Marine Pollution Bulletin 105 (2016) 286–291

Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

The role of transport processes of particulate mercury in modifying marine anthropogenic secondary sources, the case of Haifa bay, Israel

Merav M. Bareket [a,b], Revital Bookman [a,**], Regina Katsman [a], Henko de Stigter [c], Barak Herut [a,b,*]

[a] Dr. Moses Strauss Department of Marine Geosciences, Leon H.Charney School of Marine Sciences, University of Haifa, Haifa 31905, Israel
[b] Israel Oceanographic and Limnological Research, National Institute of Oceanography, Haifa 31080, Israel
[c] Royal Netherlands Institute for Sea Research (NIOZ), NL-1790 AB, Den Burg, Texel, Netherlands

ARTICLE INFO

Article history:
Received 19 September 2015
Received in revised form 29 January 2016
Accepted 4 February 2016
Available online 15 February 2016

Keywords:
Mercury
Marine sediment
Resuspension
Mediterranean
Anthropogenic pollution

ABSTRACT

We have assessed the redistribution of a secondary source of sedimentary anthropogenic mercury in the Haifa bay (HB) area (SE Levantine basin), which is the northern sink for Nile-driven sand. A long-term (30 years) ~80% decrease of the total sedimentary mercury concentrations (THg) was recorded in the inner bay, while an up to 3-fold increase was recorded in the top sediments of the outer bay. Sedimentary THg depth profiles and their temporal variability were used to model the main re-distribution processes, mainly resuspension associated with winter storm-derived transport. This mechanism transforms a secondary, sandy and well-aerated sink into a tertiary, more silty and hypoxic source at adjacent peripheral areas, affecting mercury bioavailability. We revisited the concept of environmental relaxation, i.e. the rate of return of a polluted environment to an acceptable state, showing that sedimentary transport processes may affect the associated ecological risks, mainly at shallow-water coastal sites.

1. Introduction

Anthropogenic activities have significantly increased (by several factors) mercury concentrations in the oceans and in marine animals (Mason et al., 2012; UNEP, 2013a, 2013b). Significant amounts of anthropogenic Hg are found in the environment due to its complex chemical cycles, which take place between reservoirs and its conservative behavior across the food chain (Selin, 2009). While the effects of the reduction in emissions will take some time to become apparent, and in some areas like Asia emissions still increase (Mason et al., 2012; UNEP, 2013a, 2013b), the United Nations Environment Program (UNEP) is currently leading negotiations towards a global, legally-binding instrument for reducing the emissions of global anthropogenic mercury (Hg) and its use in products (Minamata Convention; UNEP, 2013b). These international efforts to reduce releases of mercury and the associated depositions to the ocean may result in a decline in human exposure and risk.

Since in marine environments Hg is intensely absorbed by sediment pelitic particles (Covelli et al., 2001) or by detritic or live organic matter (phytoplankton, bacteria, algae), its migration and accumulation is associated with the deposition and recycling of these particles (Ullrich et al., 2001; Di Leonardo et al., 2006). Nevertheless, the evasion of elemental mercury (Hg0) from water surfaces (water–air flux) and the release of Hg from sediments into seawater due to the formation of mono-methylmercury (CH_3Hg) and dimethylmercury ($(CH_3)_2Hg$) are both key processes in biogeochemical mercury cycling that contribute to loss from marine sediments (Bouchet et al., 2011; Kuss, 2014).The Levantine basin is characterized by relatively low background concentrations of sedimentary total Hg (THg), compared to other sedimentary provinces in the Mediterranean Sea (Di Leonardo et al., 2006; Ogrinc et al., 2007), and is thus sensitive to anthropogenic impacts. Averaged sedimentary THg background concentrations of 10–25 ng g^{-1} were reported in sediments from the Israeli Mediterranean continental shelf (Herut et al., 1993; Herut and Galil, 2000; Shoham-Frider et al., 2007), 4–10 times lower than the overall mean background concentration of the Mediterranean Sea (100 ng g^{-1}).

Haifa Bay (HB), located in the SE Levantine Basin in the eastern Mediterranean Sea (Fig. 1), is considered as the northern terminal deposition basin of Nile-driven sand (Carmel et al., 1985; Zviely et al., 2009), which is transported anti-clockwise by long-shore and wind-induced currents (Zviely et al., 2007). During the last century a series of dams was built along the Nile, and since the operation of the Aswan High Dam 1965 almost all of the discharge to the southeastern (SE) Mediterranean stopped, including ~10^7 t/y of fine sediments (Halim et al., 1995). Moreover, several marine structures (ports, marinas, breakwaters, etc.) were introduced along the shoreline, impacting the transport of sand within the Nile littoral cell. This is an ongoing process that

* Correspondence to: B. Herut, Israel Oceanographic and Limnological Research, National Institute of Oceanography, Haifa 31080, Israel.
** Corresponding author.
E-mail addresses: rbookman@univ.haifa.ac.il (R. Bookman), barak@ocean.org.il (B. Herut).

http://dx.doi.org/10.1016/j.marpolbul.2016.02.014
0025-326X/

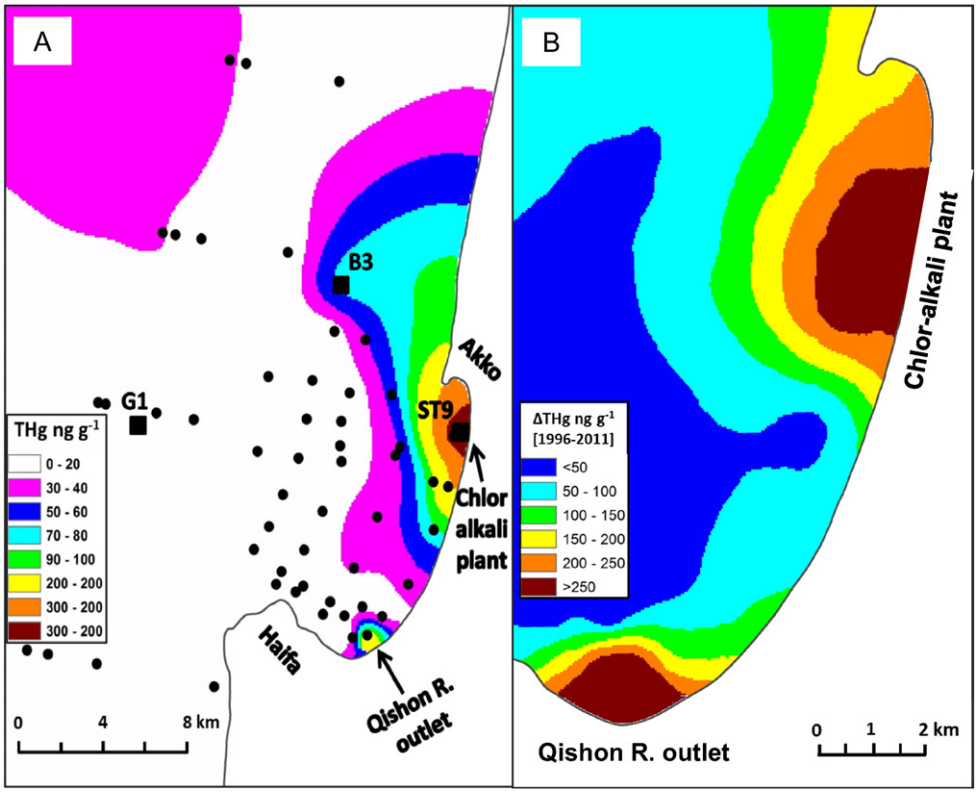

Fig. 1. A map of the Haifa bay (HB) area that shows the location of the sampling stations of surficial and sediment cores, and the spatial distribution pattern of the total Hg (THg) concentrations (μg g^{-1}) in the surface sediments during 2011 (A). A calculated differential map of the past 1994 (Herut et al., 1996) THg concentrations subtracted from the recent 2011 (this study) THg concentrations (μg g^{-1}) (B). The differential map shows that most of the reduction in THg concentrations (>200 ppb) occurred in the most polluted areas at shallow water depths, in close proximity to the location of the past effluent discharge and the Qishon outlet (representing 55–70% loss).

completely changed the hydrological regime in the SE Mediterranean. The alteration of sediment supply and transport may impact the role and reactivity of secondary sources of sedimentary pollutants.

Two main point sources of mercury pollution were found in HB: (1) a historically significant point source of elemental mercury discharge from a chlor-alkali plant (Electro-Chemical Industries) located at the northern part of the bay (Fig. 1), which operated until 2004 (Herut et al., 1996); and (2) the Kishon river estuary, located at the southern part of HB, which contains effluent from nearby industries (Herut and Kress, 1997; Herut et al., 1993, 1999). Diffusive sources of atmospheric deposition and runoff should be taken into account as well (Herut and Galil, 2000). The chlor-alkali plant was established in 1956, and it initially operated without any environmental regulation (Hornung et al., 1989; Krom et al., 1990). In 1974 effluent treatment facilities were installed, reducing the mercury flux into the bay by 90%, to less than 100 kg y^{-1}, an amount that was further reduced in 1981 to a few kg per year (Herut et al., 1996; Krom et al., 1990). The calculated total mercury influx from the plant since its establishment was approximately 20,000 kg (Herut et al., 1996; Krom et al., 1994).

The sediments in HB have been monitored annually since 1978, as part of the National Monitoring Program that is implemented along the Israeli Mediterranean coast (Herut et al., 2014). During this period, significant variations in the THg concentrations were found in the surface sediments and depth cores of the northern part of HB, while methyl Hg concentrations were very low (Shoham–Frider et al., 2012). In this study we evaluated the dynamics of the sedimentary secondary source of THg and its role in generating a tertiary source outside the bay. We have assessed the potential environmental role of this historical sediment sink by spatially and temporally mapping the sedimentary scavenged THg and by modeling the amplitude and rate of the transport processes. We were able to use the historical THg pollution in HB as a case study due to its unique conditions as the terminus of the Nile littoral cell. This allowed the re-consideration of the concept of environmental relaxation, i.e. the rate of return of a polluted environment to an acceptable state, and the role of transport processes in contaminant-derived ecological risks.

2. Methods

2.1. Sampling

Three short sediment cores and sixty-two surface sediment samples were collected during 2009–2012. The cores were retrieved from the outer northern (B3, a water depth of 36 m, core length 25.5 cm) and western (G1, a water depth of 66 m, core length 22 cm) regions off HB (Fig. 1) using a box-corer, and from the north-eastern part of HB (ST9, a water depth of 6 m, core length 23 cm), opposite the former chlor-alkali plant outfall, by diving. The latter sampling point (ST9) has a relatively long historical record of depth core measurements. Previous results of THg concentrations in short sediment cores (from years 1985, 1989, 1993, 1997, 2001, 2005, 2008; Herut et al., 1996; Herut and Galil, 2000 and the National Monitoring Program of Israel's Mediterranean coastal waters) were used here for the modeling (Fig. 2ab, sampled on 1993, 2001 and 2011-this study) and to show the long-term decrease in THg (Fig. 2c, all sampled years). The sixty-two surface samples collected by grab covered HB and the surrounding area (Fig. 1). The cores were sectioned on board the R.V. Shikmona immediately after extraction, or after being frozen and then thawed. All samples were sealed and frozen within hours, lyophilized for 48 h and then stored until further analysis was performed.

2.2. Analytical methods

Samples were sieved through a 250 μm plastic sieve in order to concentrate the dominant (>98%) finer sediment fractions. Approximately