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Mercury in sediments and *Nassarius reticulatus* (Gastropoda Prosobranchia) in the southern Venice Lagoon

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Abstract

The southern basin of the Venice Lagoon has been the focus of fewer studies concerning contamination from heavy metals than the northern and central basins. A recent increase in urban waste waters from Chioggia town, as well as dockyards, shipping and fishing activities, affect this part of the lagoon. The aim of this study was to investigate the total mercury (THg) incidence in sediments and *Nassarius reticulatus* gastropods in order to assess its distribution and evaluate the level of contamination. THg concentration measured in bottom sediments ranged between 0.1 and 3.4 mg/kg d. wt. The enrichment factor (EF) showed high values (avg. 30, max 49) near the dockyards of Chioggia; the lowest (avg. 9, max 17) were found in the coastal marine sediments near the port entrance of the southern basin. THg in marine scavenger gastropods accumulated in *N. reticulatus* with concentrations falling within the range of 0.3–1.3 mg/kg d. wt. A positive correlation was found between THg concentration in sediments and in *N. reticulatus* in all sites, excluding the dockyards. A first local cause for mercury pollution might be attributed to the antifouling paints used in great quantity in the recent past near the town of Chioggia. Moreover, fine suspended sediments associated with tidal flushing are suggested as possibly being the vehicle for pollutant dispersal from the Marghera industrial area to the whole of Venice's lagoon.

Keywords: Mercury; Sediment; *Nassarius reticulatus*; Venice Lagoon; Gastropoda

1. Introduction

The aquatic ecosystem of the Venice Lagoon has a high vulnerability due to the human impact caused by urban settlements and industrial activities. Since the end of the Second World War and during the industrial expansion of the area, pollution from several chemical compounds, including mercury (Hg), quickly increased in the Venice Lagoon, accumulating in the bottom sediments (Pavoni et al., 1992). The central basin was recognised as the most polluted area of the Venice Lagoon, as it was affected not only by atmospheric emissions and effluents from the Marghera industrial zone, but also by urban sewage from Venice and by shipping activities.

The Venice Lagoon was affected by uncontrolled discharges of Hg released from mercury cells in the chloro-alkali plant in the industrial complex of Mar-

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ghera from the 1950s to the 1980s (Locarnini and Presley, 1996; Provincia di Venezia, 2002). A decrease of Hg concentration in sediments has been more recently observed (Pavoni et al., 1992) and attributed to a greater control of urban and industrial waste waters. However, the entire lagoon bed appears to be moderately contaminated as Hg bound to suspended sediments is easily redistributed throughout the lagoon basin by tidal fluxes (Bloom et al., 2004).

Several authors have described the sediment pollution of Hg in the central Venice Lagoon (Pavoni et al., 1992; Bellucci et al., 2002) but few studies have addressed the southern area.

Biomagnification of Hg by transfer of the metal through the trophic chain has been well demonstrated (Watras and Bloom, 1992; Mason et al., 1996; Bargagli et al., 1998; Chen et al., 2000). However, compared to the large number of studies on the biomagnification of Hg in marine fish, there have been few studies on the contaminant transfer at the top of the marine benthic food chain. Circumstantial evidence indicates that concentrations of several metals (e.g. Cd, As, Cr, and Zn) in marine gastropods can be higher than those measured in marine bivalves that are frequently consumed by predatory gastropods (Wang and Ke, 2002). Due to their capacity in accumulating heavy metals, gastropods have been proposed as biomonitors of coastal contamination (Bryan et al., 1983; Langston and Zhou, 1986; Kang et al., 1999).

Nassarius reticulatus gastropod is not a commercial species but it is ubiquitous throughout the world; it may be found, for instance, in the north-eastern Atlantic and in the Mediterranean, Black and Azov Seas (Pinto and Bartoli, 2002). This species was chosen as target benthic organism since it is widespread in the investigated area, living either in sandy or muddy sediments where degradation of organic debris is relatively abundant.

The aim of this study was thus to investigate THg distribution not just in surface sediments, but also in *N. reticulatus* gastropods from the southern sector of the lagoon environment and coastal marine area, in order to assess the extent of Hg enrichment and potential sources of contamination.

2. Materials and methods

2.1. Study area and sampling

The Venice Lagoon is a transitional coastal environment, covering a surface area of 549 km^2, and it is characterised by very low average water depth (0.67 m) and tidal range (<1 m). Water and sediments exchanges through the three inlets of the Lagoon (Lido, Malamocco and Chioggia) is mainly controlled by tidal fluxes.

Sediment and gastropods were collected in the southern sector of the Venice Lagoon near Chioggia town (53,470 inhabitants), which has undergone increasing development of the dockyards, shipping and fishing activities. Sampling of 36 bottom sediments was performed by means of an Ekman grab between April and September 2003. The upper 2–3 cm was recovered and placed in acid-cleaned glass containers, then homogenised and stored at -20 °C. The sampling stations were defined as dockyards, harbour, marina, town centre, lagoon channels and coastal area on the basis of their location and the activities undertaken therein (Fig. 1).

A total amount of 25 specimens of netted dogwhelk, (*N. reticulatus*) were caught by wire basket in 18 of the sediment sampling sites. The shell length and the maximum diameter were measured. The soft tissues of the molluscs were excised with stainless-steel scalpel blades, thoroughly rinsed with MilliQ water to remove extraneous impurities and homogenised using a blender (Ultraturrax). The homogenised samples were stored at -20 °C. All samples were lyophilized for 72 h before analysis.

2.2. Analytical procedures

Sediments were treated with concentrated hydrogen peroxide for 48 h, organic debris and shell fragments were removed by sieving (<2 mm) and grain-size analyses was performed by a laser diffraction sizer (Malvern Mastersizer 2000 coupled with Hydro 2000s sampler unit). Sediments were classified in accordance with Shepard (1954).

Organic carbon (C_{org}) and nitrogen (N_{tot}) were determined in sediments by using a CHNS-O elemental analyser Fison (Italy) mod. EA1108, calibrated with acetanilide as standard. C_{org} was determined after removal of carbonates with concentrated HCl 1 N (Hedges and Stern, 1984). Weight percentages of N_{tot} were determined following the same procedure, without acidification. Analytical precision of measurements was 2.5% for both C_{org} and N_{tot}. Reference material BCSS (NRC, Canada) was used to assess the accuracy of the analytical data.

Sediments ($\cong$50 mg) and biota ($\cong$500 mg) were mineralised using a microwave oven (MLS 1200 mega, Milestone) in a teflon vessel with superpure concentrated HCl, HNO_3 and HF mixture (2 : 0.5 : 0.1

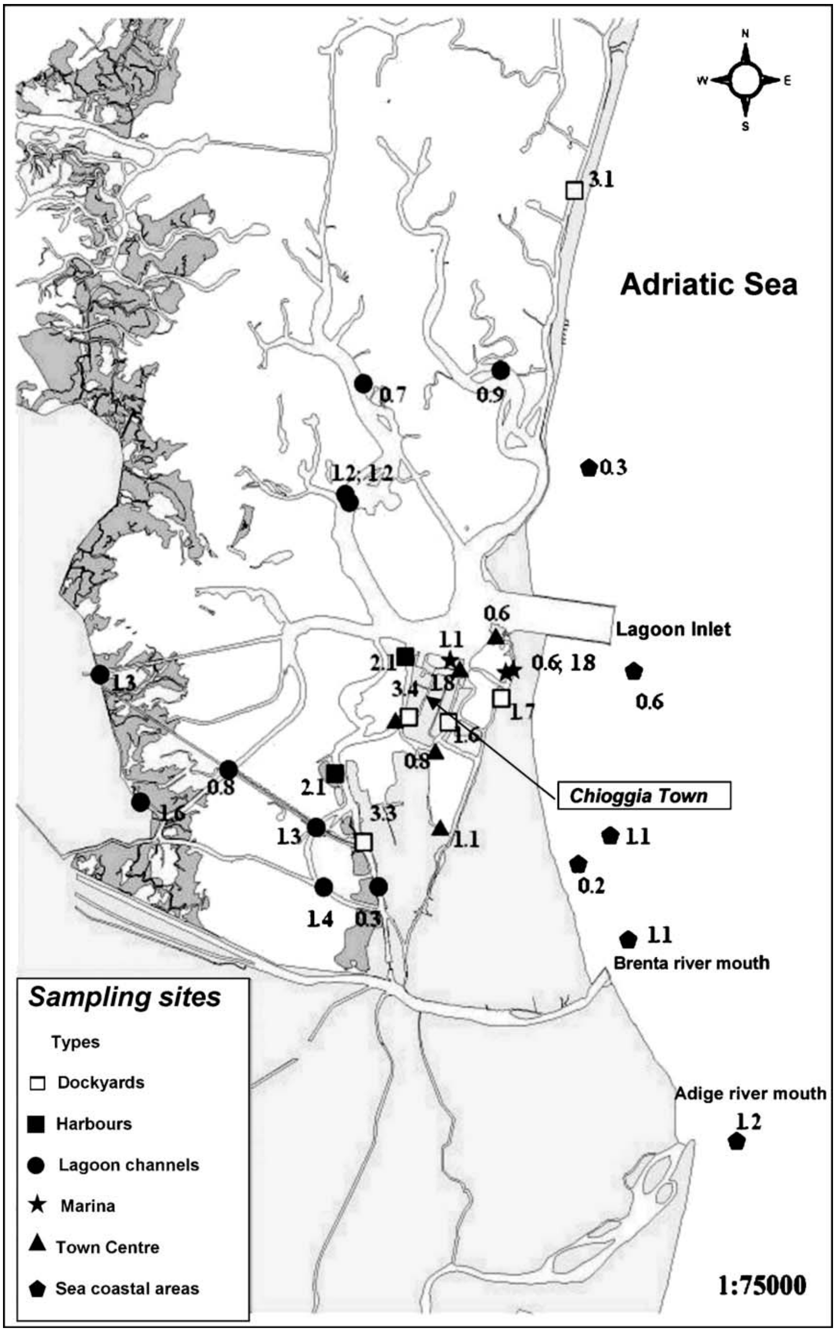

Fig. 1. Map of sampling sites and total mercury concentrations (mg/kg d. wt.) in the southern Venice Lagoon.

v/v) for dried sediments (adapted from Mester et al., 1999), and with concentrated HNO_3 (Navarro et al., 1992) for the dried biota soft tissue. All reagents were of the superpure quality (RS Carlo Erba, Italy). THg concentrations were determined by cold-vapour atomic absorption spectrometry (CVASS). Reagent blanks were processed simultaneously. The certified reference materials for marine sediment (PACS-2, NRC, Canada) and oyster tissue (1566a, NBS, USA) were used to control accuracy of the analytical method for sediments and biota. Recovery values found were 101% for sediments and 78% for biota. The precision of THg determination in sediment and biota was $\pm 3\%$ and the detection limit was 0.05 mg/kg. All THg concentrations were based on dry weight (d. wt.).

3. Results and discussion

3.1. Sediments characteristics

Sands and silty sands constituted the predominant textures in the coastal area in front of the barrier islands whereas sandy silt and silt were prevalent inside the lagoon. Clay (<2 μm) was present in reduced amounts (<7%) in all sediment samples.

Grain size progressively decreased from the tidal inlet towards the inner areas of the lagoon and in the lower energy zones. Very fine to medium sands were found only in the marine area and in some sites near Chioggia town. Most of the samples were characterised by a high percentage of 8–2 μm particles (very fine silt).

The C_{org}/N_{tot} molar ratios calculated on the basis of the C_{org} (0.3–2.0%) and N_{tot} (0.01–0.38%) contents ranged from 3.9 to 15.3. Sandy silty sediments in dockyards were the most enriched in C_{org} (Table 1), whereas the sandy sediments of coastal areas and harbours showed the lowest values of C_{org} (0.1–0.3%). It cannot be excluded that the highest C_{org}/N_{tot} values found in dockyards and centre town sediments were caused by a local source of organic matter such as the debris generated in the use of antifouling paints (Yebra et al., 2004).

Significant correlations were found between pelite (<62.5 μm) contents, C_{org} (Spearman $R=0.891$ $p<0.05$) and N_{tot} (Spearman $R=0.895$ $p<0.05$), suggesting a prevailing association and preservation of organic matter within the finer fraction of the sediments, as reported for other North Adriatic coastal environments (e.g. Faganeli et al., 1991).

3.2. THg in surface sediments

Mercury concentrations in sediments ranging between 0.1 and 3.4 mg/kg were, on average (1.3 ± 0.7, mg/kg), far higher than the natural background values proposed in previous studies for the Venice Lagoon (0.07 mg/kg from Donazzolo et al., 1982). Our results were higher than those found in the northern Venice Lagoon (avg. 0.6 mg/kg) by Bloom et al. (2004), but lower with respect to the central part – which is subject to the anthropic impact of the industrial area of Porto Marghera and the town of Venice (avg. 3.4 mg/kg) – as reported by Bellucci et al. (2002) and Bloom et al. (2004). Critto et al. (2005) reported lower average concentrations of THg (0.44 mg/kg) than our study for a smaller dataset of sediment samples in the southern sector.

The spatial distribution of THg concentration in sediments in this study showed that the highest contents, detected in dockyards and harbour sites (Figs. 1 and 2), were comparable to those found in the central part of the lagoon basin (Bloom et al., 2004).

The southern Venice lagoon, due to its shallowness and limited water exchange, acts as a trap for sediments and associated contaminants such as mercury. The accumulation of Hg in sediments was found also in the Grado and Marano lagoons, located further east along the coast, where the high THg concentrations (1–10 mg/kg) were shown to be a consequence not only of industrial discharges but also of the Isonzo River; this has been the largest source of Hg since the 16th century as a result of the extraction activity of mercury mine in Idrija, western Slovenia (Brambati, 2001; Covelli et al., 2001; Piani et al., 2005).

The resulting contamination of THg in the Venice Lagoon is probably due to extreme and near-continuous anthropogenic disturbances to the sediments resulting from the combination of dredging, salt-marsh reclamation and local techniques in clam harvesting (Bloom et al., 2004).

Although Hg behaviour depends on the chemical form introduced into the aquatic environment, Hg associated with industrial effluents was recognised to be positively correlated with <63 μm (Rae and Aston, 1981) or <20 μm (Barghigiani et al., 1996) and was

Table 1
Sediment characteristics and Hg concentrations

	Dockyards ($n=7$)	Harbour ($n=2$)	Town centre ($n=6$)	Marina ($n=4$)	Lagoon channels ($n=11$)	Coastal area ($n=8$)
Sand (%)	23.5 ± 15.8	87.3	38.9 ± 37.0	40.1 ± 35.3	31.9 ± 15.1	95.4 ± 5.8
Silt (%)	71.2 ± 15.2	11.5	56.6 ± 34.2	55.7 ± 32.7	63.2 ± 14.2	4.3 ± 5.2
Clay (%)	5.3 ± 0.9	1.2	4.6 ± 2.9	4.1 ± 2.7	4.9 ± 1.1	0.2 ± 0.6
C_{org} (%)	2.0 ± 1.1	0.3 ± 0.2	1.6 ± 1.0	1.2 ± 1.1	1.6 ± 0.8	0.1 ± 0.1
N_{tot} (%)	0.21 ± 0.11	0.05 ± 0.04	0.20 ± 0.14	0.16 ± 0.11	0.25 ± 0.08	0.02 ± 0.01
C_{org}/N_{tot} (mole/mole)	10.5 ± 2.8	6.3 ± 1.3	9.9 ± 2.1	9.0 ± 2.9	7.4 ± 1.7	7.6 ± 4.2
THg (mg/kg)	2.09 ± 0.9	1.96 ± 0.2	1.33 ± 0.4	1.18 ± 0.6	1.1 ± 0.4	0.60 ± 0.5

Average concentrations and standard deviations are noted.

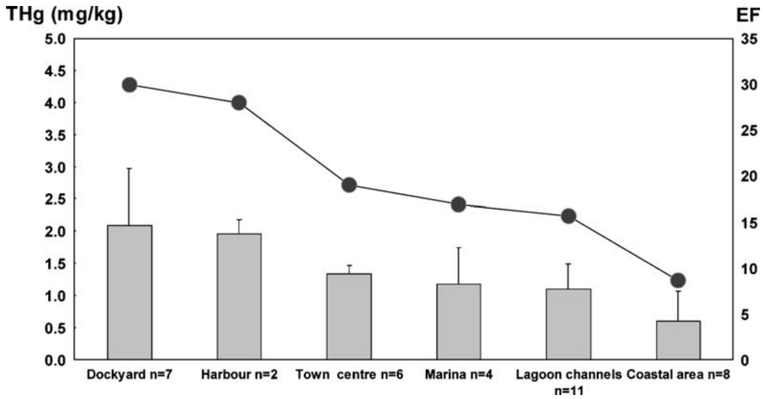

Fig. 2. Mean mercury concentration (mg/kg d. wt.), histograms, and mean Enrichment Factor (EF), black circles, for the southern Venice lagoon sediments. Vertical bars represent the standard deviations.

also reported to have a good affinity with organic matter (e.g., Smith and Loring, 1981; Baldi and Bargagli, 1982). Our results confirm these findings only partially, since THg shows a positive correlation with the <63 μm fraction (Spearman $R=0.50$; $p<0.05$) and C_{org} (Spearman $R=0.51$ $p<0.05$) in the sediments. The extremely high affinity of THg for organic matter and S-ligands (Hintelmann et al., 1995; Wallschlager et al., 1996) reduces the mobility and toxicity of the metal. This fact seems not to be very effective in the study area, suggesting that water-soluble Hg forms present in the sediments could be removed from these and easily accumulate in marine organisms.

In order to assess Hg contamination level in the lagoon sediments, the enrichment factor (EF) was calculated in accordance with Hornberger et al. (1999) as follows:

$$EF = C_i / C_{background}$$

where C_i is the metal concentration in the sample and $C_{background}$ is the THg background value (0.07 mg/kg) for the lagoon sediments estimated from the basal core levels of Donazzolo et al. (1982).

The EF showed high values (avg. 30, max 49) near the dockyards of Chioggia, whereas lower EF values were found in the channels of the southern Venice Lagoon (avg. 16, max 23) and in the coastal marine sediments (avg. 9, max 17) near the southern inlet of the lagoon (Fig. 2). The higher Hg enrichment factor in dockyards and harbours could be explained by historic or recent use of antifouling paints with Hg added as a biocide. From 1908 to 1926 several versions of paints based on red mercury oxide suspended in grade A gum shellac, grain alcohol, turpentine, and pine tar oil were used (Yebra et al., 2004). After the Second World War, important changes took place in the paints industry. The appearance of new synthetic petroleum-based resins offered improved mechanical characteristics, and increased concern about safety and health caused the abandonment of organo-mercurials and organo-arsenicals compounds (Yebra et al., 2004). However, the concentration of mercury in inorganic and organo-mercurial forms added to antifouling paints remains unclear (Johnsen and Engøy, 1999).

Reworking and dispersal of sediments due to periodic dredging operations in the contaminated areas of the central lagoon may contribute to the dispersal of mercury (Bloom et al., 2004) associated with the fine sediments, causing the more than negligible EF values found in the lagoon channels.

To assess the contamination level of sediments and in general the environmental quality of the study area, the "Effects Range-Low" (ERL) and "Effects Range-Median" (ERM) were also used. The ERL is defined as the 10th percentile of the values indicating the lowest concentration below which adverse effects rarely occur. The ERM, defined as the 50th percentile of the concentration values, is representative of concentrations above which effects frequently occur (Long et al., 1995). These "effects values" were not derived as toxicity thresholds. There is no assurance that there will be a total lack of toxicity when chemical concentrations are less than the ERL values. Similarly, there is no assurance that the samples in which ERM values are exceeded will be toxic. Toxicity, or lack thereof, must be confirmed with empirical data from toxicity tests (NOAA, 1999).

Long et al. (1995) suggested that toxic effects were rare at low Hg concentration (<0.15 mg/kg, ERL value) whereas at high values (>0.71 mg/kg, ERM value) 73% of the data entries indicated an association between toxicity and Hg in sediments. About 70% of the sam-

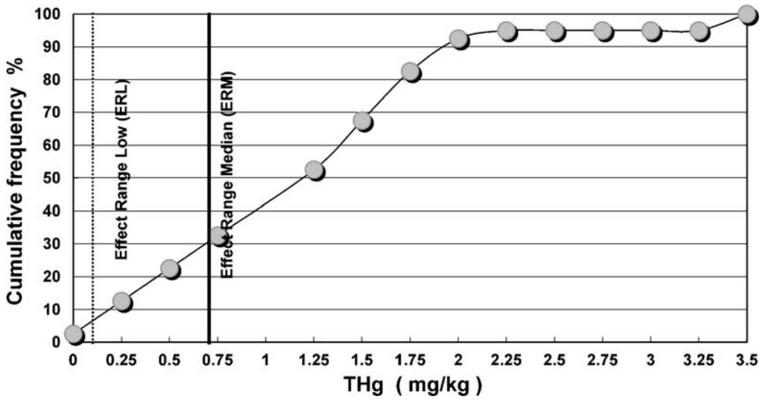

Fig. 3. Cumulative frequency (%) and mercury concentrations (mg/kg) of lagoon sediments. The "Effects Range-Low" (ERL) and "Effects Range-Median" (ERM) values were used to assess the contamination level of sediments (Sediment Quality Guidelines, NOAA).

pled sites analysed showed concentrations higher than the ERM value (Fig. 3), indicating a high probability of ecological risk to the benthic community in contaminated sediments.

3.3. THg in *N. reticulatus*

The mean length of the collected specimen was 25.5 ± 1.8 mm and the mean width was 12.0 ± 0.7 mm; all the specimens were sexually mature and were therefore presumably at least 4 years old, at which age, according to Tallmark (1980), they become sexually mature.

Mercury concentration in the gastropods ranged between 0.3 and 1.3 mg/kg d. wt. These data were higher than those found in other molluscs (mussels and clams), sampled from contaminated and non-contaminated sites in the central and southern Venice Lagoon, with THg concentrations ranging from 0.02 to 0.35 mg/kg d. wt. (Widdows et al., 1997; Di Domenico et al., 1998; Bloom et al., 2004). Furthermore, the diet of the predatory gastropods may include other prey organisms such as animal carcasses or other species of invertebrates.

High concentrations of THg (480–1220 ng/g, fresh weight) were also found in another species of gastropods, *Gibbula adriatica*, in the nearby Grado and Marano Lagoons, where a progressive eastward increase and a strong correlation with THg in bottom

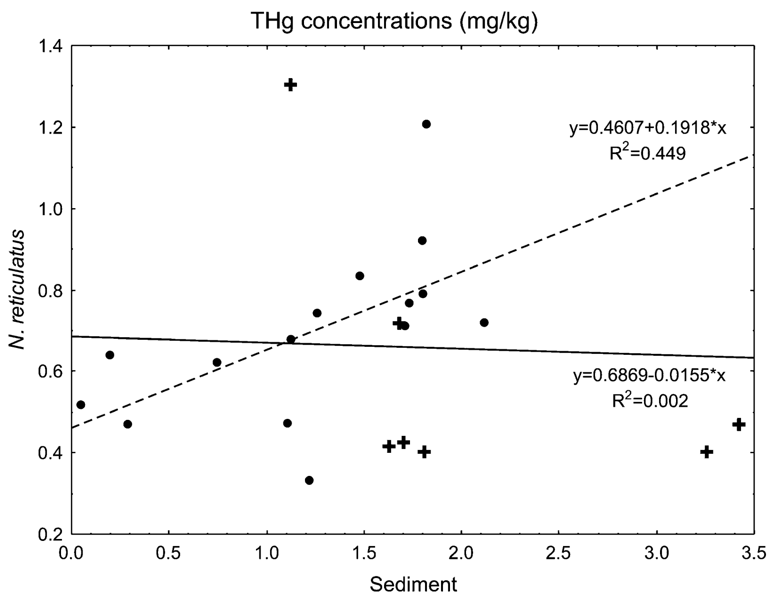

Fig. 4. Scatterplot of mercury concentration in the sediments vs. *N. reticulatus* tissues; the black line shows the linear regression for all samples, whereas the dashed line represents the linear regression excluding dockyard samples, marked with the symbol +.

sediments and in aquatic vegetation was found (Brambati, 1997). This clearly indicates Hg bioaccumulation in the lagoon's trophic chain.

The results obtained in this study revealed no correlation between THg concentration in the sediments and in *N. reticulatus* considering all the sampled stations, but a positive correlation (Spearman $R = 0.76$; $p < 0.05$) was evident when excluding the dockyard sites (Fig. 4). The dockyard sites were characterised by high concentrations of Hg in sediments and a relatively low concentration in *N. reticulatus*, probably due to a form of Hg that is not readily bioavailable.

4. Conclusions

The Venice Lagoon has experienced significant Hg discharges over the years from industrial and urban sources. Values of THg found in the sediments indicated that the southern Venice Lagoon appears to be moderately and widely contaminated, especially in the dockyard and harbour sites, where high THg concentrations and EF values may indicate a local source of pollution (i.e. antifouling paints). Thus, natural and anthropogenic factors, such as tidal fluxes, the widening of the port of Chioggia, the dredging of channels and clam harvesting, which are responsible for the resuspension of lagoon sediments, may well facilitate the transportation and spread of Hg from the northern-central area to the southern sector of the basin.

On the other hand, the bioavailability and toxicity of Hg also depend on its interaction with sediments. The relatively low correlation found between THg and C_{org} in this study might indicate that a fraction of Hg, not associated with organic matter, but with greater mobility and toxicity, can easily enter the aquatic systems and accumulate in organisms. *N. reticulatus*, a scavenger and benthic gastropod, is widespread throughout the lagoon and can provide valuable information about the bioavailability of THg and the level of biomagnification along the first steps of the benthic trophic chain.

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