
ELSEVIER
VOLUME 105 NOS. 1–2
April/May 2010
ISSN: 0375-6742
JGE
JOURNAL OF
GEOCHEMICAL
EXPLORATION
JOURNAL FOR ENVIRONMENTAL AND ECONOMIC GEOCHEMISTRY
http://www.elsevier.com/locate/jgeoexp

Journal of Geochemical Exploration 105 (2010) 19–33

Contents lists available at ScienceDirect

Journal of Geochemical Exploration

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

Geochemical baselines and risk assessment of the Bagnoli brownfield site coastal sea sediments (Naples, Italy)

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ARTICLE INFO

Article history:
Received 7 October 2009
Accepted 19 January 2010
Available online 29 January 2010

Keywords:
Geochemical baseline
Toxic elements
PAHs
PCBs
Brownfield site
Sea sediments

ABSTRACT

The Bagnoli brownfield site, one of the largest Italian dismantled industrial areas undergoing remediation, is located in the western sector of the urban territory of the city of Naples (Southern Italy) inside the active volcanic field of Campi Flegrei. The remediation project, funded by the Italian government, started in 1996 and was extended to the coastal area sea sediments facing the brownfield site in 2001.

A total of 384 sea sediment samples were collected from 128 boreholes at different depths (0–20 cm, 20–30 cm, 30–50 cm, 100–120 cm, 150–180 cm, 180–200 cm, 280–300 cm, and 380–400 cm) down the seabed along the coastline. Analyzed samples show that the coastal area facing the Bagnoli brownfield site is strongly contaminated by metals whose distribution seems to be controlled by the grain size of the sea sediments, and by the presence of organic compounds (PAHs and PCBs).

Geochemical mapping and *R*-mode factor score analysis allowed us to identify the groundwaters, emanating from the brownfield site to the sea across an artificial structure ("Colmata a mare") limited by two piers and filled by scum, slag and landfill material, as the main source of contaminants.

Since the groundwaters metal contamination was proven to be mostly dependent on a natural hydrothermal enrichment process related to the volcanic activity of Campi Flegrei, a remediation project would be needed primarily for PAHs and PCBs, which occur well above the concentration levels permitted by the Italian Law in the sea sediments facing the Bagnoli coastline.

1. Introduction

The assessment of risk to public health associated with land contamination from industrial sources is a common topic in many developed countries (US Environmental Protection Agency, 1996a; Edelgaard and Dahlström, 1999; Bieber, 2000; Quercia, 2001). In the last century, the Bagnoli urban district, located at the western outskirts of the city of Naples, on the eastern side of the active volcanic complex of Campi Flegrei, has been affected by a strong industrialization process lead by factories such as Italsider (ILVA) (steel production), Eternit (asbestos materials manufacturing), Cementir (cement production), and Federconsorzi (fertilizers production) (Fig. 1). During the decade between 1990 and 2000, these industrial activities were all closed.

With the aim of recovering and reusing the areas formerly occupied by the ILVA and the Eternit factories, the Italian Government funded in 1994 (CIPE, 1994) a remediation plan for the brownfield site, in the framework of the new urban development plan of the Naples City Council.

The Federconsorzi area was acquired by the IDIS Foundation to be converted into an exhibition centre for sciences (City of Sciences), while the area occupied by the closed Cementir factory has not yet undergone any remediation programme.

In 1996, a full-scale investigation of soils, scums, slags and landfill materials was planned by the Government for the ILVA and Eternit areas, with the purpose of characterizing the pollution and to set up a plan for recovery.

In 2000, the Bagnoli brownfield site was classified as a SNI (Site of National Interest) by the Italian Parliament (Parlamento Italiano, 2000) and in 2001 the coastal area facing the brownfield site was also included in the areas to be characterized and possibly recovered (Ministero dell'Ambiente e della Tutela del Territorio, 2001).

De Vivo and Lima (2008) discuss, in detail, the characterization of the brownfield site and the results of the remediation project. In accordance with the results of isotopic investigations carried out by Tarzia et al. (2002) on soil and landfill samples from the brownfield site, two components were identified as the source of the high concentrations of heavy metals and As found in soils and waters: 1) hydrothermal

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doi:10.1016/j.gexplo.2010.01.007

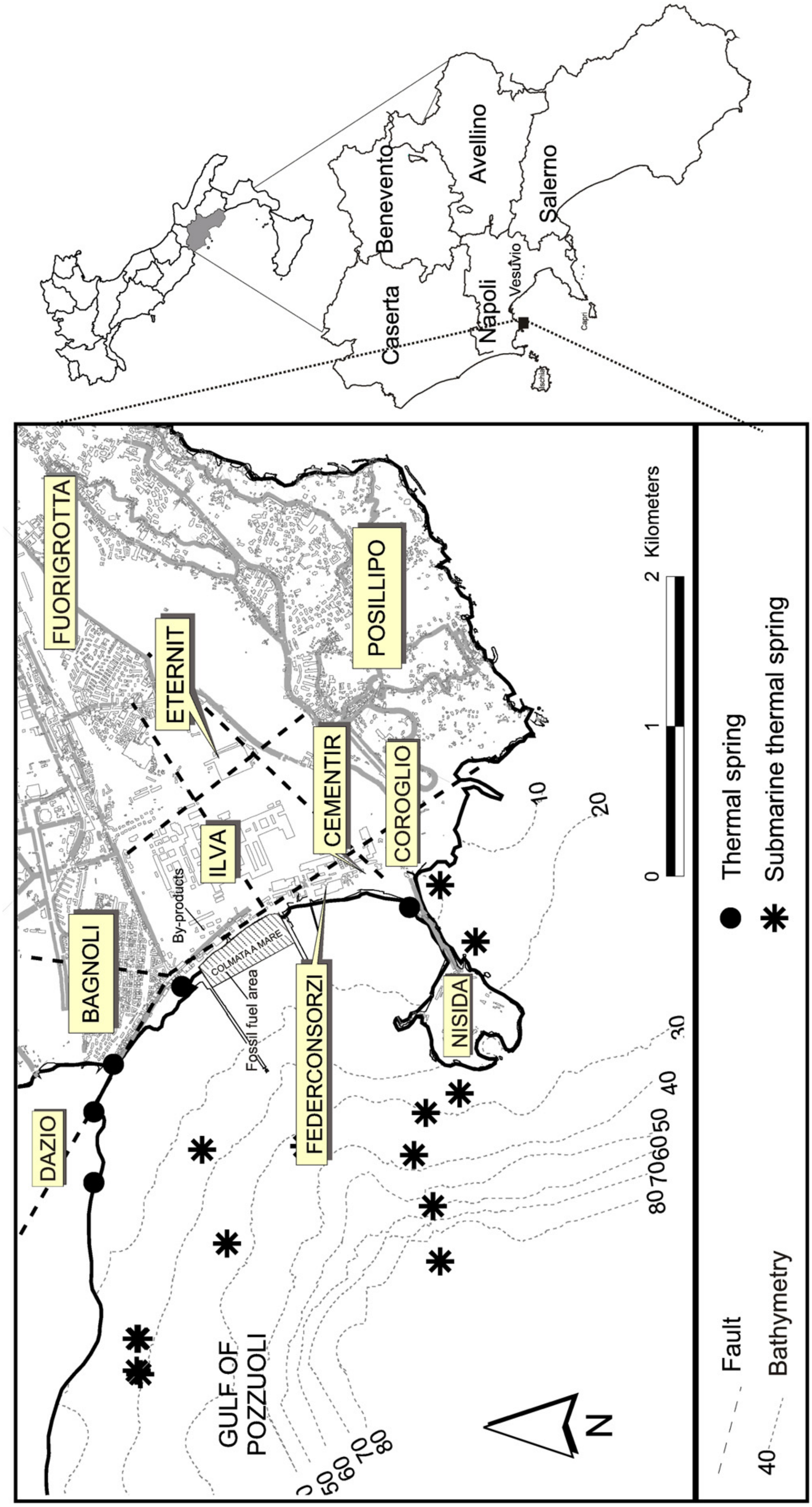

Fig. 1. The Bagnoli brownfield site and coastal area sketch map.

fluids related to the volcanic activity of the Campi Flegrei (i.e. natural), and 2) fossil fuel use, scums and slags from steel production, and emission of industrial fumes (i.e. anthropogenic).

The results of leach tests carried out on industrial materials (scum, slag and landfill material) from the brownfield site showed that their microstructural configuration is stable and heavy metals are most likely

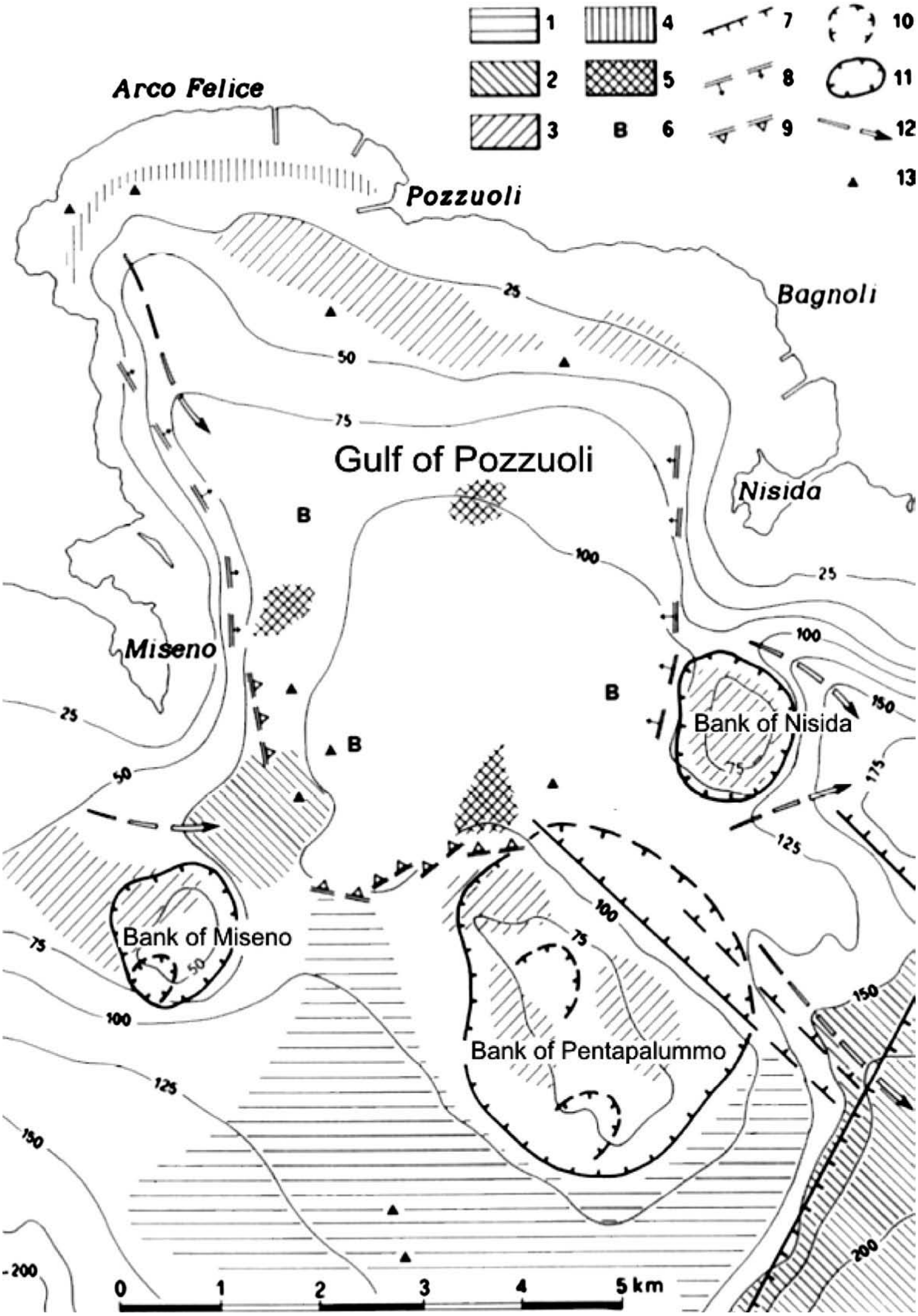

Fig. 2. Geomorphological map of Gulf of Pozzuoli (After De Pippo et al., 1988). 1) Würmian shelf; 2) sediment accumulation area; 3) post-Würmian terrace; 4) Roman age Beach; 5) basin border; 6) internal basin area; 7) regional fault scarp; 8) volcano-tectonic scarp; 9) buried volcano-tectonic scarp (probably Caldera borders); 10) crater border; 11) volcanic bank; 12) canyon; 13) gas sources.

trapped in the lattice of the insoluble microstructure phase. Since industrial materials are unlikely to contribute metallic pollution to local groundwaters, De Vivo and Lima (2008) point out the need of a remediation plan focused mainly on removing the polycyclic aromatic hydrocarbons (PAHs) found along the shoreline, whose location corresponds with a filling structure ("Colmata a mare") delimited by two piers and filled by scum, slag and landfill material from the brownfield site during the period 1962–1964.

Investigations on pollutants released to the marine environment from the brownfield site were carried on by I.C.R.A.M. (Istituto Centrale per la Ricerca scientifica e tecnologica Applicata al Mare) (ICRAM, 2005), a governmental research institute supervised by the Ministry for the Environment, Land and Sea. The ICRAM provides institutional support for the policies of the central government and local authorities in the resolution of marine environmental problems.

In this paper we will focus on the results of characterization activities carried out on the coastal sea sediments facing the brownfield site and in particular on the "Colmata a mare" structure.

The spatial variability of contaminant concentrations in sea sediments will be evaluated down to a depth of 5 m from the seabed. Univariate and multivariate geostatistical analyses will be performed on geochemical data to assess the nature (anthropic or/and geogenic) and the extent of contamination sources for the sea sediments.

2. Study area

Bagnoli is located in the south-eastern sector of the Gulf of Pozzuoli which is part of the Campi Flegrei volcanic complex (Fig. 2). Volcanic products related to Campi Flegrei activity are part of the K-series of the Roman co-magmatic province (Peccerillo, 1985; Washington, 1906) and their composition varies from trachybasalts to phonolitic and peralkaline trachytes (Di Girolamo, 1978; Armienti et al., 1983).

The coastal area in front of the Bagnoli brownfield site is part of a shelf facing the entire shoreline of the Gulf of Pozzuoli. The shelf extends from the coastline toward the 50 m bathymetry and borders a central recently collapsed area (12–10 kyr bp) (De Pippo et al., 1988). Volcanic banks (Nisida, Pentapalummo and Miseno) bordered by an external Würmian shelf are present on the southern side of the collapsed area (De Pippo et al., 1988; Fig. 2).

The presence of submarine hydrothermal springs located along a NW–SE axis, in the eastern sector of the Gulf of Pozzuoli (Sharp and Nardi, 1987), is clearly related to the secondary volcanic activity of the Campi Flegrei area. On the seabed surface sediment grain size is mostly sandy becoming silty in correspondence with some sheltered areas along the coastline, that are characterized by low energy for sediment transport (Cocco et al., 1988).

In the Bagnoli–Fuorigrotta area, the water table is found slightly above sea level and can be intercepted at shallow depths, especially on the south side of the local railroad (Fig. 3). The groundwater of the Bagnoli plain, resupplied directly by rainfall, is part of a wider groundwater body which spans the whole Campi Flegrei area and discharges directly to the sea (Celico and Habetswallner, 1999a,b, 2000).

De Vivo and Lima (2008) showed that metal pollution of the Bagnoli brownfield groundwaters is due mostly to the upwelling of hydrothermal fluids occurring for the major part along Neogenic fractures and

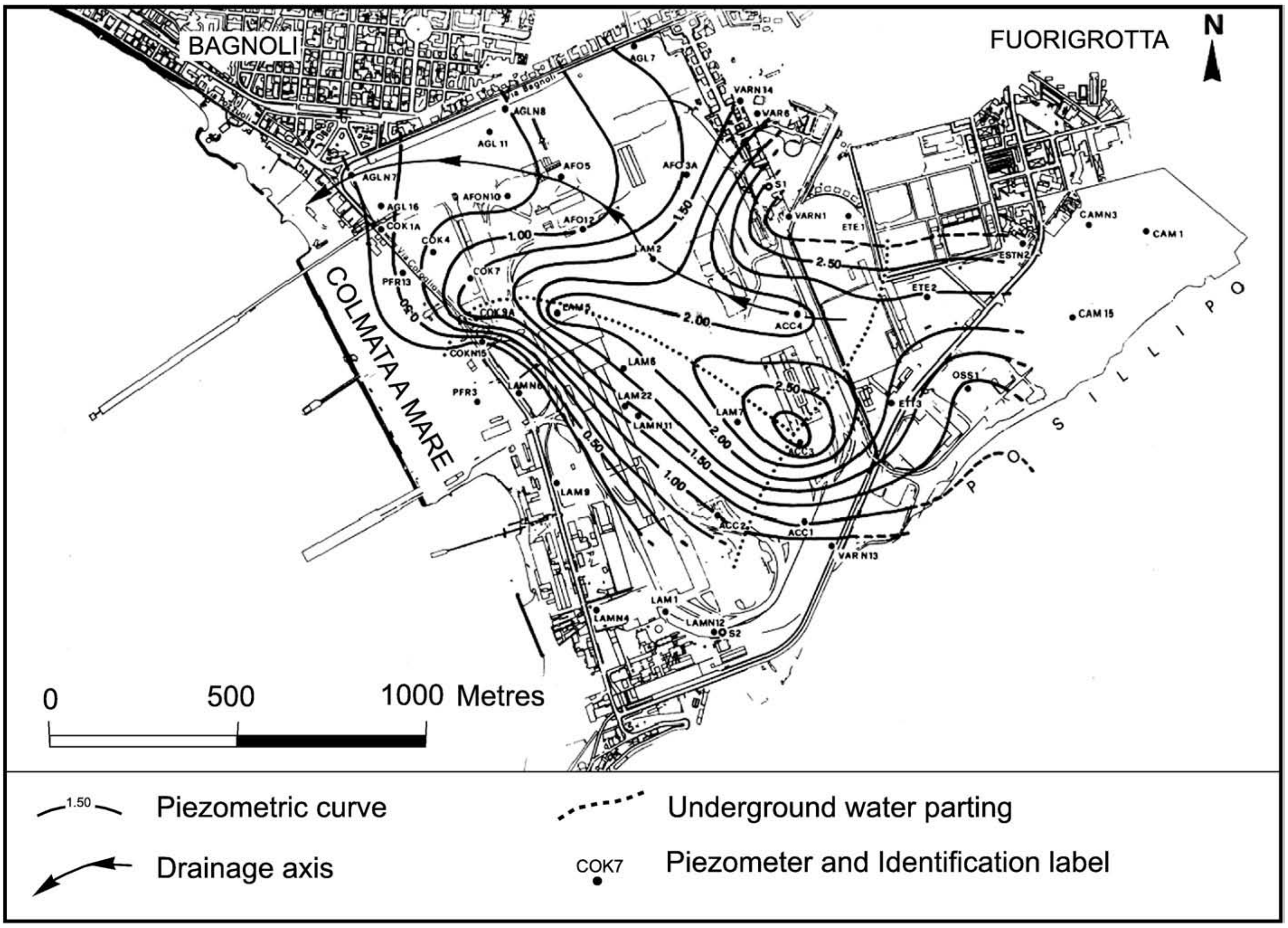

Fig. 3. Morphology of the piezometric surface obtained from surface and deep boreholes piezometers (After De Vivo and Lima, 2008).

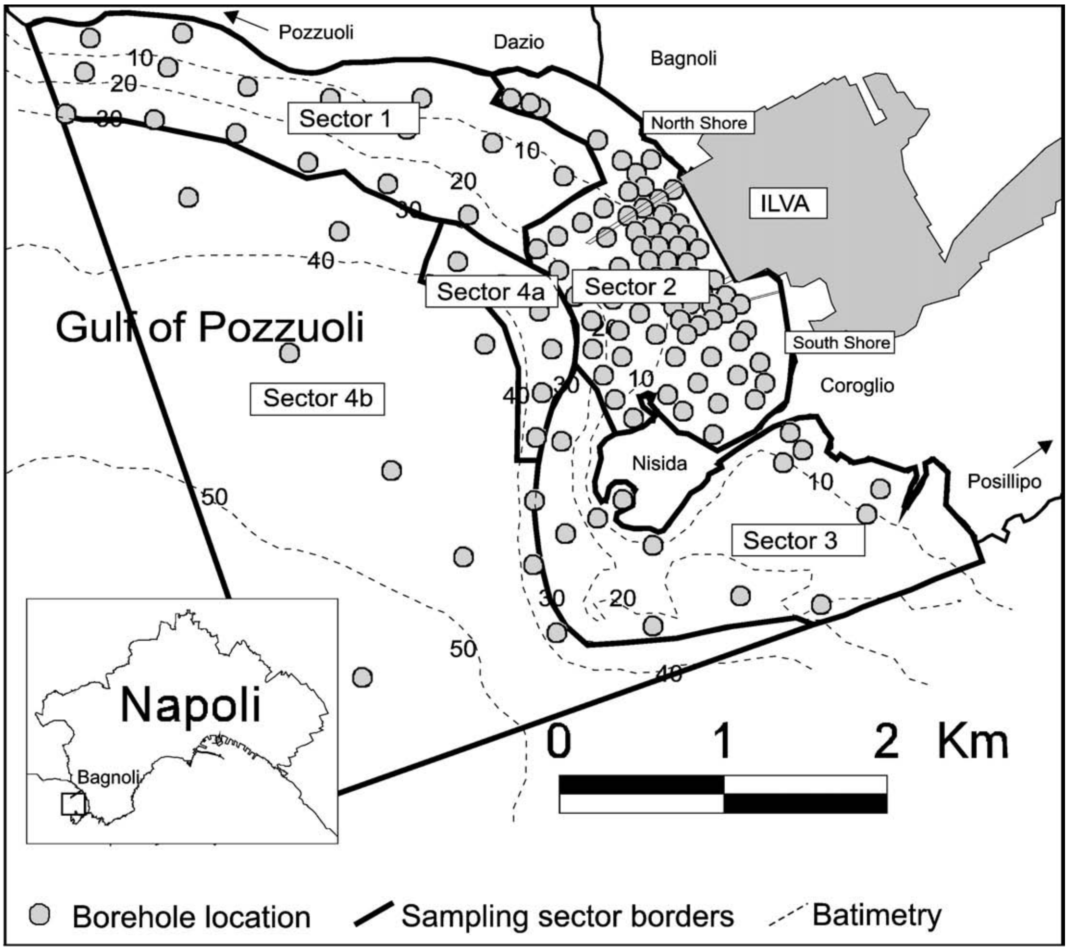

Fig. 4. Bagnoli sea sediments sampling sectors and borehole location map.

faults. These fluids contain heavy and potentially toxic metals such as As, Hg, Cu, Pb, and Cd. In contrast, hydrocarbons (mostly PAHs) are leached by rain fall percolating through soils and landfills contaminated by industrial activities.

3. Field activities and sampling

Sampling activities of sea sediments have been carried out by I.C.R.A.M. (ICRAM, 2005) in 4 distinct sectors to characterize geochemically the Bagnoli coastal area and the sea sediments facing the brownfield site.

The sectors have been delimited based on the expected contamination, as a function of the proximity to the brownfield site (Fig. 4):

- Sector 1: from the W seashore (Pozzuoli–Dazio) to 30 m sea bathymetry.
- Sector 2: from the Bagnoli–Coroglio seashore to 30 m sea bathymetry (this is the sector closest the brownfield site where the highest contamination is to be expected).
- Sector 3: from the area around Nisida Island to Posillipo coast (this area is not directly affected by industrial contamination).
- Sector 4: from 30 m to 50 m sea bathymetry coastal area in front of the brownfield site (4a) and from the 50 m sea bathymetry to the external limit defined by the Italian Government for the "Site of National Interest (SNI)" of Bagnoli (4b).

384 sea sediment samples (between November 2004 and March 2005) were collected in the 4 sectors from 128 boreholes at different depths down the seabed along the coastline (0–20 cm, 20–30 cm, 30–50 cm, 100–120 cm, 150–180 cm, 180–200 cm, 280–300 cm, and 380–400 cm) with a sampling density decreasing at increasing depths and distances respectively from the seabed surface and from the coastline. In the sector closest to the brownfield site (sector 2), a 150 m × 150 m cell sampling grid was used to locate boreholes, whereas, in the other sectors a 500 m × 500 m cell sampling grid was used (Fig. 4).

Sampling operations were carried out using a pontoon which was moved with the help of two tugboats. 3 containers were positioned on the pontoon and one of them used as a refrigerator for storage of cores.

The placement of sampling stations, established with a portable DGPS instrument, was performed prior to the arrival of the pontoon and sampling activities was carried on by means of a vibrating drill,

Table 1
Detection limits for ICP-OES and AAS analyses of metals in sea sediments.

Element	ICP-OES	AAS
	mg/kg	mg/kg
Al	3	0.2
As	3.5	0.1
Be	0.02	0.02
Cd	0.4	0.01
Cr	0.4	0.1
Co	0.4	0.1
Mn	1	0.1
Fe	1	0.1
Hg	–	0.01
Ni	1	0.1
Pb	2.5	0.1
Cu	0.3	0.1
V	0.5	0.3
Zn	0.1	0.01

with a large base ensuring its vertical position during the coring (ICRAM, 2005).

In order to collect uncontaminated sediment, an internal liner was used during drilling operations and, subsequently, collected cores were transported on the pontoon and opened lengthwise using a circular saw. From each core, subsamples were collected in correspondence of established depths (see above), labelled and stowed in the refrigerator until their daily delivery to the ICRAM laboratories.

Samples were also collected along the seashore on the northern and southern side of the “Colmata a mare”, the structure filled by scum, slags and landfill materials artificially created between two piers. The seashore characterization was carried out in a first phase by means of 20 shallow boreholes from which sediments (mostly sands) were collected at different depths. Since on the northern side of seashore, analyses carried out by ICRAM, indicated very high concentration levels of PAHs, a second phase of characterization was planned to collect 22 additional sand samples only from this area.

4. Methods

4.1. Chemical analyses and granulometry

Analyses of metals (Al, As, Be, Cd, Co, Cr, Fe, Hg, Mn, Ni, Pb, Cu, Sn, V, and Zn) and organic compounds (PAHs — Polycyclic Aromatic Hydrocarbons, both single compounds and total, and PCBs — Polychlorinated biphenyls) on the Bagnoli sea sediment samples were performed at ICRAM laboratories.

For metallic elements, microwave assisted acid digestion for a total decomposition of matrices was applied to sediment samples according to EPA Method 3052 (US Environmental Protection Agency, 1996b).

Table 2A
Statistical parameters for sea sediment sample at different sampling depths (0–20 cm, 20–30 cm, 30–50 cm).

Element/compound	*N*	Mean	Median	Min	Max	Skewness	Kurtosis	St. dev.
Depth 0–20 cm								
Al (%)	125	0.48	0.38	0.07	2.1	1.9	4.3	0.4
As (mg/kg)	125	11.7	8.5	2.2	55	2.3	6.9	8
Be (mg/kg)	125	3.6	3.3	0.01	11.7	0.71	0.97	2.1
Cd (mg/kg)	125	0.76	0.39	0.01	7.4	3	12.2	1.1
Co (mg/kg)	125	7	6.8	1.3	28	1.7	3.9	5
Cr (mg/kg)	125	16.4	12.7	4	103	3.3	16	13.2
Cu (mg/kg)	125	47	31	5.88	236	1.9	4.2	42
Fe (%)	125	4.5	4.1	0.7	10.8	1	−0.3	2.6
Hg (mg/kg)	125	0.57	0.36	0.01	3.5	2.4	5.9	0.7
Mn (mg/kg)	125	1330	940	277	8070	2.9	9.3	1293
Ni (mg/kg)	125	71	74	3.41	181	0.11	−1.4	50
Pb (mg/kg)	125	292	221	21	1773	2.44	8.7	267
Sn (mg/kg)	125	17.35	14	2.2	88	2.6	8.6	15
V (mg/kg)	125	74.44	64	26	575	6.32	44	63
Zn (mg/kg)	125	593	386	93	2829	1.8	3.9	517
PAH (mg/kg)	125	165	37	0.1	2947	4.2	20	417
PCB's (μg/kg)	125	108	23	0.8	6665	10.8	119	599
Depth 20–30 cm								
Al (%)	85	0.6	0.45	0.14	2.6	2.5	10.6	0.4
As (mg/kg)	85	9.1	7.3	1.8	26	1.6	2.6	4.7
Be (mg/kg)	85	4.4	3.9	0.05	13.3	1	1.2	2.8
Cd (mg/kg)	85	1.4	0.9	0.06	9	2.6	7.5	1.7
Co (mg/kg)	85	9.2	7.3	2.1	26	1.5	2.1	4.9
Cr (mg/kg)	85	14.5	11.5	4.8	61	2.5	7.6	9.9
Cu (mg/kg)	85	59	48	5.82	210	1.3	1	50
Fe (%)	85	5.2	4.7	0.6	11.6	0.65	−0.37	2.5
Hg (mg/kg)	85	0.72	0.37	0.01	6.3	3.2	11.7	1.1
Mn (mg/kg)	85	1803	1212	378	10,779	2.7	7.6	1920
Ni (mg/kg)	85	105	109	31.04	280	1	4.6	38
Pb (mg/kg)	85	393	316	34.69	2220	2.4	8.9	346
Sn (mg/kg)	85	20	16	0.05	79	1.5	2.5	17
V (mg/kg)	85	68	65	23	221	2.6	13.7	26
Zn (mg/kg)	85	827	613	90	3200	1.5	1.9	706
PAH (mg/kg)	85	159	48	0.08	1806	3.5	13	323
PCB's (μg/kg)	85	56	29	1	458	2.7	7.7	87
Depth 30–50 cm								
Al (%)	85	0.64	0.48	0.12	3.9	3.2	14	0.6
As (mg/kg)	85	8.9	7	3.3	37	2.6	8.4	5.6
Be (mg/kg)	85	4.6	4.2	0.03	12.1	1	0.3	3
Cd (mg/kg)	85	2.19	0.72	0.05	44	6.6	51	5.3
Co (mg/kg)	85	9	7.1	3.3	37	2.5	7.7	5.6
Cr (mg/kg)	85	13	9.2	3.3	142	5.3	31	19
Cu (mg/kg)	85	56	38	5.51	222	1.26	0.95	52
Fe (%)	85	4.6	3.5	1.1	11.2	0.87	−0.5	2.5
Hg (mg/kg)	85	1	0.3	0.01	8.3	2.6	7.8	1.5
Mn (mg/kg)	85	1703	1056	407	11,458	2.9	10	1929
Ni (mg/kg)	85	98.1	101	21	184	−0.3	−0.6	36
Pb (mg/kg)	85	412	207	35	3446	3.23	13.63	534
Sn (mg/kg)	85	23	11.1	0.05	177	2.9	9.3	34
V (mg/kg)	85	71	60	15.6	557	6.80	54	60
Zn (mg/kg)	85	886	512	102	5185	2.1	5.3	1018
PAH (mg/kg)	85	212	29	0.05	2265	2.8	7	490
PCB's (μg/kg)	85	55	6.4	0.07	726	3.7	18	109

Each sample was digested in an HCl and HNO_3 (1:3) solution for 15 min. The sample was placed in an inert polymeric vessel, sealed and heated in a microwave system. A 4-step heating cycle was performed prior to adding HF to the cooled solution. After a second heating cycle, H_3BO_3 was added to the solution. Finally, the solution was filtered and brought to a final volume of 100 ml with the addition of ultrapure water. As, Cd, Cu, Pb and Zn were analyzed by ICP-OES (Inductively Coupled Plasma-Optical Emission Spectrometry), Hg (and other elements, if characterized by concentrations below the ICP-OES detection limits) were analyzed by AAS (Atomic Absorption Spectroscopy) (Table 1).

For PAHs determinations, freeze-dried samples were extracted from the pulp using a mixture of methanol and acetone. After centrifugation the supernatant was filtered at 0.2 µm and then analyzed by HPLC-FLD (High-Performance Liquid Chromatography whit Fluorescence Detector) according to EPA Method 8310 (US Environmental Protection Agency, 1986). Detection limits are 0.5 µg/kg for each PAH. For PCBs determinations, dry samples were placed in a glass thimble with the addition of a solution of dichloromethane/hexane (1:1) and centrifuged after sonication. The organic fraction was collected in a flask. After repeating the operation, the two fractions were combined and treated with activated copper and then the solution was evaporated up to 1

Table 2B

Statistical parameters for sea sediment sample at different sampling depths (100–120 cm, 180–200 cm, 280–300 cm, 380–400 cm).

Element/compound	N	Mean	Median	Min	Max	Skewness	Kurtosis	St. dev.
Depth 100–120 cm								
Al (%)	44	0.45	0.38	0.12	1.3	1.6	2.8	0.3
As (mg/kg)	44	7.3	6.6	2.8	18	1.9	4.7	2.8
Be (mg/kg)	44	5.6	5.2	0.4	12	0.6	−0.2	3
Cd (mg/kg)	44	1	0.5	0.01	5.9	2.2	4.9	1.3
Co (mg/kg)	44	7.3	6.7	2.8	18	1.9	4.5	3
Cr (mg/kg)	44	8.6	6.2	1.9	66	5.1	30	9.7
Cu (mg/kg)	44	34	14.6	5.4	355	4.7	26	56
Fe (%)	44	3.4	2.8	1.8	8.4	2	3.4	1.5
Hg (mg/kg)	44	0.52	0.09	0.01	4.8	3.2	11	1
Mn (mg/kg)	44	991	599	433	9709	5.41	32	1449
Ni (mg/kg)	44	105	119	15	157	−0.81	−0.11	35
Pb (mg/kg)	44	212	114	30	1131	2.4	6.3	237
Sn (mg/kg)	44	11.5	5.7	2	88	3.2	11.9	16
V (mg/kg)	44	66	61	35	141	2.2	6.6	20
Zn (mg/kg)	44	500	221	93	1779	1.3	0.4	515
PAH (mg/kg)	44	105	5.2	0.05	1654	3.9	16	322
PCB's (µg/kg)	44	68	1.6	0.05	2186	6.4	41	331
Depth 180–200 cm								
Al (%)	33	0.57	0.5	0.18	1.6	1.41	2.51	0.3
As (mg/kg)	33	8.2	6.2	1.4	73	5.6	31	11.9
Be (mg/kg)	33	6.1	5.2	0.9	14.1	0.5	−0.43	3.6
Cd (mg/kg)	33	1	0.3	0.05	9.4	3.2	10.1	2.1
Co (mg/kg)	33	8.2	6.2	1.4	73	5.5	31	11.9
Cr (mg/kg)	33	7.7	6.8	2.0	38	4.1	20	6
Cu (mg/kg)	33	36	10.6	2.9	408	4.4	22	73
Fe (%)	33	3.2	2.6	1.4	10.5	2.9	9.3	1.8
Hg (mg/kg)	33	0.29	0.03	0.01	3.4	3.6	12.2	0.83
Mn (mg/kg)	33	865	526	325	6505	4.3	21	1125
Ni (mg/kg)	33	114	125	12.8	171	−1	1.1	34
Pb (mg/kg)	33	282	99	25	2061	2.7	6.2	510
Sn (mg/kg)	33	26	5.5	2.2	358	4.2	19	68
V (mg/kg)	33	62	60	35	99	0.5	1	14
Zn (mg/kg)	33	570	195	100	4229	2.7	6.8	999
PAH (mg/kg)	33	110	1	0.07	2514	5	26	457
PCB's (µg/kg)	33	76	1.46	0.34	1244	4.1	18	241

Depth 280–300 cm*

Element/compound	N	Mean	Median	Min	Max
Al (%)	7	0.69	0.7	0.43	1
As (mg/kg)	7	8.4	8.3	6.3	10.5
Be (mg/kg)	7	6.8	7.1	4.2	11.7
Cd (mg/kg)	7	0.2	0.2	0.1	0.4
Co (mg/kg)	7	8.4	8.3	6.3	10.5
Cr (mg/kg)	7	7.5	7.6	4.5	11.7
Cu (mg/kg)	7	8.4	9.1	4.9	11.2
Fe (%)	7	4.1	3.2	2.4	10
Hg (mg/kg)	7	0.02	0.02	0.01	0.07
Mn (mg/kg)	7	684	600	431	1733
Ni (mg/kg)	7	122	124	104	144
Pb (mg/kg)	7	208	120	89	755
Sn (mg/kg)	7	6.4	2.7	2.1	22
V (mg/kg)	7	80	79	62	107
Zn (mg/kg)	7	264	159	128	910
PAH (mg/kg)	7	4.0	1.5	0.1	8.4
PCB's (µg/kg)	7	1.2	0.9	0	3.6

Depth 380–400 cm*

Element/compound	N	Mean	Median	Min	Max
Al (%)	5	0.59	0.52	0.46	0.87
As (mg/kg)	5	9	9	6	12
Be (mg/kg)	5	3	3.5	0.5	4.4
Cd (mg/kg)	5	0.2	0.2	0.1	0.5
Co (mg/kg)	5	8.7	9	6	12
Cr (mg/kg)	5	9.6	8.1	4.8	18
Cu (mg/kg)	5	11	7	5.7	28
Fe (%)	5	3.3	2.8	2.2	5.8
Hg (mg/kg)	5	0.0146	0.015	0.011	0.019
Mn (mg/kg)	5	935	582	365	2731
Ni (mg/kg)	5	128	128	121	137
Pb (mg/kg)	5	238	105	75	818
Sn (mg/kg)	5	8.9	4.8	3.9	27
V (mg/kg)	5	83	80	74	98
Zn (mg/kg)	5	438	173	133	1546
PAH (mg/kg)	5	3	3.5	0.1	6.4
PCB's (µg/kg)	5	0.6	0.7	0.1	1

*Due to the lack of a statistically significant number of samples, only basic statistics are reported.

Table 3
Varimax-rotated factor (two-factor) model for 128 samples of sea sediments collected at a depth of 0–20 cm. Bold entries report loading values over 0.5; underlined entries report negative loadings below −0.5.

Element/compound	Factors	
	F1	F2
Sand	−0.02	**0.94**
Pelite	**0.75**	−0.10
As	0.02	−0.52
Cd	**0.87**	0.19
Co	0.40	**0.63**
Cr	0.37	−0.14
Cu	**0.74**	0.19
Fe	**0.53**	**0.59**
Hg	**0.83**	−0.04
Mn	0.47	0.17
Ni	−0.04	**0.90**
Pb	**0.85**	0.33
Sn	**0.85**	0.11
Zn	**0.91**	0.25
PAH	0.38	−0.03
PCBs	0.10	0.07
Variance % (Total data)	65.5	34.5

drop left. After purification, the samples were passed through an active carbon column to separate Dioxins and Furans from PCBs and analyzed by means of a gas chromatograph equipped with an electron capture detector (according to EPA Method 8082 (US Environmental Protection Agency, 1996c) with a detection limit of 0.1 µg/kg for each PCB congener.

For the evaluation of granulometry, the sediment samples were prepared following the method of Romano and Gabellini (2001). After sieving, the finest fraction (<63 µm) was dried at 40 °C and suspended in a solution with distilled water and sodium hexametaphosphate (0.05%) to be analyzed with an X ray sedigraph (Micromeritics Sedigraph 5.100). The coarser fraction (>63 µm) was examined under a stereo microscope to qualitatively determine the presence of non-geological materials. Granulometry data were expressed as percent of gravel (>2 mm), sand (2 mm>×>0.063 mm), silt (0.063 mm>×>0.004 mm), clay (<0.004 mm) and pelite (<0.063 mm), which includes both silt and clay fractions.

The quality of all data was assessed by estimations of accuracy and precision. Instrument calibration utilised calibration solutions included at the beginning and end of an analytical run. Spiking of the digestion solution with a tracer element controlled internal standardization. Precision was calculated on in-house replicates. Precision is ±100% at the detection limits and increases to better than ±10% at concentrations of 50 times the detection limit or higher.

The certified reference materials for marine sediment (PACS-2, NRC, Canada) were used to control accuracy of the analytical method for sediments. Recovery values found were 101% for sediments (ICRAM, 2005).

4.2. Statistical analysis and geochemical mapping

Chemical and granulometry data were stored in 7 separate databases, depending on the sampling depth of the corresponding sample. In order to show the single-element geochemical distribution, a detailed univariate analysis was performed on each of produced databases. A value corresponding to 80% of the instrumental detection limit (IDL) was assigned to all data reported as below the IDL in order to allow their inclusion in the statistical analysis. Table 2A and 2B present all the calculated statistical parameters for each sampling depth.

Maps showing chemical data distributions were generated using the ArcView™ and GeoDas™ softwares (Cheng, 2003). The multifractal inverse distance weighted (MIDW) algorithm was used as the interpolation method to compile the geochemical maps (Cheng, 1999a, b; Lima et al., 2003) and the C-A (Concentration-Area) method was used to define concentration intervals on maps (Cheng et al., 1994; Lima et al., 2003). Intervention maps were also produced reclassifying the interpolated values surfaces using the intervention limits (I.L.) established by the Italian Environmental Law for sea sediments (Ministero dell'Ambiente e della Tutela del Territorio, 2003) and for soils (Parlamento Italiano, 2006) (Table 3). *R*-mode factor analysis was performed on data produced for the 0–20 sampling depth, using G-RFAC (Miesch Programs, 1990) software on a matrix containing 12 elements, 2 organic compounds and granulometric fraction data for sand and pelite.

In order to facilitate the interpretation of factor analysis results, a Varimax rotation was applied to minimize the number of variables with high loadings on each factor. This process simplifies the transformed data matrix and eases the final interpretation of the data. A two-factor model which accounts for 56% of data variability was deemed appropriate for the sea sediments facing the Bagnoli brownfield area (Table 4) Variables with loadings over 0.5 were considered to describe quite effectively the composition of each factor. The associations of the two-factor model are F1: Zn, Cd, Sn, Pb, Hg, pelite, Cu, Fe; F2: Sand, Co, Fe, Ni, (− As).

5. Results and discussion

Granulometric data reclassified according to Nota (1958) show a wide spatial variability which is clearly evident in Fig. 5 that also shows the spatial distribution of granulometric classes for the depth interval from 0 to 50 cm below the seabed. Sand and pelitic sand are

Table 4
Intervention limits (I.L.) established by Italian Law for soils (D.M. 152/06) (Parlamento Italiano, 2006) band sea sediments (D.M. 367/03) (Ministero dell'Ambiente e della Tutela del Territorio, 2003). Geochemical background values determined for sea sediment of the Pozzuoli Bay (Damiani et al., 1987) (Column A), for stream sediments on volcanoclastic deposits (Albanese et al., 2007) (Column B) of Campania region and for neapolitan volcanic soils (Cicchella et al., 2005) (Column C) are reported, where possible.

Element/compound	I.L. soils D. Lgs 152/06		I.L. sea sediments D.M. 367/03	A	B	C
	Residential use	Industrial use		Damiani et al., 1987	Albanese et al., 2007	Cicchella et al., 2005
As (mg/kg)	20	50	12	–	10.2–13.9	5–15
Be (mg/kg)	2	10	–	–	–	–
Cd (mg/kg)	2	15	0.3	–	0.43–0.69	0.1–0.5
Co (mg/kg)	20	250	–	–	–	–
Cr (mg/kg)	150	800	50	30	12–28	5–15
Cu (mg/kg)	120	600	–	20	19–40	20–120
Hg (mg/kg)	1	5	0.3	0.25	0.067–0.2	0.05–0.2
Ni (mg/kg)	120	500	30	20	–	–
Pb (mg/kg)	100	1000	30	60	29–51	30–85
Sn (mg/kg)	1	350	–	–	–	–
Zn (mg/kg)	150	1500	–	80	82–156	–
V (mg/)kg	90	250	–	–	–	–
PCB's (µg/kg)	0.06	5	4	–	–	–
PAH (mg/kg)	10	100	0.2	–	–	–

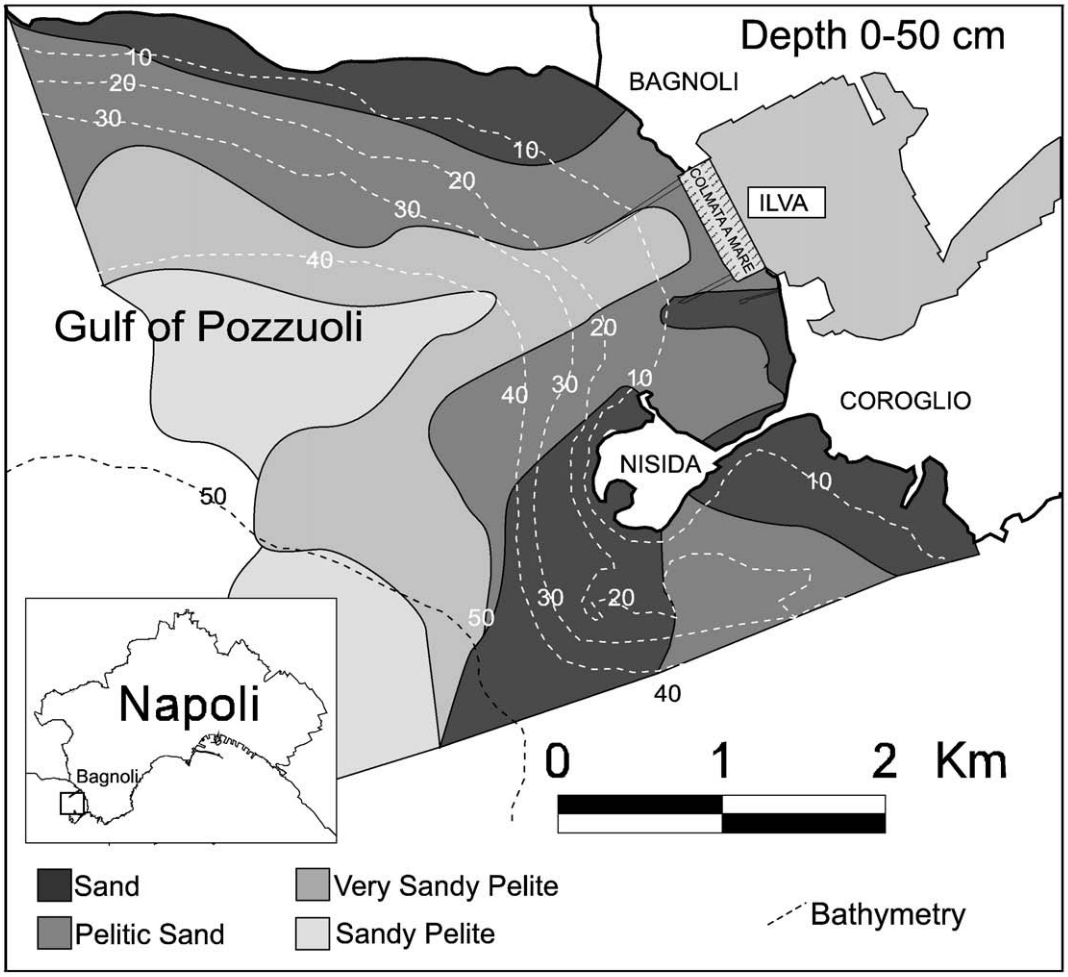

Fig. 5. Map of granulometric data distribution in the 0–50 sampling depth interval, reclassified according to Nota (1958).

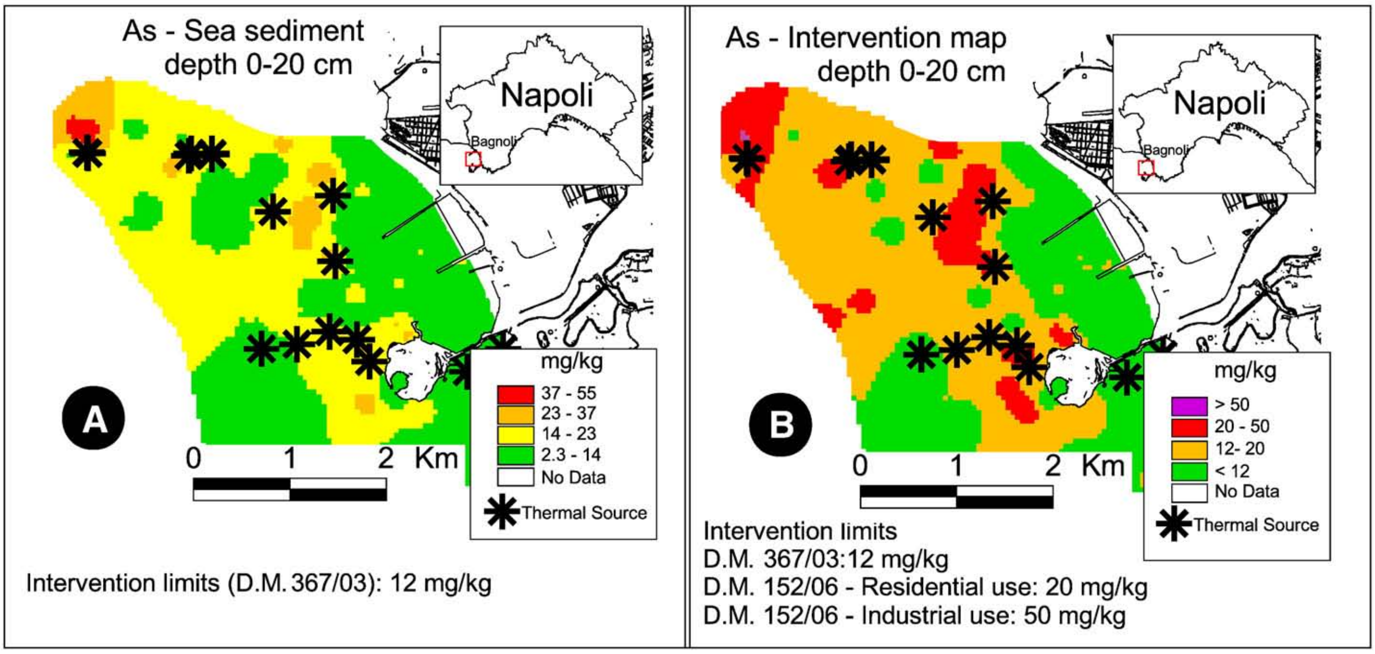

Fig. 6. Geochemical map of As in sea sediments. Baseline (A) and intervention (B) map for the 0–20 sampling depth.

mostly concentrated along the shorelines and in a wide strip oriented N–S on the southern side of Nisida.

Finer sediments, mostly pelitic with a considerable amount of sand (decreasing toward the open sea), are concentrated in proximity to the front side of the filling structure ("Colmata a mare"), due to the low energy of marine currents locally diverted by the presence of the two bordering piers and influenced by the presence of the Nisida promontory. Microscopic observation of the coarser fraction of sediments sampled in proximity of the "Colmata a mare" and along the shoreline facing the Bagnoli brownfield site revealed the presence of a large amount of anthropogenic grains mostly made of tar and oil residues, blast furnace slag and iron pellets.

We discuss, in detail, only the distribution of As, Pb and Ni, as these elements have the most relevance to potential human health issues. Maps showing the distribution of the balance of studied elements are given as a file repository (Appendix A).

Arsenic concentration values in the upper sampled layer (0–20 cm) range between 2.3 and 55 mg/kg (Fig. 6A). Higher concentrations (> 14 mg/kg), as established by the C-A method applied to interpolated map, show a strong spatial correspondence with the presence of some submarine thermal springs scattered along a NW–SE axis connected to the active volcanic field of Campi Flegrei. Lower concentrations (between 2.3 and 14 mg/kg) are mostly concentrated in the sea sediments facing the "Colmata a mare" and the shorelines of the brownfield site.

In the upper sampled layer (0–20 cm) As I.L. for sea sediments (12 mg/kg) are exceeded in correspondence with the submarine thermal springs in the Pozzuoli Bay (Fig. 6B), as well.

Arsenic concentrations decrease in the lower sampled layers (Table 2A and 2B), probably due to the higher temperatures of the

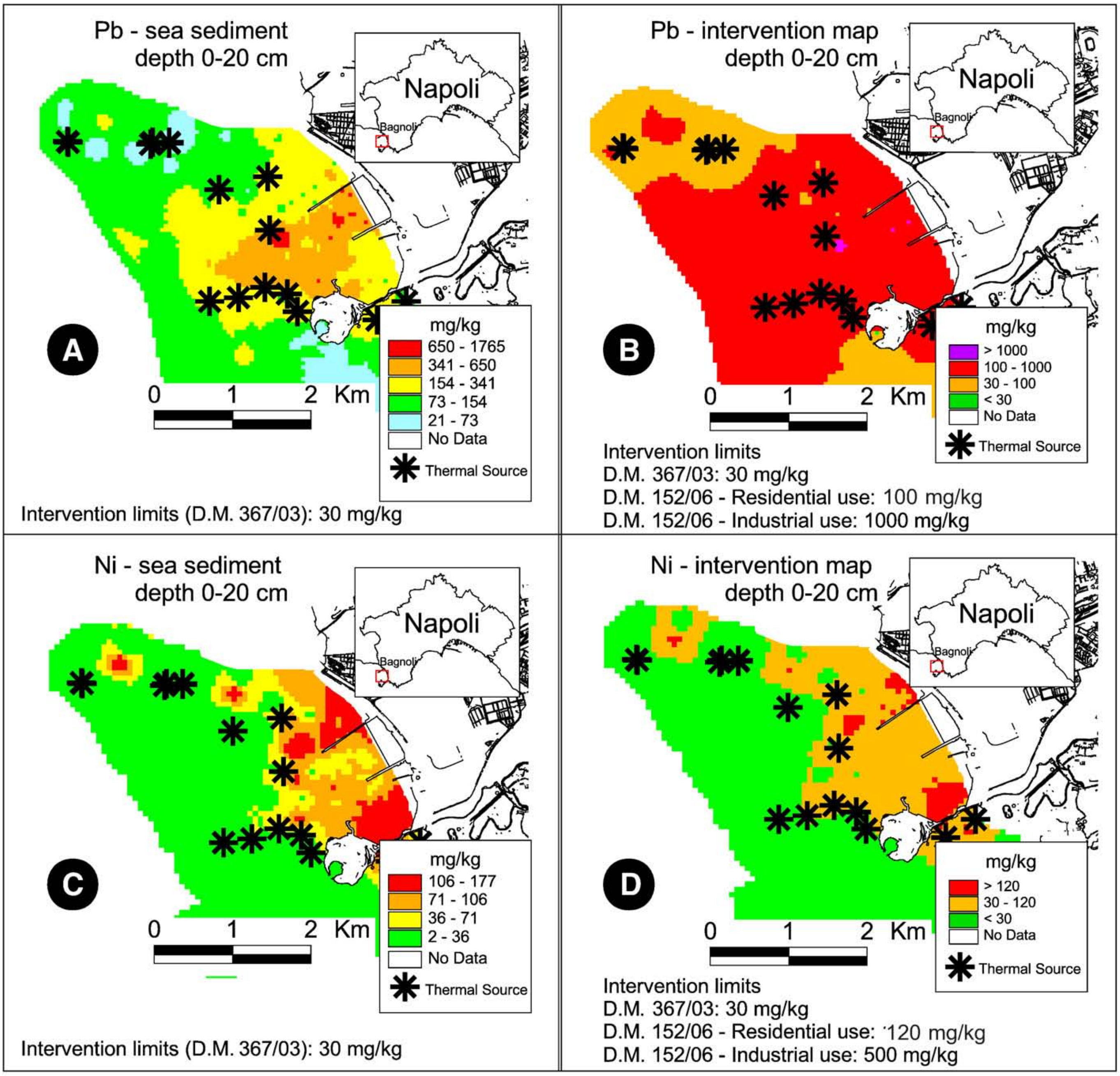

Fig. 7. Geochemical map of Pb and Ni in sea sediments. Baseline (A, C) and intervention (B, D) maps for the 0–20 sampling depth.

upwelling thermal fluids limiting the precipitation process. At the 30–50 cm sampling depth layer, As concentrations range between 1.8 and 26 mg/kg and the I.L. for sea sediments (Ministero dell'Ambiente e della Tutela del Territorio, 2003) are exceeded in the sediments adjacent to Nisida. At a 100–120 cm sampling depth layer, As concentration ranges between 3.3 and 18 mg/kg and I.L. (Ministero dell'Ambiente e della Tutela del Territorio, 2003) are exceeded only in a small area in front of the Dazio coastline. In the lower sampled layers, As concentrations are generally below the I.L. established by the Italian Law (D.M. 367/03) and only at the 180–200 cm sampling depth a single sea sediment sample, collected in front of the "Colmata a mare" structure, reached the concentration of 73.5 mg/kg. Although As concentrations in the topmost sampling layers tend to exceed the I.L. established by Italian Law (Ministero dell'Ambiente e della Tutela del Territorio, 2003) for sea sediments (Fig. 6B), any remediation concerning As would be of little use in the study area, due to the source of As from natural thermal springs. This holds true for all other metals associated with thermal volcanic activity in the Campi Flegrei (De Vivo and Lima, 2008).

Lead concentrations in the upper sampling layer (0–20 cm) (Fig. 7A), range between 21 and 1765 mg/kg. According to the I.L. established by the Italian Law (Ministero dell'Ambiente e della Tutela del Territorio, 2003) for sea sediments (30 mg/kg), almost the entire study area should undergo remediation (Fig. 7B). The latter condition is also true for the remaining of the sampled layers (Table 2A and 2B) since at the 30–50 sampling depth, Pb concentrations range between 50 and 2739 mg/kg; at 100–120 cm sampling depth they range between 30 and 1131 mg/kg; at the 180–200 cm sampling depth they range between 25 and 2061 mg/kg; and at the lower sampling depths (280–300 cm and 380–400 cm) they range, respectively, between 89 and 755, and 75 and 818 mg/kg . For all sampling layers, the higher concentrations of Pb in the sea sediments are located in relation to the "Colmata a mare" structure. Thus, it is reasonable to assume that the higher Pb concentrations are due probably to the discharge of the brownfield groundwaters to the sea across the filling structure.

If dredged sediments from sector 2 were to be reused inland, the major part could be transferred to an industrial area, in accordance with I.L. established by Italian law (Parlamento Italiano, 2006) for industrial land use (<1000 mg/kg); however, only a small part would be suitable for residential/recreational land use (I.L.<100 mg/kg) (Fig. 7B) such as the urban park planned to be located in the Bagnoli brownfield area after its remediation).

The highest concentrations of Ni found in the upper sampled layer (0–20 cm) (Fig. 7C) are mostly located in sediments sampled on both the northern and southern side of the "Colmata a mare" structure occupying the Bagnoli–Coroglio coastline. Values for Ni range between 2 and 177 mg/kg and strict application of the I.L. for sea sediments (30 mg/kg) (Ministero dell'Ambiente e della Tutela del Territorio, 2003) would require the whole area facing the brownfield to undergo remediation (Fig. 7D). At the other sampling depths (Table 2A and 2B) Ni concentrations range respectively between 31 and 280 mg/kg in the 20–30 cm sampling layer, between 21 and 184 mg/kg in the 30–50 sampling layer, between 15 and 157 mg/kg in the 30–50 sampling layer, between 13 and 171 mg/kg in the 180–200 cm sampling layer, between 104 and 144 mg/kg in the 280–300 cm sampling layer, and between 121 and 137 mg/kg in the 380–400 cm sampling layer. Although Ni concentrations show a general decrease at increasing depths suggesting the anthropogenic nature of the source, sediments sampled at lower depths in the area that face the entire Bagnoli–Coroglio shoreline exceed the I.L. (30 mg/kg) established by Italian Law for sea sediments (Ministero dell'Ambiente e della Tutela del Territorio, 2003).

Metallic elements in the Bagnoli coastline sea sediments follow two different distribution patterns, as the two-factor model generated by the *R*-mode factor analysis clearly shows (Table 4).

The factor scores of association F1 accounts for 65.5% of total data variability and show that Zn, Cd, Sn, Pb, Hg, Cu and Fe have a strong spatial relationship with pelitic sea sediments facing the "Colmata a mare" structure and the Bagnoli–Coroglio coastline (Fig. 8A). Since the mobility of metals is generally strongly influenced by adsorption processes linked to the presence of fine-grain sized sediments, it is very likely that concentration values of these metals are significantly increased by the presence of pelite along the wide belt (E–W oriented) which connects the "Colmata a mare" structure to the open sea (Fig. 5).

In the northern and southern sector of Bagnoli–Coroglio coastline, the association F2, accounting for 34.5% of total data variability, apparently correlate the presence of Co, Fe and Ni to the sea sediment sandy texture (Fig. 8B). Actually this element association is more influenced by the anthropogenic origin of sands (from tar and oil residues, blast furnace slag and iron pellets) than by the grain size. The

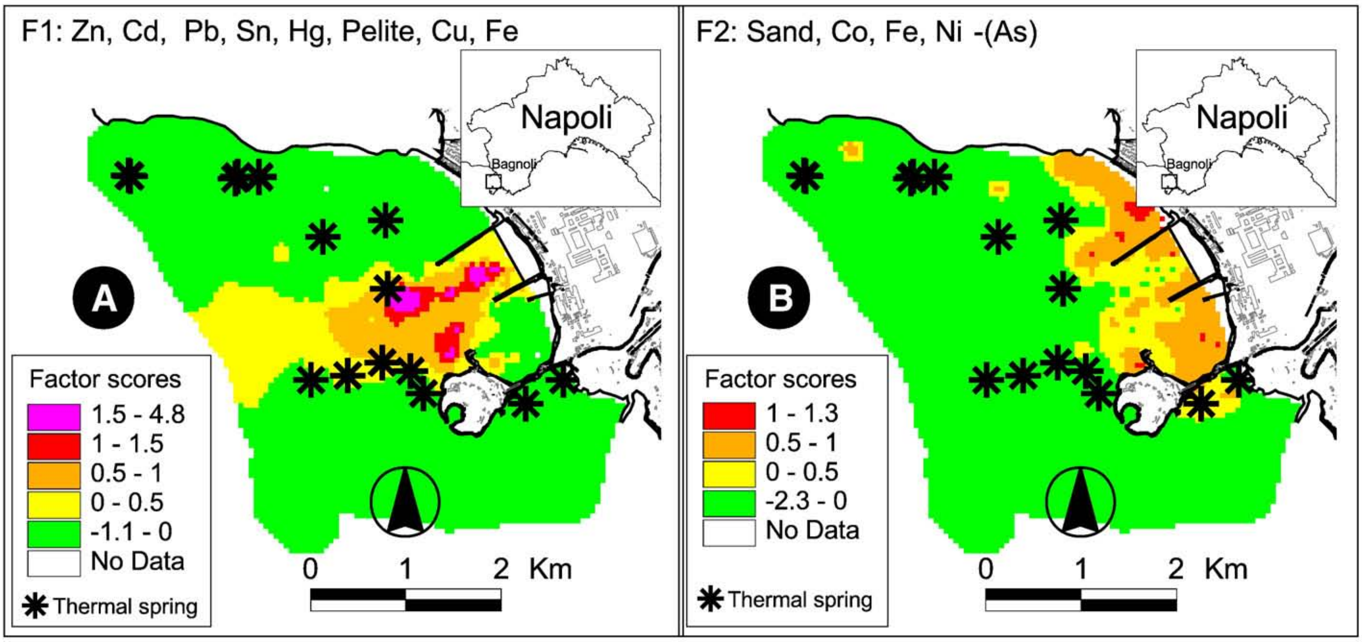

Fig. 8. Factor score association map for the 0–20 sampling depth.

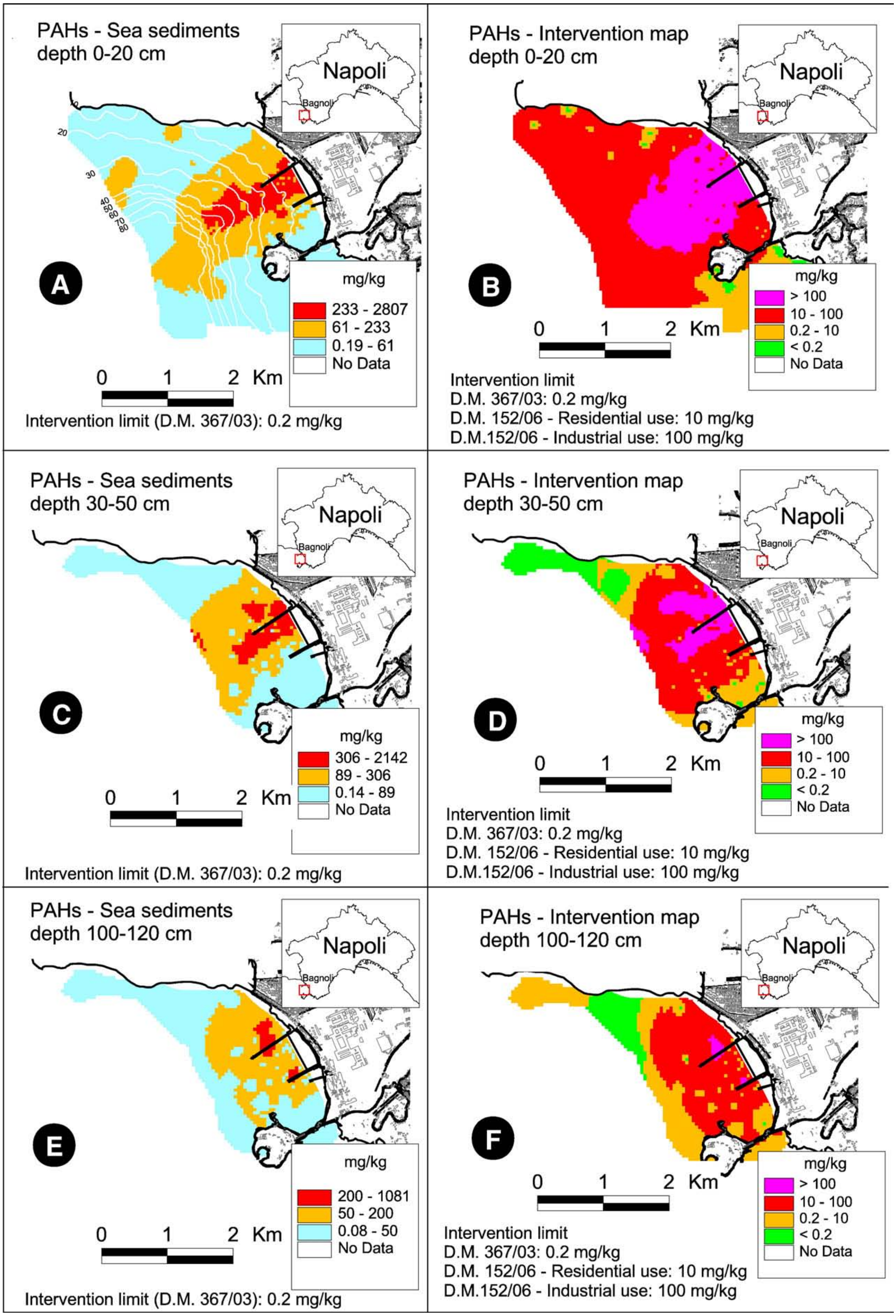

Fig. 9. Geochemical map of PAHs in sea sediments. Baseline (A, C, E) and intervention (B, D, F) maps, respectively, for the 0–20, 30–50 and 100–120 sampling depths.

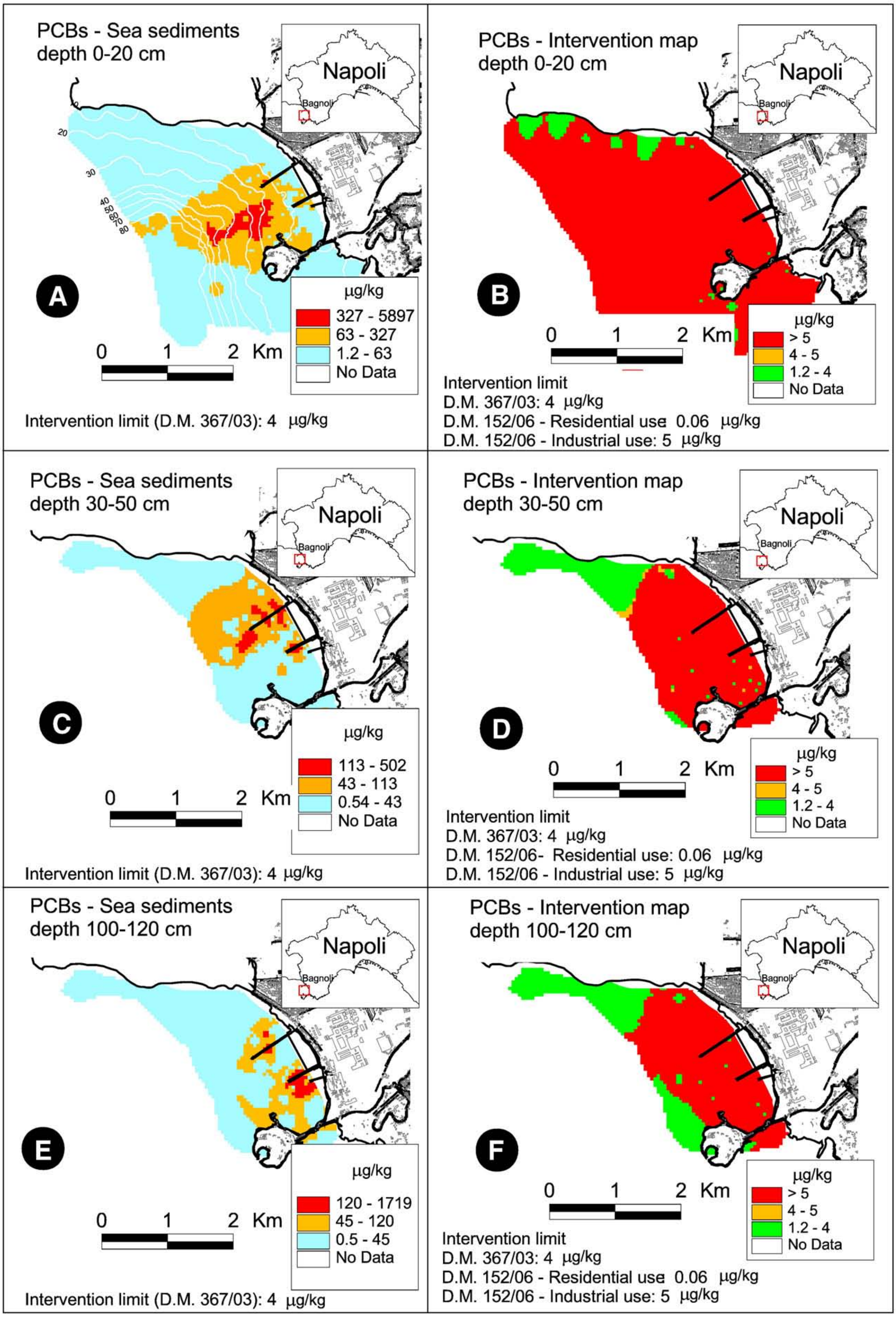

Fig. 10. Geochemical map of PCBs in sea sediments. Baseline (A, C, E) and intervention (B, D, F) maps, respectively, for the 0–20, 30–50 and 100–120 sampling depths.

negative loading of As (−0.52) in this element association also can be assumed to represent a marker that distinguishes the anthropogenic nature of these sediments.

PAHs (Fig. 9A, C, and E) and PCBs (Fig. 10A, C, and E) spatial distribution patterns at the different depths in the sea sediments of the study area are characterized by the highest concentration values in sea sediments facing both the "Colmata a mare" structure and the fossil fuel area located on the north-western sector of the brownfield site (Sector 2 of the sampling plan) (Fig. 4). PAHs and PCBs are manmade, hence the logical source for these compounds found in the sea sediments in our area, are the groundwaters from the brownfield site. In fact, the groundwater drainage axis goes across the fossil area and the oil by-product deposits (Fig. 3). In the topmost sampling layer (0–20 cm) (Fig. 9A) PAHs concentrations range between 0.19 and 2087 mg/kg. The PAHs intervention map for this layer (Fig. 9B) clearly show that the whole study area should undergo remediation according to I.L. (0.2 mg/kg) established by Italian Environmental Law for sea sediments (Ministero dell'Ambiente e della Tutela del Territorio, 2003). Reuse of dredged sediments in residential or industrial areas with respect to the I.L. established by Italian Environmental Law (Parlamento Italiano, 2006) is unfeasible, since the sea sediments nearby the coastline (Sector 2, Fig. 4) are characterized by PAHs concentrations >100 mg/kg (Fig. 9B). This is true also for the 20–30 cm, 30–50 cm (Fig. 9C), 100–120 cm (Fig. 9E) and 180–200 cm depth sampling layers. In all these layers in most of Sector 2 the I.L. (10 mg/kg) for a potential inland reuse in residential areas (Parlamento Italiano, 2006) of sea sediments are generally exceeded, as shown for both the 30–50 cm depth (Fig. 9D) and the 100–120 cm depth sampling layers (Fig. 9F), even though a general decrease of concentrations occurs (Table 2A and 2B). PAHs concentrations in the 280–300 cm and the 380–400 cm depth sampling layers range between 0.1 and 8.4 mg/kg and between 0.1 and 6.4 mg/kg, respectively (Table 2A and 2B), not exceeding the I.L. established by Italian Law for a potential inland reuse (residential or industrial) (Parlamento Italiano, 2006) (Table 3) of sea sediments.

PCBs concentrations range between 1.2 and 5897 µg/kg in the upper sampling layer (0–20 cm) (Fig. 10A). The intervention map for this layer (Fig. 10B) shows that the whole study area should undergo remediation regardless of the I.L. established for sea sediments (4 µg/kg) (Ministero dell'Ambiente e della Tutela del Territorio, 2003), for soil residential (0.06 µg/kg) or industrial land use (5 µg/kg) (Parlamento Italiano, 2006). In the 20–30 cm depth, 30–50 cm depth (Fig. 10C and D), 100–120 cm depth (Fig. 10E and F) and 180–200 cm depth sampling layers, sea sediments in Sector 2 (Fig. 4) are characterized by PCBs values higher than 5 µg/kg and must undergo remediation since they are not suitable even for an industrial inland reuse. PCBs concentrations in the 280–300 cm depth and in the 380–400 cm depth sampling layer range between 0 and 3.6 µg/kg and between 0.1 and 1 µg/kg, respectively. I.L. established for sea sediments by Italian Law (Ministero dell'Ambiente e della Tutela del Territorio, 2003) are not exceeded for these levels.

6. Conclusions

In the studied area, metallic elements, some potentially harmful (Al, As, Be, Cd, Co, Cr, Fe, Hg, Mn, Ni, Pb, Cu, Sn, V, and Zn), and organic (PAHs and PCBs) compounds in sea sediments are characterized by high concentration values that exceed both local and regional geochemical backgrounds and, in most cases, I.L. established by Italian Law (Ministero dell'Ambiente e della Tutela del Territorio, 2003) for sea sediments (Table 4).

Except for As (mostly introduced in the environment by submarine thermal springs), metallic elements and organic compounds (PAHs and PCBs) are brought to the sea by the discharge of the brownfield groundwaters across the "Colmata a mare" structure facing the brownfield site.

The physical transport processes of wind or human activities of metallic slag and production scoria particles from the inland towards the coastline plays an important role in sediment contamination related to Co, Fe and Ni distribution.

Sea sediments closer the Bagnoli–Coroglio coastline are generally more contaminated than the open sea sediments. They are characterized by concentration values of metals and organic compounds exceeding not only the I.L. established for sea sediment (Ministero dell'Ambiente e della Tutela del Territorio, 2003) but also the I.L. for soils (Parlamento Italiano, 2006), excluding a potential inland reuse in residential and industrial areas.

Although metallic elements have been proven to be enriched in groundwaters from hydrothermal sources occurring both in the brownfield site and in the sea shelf facing the brownfield site (De Vivo and Lima, 2008 and ref. therein), contamination of sea sediments by PAHs and PCBs has to be ascribed solely to the past industrial activities in the Bagnoli–Coroglio area.

Generic Hazard Quotient (HQ) (maximum measured pollutant concentration divided by a selected reference value representing the predicted "no effect concentration") has been calculated for As, Cd, Cr, Hg, Ni, Pb, PAHs and PCBs in seabed sediments (0–20 sampling depth) (Table 5) with the aim of performing an Ecological Risk Assessment (ERA). ERA evaluates the possibility and the grade of occurrence of adverse ecological effects due to environmental contamination with the purpose of protecting populations and communities belonging to different species (Campbell and Bartell, 1998; Khairy et al., 2009).

I.L. established for sea sediments by Italian law (Ministero dell'Ambiente e della Tutela del Territorio, 2003) were assumed as predicted "no effect concentrations" (PNEC) (Maggi et al., 2007).

Result obtained showed that the Bagnoli brownfield site coastal sea sediments are always characterized by HQ>1 up to the remarkable values of 14,737 and 1666 respectively for PAHs and PCBs.

Since the higher the HQ, the more frequent adverse ecological effects are expected, a remediation project should be planned for the PAHs and PCBs found in the sea sediments facing the coastline, since the area in the summer time is crowded by family and children. This area should be closed to any bathing activity until a recovery of the entire area is completed.

Removal of contaminated sediments from the seabed down to a depth of 2 m, only from the area closer to the Bagnoli–Coroglio coastline (Sector 2, Fig. 4) could be considered as a solution. This operation would require dredging and moving a volume of 1,660,000 m^3 of material. After removal, the contaminated sediments should either undergo remediation or be stored indefinitely in a special waste depository. The option of inland reuse of dredged sediments is not be feasible because of the very high PAHs and PCBs contamination levels.

Before any action is taken to remediate the sea sediments it is necessary to check the permeability/impermeability conditions of the "Colmata a mare" structure and to check the release of organic

Table 5
Ecological hazard quotient (HQ) calculated for seabed sediments (0–20 sampling depth) assuming as reference value for the "no effect concentration" (PNEC) Intervention Limits (I.L.) established by Italian Law D.M. 367/03 (Ministero dell'Ambiente e della Tutela del Territorio, 2003).

Element/compound	PNEC I.L. sea sedimets	Max (Depth 0–20 cm)	HQ
As (mg/kg)	12	55	4.5
Cd (mg/kg)	0.3	7.4	25
Cr (mg/kg)	50	103	2.1
Hg (mg/kg)	0.3	3.5	11.5
Ni (mg/kg)	30	181	6
Pb (mg/kg)	30	1773	59
PAH (mg/kg)	0.2	2947	14,737
PCB's (µg/kg)	4	6665	1666

pollutants (PAHs and PCBs) from the fossil and oil by-product area of the brownfield site to the underlying groundwaters.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.gexplo.2010.01.007.

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