

Mercury in lagoons: An overview of the importance of the link between geochemistry and biology

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ABSTRACT

Shallow-water lagoons, which are common features along coastlines, are important sites for elemental cycling in this environmentally-sensitive terrestrial–marine interface. Factors governing mercury (Hg) cycling in these lagoons are poorly characterized, but critical to understanding the links between sources and higher trophic levels, that are ultimately vectors of human exposure in lagoon environments. This article discusses the processes controlling the fate of Hg from various sources, including methylation of Hg, demethylation of methylmercury, and benthic fluxes of Hg species in three of the most thoroughly studied lagoons worldwide, namely Thau (France), Venice (Italy) and Marano and Grado (Italy). Although each lagoon system experiences differences in sources of Hg and details of how Hg is transformed and transported, Hg in each system is strongly affected by biogeochemical transformations of other elements, especially redox sensitive, microbially important elements such as sulphur, iron and manganese, and their interaction with organic matter. The shallow nature of lagoons and the rapid rates of microbially mediated organic matter decomposition result in seasonally dynamic processes that influence Hg bioavailability. Despite considerable work to date, the current understanding of Hg dynamics in lagoon ecosystems, through Hg distribution, MeHg production and degradation, and trophic transfer, is still limited and more research is needed to link all subparts into a general coherent picture.

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1. Introduction

Coastal lagoons are relatively protected shallow marine areas along the coastal margins that occupy about 13% of the world's coastline. They are characterized by poor biodiversity and high productivity, and are important commercially due to their extensive fisheries and mariculture and their role in protecting harbours. Human influences impose stress from physical changes in the lagoon and hinterland (run-off) and from chemical contamination. Lagoons are very sensitive to chemical contamination because of the long hydrologic residence times (Kjerfve, 1994) and consequently long-term retention of pollutants (Mee, 1978). This is especially acute for contamination by heavy metals, for example Hg, which is often present in industrial and municipal effluents. In this overview, we compare three lagoons, namely the Thau, Venice and Marano-Grado (Fig. 1), where, to our knowledge, the

distribution of Hg and MeHg, and Hg methylation rates are known and we are able to decipher the basic mechanisms influencing the MeHg production in a lagoon environment.

2. Distribution and fluxes of mercury in lagoons

2.1. Thau Lagoon

In the eutrophic Thau Lagoon (French Mediterranean), Hg biogeochemistry was investigated primarily in terms of the Hg and MeHg contents in the sediment solid phase, pore-waters and overlying waters (Muresan et al., 2007) but Hg methylation potential was also determined (Monperrus et al., 2007). Total Hg (THg) and reactive Hg (RHg) in the water column varied between 0.3 and 1.11 ng/L and between 0.1 and 0.6 ng/L, respectively, with summer values approximately double those in winter. Depth profiles followed temperature profiles, with maxima at the surface in winter, and at 1–2 m depth in summer. Conversely, the highest MeHg and dissolved gaseous Hg (DGM) concentrations occurred in the bottom water layer and varied from 0.02 to 0.13 ng/L and

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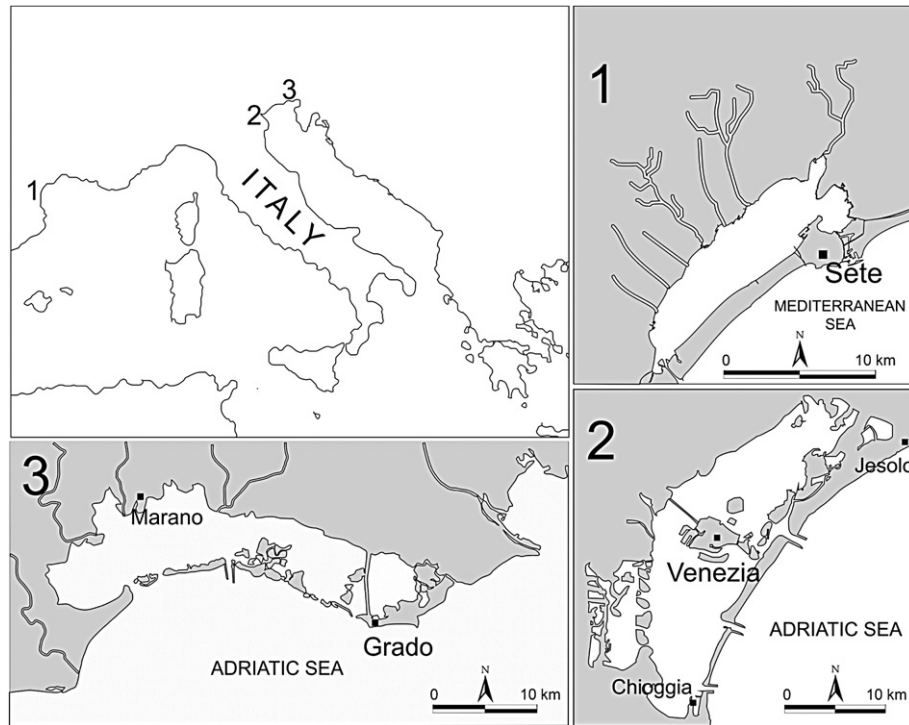


Fig. 1. Location of three coastal lagoons considered in this review.

between 0.02 and 0.14 ng/L, respectively. The sediment solid phase THg in Thau Lagoon sediments (Table 1) was the result of more than 100 years of contamination due to agricultural and municipal sources, the harbour of Sète and a highly industrialized area (Pena and Picot, 1991; Szefer et al., 1999), with a slight increase with depth due to a decrease in anthropogenic Hg deposition over time. Conversely, total MeHg decreased with depth to a constant

concentration below 30 mm, indicating net MeHg production near the sediment–water interface, but net degradation in deeper sediments (Muresan et al., 2007). The horizontal distribution of THg and MeHg was similar with higher MeHg levels noted near areas of oyster farms. Negative correlations between Hg and sedimentary organic matter suggested diagenesis was not controlling the sedimentary distribution of Hg, and that historical loading is the most important factor controlling the vertical distribution of THg (Muresan et al., 2007).

Wide variations of pore-water THg and MeHg concentrations were encountered among various cores collected in the Thau Lagoon (Table 1). Partitioning between solid and dissolved phases resulted in equilibrium in lower strata where solid phase S minerals, including HgS control dissolved Hg levels. Conversely, in surficial suboxic sediments, solid phases act as sources of dissolved Hg and the depth distribution of solid and dissolved Hg species mirror each other (Muresan et al., 2007). The bimodal vertical distribution of the pore-water dissolved MeHg implies its formation by methylation at the sediment–water interface, as noted above, and by sulphate reducing bacteria (SRB) in the deeper accumulating zone, but the seasonal variation in the depth of the redox gradient, and subsequent variations in the distribution of neutral Hg sulfide complexes (Benoit et al., 2001), affected the vertical distribution of MeHg. At the sediment–water interface, oxyhydroxides of Fe, and especially Mn, also strongly affect the mobility of THg and MeHg.

Diffusive fluxes of Hg and MeHg, measured in summer, were much lower than those measured with *in situ* flux chambers (Point et al., 2007). Discrepancies, caused by diagenetic processes occurring at the sediment–water interface and seasonal changes in bioturbation/bioirrigation and photosynthesis/respiration, all affected the exchange of Hg species between sediments and overlying waters. Hg methylation in sediments, averaging $12 \text{ mmol m}^{-2} \text{ d}^{-1}$, was considered a major process controlling Hg cycling despite the fact that methylation was also detected in the water column, albeit at a lower

Table 1

Hg speciation in surface sediments (THg, MeHg) and pore waters (DHg, DMeHg) with respective distribution coefficients (K_d), pore water sulphide and iron concentrations, potential methylation rates (k_{meth}) and mean diffusive benthic fluxes of Hg and MeHg in Thau, Venice and Marano and Grado lagoons.

	Thau	Venice	Marano sector	Grado sector
THg ($\mu\text{g/g}$)	0.3–0.46 [†]	0.5–2.51 [†]	1.22–4.49 ^{††}	9.5–14.4 ^{†††}
MeHg (ng/g)	0.76–2.75 [†]	0.31–1.7 [†]	1.14–1.7 ^{††}	3.11–21.9 ^{†††}
%MeHg	0.02–0.8 [†]	0.1–0.3 [†]	0.04–0.09 ^{††}	0.001–0.2 ^{†††}
DHg (ng/L)	3.0–17.1 [†]	0.38–29.7 [‡]	9.6–675.1 ^{‡‡}	5.76–231 ^{†††}
$\log K_d$	4.92 [†]	4.56–5.01 [‡]	3.34–5.6 ^{‡‡}	5.31 ^{†††}
DMeHg (ng/L)	0.01–0.43 [†]	0.04–3.24 [‡]	1.2–6.5 ^{‡‡}	1.61–7.9 ^{†††}
$\log K_d$	2.96–4.36 [†]	1.96–3.3 [‡]	2.84–4.11 ^{‡‡}	2.01–3.72 ^{†††}
Corg. (%)	2.8–6.7	1–2 [‡]	0.8–1.1 ^{††}	1.2–2.7 ^{†††}
H ₂ S (mM)	0.02–24.2 [†]	up to 10 [‡]	0.001–0.01 ^{††}	0.002–0.01 ^{†††}
Fe _{diss.} (μM)	up to 200 [†]	up to 60 [‡]	5–20 ^{††}	5–20 ^{†††}
k_{meth} (% day ⁻¹)	0.2–1.32	0–4.8 [‡]	0–2.2 ^{§§}	0–5.4 ^{§§}
Diffusive fluxes:				
Hg ($\text{ng m}^{-2} \text{ day}^{-1}$)	8 ^{**}	9.45 [§]	171 ^{††}	57 ^{†††}
MeHg ($\text{ng m}^{-2} \text{ day}^{-1}$)	0.9 ^{**}	1.1 [§]	0.7 ^{††}	7.8 ^{†††}

[†] Bloom et al., 2004.

[‡] Han et al., 2007 (using ²⁰⁰Hg).

[§] Guedron et al., 2012.

^{††} Muresan et al., 2007.

^{||} Metzger et al., 2007, Monperrus et al., 2007 (using ¹⁹⁹Hg).

^{**} Point et al., 2007.

^{†††} Acquavita et al., 2012.

^{§§} Emili et al., 2012.

^{§§§} Hines et al., 2012 (using ²⁰³Hg).

^{††††} Covelli et al., 2008.

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