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Historical flux of mercury associated with mining and industrial sources in the Marano and Grado Lagoon (northern Adriatic Sea)

Stefano Covelli [a,*], Leonardo Langone [b], Alessandro Acquavita [a,c], Raffaella Piani [a], Andrea Emili [a]

[a] Dipartimento di Geoscienze, Università di Trieste, Via Weiss 2, 34128 Trieste, Italy
[b] CNR, Istituto di Scienze Marine (ISMAR), via Gobetti 101, 40129 Bologna, Italy
[c] Osservatorio Alto Adriatico, Agenzia Regionale per la Protezione dell'Ambiente del Friuli Venezia Giulia (ARPA-FVG), Via Cairoli 14, 33057 Palmanova, Italy

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ABSTRACT

The "MIRACLE" Project was established in order to assess the feasibility of clam farming and high levels of sediment mercury (Hg) contamination coexisting in the Marano and Grado Lagoon, Italy. This lagoon has been subjected to Hg input from both industrial waste (chlor-alkali plant) and long-term mining activity (Idrija mine, NW Slovenia). One of the subtasks of the "MIRACLE" Project was to determine the historical evolution of Hg accumulation in the lagoon's bottom sediments. Thirteen 1-m deep sediment cores were collected from the subtidal and intertidal zones, plus one in a saltmarsh, all of which were then analyzed for total Hg content and several physicochemical parameters. Sedimentation rate assessments were performed by measuring short-lived radionuclides (excess ^{210}Pb and ^{137}Cs). For most of the analyzed cores, natural background levels of Hg were observed at depths of 50–100 cm. In the eastern area, Hg contamination was found to be at its maximum level at the core top (up to 12 μg g^{-1}) as a consequence of the long-term mining activity. The vertical distribution of Hg was related to the influence of the single-point contamination sources, whereas the grain-size variability or organic matter content seemed not to affect it. In the western area, Hg content at the surface was found not to exceed 7 μg g^{-1} and contamination was recorded only in the first 20–30 cm. Geochronological measurements showed that the depositional flux of Hg was influenced by anthropogenic inputs after 1800, when mining activity was more intense. After 1950, Hg in the surface sediment, most remarkable in the central-western sector, seemed to also be affected by the discharge of the Aussa River, which delivers Hg from the chlor-alkali plant. In 1996, Hg mining at Idrija ceased, however the core profiles did not show any subsequent decreasing trend in terms of Hg flux, which implies the system retaining some "memory" of contamination. Thus, in the short term, a decrease in Hg inputs into the nearby Gulf of Trieste and the lagoon seems unlikely. A preliminary rounded-down gross estimate of total Hg "trapped" in the lagoon's sediments amounted to 251 t. Such a quantity, along with the complexity of the lagoon ecosystem, suggests that an *in toto* reclamation of the sediments at the lagoon scale is unfeasible, both economically and environmentally.

1. Introduction

Sediments in shallow coastal waters act as sinks for metals originating from both natural and anthropogenic sources. Fine-grained sediments in particular show an ability to accumulate metals from atmospheric deposition, from soil erosion, through transport from watersheds downstream to the aquatic environment, or directly from wastewater discharge. Among the metals found in coastal sediments, mercury (Hg) is of particular concern with regards to contamination due to both its high mobility (Fitzgerald et al., 2007) and the toxicity and bioavailability of its organic form (methylmercury) within the food web (Morel et al., 1998; Benoit et al., 2003). Many coastal ecosystems act as large reservoirs of Hg through their sediments (Covelli et al., 2001; Sager, 2002; Canário et al., 2005), and a number of controlling physical and chemical factors, such as grain-size, organic matter content (e.g. Benoit et al., 1998; Mason and Lawrence, 1999), co-precipitation with iron (Fe) and manganese (Mn) oxides (Gagnon et al., 1997; Muresan et al., 2007), and binding to sulphides (Gagnon et al., 1997; Fabbri et al., 2001), have been shown to affect the distribution of Hg within those sediments.

* Corresponding author. Present address: Dipartimento di Geoscienze, Università di Trieste, Via Weiss, 2, 34128 Trieste, Italy.
E-mail address: covelli@units.it (S. Covelli).

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