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ENVIRONMENTAL ASSESSMENT OF POTENTIALLY TOXIC TRACE ELEMENTS IN SEDIMENTS OF FILIPPOS B PORT, NORTHERN AEGEAN SEA – A COMPARISON WITH OTHER NATIONAL AND INTERNATIONAL COASTAL REGIONS

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Abstract: Nine sediment samples from Filippos B port, Kavala, northern Greece were collected, sieved under 200 μm and analyzed for their content in 14 potentially toxic trace elements (Ag, As, Ba, Cd, Co, Cr, Cu, Hg, Mn, Ni, Pb, U, V and Zn). The results indicate that the majority of the elements are found with concentrations similar to other national and international coastal regions. However, Cd seems to be highly enriched in the sediments of the present study. The samples with the highest concentrations of Cd, as well as for the rest of the elements, are found in front of the local, anthropogenic activities. According to their distribution, the elements of the present study can be divided into two groups; group A includes the elements Ag, As, Cd, Hg, Pb and U, group B the elements Ba, Co, Cr, Cu, Mn, Ni, V and Zn. The former are influenced mainly by the activities of a fertiliser plant, while the latter by all the local anthropogenic activities.

Keywords: sea sediment; trace elements; geochemistry; environment; Kavala; Greece.

1. Introduction

Natural and anthropogenic activities are the reasons why many potentially toxic elements, but especially trace elements, are found in aquatic environments. These contaminants may accumulate in bottom sediment and, thus trigger toxic effects on organisms living in the adjacent area (e.g., Michel and Zengel, 1998; Valette-Silver et al., 1999). In fact, the accumulation of potentially toxic and persistent substances in the coastal environment continuously increases due to anthropogenic activities as the estuaries and their nearby coasts are the focus of many economic activities (e.g., Ramessur, 2004; Grigoriadou et al., 2008a,b). Consequently, monitoring the geochemical status of sea sediments is an important and, at the same time, complex task. This was the objective of the present research, as well. En effort was made to assess the environmental status of the sediments of Filippos B port by comparing the concentrations of the elements determined with other coastal regions, national and international.

2. Study area

The study area is located in northern Greece and includes the coastal part of the industrial zone of the city of Kavala (Fig. 1). The climate of the area has general Mediterranean characteristics with mild winters and warm summers. Land uses in the area can be divided into four categories: agricultural, uncultivated, industrial and residential (Fig. 1). The main industrial activities are the Phosphoric Fertilizer Industry (PFI) and the Kavala Oil land facilities. The former industry produces phosphoric fertilizers, pesticides and other similar products and its main by-product is phosphogypsum, while in the latter oil desulphurization takes place. Some contaminants associated with the production of phosphoric fertilizers and similar products (i.e., pesticides), as well as phosphogypsum, are Ba, Cd, Cu, P, Pb, Th, U, and Y (Rutherford et al., 1996; Komnitsas et al., 1999; Chen et al., 2001; Kabata-Pendias and Pendias, 2001; Carbonell-Barrachina et al., 2002; Villa et al., 2009); contaminants associated with the oil industry are Co, S, Th, and V (Kabata-Pendias and Pendias, 2001). Other activities include some small enterprises that exploit and market local marbles and the Xifias Fishery enterprise which terminated its activities during the undertaking of the present research. Furthermore, near the Kavala Oil land facilities and the Xifias Fishery there are several uncontrolled landfill sites.

The main rock types in the area are (Fig. 2): (a) gneisses, schists and amphibolites, (b) marbles and limestones, (c) granitic and granodioritic rocks and (d) sedimentary deposits (Christofides et al., 1998; 2001; Kilias et al., 1999). Additionally, in the adjacent area, several Pyrite-Blende-Galena (PBG), Mn, and Fe mineralizations exist (Filippidis et al., 1996; Vavelidis et al., 1996;1997). The mineral characteristics of the surface sea sediments in the study area are attributed to the delta of Nestos River and to local sources. The sand fraction mainly consists of quartz, feldspars, micas, amphiboles, biogenic carbonate and pyroxenes, while the respective composition of the clay fraction is illite, kaolinite, smectite and chlorite. The predominant source of clay minerals is Nestos River (Conispoliatis and Lykousis, 1986).

3. Materials and methods

Nine sampling sites in Filippos B port and adjacent areas to the east and west were selected, at variable distances from the coastline (Fig. 1). Surface sediment samples were collected, using a sediment grab sampler, in front of the major industrial activities of the area (S100, S102, S104, S105, S106, S107, S108), but also away from them (S101, S103) (Fig. 1). Samples S100, S106, S107 and S108 were collected in front of the PFI facilities and the phosphogypsum dumps, while samples S102, S104 and S105 were taken in front of the Kavala Oil land facilities and the Xifias Fishery. Sample S103, although it was collected away from the industrial activities of the study area, is located at a point were a water canal connects the aquacul-

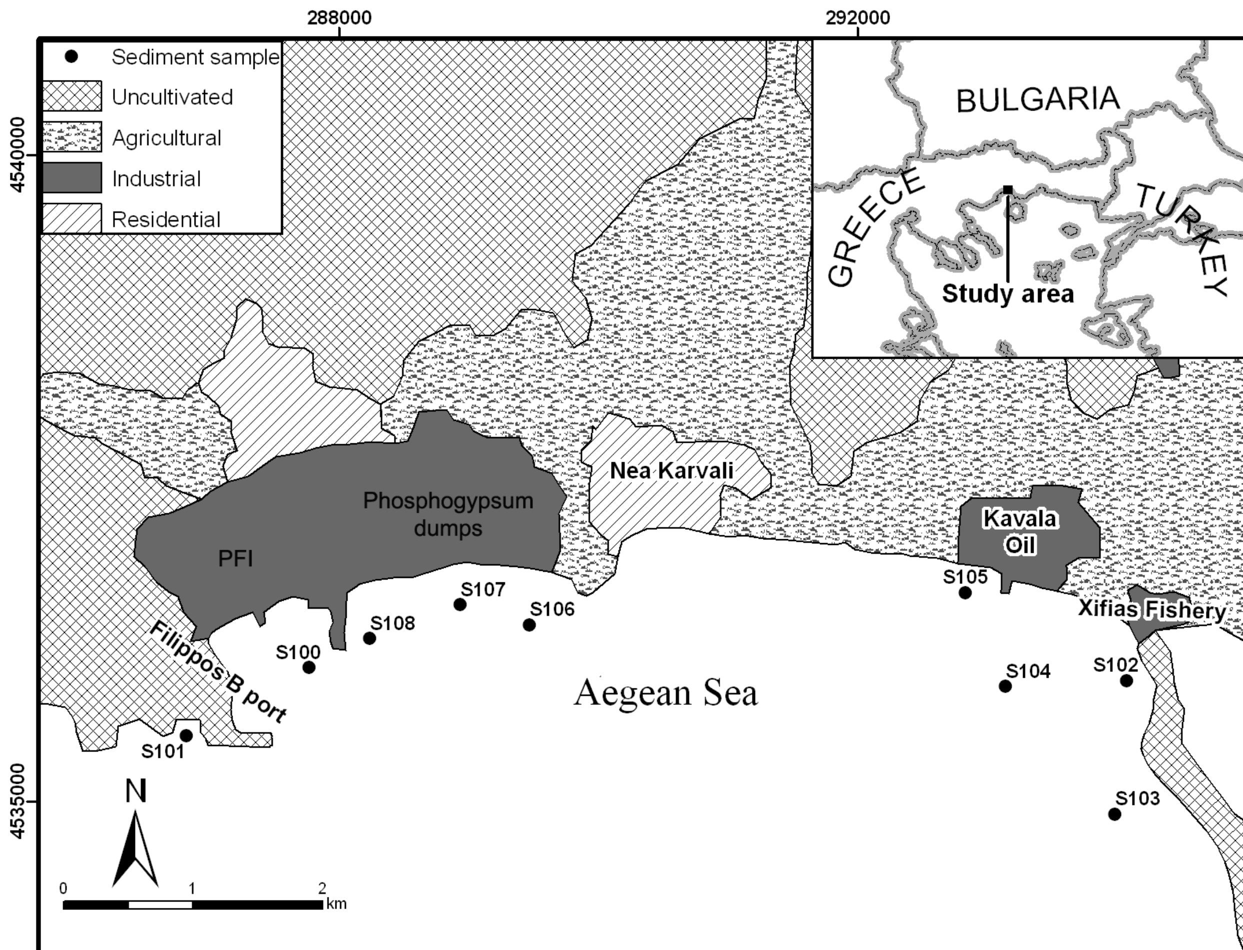

Fig. 1. Map showing the Filippos B port, the sampling locations, the industrial activities and the land use/cover types of the study area (modified after Papastergios 2008).

tures of the Xifias Fishery with the Aegean Sea. Although the present research would benefit from the collection of more samples, unfortunately this was not possible due to various limitations (i.e., accessibility of the area and others). Still, valuable information regarding the status of the sediments in Filippos B port compared to several national and international coastal regions is presented.

All samples were dried in an oven at 40°C, sieved under the 200 μm fraction and the elements determined were extracted using analytical grade, concentrated (65%) HNO_3 (Fernandez-Turiel et al., 2001; Papastergios et al., 2009a; 2010a). The extraction is conducted under extreme analytical conditions, in this way, while at the same time, the compatibility of the leachate with the input solution in Inductively Coupled Plasma – Mass Spectrometry is maintained (direct determination after dilution). Furthermore, the polyatomic interferences produced on analytes by other extraction agents commonly used [e.g., the Cl of aqua regia (3:1, HCl:HNO_3) interferes with elements such as As and V], are avoided.

The concentrations of 14 trace elements (Ag, As, Ba, Cd, Co, Cr, Cu, Hg, Mn, Ni, Pb, U, V and Zn) were determined in all samples by ICP–MS. A Perkin Elmer Sciex Elan 6000 with a Perkin Elmer AS-91 automatic sampler was used. The analyses were performed at the Scientific Technical Services of the University of Barcelona (SCT–UB), Spain. Details on ICP–MS analysis can be found in Fernandez-Turiel et al. (2000). The quality of the results was checked by applying the same procedure to a reference material (CANMET NRCC-MESS-2) as well as to a blank and four replicates of a randomly selected sample. The methodology used, achieved analyses that were very satisfactory, for the work objectives. Many elements exhibited analytical reproducibility values (percent relative deviation-RSD, n=3) lower than 3%. In regard to the extraction procedure's reproducibility, the majority of the elements exhibited RSD values lower than 5% (Papastergios, 2008).

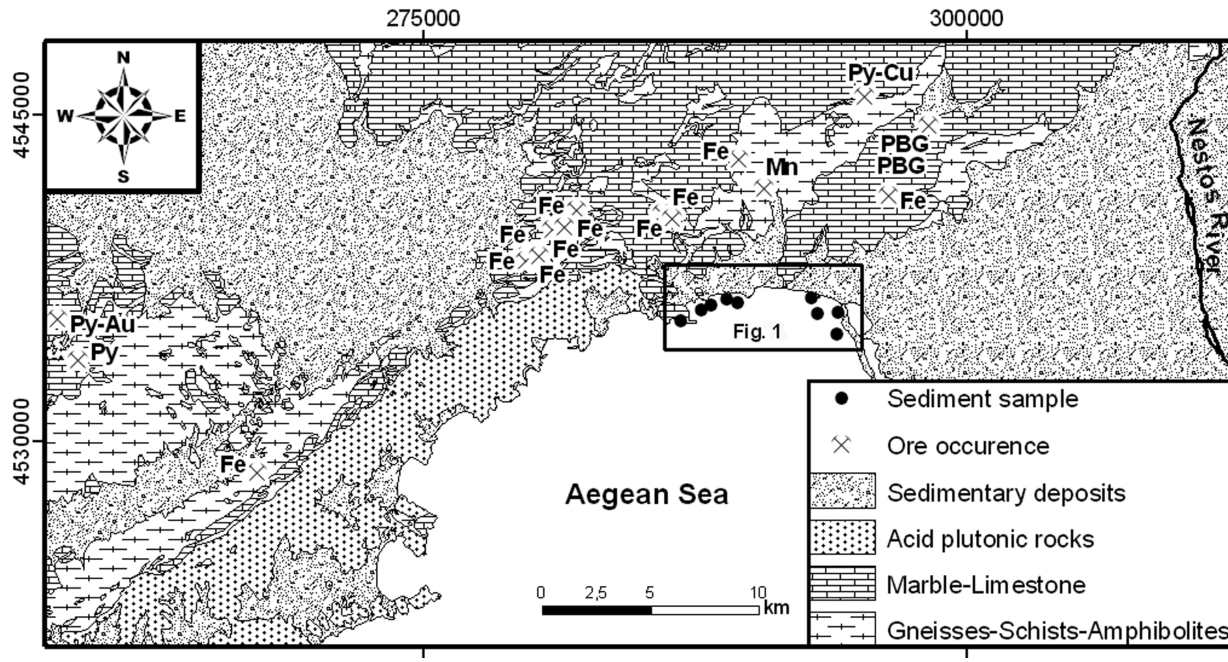

Fig. 2. Simplified geological map of the study area (modified after Papastergios 2008).

4. Results and discussion

Summary statistics regarding the elements determined in the present work are given in Table 1. For the calculation of the descriptive statistics, values that were below the detection limit, for each element, were substituted by half its detection limit. Elements, with average concentrations above 100 mg kg^{-1} are Mn and Zn. Average concentrations between 1 and 100 mg kg^{-1} are noted for Pb, Ba, Cu, Cr, Ni, V, As, Cd, Co and U, while Ag and Hg have average concentrations below 1 mg kg^{-1}.

The concentrations of the elements determined in the present study were compared with the concentrations cited in the literature regarding national and international marine areas (Tab. 2). The average Ag concentration is 0.3 mg kg^{-1}, which is lower than the values cited for Thermaikos gulf and Sydney, and approximately the same with San

Francisco. The two largest values (S100: 0.7 mg kg^{-1} and S108: 0.9 mg kg^{-1}) are noted in front of the PFI facilities. Arsenic's mean concentration is above 15.0 mg kg^{-1}. This value approximates the values given for ports such as Barcelona, Sydney and Thermaikos. The distribution of this element is discussed in detail elsewhere (Papastergios et al. 2010a). The mean concentration of Cd is 7.1 mg kg^{-1}, a value much higher than those reported for both, national and international ports. However, Zabetoglou et al. (2002) have reported similar values (6.2 and 6.3 mg kg^{-1}) of Cd for a specific location (in front of the White Tower) of Thermaikos Bay. The authors attributed the elevated concentrations to anthropogenic sources. The samples with the highest concentrations are found in front of the PFI facilities (S100: 7.8 mg kg^{-1}, S107: 12.3 mg kg^{-1} and S108: 39.2 mg kg^{-1}). Mercury has an average concentration of 0.1 mg kg^{-1}. This value is the same with the one reported for Lesvos' coastal area, lower than the ones reported for San Francisco, Turkey and Italy, and considerably lower than the average values cited for Sydney and Barcelona. However, samples S100 (0.3 mg kg^{-1}), S107 (0.2 mg kg^{-1}) and S108 (0.4 mg kg^{-1}) have similar concentrations with the former, international coastal areas. The average concentration of Pb is above 88.0 mg kg^{-1}. This value is similar or larger to some national or international coastal regions but with Sydney and Barcelona, however, having much larger average concentrations. The largest Pb concentration is noted in front of the phosphogypsum dumps (S107: 274.5 mg kg^{-1}), suggesting that Pb and P are interacting and, possibly forming pyromorphite [$Pb_5(PO_4)_3Cl$], which acts as an important buffer mechanism controlling the migration and fixation of Pb in water, soils, sediments and wastes (Ryan et al., 2001; Manecki et al., 2006). Finally, U has an average concentration of 1.2 mg kg^{-1}. The two largest values (S100: 4.9 mg kg^{-1} and S108: 2.5 mg kg^{-1}) are found in front of the PFI facilities. Unfortunately no values were available for comparison with the other selected coastal sites. All the former elements have been associated with the production of (phosphate) fertilizers, pesticides and other similar products (i.e., Arocena et al., 1995; Martin et al., 1999; Kabata-Pendias and Pendias, 2001; Bolivar et al., 2002; Villa et al. 2009) and because they have their largest concentrations in front of the PFI facilities (samples S100, S107 and S108), it is rather obvious that its activities must play an important role in the distribution that these elements have in Filippos B port.

On the contrary, the rest of the elements have such distributions that they must be influenced by all the industrial activities of the study area, as they have elevated concentrations in front of the PFI and the Kavala Oil facilities and the channel that connects the Xifias Fishery aquacultures with the open sea, as well. The mean concentration of Ba is 45.6 mg kg^{-1}, which is considerably larger than the value reported for Thermaikos gulf. The samples with the largest concentrations are S108 (121.5 mg kg^{-1}), S104 (63.6 mg kg^{-1}) and S103 (46.7 mg kg^{-1}) located in front of the PFI, the Kavala Oil and the Xifial Fishery, respectively. Cobalt's mean concentration approximates 5.0 mg kg^{-1}. This value is much lower than those given for national and international coastal regions. Nonetheless, the samples with the highest concentrations are found in front of the Kavala Oil (S104: 8.6 mg kg^{-1}), the PFI (S108: 8.2 mg kg^{-1}) and the Xifias Fishery (S103: 6.8 mg kg^{-1}). The average concentration of Cr is almost 40 mg kg^{-1}. This value is similar to some

Table 1. Summary statistics for the elemental concentrations of the present study (n=9).

Element (mg kg^{-1})	median	mean	minimum	maximum	std deviation
Ag	0.1	0.3	0.1	0.9	0.3
As	7.4	15.3	2.2	44.8	15.2
Ba	37.9	45.6	11.4	121.5	32.2
Cd	0.4	7.1	0.1	39.2	12.1
Co	4.5	4.8	1.1	8.6	2.7
Cr	33.6	38.0	5.2	76.3	23.9
Cu	18.9	40.9	2.3	119.1	42.1
Hg	0.1	0.1	0.03	0.4	0.1
Mn	130.1	143.9	45.6	264.2	66.1
Ni	22.3	18.7	3.5	40.9	11.6
Pb	49.0	88.3	8.5	274.5	79.0
U	0.5	1.2	0.3	4.9	1.5
V	18.8	17.5	4.0	36.5	10.4
Zn	59.8	133.9	11.8	538.2	157.2

national and international areas. Still, there are ports reported with higher Cr concentrations. Once again, sample S104 (76.3 mg kg^{-1}) in front of the Kavala Oil, samples S100 (61.3 mg kg^{-1}) and S108 (59.9 mg kg^{-1}), in front of the PFI and sample S103 (33.6 mg kg^{-1}) near Xifias Fishery, are the samples with the highest concentrations. Copper has a mean value of nearly 41.0 mg kg^{-1} and has, relatively, similar values with other national ports. Sydney and Barcelona, on the other hand have been reported with much higher concentrations of Cu. Sample S108 (119.1 mg kg^{-1}) has the highest concentration and is followed by samples S100 (103.2 mg kg^{-1}) and S104 (67.9 mg kg^{-1}). The mean concentration of Mn is above 140.0 mg kg^{-1}, but this concentration is lower for almost all other coastal regions used for comparison in the present study, Sydney being the only exception. Samples S104 (264.2 mg kg^{-1}), S103 (229.2 mg kg^{-1}) and S100 (172.8 mg kg^{-1}) have the largest values.

Nickel has an average concentration that is below 20.0 mg kg^{-1}. This value is similar to San Francisco, Sydney and Barcelona but much smaller than those given for Italy or Thermaikos. Again, the highest concentrations are found in front of the local industrial activities (S104, S100 and S103). The mean concentration of V is almost 18.0 mg kg^{-1}. The distribution of this element is very similar to Ni. Finally, Zn has an average concentration above 130 mg kg^{-1}. This concentration is similar to most national coastal areas but notably smaller than the average concentrations of Zn in Barcelona and Sydney. However, the samples with the highest concentrations, especially near the PFI (S108: 538.2 mg kg^{-1}), are found in front of the anthropogenic activities. All the former elements have been associated with the type of activities occurring in the area (Rutherford et al., 1996; Kabata-Pendias and Pendias 2001). Furthermore, the distribution that the elements of the present study have in the sea sediments of Filippos B port is very similar to their distribution in the sea water of the study area (Georgakopoulos et al., 2002).

Research conducted recently in the area of Kavala

Table 2. Average concentrations of the Filippos B port and several national and international coastal areas.

Element (mg kg^{-1})	Filippos B port	Thermaikos gulf (1)	Thermaikos gulf (2)	Evros prodelta (3)	Lesvos coastal area (4)
Ag	0.3	3.1			
As	15.3	19.0			
Ba	45.6	0.4			
Cd	7.1	1.8		0.4	0.1
Co	4.8	33.0			
Cr	38.0	294.0	47.0	76.0	137.8
Cu	40.9	72.0	80.0	39.0	18.3
Hg	0.1				0.1
Mn	143.9	770.0		561.3	294.5
Ni	18.7	98.0		45.3	
Pb	88.3	87.0	77.0	42.7	30.9
V	17.5	160.0			
Zn	133.9	239.0	184.0	104.3	63.8

Element (mg kg^{-1})	San Francisco (5)	Sydney (6)	Barcelona (7)	Turkey (8)	Italy (9)
Ag	0.1	1.8			
As		21.0	21.5		
Ba					
Cd	0.2	2.8	1.5	0.2	0.4
Co					55.3
Cr	21.1	81.0	90.6	216.0	1194.0
Cu		200.0	234.5		
Hg	0.2	1.4	1.8	0.3	0.6
Mn	480.6	120.0			
Ni	39.3	20.0	25.6		1325.0
Pb	21.8	360.0	206.0	58.5	44.5
V					
Zn	65.2	1000.0	515.6		

(1) Violintzis et al 2009, (2) Christoforidis et al. 2009, (3) Kanellopoulos et al. 2006, (4) Aloupi et al. 2007, (5) Lu et al. 2005, (6) McReady et al. 2006, (7) Casado-Martinez et al. 2006, (8) Kucuksezgin et al. 2006, (9) Lafabrie et al. 2007.

has revealed the influence of the local anthropogenic activities, for organic contaminants, on soils, sediments and sea water (Grigoriadou et al., 2008a, b), for heavy metals in street dust and roadside soil along the major national road in Kavala's region (Christoforidis and Stamatis, 2009) and As in sediments of the Filippos B port (Papastergios et al., 2010a). Although there are several ore occurrences (i.e., PBG, Mn, Fe) in the wider study area, which could contribute, as metal sources, to the observed high concentrations, the fact that Nestos River (and, to a lesser extent, local sources) is the predominant source of minerals seems to weaken this alternative, especially since the elemental content of Nestos River sediments and of the surrounding rocks has been found to be within normal ranges (Papastergios et al., 2009a,b; 2010b).

5. Conclusions

All the elements of the present study have their lowest concentrations in either sample S101 or sample S102 and their highest concentrations in either sample S108 or S104. The latter samples are situated in front of the PFI and the Kavala Oil facilities, respectively, indicating that the anthropogenic activities of the study area have modified the geogenic distribution of these elements. However, the contribution of the anthropogenic activities seems to be restricted only in small areas, near their facilities. The concentrations of the elements determined in the present study are similar to many national and international coastal areas but, at the same time, differ as well, reflecting the differences in the local geology and industrial activities. Additionally, some of the differences noted between the present study and those used for comparison purposes could be due to differences in the methodologies applied. Nonetheless, Cd concentrations are significantly larger than compared to any other coastal region used for comparison in the present work, especially for the samples that are found in front of the PFI facilities. A further monitoring of the sediments in the area with additional parameters such as mineralogy of the sediments, pH, redox potential, bioavailability and toxicity of the contaminants etc. is suggested in order to control the evolution of the elemental concentrations in the sediments of this area of Aegean Sea.

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