models with Pond only or Pond + Depth, + Hg_t, or + Salinity, indicating that pH did not improve model likelihood over Pond only or these other factors. In contrast, the evidence ratio for models Pond + pH:pH was 34.7, indicating that the Pond effect was the primary factor that improved model likelihood, and not pH. Evidence ratios for competing models of Pond + Depth:Depth (370.4), Pond + Hg_t:Hg_t (18.2), or Pond + Salinity:Salinity (11.0) further substantiated that these covariates had little influence on MeHg in Year 1.

The role of covariates in Year 2 differed from that concluded in pre-restoration (Table 5 [set 2, Year 2]). Similar to Year 1, the models containing Pond + Depth and Pond + pH had ΔAIC_c values ≤ 1.0; however, in this set the ΔAIC_c for all other models including the Pond model was >4.0. The top model was only 1.3 times better than the next competing model. Evidence ratios of the top models versus the Pond model (Pond + Depth:Pond = 27.1 and Pond + pH:Pond = 20.1) indicated that depth and pH improved model likelihood while accounting for variation in MeHg among ponds in Year 2. When Pond effects were accounted for, depth was inversely related to MeHg while pH was positively related to MeHg (Fig. 4A). However, when Pond effects were not controlled, the MeHg relationship with pH was weakened and Pond A12 was a notable outlier in the relationship with depth (Fig. 4B).

3.4. Alviso time series ponds: spatially explicit Hgt and MeHg concentrations

Specific hotspots of sediment Hg_t (mean > 2 μg/g) were most evident in Ponds A7 and A12, and concentrations were lower and consistent over time at sites in A3N, A9, and A16 (Fig. 5), whereas MeHg hotspots (mean > 2 ng/g) were evident in all Alviso time series ponds (Fig. 6). There was little consistency of elevated hotspots between Hg_t and MeHg in sediment except within Pond A12 and to a lesser degree in Pond A9. Typically, MeHg concentrations were elevated and temporally variable in Ponds A3N, A7, and A16, with indications of decline particularly in Ponds A3N and A7 from summer 2005 to winter 2007. In particular, concentrations of MeHg at the northern end of Pond A7 declined substantially over time.

3.5. Alviso and Eden Landing ponds: AIC–Hg_t and MeHg

The Complex + Season model best explained variation in Hg_t ($\Delta AIC_c = 0.0$, $w_i = 0.67$) (Table 6). Sediment Hg_t levels were approximately 10 times higher in Alviso (ls mean: 0.82 μg/g, 95% CI: 0.42–1.57) than those in Eden Landing ponds (0.09 μg/g, 0.04–0.18), and overall were slightly higher during winter (0.31 μg/g, 0.19–0.50) than summer (0.24 μg/g, 0.15–0.39). The model containing Complex only best explained variation in MeHg ($\Delta AIC_c = 0.0$, $w_i = 0.64$) (Table 6). MeHg levels were approximately 3 times higher in Alviso (2.41 ng/g, range 1.97–2.95 ng/g) compared to Eden Landing ponds (0.81 ng/g, range 0.65–1.00 ng/g).

4. Discussion

Salt ponds have been common to the South San Francisco Bay landscape since the 1860s (Ver Planck, 1958). The timing of the creation and large-scale functioning of salt ponds coincided with that of mercury mining in the south Bay watershed (Cargill et al., 1980). Historically, mining in the Santa Clara Valley south of the Bay produced more mercury than anywhere else in North America (Thomas et al., 2002). The release of Hg into the south bay watershed apparently influenced sediment in the Alviso salt ponds but was not as apparent in the Eden Landing pond sediment, just ~15 km to the north. Even among the Alviso ponds, in-depth spatial sampling revealed high variability that further emphasized the necessity for adaptive management of water by pond between and within seemingly similar systems. For example, of the 22 Alviso ponds, sediment Hg_t concentrations were consistently <0.50 μg/g in 9 of those ponds and >0.50 μg/g in 8 others.

Our study derived a higher grand (arithmetic) mean for sediment Hg_t at the Alviso salt ponds than the 0.50 μg/g derived from data collected Bay-wide (Long et al. 1988, 1995). The grand mean of sediment samples collected early in the study before any restoration at Alviso ponds was 0.74 μg/g and increased to 1.03 μg/g after restoration was instituted at these ponds. These concentrations also exceeded the suggested 0.71 μg/g Hg_t mid-biological effects criteria for sediment (Long et al., 1995) and the suggested ambient concentrations of 0.25 (fine-grained) to 0.43 (coarse-grained) Hg_t μg/g (SFEI, 1997). Grand mean sediment Hg_t concentrations from the Eden Landing ponds were low and the same pre- and post-restoration (0.11 μg/g), and only 2 times higher than historical pre-mining levels (0.05 μg/g; Hornberger et al., 1999). Sediment Hg_t levels tended to be relatively stable over time as demonstrated in this and other estuarine studies (Miles and Tome, 1997; Chapman, 1989), thus the implication is that levels in certain Alviso salt ponds have been elevated >1.0 μg/g long-term. Events since the creation of the ponds that may have contributed to elevated Hg_t levels, e.g., inflow of contaminated water-borne particle matter, periodic resuspension from dredging for levee maintenance, or atmospheric deposition probably were chronic but not acute influences.

Overall (arithmetic mean) sediment MeHg concentrations in Alviso restoration ponds increased (2.58 to 3.03 ng/g) while they decreased (2.20 to 1.03 ng/g) in Eden Landing restoration ponds during the study. Methylmercury comprised from <1 to 3% of the Hg_t detected across all ponds. No definitive criteria have been established for possible biological effects due to sediment MeHg, but all concentrations we observed were higher than the U.S. Environmental Protection Agency suggested recommended daily intake of 0.1 ng/g MeHg per kg body weight. This threshold has not been established as detrimental to birds or fishes but we still know little about chronic long-term exposure to sublethal concentrations to these vertebrates (Burger and Gochfeld, 1997; Scheuhammer, 1987). Sediment (or soils) represents 1 of 3 (water, air) media that form the physical basis of food chains, and MeHg at succeeding trophic positions would be expectedly higher than in sediment due to bioaccumulation. Infaunal or epifaunal brackish or salt water organisms (e.g., polychaete worms, *Macoma* clams) ingest sediment. These organisms as well as small amounts of sediment are then ingested by waterbirds, thus the link up the food chain from sediment is direct as well as indirect (Hui and Beyer, 1998; Beyer et al., 1999).

Initial changes in water management strategies on certain Alviso ponds instituted to reduce salinity probably had some effect on sediment MeHg concentrations. These changes included new or increased water movement between ponds and tributaries or between ponds by means of a culvert and gate for direction of flow (i.e., inlet [I] or outlet [O]) (Mruz, Ransom, Peterson, personal communication). Sediment MeHg concentrations declined >2-fold in 6 of 10 Alviso ponds (Table 2) that were directly connected hydrologically with a Bay tributary during July 2004 (A1, A7, A2W [I gate], and AB1 [O]) or March 2005 (A9 [I] and A17 [I/O]). In the remaining 4 ponds, sediment MeHg levels remained stable in A14 [I] and A16 [I/O], increased in A5 [I] in winter 2005, and increased >2-fold in A3W [O]. The higher MeHg level

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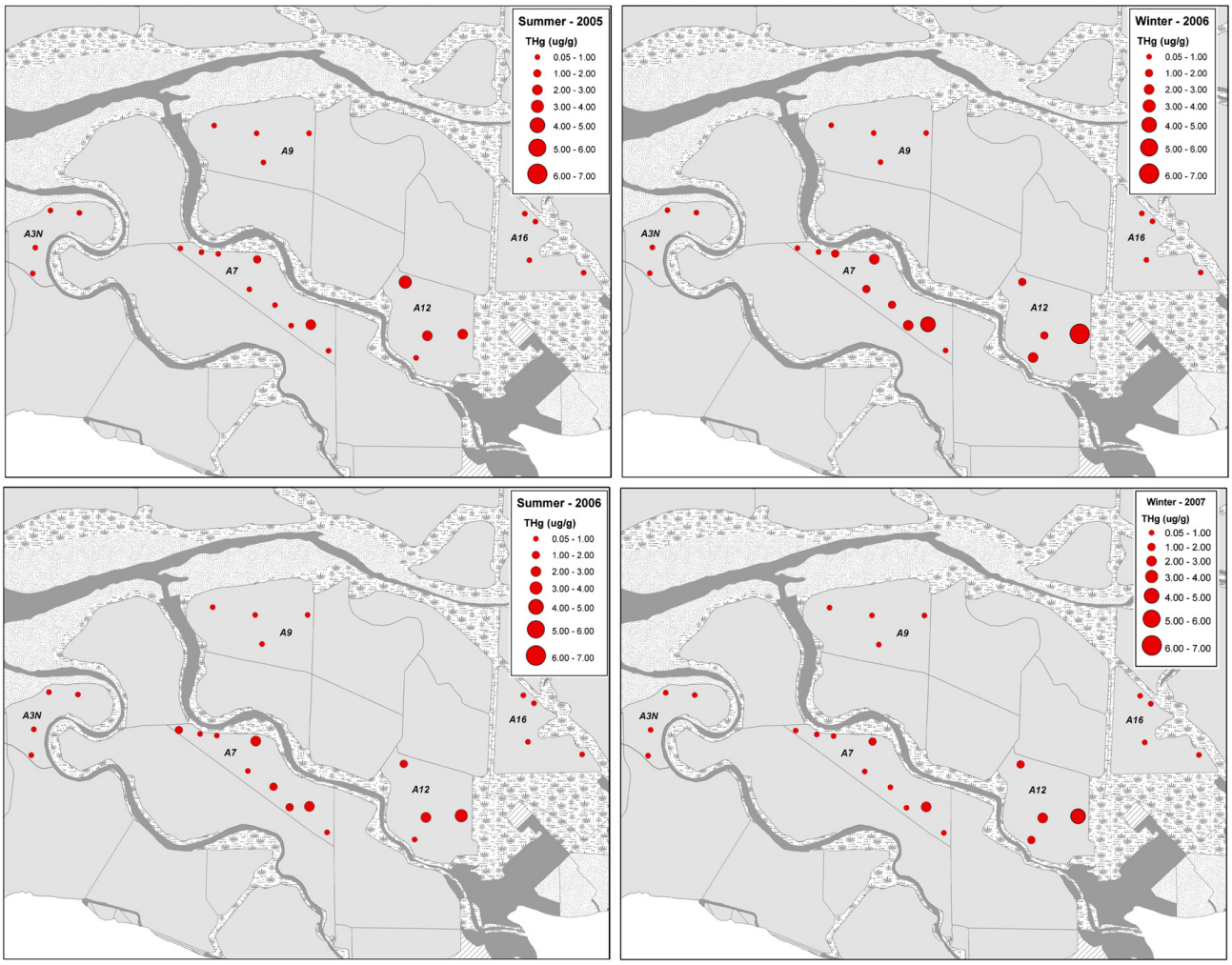

Fig. 5. Spatially explicit time series of sediment total (Hg_t) mercury concentrations in Alviso Ponds A3N, A7, A9, A12, and A16; summer 2005, 2006 – winter 2006, 2007.

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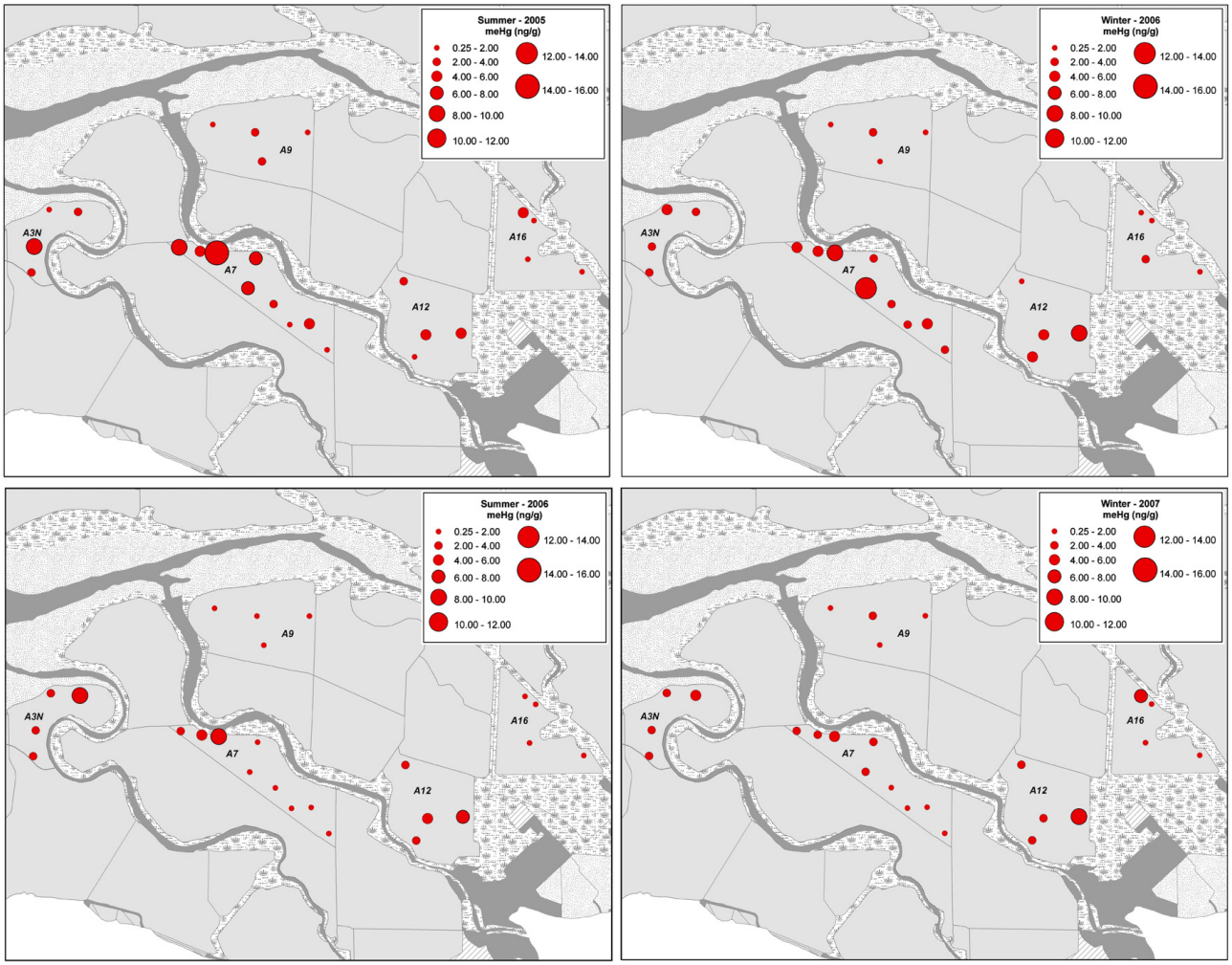

Fig. 6. Spatially explicit time series of sediment methylmercury (MeHg) concentrations in Alviso Ponds A3N, A7, A9, A12, and A16; summer 2005, 2006 — winter 2006, 2007.

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Table 6
Akaike's Information Criterion (AIC) used to determine the model that best defines total mercury (Hg_t) and methylmercury (MeHg) differences in sediment between Alviso and Eden Landing pond complexes, San Francisco Bay, CA, sampled summer 2005 and winter 2006.

Response	Model	K[a]	$(-\text{Log } \mathcal{L})$[b]	AIC_c	ΔAIC_c	(w_i)[c]
Hg_t	Complex + Season	6	147.0	160.0	0.0	0.67
	Complex + Season + Complex*Season	7	146.3	161.7	1.7	0.29
	Complex	5	155.2	165.9	5.9	0.04
	Season	5	159.3	169.9	9.9	0
	Intercept	3	242.4	248.6	106.6	0.0000
MeHg	Complex	5	203.8	214.5	0.0	0.64
	Complex + Season	6	203.7	216.7	2.2	0.21
	Complex + Season Complex*Season	7	202.4	217.7	3.2	0.13
	Season	5	221.9	222.6	8.1	0.01
	Intercept	3	240.0	248.4	33.9	0.0000

$N = 92$. See methods for explanation of AIC values.
[a] K = parameters.
[b] $(\mathcal{L})$ = Likelihood.
[c] w_i = Akaike weight.

in A5 was not associated with any known dredging during the time period under study. However, A5 was the only Alviso pond where a gate was added to allow inflow from Guadalupe Slough during summer 2004. This slough connects to the Guadalupe River that received effluent from the largest producing Hg mine (New Almaden) in the watershed (Thomas et al., 2002); the higher MeHg concentration was observed in the winter (2005) season when precipitation and subsequent flow in the Guadalupe River were greater than during the summer sampling. Methylmercury in A5 then notably returned to pre-restoration levels in summer 2006.

Changes in MeHg concentrations were most notable on Ponds A19 and A20, which including A21, were the only ponds that had levees actually breached to an adjacent tributary during the study. These inter-connected ponds were subjected to increased tidal flow and subsequent variation in water depth and wetting–drying cycles. Sediment MeHg concentrations increased >5-fold in A19 and A20 but only slightly in A21 from pre- to post-breach. Sediment MeHg concentrations also increased notably in Ponds A8 and A10 that had no restoration activity instituted during the study and in A11 that was gated to deliver water to A14. Pond A8 received water from A5 (highly elevated one time-interval) and A7 (decreasing MeHg). Pond A10 received water from A9 (decreasing MeHg) and delivered to A11. From a resource manager's perspective, if a relationship was expected based on pond interconnectivity then sediment MeHg levels would be lower in A8 (based on A5 and A7) and A10 (based on A9) than observed. Only MeHg levels in Pond A11 might be predicted from those measured in A10. However, sediment MeHg levels were likely influenced more by within pond conditions (e.g., water depth, phytoplankton levels) than by between pond connectivity. Discrete measures taken fall 2002, spring 2003, and winter 2004 in the Alviso ponds indicated increasing but seasonally variable chlorophyll concentrations from lower to higher salinity ponds (e.g. A9~5–9 mg/m^3, A11~20–84 mg/m^3, A12, A14, A15, A16~79–340 mg/m^3; Miles et al. unpublished USGS report). The spatial and temporal variabilities of chlorophyll probably influenced similar variability in MeHg levels. A system-wide assessment of how restoration activities may have affected MeHg concentrations is difficult because different management regimes were applied to ponds of different physical characteristics (particularly depth and salinity) during the study.

In the remaining Alviso ponds, the trend of sediment MeHg levels was: elevated (i.e., ≥2 ng/g) but stable in Pond A12 that received water from Pond A11 (elevated then increased); elevated then decreased in Pond A13 that received water from Pond A12; elevated then decreased in Pond A15 that received water from A13 and also from a tributary; substantially decreased in Pond A2E (received water from Pond AB1 that had stable sediment concentrations); and stable in Pond AB2 (received water from Bay).

Results from our time series sampling of the Alviso ponds between sampling interval Year 1 and Year 2 indicated that concentrations of Hg_t, (and to a lesser degree MeHg) varied more spatially than temporally. Spatial variation in sediment Hg_t from Alviso ponds was probably related to spatial heterogeneity of Hg in original parent material Bay sediment (Hornberger et al., 1999) as well as periodic dredging of sediment for levee maintenance in the pond complexes. Concentrations of MeHg did not vary substantially across Alviso ponds between Year 1 and Year 2, and the effect indicated due to time was probably attributed largely to the 2-fold change in MeHg concentrations in (time series) Pond A7. The sloped floor of Pond A7 was subjected to routine wetting and drying associated with a water management strategy designed to benefit shorebird foraging. Also, the decline in MeHg between Year 1 and Year 2 was associated with lower MeHg levels at sampling points at the northern end of Pond A7 where an inflow gate was installed in July 2004, which may indicate a delayed reduction in MeHg levels following an initial increase after management actions.

Concentrations of MeHg did not relate strongly to water depth and salinity or sediment pH and Fe_t in Year 1, but patterns were apparent by Year 2. Generally increased water depths across Ponds A3N, A7, A9, and A16 correlated with decreased sediment MeHg concentrations; Pond A12 was an exception. Cooler temperatures in deeper water might result in slower microbial rates and thus decreased MeHg production (Korthals and Winfrey, 1987; Gustin et al., 2006). Alternatively, the seasonal filling or draining of Pond A3N and sloped bathymetry of Pond A7 resulted in periodic wetting and drying of sediment conducive to MeHg production (Gilmour et al., 2004).

The positive relation between MeHg and sediment pH was unexpected because some studies have reported decreased MeHg production in acidified subsurface sediment (Winfrey and Rudd, 1990). However, most pH measures in our study ranged from ~7.0 to 8.0, and this narrow range limits inference on MeHg patterns relative to pH levels. Sediment Fe_t had little influence on MeHg concentrations even though Fe amendment in natural wetland sediment can reduce levels of bioavailable Hg complexes conducive for methylation (Mehrotra and Sedlak, 2005). However, Fe_t was measured in our study and not Fe (II) that Mehrotra and Sedlak (2005) found reduced net mercury methylation in their experiments. Thus it was not surprising that we did not find a relationship of Fe_t to MeHg concentrations in our study.

More importantly, levels of Hg_t did not covary with MeHg levels which probably indicated available Hg_t was not a limiting factor for Hg methylation (Lambertsson and Nillson, 2006). Conaway et al. (2003) reported a similarly weak correlation between Hg_t and MeHg in Bay sediments. Although outweighed by discrete Pond effects in our study, biogeochemistry likely exerts a stronger influence on Hg

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methylation than available concentrations of Hg_t in Bay salt pond sediment. Biogeochemical processes are highly complex (Ulrich et al. 2001) and factors such as organic matter (Lambertsson and Nillson, 2006) and sulfide activity and speciation (Mehrotra and Sedlak, 2005), which can have a strong positive effect on Hg methylation, were not determined in our study. Furthermore, the strength of relationships between physical covariates and MeHg generally weakened when spatial effects (i.e. Pond) were not controlled (Fig. 4B), which further indicates the complexity of factors affecting MeHg within specific salt ponds.

The Alviso salt pond system is unique from other similar systems because of elevated sediment Hg levels and the potential for continual cycling of Hg. Mercury is toxic with no known beneficial qualities and MeHg is the most toxic form of this element. Most studies of effects of Hg on natural systems or biota have been definitive in the laboratory but generally only suggestive in the environment (Burger and Gochfeld, 1997). Long-term study of Hg and natural resources in the Alviso pond system is an important and unique opportunity to understand potential effects on wildlife and their habitat. The Eden Landing salt ponds are suitable as reference sites for ecological studies of Hg because of comparatively low Hg levels in the physical environment and proximity to the Alviso system. These impounded, semi-closed water circulation systems are ideal for experimentation. Recent studies of San Francisco Bay salt pond systems are providing a wealth of information on potential effects of Hg on vertebrates in a natural setting (Ackerman et al., 2007, Eagles-Smith et al., 2008), but more intense and longer-term studies are necessary as these ponds transition from commercial to natural resource systems.

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