Δελτίο της Ελληνικής Γεωλογικής Εταιρίας, τομ. XLVII , 2013
Πρακτικά 13[ου] Διεθνούς Συνεδρίου, Χανιά, Σεπτ. 2013

Bulletin of the Geological Society of Greece, vol. XLVII 2013
Proceedings of the 13[th] International Congress, Chania, Sept. 2013

ASSESSMENT OF HEAVY METALS CONTAMINATION IN THE COASTAL SEDIMENTS OF THE BROADER AREA OF CHIOS HARBOR (AEGEAN SEA)

Tsoutsia A.[1], Kapsimalis V.[2], Poulos S.[1], Paraskevopoyloy V.[3] and Dassenakis E.[3]

[1] University of Athens, Faculty of Geology and Geoenvironment, Department of Geography & Climatology, Panepistimioupoli, Zografou 15784, Athens, Greece

[2] Hellenic Centre for Marine Research (HCMR), Institute of Oceanography, 46.7[th] km Athens - Sounio Ave., P.O Box 712, P.C. 19013, Anavyssos, Greece

[3] University of Athens, Faculty of Chemistry, Laboratory of Environmental Chemistry, Panepistimioupolis-Zografou, 15784, Greece.

Abstract

The present study evaluates the pollution level of the surficial sediments of the coastal area around the city of Chios (east Aegean Sea), with particular emphasis on the Port of Chios, fishing shelters, the waste water treatment plant and the electric power plant. For this reason, the metals/trace metals (Pb, Fe, Zn, Cu, Cr, Mn, Al, Ni) along with total organic carbon (TOC) and $CaCO_3$ were analyzed in 23 sediment samples. The results showed that the highest concentrations are observed in the main Port, offshore the power plant station and in a small fishing shelter, without being considered as dangerous for the marine organisms and human health, according to metal pollution indices and Sediment Quality Guidelines.
Key words: *trace metals, seabed sediments' pollution, Chios Island, North Aegean.*

Περίληψη

Η παρούσα μελέτη διερευνά τη ρύπανση των επιφανειακών ιζημάτων στο παράκτιο μέτωπο της πόλης της Χίου (Ανατολικό Αιγαίο, Ελλάδα), συμπεριλαμβανομένων του κεντρικού λιμένα της Χίου, μικρότερων αλιευτικών καταφυγίων, της εγκατάστασης παραγωγή ηλεκτρικής ενέργειας και της εξόδου του αγωγού του βιολογικού καθαρισμού. Είκοσι τρία δείγματα ιζημάτων αναλύθηκαν ως προς τη συγκέντρωσή τους σε βαρέα μέταλλα (Pb, Fe, Zn, Cu, Cr, Mn, Al, Ni) καθώς και σε οργανικό άνθρακα (TOC) και ανθρακικό ασβέστιο ($CaCO_3$). Σύμφωνα με τα αποτελέσματα των αναλύσεων οι περιοχές που παρουσίασαν τις σχετικά μεγαλύτερες τιμές συγκεντρώσεων, είναι ο κεντρικός λιμένας, μια θέση πλησίον του εργοστασίου παραγωγής ενέργειας (ΔΕΗ) και ένα αλιευτικό καταφύγιο. Οι τιμές αυτές δεν θεωρούνται επικίνδυνες για τους θαλάσσιους οργανισμούς και κατ' επέκταση για την ανθρώπινη υγεία, σύμφωνα με γεω-περιβαλλοντικούς δείκτες ρύπανσης των ιζημάτων.
Λέξεις κλειδιά: *Βαρέα μέταλλα, ρύπανση επιφανειακών ιζημάτων πυθμένα, Χίος, Β. Αιγαίο.*

1. Introduction

Marine pollution may be related to natural factors (e.g. increased concentration of various chemical and radioactive elements, ash) and to a variety of human activities (e.g. industry, marine transport, oil spills, untreated sewage, agriculture). The main categories of marine pollution are (Clark, 1993): (1) organic contaminants, which cause the reduction of dissolved oxygen; (2) increased nutrient content; (3) microbial pollution; (4) oil pollution; (5) pollution by halogenated hydrocarbons; (6) solid waste (garbage); (7) heavy metals; (8) radiation; and (9) thermal pollution.

The effects of heavy metal pollution to aquatic life can be estimated by: (a) the application of a variety of geostatistical indices, such as the Geoaccumulation Index (Igeo) (Muller, 1969; Főrstner et al., 1990), the Combined Contamination Index (W) (Widianarko et al., 2000), the Modified Contamination Degree (mCd) (Abrahim and Parker, 2008); and (b) the comparison with effect-based numerical Sediment Quality Guidelines (SQG), which provide tolerable concentrations of sediment-bound contaminants in order to protect the organisms living in or near sediments. The most common set of SQGs is the ERL/ERM approach (Long et al., 1995) that provides: (i) a lower threshold value (ERL: Effect Range Low) below which adverse effects on sensitive life stages and/or species occurred only infrequently and (ii) an upper threshold value (ERM: Effect Range Median) above which adverse effects were frequently observed.

The purpose of the present investigation is to assess the degree of heavy metal contamination in surface sediments of the coastal zone of the city-harbour of Chios Island. Particular attention is given to locations with intense human activities, such as the Commercial harbour of Chios, fishing shelters and coastal areas in the vicinity of industrial plants, in order to identify the potential sources of pollution. In addition, the probable adverse effects of sediments to aquatic organisms were assessed through the application of widely used sediment pollution indices and quality guidelines.

2. The Study Area

Chios Island lies in the Eastern Aegean Sea (Fig. 1) and geologically belongs to the Pelagonian isotopic zone. Lithologically, the NE - SW Chios consists of limestones and dolomites (middle Triasic or Triassic to Lower Jurassic in age) its NW part of schists, grauwackes, conglomerates with lenses of limestone (Paleozoic), while its ESE part comprises of marl, clay rocks, limestones and sandstones (lower Miocene - lower Pliocene) (Bornovas and Rondogianni -Tsiambaou, 1989). The wave climate (Tsoutsia, 2012) of the narrow passage between the east coast of Chios and the Asia Minor (Turkey) is characterized by a moderate wave regime with the north waves to be the dominant (21.9%, annually) ones. The most frequently occurring north waves (4.4%) have significant wave height of only 0.2 m and period 0.2 sec. On the other hand, the highest observed waves are the easterly ones having significant heights <5 m and period <8 sec, but occurring very rarely (<0.01%). Parthenis and Kokkalas are the main rivers debouching in the study area; the former covers an area of 23.7 km^2, reaching the 800 m in altitude, while the latter drains an area of 36.3 km^2, being in altitude up to 707 m (Paidas, 2011). In addition, along the east coast of the Chios Island few ephemeral streams debouch also. The main human activities with significant potential to pollute coastal environment are the harbour and the Electric Power Plant (Fig. 2).

3. Materials and Methods

The present investigation is based on the granulometric and geochemical analyses of 23 samples, which were collected in October of 2011 with the use of a van Veen grab. The grain size treatment included: (a) wet separation of the bulk sample through a 63 μm sieve; (b) dry sieving of the coarse-grained fraction (particle size >63 μm); and (c) laser diffraction analysis of the fine-grained fraction (particle size <63 μm) using a Malvern Mastersizer 2000.

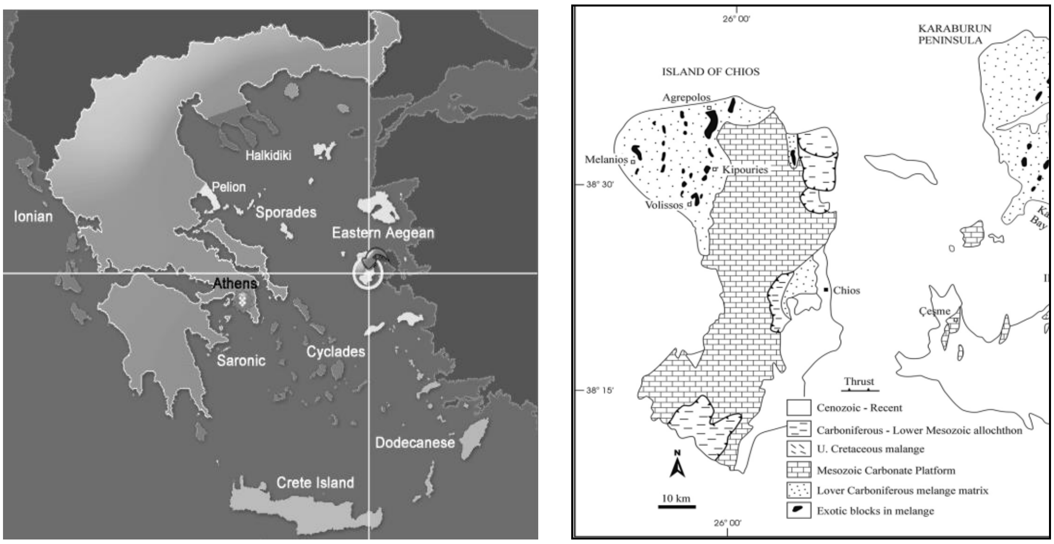

Figure 1 - Location of Chios Island (left); Simplified lithological map of Chios Island (right) (abstracted from Robertson & Ustaömer, 2009).

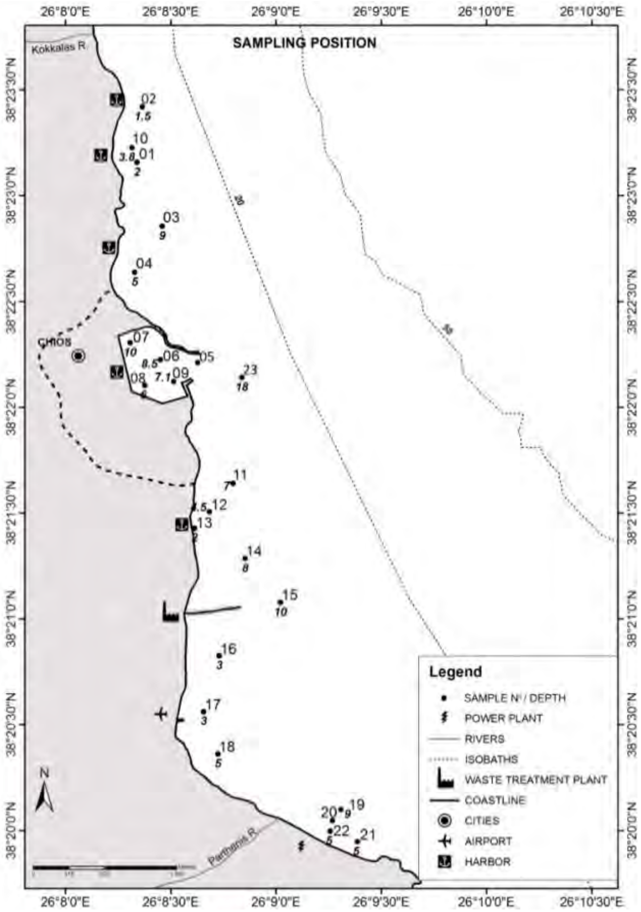

Figure 2 – The coastal area under investigation together with sampling locations and major human activities.

The concentrations of major (Al and Fe) and trace (Cr, Cu, Mn, Ni, Pb and Zn) elements (in the dry weight) were determined both in the bulk sample and in the fine-grained fraction (when its content was more than 10% of the total sample) using a Graphite Furnace Atomic Absorption

(GF-AAS) and/or Flame Atomic Absorption (FAAS). The organic Carbon content was estimated in accordance to Walkey and Black (1934) method, while the carbonate concentration was calculated after treatment of each sample with solution of HCL 6M of each sample and with the use of the following equation:

Equation 1

$$\% \ Carbonates = \frac{m_1 - m_2}{m_s}$$

where: m_1: mass before treatment; m_2 mass after 1.5 minute treatment and m_s: total sediment mass.

The indices that were used to describe the environmental status were: Igeo (Geoaccummulation index, after Muller (1969) and Förstner et al. (1990)); mCd (modified Contamination degree, after Abrahim and Parker (2008)); and W (combined contamination index, after Abrahim and Parker (2008)).

Equation 2

$$Igeo = \log_2\left(\frac{c_i}{1.5B_i}\right)$$

where: c_i: the concentration of element in the sediment and B_i: the concentration of element in the reference sediment.

Equation 3

$$mCd = \frac{\sum_{i=1}^{n} cf_i}{n}$$

where: n: number of analyzed samples (Σ: algebraic sum), cf : the contamination factor.

Equation 4

$$W = \log \prod_i cf_i$$

where: cf : values of the ratios of concentrations of heavy metals in sediment samples with the corresponding present in natural sediment reference (where Π: the algebraic product).

The incidence of adverse biological effects was investigated utilizing the ERM (Effects Range Low), ERL (Effects Range Median), (Long et al., 1995), and TEL (Threshold Effects Level), PEL (Probable Effects Level) (Macdonald et al., 1996) guidelines values that are associated with ranges of trace metal chemical concentrations; the latter are provided by Macdonald et al. (1996; table 3 p. 266) and by Long et al. (1995; table 3 p. 92), respectively.

4. Results and Discussion

The sand fraction varies from 17.94 % (sample 13) to 99.98 % (sample 17), with samples 1 (99.59%), 12 (98.31%), 17 (99.98 %) and 21 (99.05%) exceeding the 98% (Table 1); samples 4 (21.20%), 9 (21.45 %) and 13 (17.94%) consists of <25% of sand, whereas the average value of sand is equal to 52.74%. Silt varies from 0.00 % (sample 22) to 62.13 % (sample 4), having an average value of 31.85 %. Clay, having smaller contributions, varies from 0% (samples: 1, 12, 17, 20, 21 & 22) up to 20.18% (sample 7), with an average value of 9.58%. According to Folk (1974), most of the samples are characterized as slightly sandy mud (s)M.

In terms of heavy metals, the average value for zinc (an element of both anthropogenic and natural origin) is 55 mg/kg, while the maximum and the minimum values were 167 mg/kg (sample 13) and 13 mg/kg (sample 1), respectively (see Table 2). The average value of copper is 36 mg/kg, presenting the maximum value in sample 13 (119 mg/kg) and the minimum in sample 1 (12 mg/kg); the increased value of Cu in sample 13, taken for a small shelter with water depth <2 m, may be attributed to the use of Cu-based antifouling agents in boats and fishing nets. The lowest lead value exhibits in sample 1 (4 mg/kg) and the highest in the sample 18 (73 mg/kg), with average value being equal to 26.1 mg/kg. For nickel, the highest value (95 mg/kg) is found in sample 19, which presents also the maximum value for Mn (456 mg/kg) and Cr (237 mg/kg); these increased values could be attributed to the operation of the nearby electric power plant. Aluminium presents its maximum value in sample 9 (47.1 mg/kg), while the iron in sample 8 (25.5 mg/kg); these values are most likely related to harbour activity.

Table 1 - Gravel (G), sand (S), silt (Z) and clay (C) percentages and sediment texture of the seabed samples.

Station	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
G	0.40	1.89	2.05	1.10	2.84	7.55	11.74	0.71	2.75	1.71	1.11	1.11
S	99.59	32.21	39.55	21.2	50.66	27.45	31.66	25.49	21.45	49.31	52.99	98.31
Z	0.01	52.69	41.27	62.13	37.96	48.86	36.42	54.7	59.31	40.62	35.04	0.58
C	0.00	13.21	17.13	15.57	8.54	16.14	20.18	19.1	16.49	8.36	10.86	0.00
Text.	(g)S	(g)sM	(g)sM	(g)sM	(g)mS	gM	gM	(g)sM	(g)sM	(g)mS	(g)mS	(g)S
Station	**13**	**14**	**15**	**16**	**17**	**18**	**19**	**20**	**21**	**22**	**23**	
G	14.89	4.45	6.25	4.76	0	3.81	0.86	11.71	0.74	49.49	2.00	
S	17.94	45.9	42.1	70.14	99.98	53.96	58.06	87.98	99.05	50.51	37.63	
Z	55.49	36.00	38.93	19.84	0.02	33.18	31.48	0.31	0.21	0.00	47.44	
C	11.68	13.65	12.72	5.26	0.00	9.05	9.60	0.00	0.00	0.00	12.93	
Text	Gm	(g)sM	gM	(g)mS	S	(g)mS	(g)mS	gS	(g)S	sG	(g)sM	

Key. (g)S: slightly gravely Sand; (g)sM: slightly gravely sandy Mud; (g)mS: slightly gravelly muddy Sand; gM: gravelly Mud; (g)S: slightly gravelly Sand, Gm: muddy Gravel; gS: gravelly Sand; sG: sandy Gravel.

TOC ranged from 0.1% to 6.9%, with its maximum value found in sample 13; the latter may be due to increased seaweed presence and/or to the influence of the nearby waste water treatment outfall. Finally, $CaCO_3$ ranges from 17% (sample 12) to 67% (sample 11), being associated with the coarser grained samples and the presence of shell fragments.

The examined chemical element average values of the Chios Harbour coastal area are relatively lower compared to the other coastal areas of Greece (presented in table 3), with minor exceptions of Cu (due to the existence of small shelters and the main harbor) and Cr (due to the electric power plant) .

According to geoaccumulation (Igeo) and combined contamination index (W), the sediments are characterized as either unpolluted or slightly polluted (Table 4), although there are some samples considered as polluted according to the Igeo index for specific trace metals (indicatively mentioned sample 2 and 13 for Cu and sample 8 for Ni). Moreover, according the modified contamination degree (mCd), the samples are characterised as low to very low polluted, with the exception of stations 2 and 19 that present a moderate degree of pollution (see table 4).

Table 2 - Metal / trace metals concentrations and percentages of $CaCO_3$ and total organic carbon (TOC) of the seabed samples.

Sample	CONCETRATIONS IN TOTAL FRACTION (mg/kg)								% $CaCO_3$	% TOC
	Zn	Cu	Mn	Pb	Cr	Ni	Fe $x10^3$	Al $x10^3$		
01	12.9	11.8	125.1	4.2	40.1	47.9	16.7	21.7	20	0.2
02	79.2	82.2	176.0	20.2	86.1	54.8	15.4	32.0	25	2.9
03	28.6	17.3	149.1	10.0	67.5	38.0	12.2	31.1	28	1.9
04	114.6	56.1	164.0	60.7	84.4	39.1	18.0	35.1	35	1.8
05	32.7	15.1	132.6	16.8	50.5	25.0	11.8	29.9	22	0.6
06	37.3	24.7	202.1	6.5	95.3	73.6	19.5	35.5	33	3.2
07	38.1	34.6	403.8	3.9	114.1	25.6	25.3	46.5	28	0.1
08	59.4	44.0	265.1	27.5	108.1	80.3	25.5	43.3	23	1.6
09	66.4	46.3	277.3	44.2	115.7	19.2	24.5	47.1	27	1.7
11	26.9	13.4	106.7	12.2	52.8	26.1	10.8	41.4	67	2.4
12	73.8	21.4	207.6	22.8	48.1	26.5	22.8	42.1	17	0.3
13	166.6	119.1	161.5	68.3	99.6	61.9	22.4	38.1	23	6.9
15	33.4	22.6	61.8	6.9	106.4	61.8	15.0	28.9	35	2.9
18	30.4	15.6	181.7	72.8	87.6	44.3	13.6	31.3	18	1
19	27.6	25.2	456.2	8.4	237.0	95.0	22.5	36.9	30	0.3
23	44.1	23.3	195.4	31.7	96.5	46.2	15.8	36.0	30	1.5
AVG	54.5	35.8	204.1	26.1	93.1	47.8	18.2	36.1	29	1.8

Note: shaded are the samples from the Chios harbour basin

Table 3 - Concentrations of chemical in elements, in different Greek coastal areas.

AREA	References	Element (mg/kg)				
		Zn	Cu	Pb	Ni	Cr
S. Evoikos Gulf	Anagnostou et al. (1998)	111	17	36	308	378
Elefsina		523	99	194	118	181
Maliakos		89	41		234	
Pagasitikos		172	72	60	174	401
Thermaikos		158	43	190	106	190
Thessaloniki		296	79	64	95	221
NW Saronikos	Paraskevopoulou (2009)	74	23	31	882	2911
Isthmia-Kehries	Dima (2000)	180	28	29	421	
W. Saronikos	Giannopoulou (2005)	134	24	38	277	154
Elefsina		647	108	128	112	91
Psitaleia		460	124	138	150	122
E. Saronikos		436	86.5	86	93	157
S.Saronikos		164	23	49	92	80
Oropos	Botsou (2007)	72	68	27	453	309
N. Aegean Sea	Karageorgis et al.(2005 a, b)	231	55	141	221	249
Average of all areas above		**250**	**57**	**86**	**250**	**420**
Chios (east coast)	This study	54.5	35.8	26.1	47.8	93.1

The percent incidence of adverse biological effects related to concentration, ranges according to TEL, PEL (Macdonald et al., (1996): table 3 p. 266) and ERM, ERL (Long et al., (1995): table 3 p. 92) guideline values are generally in low levels (Table 5). Specifically, according to TEL, PEL range of values, higher possibilities of adverse biological effects have Cr (53%) and Cu (56%), lower Pb (26%) and below 10% Ni (9.6%) and Zi (4%). Moreover, according to ERM, ERL range of values, the largest possibility of harmful effects on living organisms is related to Zn (47%), and to a lesser degree to Pb (35.8%), Cu (29.1%), Cr (21.1%) and Ni (16.7%).

Table 4 - Environmental status of seabed sediments, according to Geoaccumulation Index (Igeo), modified Contamination degree (mCd) and Combined Contamination index (W).

Igeo						mCd	W
Stat.	**Cr**	**Cu**	**Ni**	**Pb**	**Zn**		
01	UP	UP	SP	UP	UP	very low	UP
02	SP	P	SP	UP	UP	moderate	SP
03	SP	UP	SP	UP	UP	very low	UP
04	SP	P	SP	UP	UP	low	UP
05	SP	UP	UP	UP	UP	very low	UP
06	SP	SP	P	UP	UP	very low	UP
07	SP	SP	UP	UP	UP	very low	UP
08	SP	SP	P	UP	UP	Low	SP
09	SP	P	UP	UP	UP	Low	SP
11	SP	UP	UP	UP	UP	very low	UP
12	SP	UP	UP	UP	UP	very low	UP
13	SP	HP	P	SP	UP	voderate	UP
15	SP	UP	P	UP	UP	very low	UP
18	SP	UP	SP	SP	UP	very low	SP
19	P	SP	P	UP	UP	moderate	SP
23	SP	SP	SP	UP	UP	very low	SP
Key. HP: heavily polluted, P: polluted, SP: slightly polluted and UP: unpolluted							

5. Conclusions

The broader coastal area of Chios harbour consists of fine grained sediment that are generally characterized as slightly gravelly sandy mud ((g)sM), according to Folk (1974). The concentrations of the identified heavy metals Zn, Cu, Pb, Ni, Fe, Mn, Al, Cr, as well as those of TOC and $CaCO_3$ vary within normal ranges, while their comparison with other Greek coastal areas showed that the study area is less polluted, although some metals' slightly increased values are found locally; the latter are related to the harbour of Chios, fishing shelters, the offshore area close to the electric power plant and nearby the outflow of the waste treatment plant.

The application of Igeo, mCd and W indices, showed that in most locations the bottom sediments are non-polluted, while marginally to slightly polluted found most of the samples in chromium, samples 2, 4, 6, 8, 19 & 23 both in copper and in nickel, sample 7 only in copper, samples 1, 3, 13, 15, 18 & 19 in nickel and samples 13 & 18 in lead. Nevertheless, all samples appeared not to have any significant environmental impact to benthic organisms, according to TEL, PEL and REM, ERL guideline values.

Table 5 - Percent incidence of adverse biological effects, in concentration ranges, according to TEL - PEL) (Macdonald et al., 1996) and ERM – ERL guidelines values (Long et al., 1995).

	TEL – PEL					ERM- ERL				
	Cr	**Cu**	**Ni**	**Pb**	**Zn**	**Cr**	**Cu**	**Ni**	**Pb**	**Zn**
1	4	9	9.4	6	4	2.9	2.9	16.7	8	6.1
2	15	22	9.8	6	4	21.1	29.1	16.9	8	6.1
3	15	9	8.4	6	4	2.9	2.9	16.7	35.8	6.1
4	15	22	8.4	26	4	21.1	29.1	16.7	35.8	6.1
5	4	9	8.4	6	4	2.9	2.9	16.7	8	6.1
6	15	22	9.4	6	4	21.1	2.9	16.9	8	6.1
7	15	22	8.4	6	4	21.1	29.1	16.7	8	6.1
8	15	22	9.4	6	4	21.1	2.9	16.9	8	6.1
9	15	22	8.4	26	4	21.1	29.1	1.9	8	6.1
11	15	9	8.4	6	4	2.9	2.9	16.7	8	6.1
12	4	22	8.4	6	4	2.9	2.9	16.7	8	6.1
13	15	56	9.4	6	4	21.1	29.1	16.9	8	6.1
15	15	22	9.4	6	4	21.1	2.9	16.9	8	6.1
18	15	9	9.4	26	4	21.1	2.9	16.7	35.8	6.1
19	53	22	9.5	6	4	21.1	2.9	16.9	8	6.1
23	15	22	9.6	26	4	21.1	2.9	16.7	35.8	47

6. Acknowledgements

The authors A. Tsoutsia, V. Kapsimalis and S. Poulos, during this work have been supported also by the Project MARE (MIS 375655), funded by the Operational Programme: “Education and lifelong learning, 2007-2013” of the Ministry of Education and Religious Affairs, Culture and Sports, co-funded by the European Union and the Greek Government. Mrs A. Tsoutsia was supported, also, by Papadakis Antonios Legacy. Finally, thanks are extended to Dr I. Panagiotopoulos (HCMR), Mr G. Pappas (HCMR) and to Mr. S. Petrakis (PhD candidate of University of Athens) for their contribution during the field trip and laboratory analyses.

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