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Mercury Enrichment in Sediments of the Coastal Area of Northern Latium, Italy

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Abstract The purpose of this study was to evaluate the extent of the Hg geochemical anomaly arising in the Amiata and Tolfa complex to the coastal area of northern Latium and to examine the possible influence on this area by the Mignone River, and by the small coastal basins, which are characterized by both previous mining activities and decades of past industrial impact. The results confirm the extension of the anomaly of concentrations of Hg in the coastal area of northern Latium, with the northern sector influenced by the contributions of the Fiora and Mignone Rivers and the southern sector influenced by the contributions of minor basins. The results show high values of the Adverse Effect Index throughout the considered area and highlight the need for further investigation in order to assess the impact of human activities on the present and past values of Hg in marine sediments.

Keywords Mercury · Sediments · Tyrrhenian sea · Geochemical anomaly · Anthropic impact

Today, mercury is re-emerging as a pollutant of concern for both environmental and human health. Current global atmospheric deposition of mercury (Hg) has been estimated to be three times higher than preindustrial levels, reaching values of potential toxicity for many living organisms (Goodsite et al. 2004; Mirlean et al. 2005; Di Leonardo et al. 2006; Selin et al. 2008).

Trace elements, such as Hg, in marine sediments have a particular affinity for finer particle grain sizes, binding to the surface of fine particles by different processes (Ligero et al. 2001) and following fine particles' sedimentary fate. Sediment can receive Hg from both natural and anthropogenic sources such as river basin runoff, atmospheric deposition, discharge of sewage, and industrial emissions.

Rapid industrialization, urbanization, and the increase of human activity in coastal areas has resulted in an increased quantity of pollutants, such as Hg, that accumulate in air, water, soil, and sediment, as described in several works (Nriagu and Pacyna 1988; Varol 2011; Chaudhari et al. 2012; Bastami et al. 2014). Therefore, the study of Hg distribution in coastal sediments is a realistic way to understand the Hg cycle in a marine environment and to explore the history of the possible anthropogenic Hg emissions at the land–ocean interface zone.

In the Mediterranean area, there are some studies of Hg related to the atmospheric compartment (Pyle and Mather 2003), as well as several studies about the Hg distribution in aquatic environments and in dissolved gaseous Hg concentrations (Ferrara et al. 2003; Horvat et al. 2003; Maffucci et al. 2005). But, there is a lack of information about the Hg spatial and temporal distribution in Latium coastal sediments. Northern Tyrrhenian Sea sediments were previously studied by some authors (Leoni and Sartori 1996; Barghigiani et al. 1996; Baldi and Bargagli 1982) and an anomaly of Hg concentration was identified.

As the southern Tuscany region has had an extended history as associated with Hg contamination (Rimondi et al. 2012), this study aimed to investigate the

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concentrations of Hg in the northern Latium coastal area, with particular reference to: (1) the spatial distribution of Hg associated with marine sediments in order to assess the extent of the Hg anomaly, (2) the degree of Hg in sediments and (3) the potential toxicity and adverse effects of Hg in biota.

The study area is located in the Tyrrhenian Sea, in northern Latium, and it included the coastal platform that extends from Santa Severa (42.01676N–11.95604E), located south of Capo Linaro, up to the Montalto di Castro coastal area (42.33215N–11.56786E), in the physiographic unit Monte Argentario-Capo Linaro (Fig. 1). Regarding the geomorphological features of this Physiographic Unit, a series of stratigraphic marine and/or riverine units that define a series of terraces were identified. This geomorphological features are the result of interactions between lifting of the zone during the Pleistocene and sea-level glacial fluctuations. (De Rita et al. 2002; Aureli et al. 2012). The seaward margin in this area (begin of the continental slope), can be found at depths between 120 and 150 m. Moving from the north to the south, the seabed becomes shallower and particularly in the section between Sant'Agostino and Capo Linaro the slope of the continental platform increases (Chiocci and La Monica 1996; La Monica and Raffi 1996). The sediment transport along the shore occurs in a southeast to northwest direction (Anselmi et al. 1976; Berriolo and Sirito 1985; Noli et al. 1996) and it's also confirmed by the northern orientation of the Marta and Mignone rivers.

The study area is characterized by the sedimentary contributions of three important river basins (Fiora, Marta, and Mignone) and, subordinately, from the contributions of other smaller rivers, which influence marine sedimentation only locally and during strong flood phases (Angelucci et al. 1979; Carboni et al. 1980).

The geological features and the sedimentary productivity of these river basins have been exhaustively described in several works (Spadoni et al. 2005; Scanu 2012; Scanu et al. 2015a). The Fiora River basin (which springs from the southern flank of the Monte Amiata) is mainly situated on tuff and volcanic formations, respectively characterized by predominantly sand and clay type sedimentary production. Furthermore, the Amiata district is characterized by several inactive Hg mines (Rimondi et al. 2012). The Marta river basin originates from Lake Bolsena and is mainly covered by volcanic formations, tuff reliefs, lakes and marl. The sedimentary production is clayey for the volcanic formations and sandy for the tuffaceous formations. The Mignone River basin is largely set on hilly topography consisting of marl, arenaceous conglomerate, and clay materials and it includes the presence of many sulphide mineral ores linked to the upwelling of hydrothermal fluids.The sedimentary production is sandy for the conglomeratic formations and

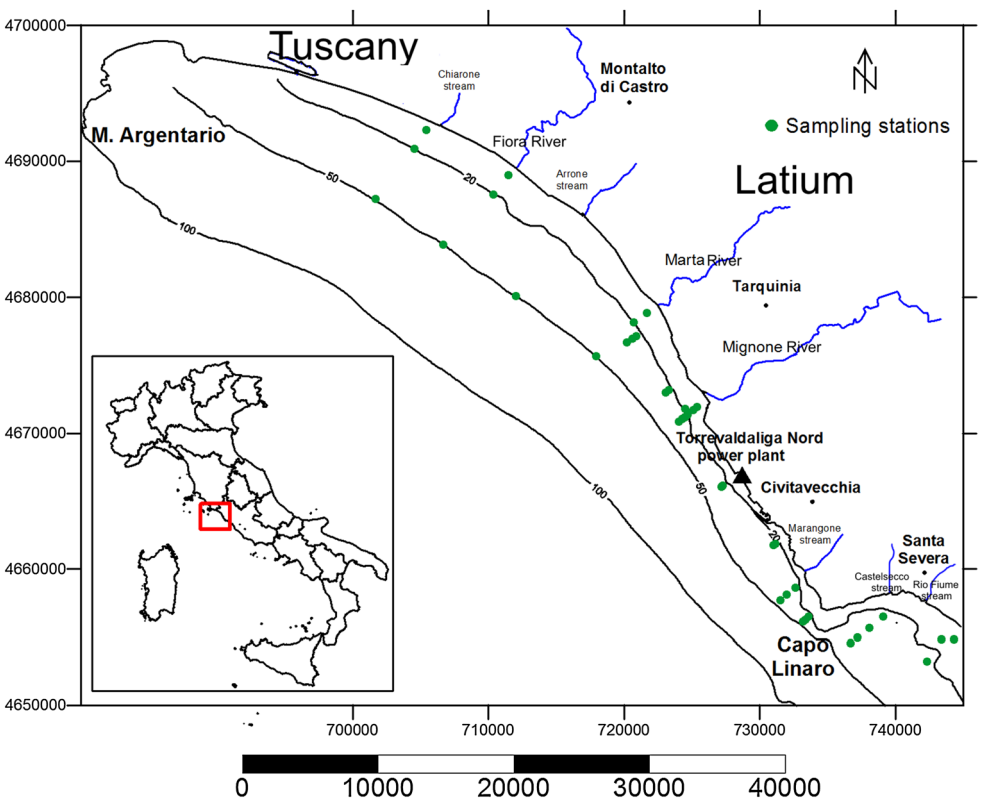

Fig. 1 Study area and sampling locations

clayey for the marl formations (Spadoni et al. 2005; Scanu 2012). The most important local contributions come from the Marangone basin which has the same geologic characteristics of the Mignone basin. The Marangone small basin is characterized by abandoned iron sulphide (pyrite, marcasite) and galena mines. Furthermore, marcasite mine waste deposits and galena small pits are presents (Kreidie et al. 2011).

Regarding Hg levels in these river basins, several studies have shown that the Fiora River area is affected by cinnabar (HgS), a geologic anomaly of Mt. Amiata, but more so by the Hg extraction that lasted from Etruscan times (the eight to the first century B.C.) to 1980. This gave rise to high soil and vegetation contamination in much of the Amiata area (Barghigiani et al. 1981; Barghigiani and Bauleo 1992; Barghigiani and Ristori 1994, 1995; Rimondi et al. 2012).

Materials and Methods

Sediment sampling was performed using a Van Veen grab (18 L). A total of 39 surface sediment samples, located between a depth of −50 and −10 m, were sampled between 2011 and 2013 (Fig. 1). The surface layer (1 cm) of each sample was subsampled, homogenized, and placed into a polyethylene bag. Once transported (at 4°C) to the laboratory, sediments were dried in an oven at 40°C for 48 h, crushed, and stored in polyethylene bags for subsequent analysis. Grain-size analysis was performed through wet sieving on sediment samples previously treated with 16 % solution of H_2O_2 (Spagnoli et al. 2014).

Hg extraction from sediments and analytical determination was performed according to EPA 3051A:2007 and 6020A:2007, respectively. In particular, for the lab tests, 1 g of each analytical sample group was weighed with an analytical scale (Mettler Toledo xs105) and mineralized in a microwave system, Ethos Touch Control (Milestone S.r.l-Italy), with a digestion solution prepared with 9 mL of 65 % nitric acid (HNO_3) (Carlo Erba) and 3 mL of 30 % chloridric acid (HCl) (Carlo Erba). After mineralization, ultrapure water (Merck) was added to the samples up to 50 mL. The test for Hg analysis was performed with an ICP-MS DRC-e (Perkin Elmer-USA). Standards for the instrument calibration were prepared on the basis of a certified reference solution ICP Standard (Merck). The system was calibrated using a calibration line made up of five points of increasing concentration for each element, with a linear regression (R^2) for the calibration line between 0.9972 and 0.9989. Analytical blanks and recovery samples were run in the same way as the real samples, and concentrations were determined using standard solutions prepared in the same acid matrix to validate the calibration. Analytical quality control was performed with standard reference material NIST-2711a-Montana II Soil, with recovery ranging between 95 % and 110 %. The cartographic elaborations were made using Surfer 8 (Golden Software, USA) and the Kriging interpolation method was used.

To assess the Hg enrichment degree in sediments, enrichment factor (EF) has been used in other studies. It has been previously described by several authors (Morillo et al. 2004; Adamo et al. 2005; Vald'es et al. 2005) and is widely used to study sediment-contamination degree. For the normalization of Hg concentrations in EF aluminum was used, as previously described by Huang and Lin (2003) and Woitke et al. (2003). The enrichment factor is determined as:

$$EF = (H_s/Al_s)/(H_c/Al_c),$$

where H_s and H_c are the sample and background trace metal concentrations, respectively, and Al_s and Al_c represent the sample and background reference of aluminum contents.As background concentrations, the values described by Turekian and Wedepohl (1961) were used because of the different sedimentary matrix (Sandstone or shale). Values of EF less than 1 indicate no enrichment, of less than 3 indicate minor enrichment, of equal to 3–5 indicate moderate enrichment, of equal to 5–10 indicate moderately severe enrichment, of equal to 10–25 indicate severe enrichment, of equal to 25–50 indicate very severe enrichment, and of greater than 50 indicate extremely severe enrichment (Sakan et al. 2009).

In order to assess the potential ecological risk and toxicity of Hg, the measured concentrations in the sediment samples were compared with the effects range low (ERL) and the effects range medium (ERM) values. These numerical values are sediment quality guidelines (SQGs) originally determined by Long and Morgan (1990) and later refined by Long et al. (1995). The ERL was calculated as the lower 10th percentile of "effects" concentrations and the ERM as the 50th percentile of "effects" concentrations. Moreover, AEI values described by Muñoz-Barbosa et al. (2012) were calculated using the following equation:

$$AEI = [X]/[TEL],$$

where X is the metal concentration, and TEL is the Threshold Effect Levels derived by Long et al. (1995). If AEI is less than or equal to 1 then the metal concentration in the sample is not high enough to produce adverse effects in biota. However, if AEI is greater than or equal to 1 then the metal concentration in the sample could produce adverse effects (Muñoz-Barbosa et al. 2012).

Table 1 Hg, pelitic fraction percentage, Al, Hg/Al, EF, and AEI values in the study area

Sampling stations	Location		Pelitic %	Al (mg/kg)	Hg (mg/kg)	(Hg/Al)	EF	AEI
	x	y						
M1	705,441	4,692,270	8.2	49,800	0.9	1.8E−05	15.1	6.9
M2	704,539	4,690,881	36.8	33,900	0.59	1.7E−05	14.5	4.5
M3	701,682	4,687,213	92.8	3200	0.63	2.0E−04	39.4	4.8
M4	711,487	4,688,958	8.9	28,400	1.44	5.1E−05	42.3	11.1
M5	710,384	4,687,529	86.5	64,600	1.69	2.6E−05	5.2	13
M6	706,688	4,683,882	86.0	48,300	0.79	1.6E−05	3.3	6.1
M7	712,012	4,680,106	83.3	50,000	0.44	8.8E−06	1.8	3.4
M8	721,657	4,678,853	20.7	36,100	0.75	2.1E−05	17.3	5.8
M9	720,710	4,678,152	61.8	48,500	0.31	6.4E−06	1.3	2.4
M10	717,923	4,675,650	97.1	48,600	0.61	1.3E−05	2.5	4.7
M11	724,509	4,671,761	61.8	18,000	2.2	1.2E−04	24.4	16.9
M12	720,912	4,677,160	15.2	51,545	0.63	1.2E−05	10.2	4.9
M13	720,629	4,676,951	45.0	79,926	0.34	4.3E−06	3.6	2.6
M14	720,184	4,676,671	43.9	61,344	0.36	5.8E−06	4.8	2.7
M15	723,277	4,673,178	15.4	54,922	0.39	7.1E−06	5.9	3.0
M16	723,068	4,672,993	54.5	51,804	0.33	6.5E−06	1.3	2.6
M17	725,352	4,671,921	6.8	22,015	0.46	2.1E−05	17.2	3.5
M18	725,086	4,671,702	3.6	35,464	0.24	6.8E−06	5.7	1.9
M19	724,683	4,671,377	28.3	38,841	0.52	1.3E−05	11.2	4.0
M20	724,473	4,671,193	58.3	46,020	0.66	1.4E−05	2.9	5.1
M21	724,251	4,671,036	20.8	48,387	0.55	1.1E−05	9.5	4.2
M22	724,050	4,670,857	26.9	52,024	0.41	8.0E−06	6.6	3.2
M23	727,283	4,666,127	42.9	68,006	0.38	5.6E−06	4.7	2.9
M24	727,187	4,666,046	25.6	68,273	0.41	6.0E−06	5.0	3.2
M25	731,155	4,661,870	11.9	12,596	0.03	2.0E−06	1.7	0.2
M26	731,002	4,661,755	15.4	41,731	0.14	3.4E−06	2.9	1.1
M27	732,612	4,658,628	49.6	47,629	0.43	9.1E−06	7.6	3.3
M28	731,958	4,658,107	30.1	99,119	0.61	6.2E−06	5.1	4.7
M29	731,491	4,657,693	71.2	57,472	0.81	1.4E−05	2.8	6.2
M30	733,582	4,656,476	43.7	72,603	0.27	3.7E−06	3.1	2.1
M31	733,361	4,656,272	33.0	120,297	0.46	3.8E−06	3.2	3.6
M32	733,171	4,656,122	30.5	61,017	0.42	6.9E−06	5.7	3.2
M33	739,090	4,656,476	60.9	46,623	0.58	1.3E−05	2.5	4.5
M34	738,055	4,655,686	18.6	64,626	0.64	9.9E−06	8.3	4.9
M35	737,184	4,654,956	51.5	75,499	0.54	7.2E−06	1.4	4.2
M36	736,701	4,654,551	93.4	44,381	0.58	1.3E−05	2.6	4.4
M37	744,302	4,654,853	36.8	52,342	1.09	2.1E−05	17.4	8.4
M38	743,374	4,654,821	48.2	35,351	0.79	2.2E−05	18.7	6.1
M39	742,310	4,653,217	51.7	31,455	0.62	2.0E−05	4.0	4.8
Range					0.03–2.2			
Average ± SD					0.62 ± 0.41			
ERL					0.15			
ERM					0.71			
TEL					0.13			

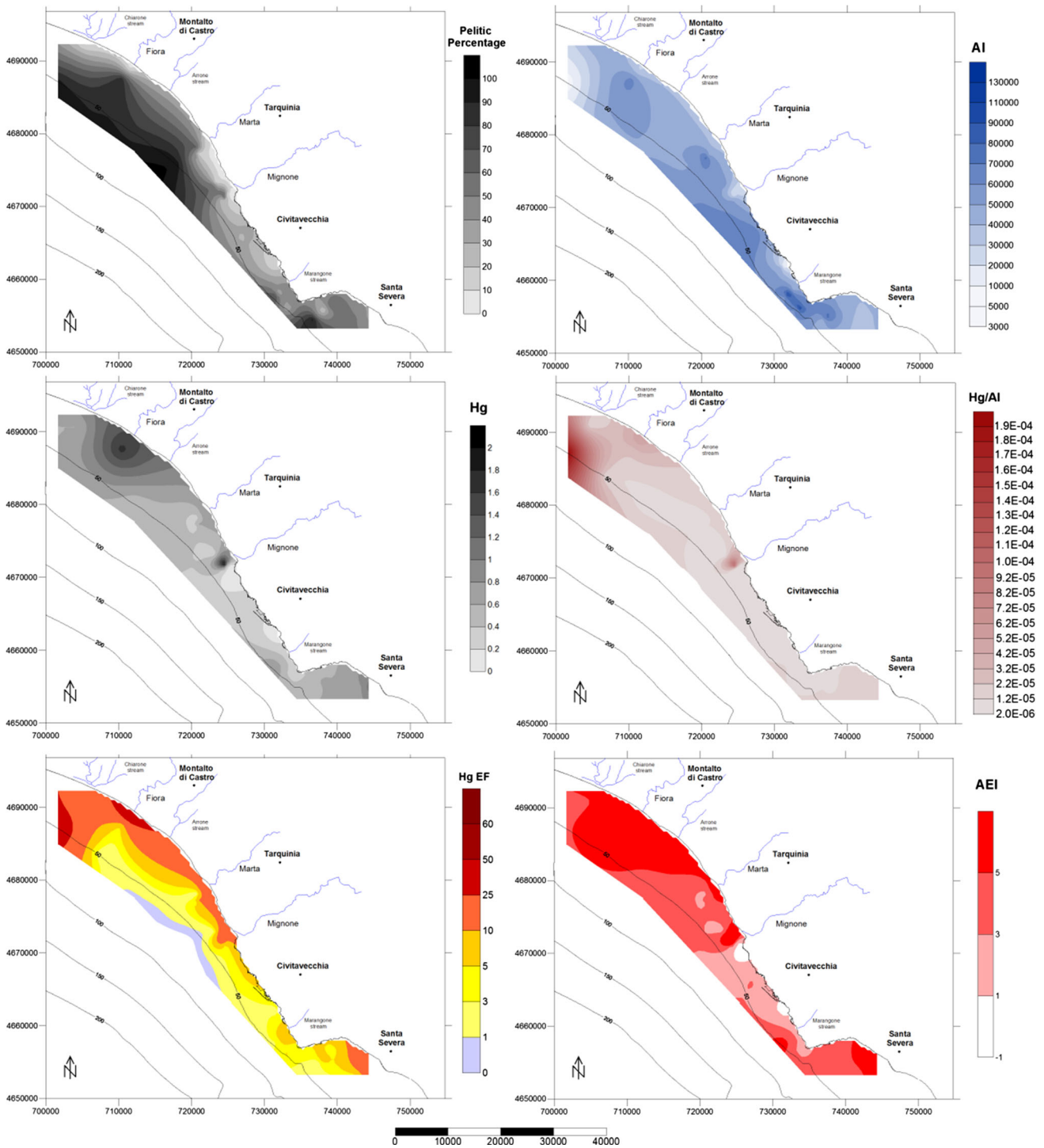

Fig. 2 Distribution pattern of pelitic percentage, Al (mg/kg), Hg (mg/kg), Hg/Al, EF, and AEI in coastal sediments of northern Latium

Results and Discussion

The results are shown in Table 1 and the superficial distribution patterns of pelitic fraction, Al, Hg, Hg/Al, EF, and AEI are reported in Fig. 2. The maximum concentration is 2.20 mg/kg (M11), while the minimum is concentration is 0.03 mg/kg (M25). The average value for the area is 0.62 mg/kg, but there is great variability in the data. Overall, the mean Hg concentration is above the ERL value. Moreover, eight sampling stations exceed the ERM value.

The distribution pattern of Hg clearly shows the existence of two Hg anomalies. The first occurs in the northern

part of the study area, near the Fiora River delta, that extends to the coastal area of the Mignone River delta, north of Civitavecchia. The second occurs in the coastal area south of the Port of Civitavecchia, from the Marangone Stream delta to the south of Capo Linaro. The central stretch of coastline, corresponding to the urban area of Civitavecchia, shows rather low values of Hg. There is also a decreasing gradient in Hg values with increasing depth.

The distribution of EF values shows that the northern coastal area is characterized by severe enrichments, with maximum values at the Fiora River delta (very severe enrichment, EF >60), while the southern area has enrichments from severe to moderately severe, corresponding to the coastal area south of the Port of Civitavecchia towards Santa Severa. In general, enrichment values show a distribution with maximum values at the north and south end of the study area and minimal values in the central coastal area of Civitavecchia.

The distribution of AEI values shows a similar pattern to EF values in the coastal area of Civitavecchia that is enclosed between two areas having values well above the limit of possible adverse effect.

The concentrations of Hg and the other indices show that the anomaly of Hg of the southern Tuscany extends further south than Cape Linaro. More specifically, the coastal area south of the Port of Civitavecchia is characterized by a second geochemical anomaly of Hg, spatially smaller than the northern one.

Considering the coastal area south of the Port of Civitavecchia, it is reasonable to assume that the Hg anomaly is due to the geological characteristics of the minor local basins that are all characterized by enrichment of sulfides and past mining activities (Kreidie et al. 2011; Scanu et al. 2015b; Spadoni et al. 2005; Piazzolla et al. 2015). The same geogenic origin is presumable for the northern Hg anomaly from the Fiora River delta to Mignone River mouth. The Amiata distric, that is part of the "circum-Mediterranean Hg belt" (Bargagli et al. 1986), has several inactive Hg mines with the extraction of cinnabar being completely stopped by the 1980s. Assuming the geogenic nature of Hg anomalies it is appropriate to consider the possibility that the concentrations are also influenced by anthropogenic inputs which overlap with the already high baseline values. The main potential anthropogenic source of Hg in the area is the coal fired power plant of Torrevaldaliga Nord with an installed capacity of 1980 MW and more than 4 million tons of coal burned per year (National Entity for Electricity 2014). Hg is a volatile element emitted to the atmosphere through flue gas generated in coal power plants and it tends to be adsorbed onto the atmospheric dust particles, which are subsequently deposited in the aqueous and terrestrial systems by dry or wet deposition (Haykiri-Acma et al. 2011; Tang et al. 2013). This fallout can affect hydrographic basins and can lead to an enrichment of Hg in coastal marine environment. In this context, the main river basins of the area (Fiora, Marta and Mignone), may be subject to emissions from the Torrevaldaliga Nord coal fired power plant also considering the prevailing wind direction and the coastal sediment transport (Scanu et al. 2015a, b; Piazzolla et al. 2015).

The study area characteristics imply to deepen the knowledge of the source of the enrichment of Hg in marine sediments through the quantification of the real weight of the anthropogenic sources even in the high values of natural origin.

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