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Mercury cycling in agricultural and managed wetlands: A synthesis of methylmercury production, hydrologic export, and bioaccumulation from an integrated field study

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HIGHLIGHTS

- Ecosystem MeHg fluxes were studied in replicate wetlands managed for rice or wildlife.
- MeHg export from rice-growing wetlands was higher in winter compared to summer.
- MeHg production was similar among wetlands but export and soil MeHg varied.
- Contrasting effects of evaporation and transpiration were observed for water MeHg.
- Holding water on rice-growing wetlands may enhance in situ MeHg bioaccumulation.

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ABSTRACT

With seasonal wetting and drying, and high biological productivity, agricultural wetlands (rice paddies) may enhance the conversion of inorganic mercury (Hg(II)) to methylmercury (MeHg), the more toxic, organic form that biomagnifies through food webs. Yet, the net balance of MeHg sources and sinks in seasonal wetland environments is poorly understood because it requires an annual, integrated assessment across biota, sediment, and water components. We examined a suite of wetlands managed for rice crops or wildlife during 2007–2008 in California's Central Valley, in an area affected by Hg contamination from historic mining practices. Hydrologic management of agricultural wetlands for rice, wild rice, or fallowed — drying for field preparation and harvest, and flooding for crop growth and post-harvest rice straw decay — led to pronounced seasonality in sediment and aqueous MeHg concentrations that were up to 95-fold higher than those measured concurrently in adjacent, non-agricultural permanently-flooded and seasonally-flooded wetlands. Flooding promoted microbial MeHg production in surface sediment of all wetlands, but extended water residence time appeared to preferentially enhance MeHg degradation and storage. When incoming MeHg loads were elevated, individual fields often served as a MeHg sink, rather than a source. Slow, horizontal flow of shallow water in the agricultural wetlands led to increased importance of vertical hydrologic fluxes, including evapoconcentration of surface water MeHg and transpiration-driven advection into the root zone, promoting temporary soil storage of MeHg. Although this hydrology limited MeHg export from wetlands, it also increased MeHg exposure to resident fish via greater in situ aqueous MeHg concentrations. Our results suggest that the combined traits of agricultural wetlands — slow-moving shallow water, manipulated flooding and drying, abundant labile plant matter, and management for wildlife — may enhance microbial methylation of Hg(II) and MeHg exposure to local biota, as well as export to downstream habitats during uncontrolled winter-flow events.

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1. Introduction

Mercury (Hg) is widely regarded as a toxic metal with no known beneficial function for living organisms. Exposure of humans and wildlife to Hg is largely mediated through dietary consumption of methylmercury (MeHg), a chemical compound that is bioconcentrated in organisms and bioaccumulated in food webs. Decades of research have shown that anoxic sediment, especially surface sediment in wetland environments, promotes the microbial methylation of divalent inorganic mercury (Hg(II)) to MeHg (Hall et al., 2008; Lacerda and Fitzgerald, 2001; Marvin-DiPasquale et al., 2003; Windham-Myers et al., 2009; Zillioux et al., 1993). The production of MeHg in sediment is enhanced when both organic matter and Hg(II) are bioavailable to anaerobic bacteria (Marvin-DiPasquale et al., 2009a, 2009b), a combination of conditions that is commonly characteristic of wetland habitats (Merritt and Amirbahman, 2009). Furthermore, the large pool of dissolved organic matter (DOM) typical of wetlands can promote aqueous MeHg export to receiving surface water (Hall et al., 2008; Wallschläger et al., 1996). Wetlands also tend to be highly productive and provide important foraging habitat for fish and wildlife (Elphick, 2000). Thus, MeHg production associated with wetland habitats may be directly associated with biotic Hg exposure (Brumbaugh et al., 2001). In this study, we sought to compare patterns and processes of MeHg cycling among neighboring wetland habitats with different management practices, specifically wetlands managed for agriculture, and those managed for wildlife.

Among wetland habitat types, rice agriculture is predominant in temperate and tropical latitudes, covering 162 million ha globally (Leff et al., 2004; Mitsch et al., 2010). In California, rice fields — including wild rice (*Zizania* spp.) and domesticated or white rice (*Oryza* spp.) — represent a 2.5-fold greater amount of area than the remaining natural (non-agricultural) United States (U.S.) Fish and Wildlife Service-delineated wetland habitats (U.S. Department of Agriculture National Agricultural Statistics Service, 2007). Rice farming creates seasonally flooded agricultural wetlands (a.k.a. rice paddies) that typically are characterized by productive, shallow-water habitats. The microbial processes and biogeochemical conditions that develop in rice-field sediment are largely a function of the pulsed-flooding management regime that provides abundant water and nutrients, and in turn, the rapid production of relatively labile carbon by rice plants. The effect of rice agriculture on MeHg exposure to biota has not been rigorously addressed. Yet, recent data from China's Guizhou province have documented high MeHg bioaccumulation in white rice grains grown in mining impacted regions (Zhang et al., 2010), and highlighted the potential for MeHg exposure to humans and wildlife through rice agriculture (Feng et al., 2008). Recent research has demonstrated that atmospherically deposited Hg is especially susceptible to methylation in rice fields (Liu et al., 2012), and is thus a prime MeHg source for uptake into rice grains (Meng et al., 2011). Beyond MeHg in edible rice grains, the on-site production of MeHg within rice fields and the hydrologic export of MeHg from these systems may be an important source of MeHg contamination to local in situ aquatic food webs, as well as waterfowl, invertebrates, mammals, and other organisms downstream.

Agricultural wetlands provide important habitat for many fish and wildlife species (e.g., Elphick, 2000). A high density of wildlife, such as migratory waterbirds, may thus be exposed to elevated MeHg concentrations produced within these habitats. Even without contamination from local mining sources, the increasing global dependence on fossil fuels (particularly coal) in both industrial and developing countries may be increasing the rate of atmospheric Hg transport and deposition to rice-growing regions, such as southeastern Asia, the western United States, the Gulf of Mexico, Brazil, and India (Selin, 2009). The result may be that agricultural wetlands, especially rice fields, could become an increasingly important source of MeHg to wildlife and humans.

Given the regional and global significance of agricultural wetlands, and the potential for episodic and site-specific pulses in MeHg production, export, and bioaccumulation, we examined Hg pools and fluxes, and associated biogeochemical factors, in a suite of wetland habitats managed for either agriculture or wildlife purposes. Our primary goals were to: 1) quantify MeHg sources and sinks at the field scale; 2) relate sources and sinks to key biogeochemical processes; and 3) assess the relative importance of different wetland conditions that could be managed to mitigate MeHg production, export, and bioaccumulation. Herein, we synthesize the findings of seven independent research papers in this special section, focused on describing how Hg and MeHg pools and fluxes are linked in wetlands managed for agriculture (rice growing) and wildlife in California's Central Valley, U.S. In this synthesis, the value of multi-disciplinary, integrated datasets is examined for the elucidation of the relationships and factors driving Hg biogeochemistry in agricultural and managed, non-agricultural wetland settings.

2. Methods

2.1. Field setting

The Yolo Bypass (YB) is a low-elevation depression that has been engineered as a component of the Sacramento River Flood Control Project, routing high flows in the Sacramento River around the city of Sacramento, California (Fig. 1). The Yolo Bypass Wildlife Area (YBWA) is a 6800 ha preserve representing about one-quarter of the total acreage. The YBWA is owned by the State of California, and managed by the California Department of Fish and Game (CDFG) with the primary goal of flood control, and secondary goals of promoting wildlife habitat (especially for wintering waterfowl) and recreational opportunities, in a landscape mosaic comprised of diverse upland and wetland settings. Because the primary management goal is flood control, there are restrictions on the type and density of vegetation allowed to accumulate so that flows are not impeded. One wetland habitat type that serves all land management goals, as well as providing economic return, is rice cultivation. Approximately 34% of the YBWA is managed as agricultural wetlands for production of both white rice (*Oryza sativa*) and wild rice (*Zizania palustris*). Rice cultivation along with other agricultural operations within the YBWA (e.g. livestock grazing) contributes to the economic sustainability of the preserve by providing lease income to the State. Non-agricultural wetlands within YBWA consist of duckling brood ponds, open-water ponds, and winter-flooded seasonal wetlands that are mowed often to promote higher flow velocities when the YB is inundated by high river flows from snowmelt and reservoir releases (CDFG, 2008).

The YB resides in a Mediterranean (xeric) climatological zone which experiences hot, dry summers and cool, wet winters. Average daily air temperature ranges from 5 °C to 15 °C in January to 20 °C to 35 °C during July and August. The area has average annual precipitation of approximately 50 cm, which typically occurs between October and April. Despite high evaporation rates through the summer months, due to intense solar radiation and low-humidity winds, wetlands naturally persist within the low-lying YB naturally due to surrounding flows of the large delta watershed that integrates surface flows from the Sierra Nevada, the Cascades and the Coast Range Mountains.

Water management within the YBWA is complex. During summer low-flow conditions, the dominant sources of water are agricultural drains from the north and west (Toe and Davis Drains). This water is made available by pumping based on Pacific Ocean tides, from the Sacramento–San Joaquin Delta (Delta) to the south (Fig. 1). This area is considered the northernmost extent of the statutory San Francisco Bay–Delta since it is affected by ocean tides. Water management during summer dry conditions is primarily for agriculture and includes extensive within-basin water recycling. The primary conduit of source water in summer is the Toe Drain on the eastern edge of the YB along the Sacramento River levee (Fig. 1). Toe Drain water is pumped into the YBWA via several lateral ditches running east–west (South Supply, and Central Unit Supply Ditch). The lateral ditches transport and distribute the Toe Drain water west to the agricultural fields that were part of