



Reprint of “Methylmercury production in and export from agricultural wetlands in California, USA: The need to account for physical transport processes into and out of the root zone”[☆]



P.A.M. Bachand^{a,*}, S.M. Bachand^a, J.A. Fleck^b, C.N. Alpers^b, M. Stephenson^c, L. Windham-Myers^d

^a Bachand & Associates, Davis, CA 95618, United States

^b U.S. Geological Survey, California Water Science Center, Sacramento, CA 95819, United States

^c California Department of Fish and Game, Moss Landing, CA, United States

^d U.S. Geological Survey, Western Region Bureau of Regional Research, Menlo Park, CA, United States

HIGHLIGHTS

- Cl^- was used to distinguish MeHg root zone loading from ecosystem production.
- Comparing [MeHg] to [Cl] enables identification of net MeHg ecosystem production.
- Summer transpiration moves MeHg into the root zone; winter diffusion releases.
- Winter MeHg exports result from upward benthic flux and net ecosystem production.
- Internal hydrologic pathways decouple processes of MeHg production and export.

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ABSTRACT

Concentration and mass balance analyses were used to quantify methylmercury (MeHg) loads from conventional (white) rice, wild rice, and fallowed fields in northern California's Yolo Bypass. These analyses were standardized against chloride to distinguish transport pathways and net ecosystem production (NEP). During summer, chloride loads were both exported with surface water and moved into the root zone at a 2:1 ratio. MeHg and dissolved organic carbon (DOC) behaved similarly with surface water and root zone exports at ~3:1 ratio. These trends reversed in winter with DOC, MeHg, and chloride moving from the root zone to surface waters at rates opposite and exceeding summertime root zone fluxes. These trends suggest that summer transpiration advectively moves constituents from surface water into the root zone, and winter diffusion, driven by concentration gradients, subsequently releases those constituents into surface waters. The results challenge a number of paradigms regarding MeHg. Specifically, biogeochemical conditions favoring microbial MeHg production do not necessarily translate to synchronous surface water exports; MeHg may be preserved in the soils allowing for release at a later time; and plants play a role in both biogeochemistry and transport. Our calculations show that NEP of MeHg occurred during both summer irrigation and winter flooding. Wild rice wet harvesting and winter flooding of white rice fields were specific practices that increased MeHg export, both presumably related to increased labile organic carbon and disturbance. Outflow management during these times could reduce MeHg exports. Standardizing MeHg outflow:inflow concentration ratios against natural tracers (e.g. chloride, EC) provides a simple tool to identify NEP periods. Summer MeHg exports averaged 0.2 to 1 $\mu\text{g m}^{-2}$ for the different agricultural wetland fields, depending upon flood duration. Average winter MeHg exports were estimated at 0.3 $\mu\text{g m}^{-2}$. These exports are within the range reported for other shallow aquatic systems.

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1. Introduction

Mercury (Hg) contamination in aquatic systems is considered as one of the most difficult environmental issues to understand and to manage.

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* Corresponding author.

Over the last 25 years, an improved understanding of Hg cycling in wetlands has developed, including the mechanisms and processes that lead to its methylation and ultimate bioaccumulation in the food chain (see reviews by Merritt and Amirbahman, 2009; Hsu-Kim et al., 2013; Driscoll et al., 2013). Production of MeHg involves complex relationships between electron donors such as dissolved organic matter, electron acceptors including sulfate and ferric iron, and Hg availability for methylation (Marvin-DiPasquale et al., 2013). Sulfate (SO_4^{2-}) can

play a complex role in MeHg production, stimulating MeHg production at lower concentrations and inhibiting MeHg production as reduced S compounds reach higher concentrations (Gilmour et al., 1992; Marvin-DiPasquale et al., 2013). Although methylation of reactive divalent mercury (Hg(II)) by sulfate-reducing bacteria is considered the primary source of MeHg in most wetlands (King et al., 2002; Branfireun et al., 1999), in the agricultural wetlands in the Yolo Bypass, located along the Sacramento River in California's Central Valley, the energy flow which appears dominated by iron (Fe) reduction, is an additional or alternative source of MeHg (Marvin-DiPasquale et al., 2013). Furthermore, organic matter may also play conflicting roles depending on its source and character, with labile forms stimulating MeHg production and other forms limiting it (Guimaraes et al., 2000; Hall et al., 2008; Lambertsson and Nilsson, 2006; Windham-Myers et al., 2014b–in this issue, 2014c–in this issue). In addition to the production processes, MeHg degradation through biotic and abiotic processes occurs at varied temporal and spatial scales, ultimately affecting ecosystem net production rates (Fleck et al., 2013; Marvin-DiPasquale et al., 2013; Windham-Myers et al., 2014a–in this issue). Because of the favorable conditions for microbial methylating activity and Hg bioavailability, wetland sediments are often identified as principle sites for the production of methylmercury (MeHg) within watersheds (e.g. Hall et al., 2008).

Less work has been conducted in linking Hg and MeHg contamination and net ecosystem production to hydrology. Hydrology is often viewed primarily with regard to spatially large-scale transport pathways. Hydrologic budgets are integrated with water quality concentrations to calculate mass budgets and ascertain if systems are net constituent sinks or sources (Carleton and Montas, 2009; Kadlec, 1994; Kadlec and Knight, 1996; Liu et al., 2008; Martinez and Wise, 2003; Miles and Fink, 1998; Persson and Wittgren, 2003; Werner and Kadlec, 2000). Under these approaches, evapotranspiration, which combines evaporation and transpiration into a single term, is typically treated similarly to evaporation in shallow aquatic systems, providing a means to concentrate constituents within surface waters.

This hydrologic approach is fundamentally too simplistic when considering shallow aquatic systems. In all systems, root hairs provide a large surface area for plants to move water from the soil into the plants (Raven et al., 1999). During periods of rapid transpiration, soils adjacent to the root zone may have insufficient water, causing water from further distances away to move towards the root hairs (Raven et al., 1999). This bulk flow, an advective process, is essentially driven by a hydraulic pressure gradient with root hairs causing suction by depleting the pressure near the root surface and increasing the pressure gradients relative to other neighboring areas with higher pressure values (Taiz and Zeiger, 1991). This process also serves to move nutrients and other dissolved constituents towards plant roots through the soil matrix (Brady and Weil, 2002; Taiz and Zeiger, 1991; Kadlec, 1999; Reddy et al., 1999). It is estimated that transpiration accounts for 50–90% of evapotranspiration (ET) in shallow aquatic systems (Bachand et al., 2014–in this issue; Bouman et al., 2005; Herbst and Kappen, 1999; Sanchez Carrillo et al., 2004), which drives the transport of surface waters past the soil–water interface and into the root zone for uptake by the plants (Howes et al., 1986; Van der Kamp and Hayashi, 2009; Parsons et al., 2004; Whitmer et al., 2000). Not accounting for this advective transport can result in misinterpretation of the processes leading to changes in surface water outflow concentrations (Bachand et al., 2014–in this issue), including seasonal transport from the surface water to the root zone (summer) and back (winter), and changes in concentrations of some constituents in the root zone. Further, transpiration may result in seasonal exchanges between soils and water (Bachand et al., 2014–in this issue). When transpiration ceases, diffusive fluxes would be expected to release Hg and other water quality constituents from the soils into the water column. Transport of MeHg from sediments into the water column potentially enhances biotic exposure to Hg in aquatic food webs in situ, as well as downstream (e.g. Chasar et al., 2009). Given the interrelationships between biogeochemistry and hydrology, our

abilities to understand the impacts of controls on MeHg fate and transport are challenged (e.g. Marvin-DiPasquale et al., 2013; Windham-Myers et al., 2014a–in this issue).

In this paper, the transport and cycling of MeHg and other water quality constituents through agricultural wetlands were assessed in two ways: 1) by considering MeHg concentrations from inflow to outflow, and 2) through the development of mass budgets for Hg and other water quality constituents. The agricultural wetlands studied included conventional (hereafter, white) rice, wild rice, and fallowed fields located in the Yolo Bypass adjacent to the Sacramento River in northern California. Chloride (Cl^-) was used as a natural tracer (Bachand et al., 2014–in this issue) in order to separate effects associated with hydrologic transport from those associated with biogeochemical cycling and processes. Using Cl^- as a tracer is consistent with other studies in which bromide or electrical conductivity (EC) were used as natural tracers (Hayashi et al., 1998; Parsons et al., 2004; Pellerin et al., 2007; Whitmer et al., 2000). Concentration changes of all water quality constituents occur because of hydrologic transport and effects. These effects are captured through assessing tracers. The concentration changes of MeHg from inflow to outflow when standardized against concentration changes of Cl^- provide a means of identifying concentration changes resulting from biogeochemical cycling and processes, and in effect, enable us to identify periods in which net ecosystem production or consumption of MeHg are occurring. Our mass balance analyses, standardized against Cl^- , enable us to quantify MeHg loading to and from the root zone due to hydrologic transport as compared to from biogeochemical cycling causing net ecosystem production or consumption of MeHg. We hypothesized that hydrologic analyses and use of Cl^- as a conservative tracer would provide a framework for understanding biogeochemical processes that affect MeHg and other constituent flux and storage.

2. Methods

In this section, we describe the study site and its management; the experimental design and associated field and laboratory methods; and data management, data analyses and standardization of those analyses to Cl^- . Supplemental information contains maps, conceptual models, tables on system cultural and hydrologic practices, field hydrology, water quality, and concentration and loading changes for Hg and other water quality constituents.

2.1. Study site

Six agricultural wetlands within the Yolo Bypass Wildlife Area (YBWA), California (USA), were monitored for water flow and chemistry between June 2007 and May 2008 (Supplemental information, Fig. S1; Windham-Myers et al., 2014a–in this issue). The wetlands included two white rice fields (R31, R64), two wild rice fields (W32, W65), and two fallowed fields (F20, F66), ranging in size from 30 to 80 ha. Area soils are characterized as silty clay loam (California Department of Fish and Game et al., 2008) and contain elevated Hg concentrations. Legacy gold and Hg mining in the upstream watersheds has resulted in elevated soil Hg concentrations. For instance, surface sediment (0–2 cm) THg concentration data for the study area had a range of 107–434 ng/g dry wt., with a median of 296 ng/g dry wt. and a mean (\pm std. dev) of 267 \pm 104 (N = 56), as calculated from data provided in Marvin-DiPasquale et al. (2013). Agricultural operations and hydrologic management are described in detail elsewhere (Windham-Myers et al., 2014a–in this issue; Bachand et al., in this issue). In summary, irrigation water was delivered onto the fields through gate valves or weir structures from nearby irrigation supply ditches. Each field was subdivided into a series of check ponds/fields (checks) separated by berms set along elevation contours (Fig. S2). Surface water exited the fields through weirs into outflow ditches and drains. During winter, water management was more problematic due

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