OIL AND HEAVY METALS IN THE SEDIMENTS OF THERMAIKOS GULF AND THE SARONIC GULF, GREECE

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INTRODUCTION

The distribution of oil and heavy metals in surface sediments of marine coastal environments is influenced considerably by anthropogenic inputs. Many studies have been carried out using sediments as an indicator of the distribution and therefore environmental impact of these pollutants.

This study was commissioned by the environmental group, Greenpeace Greece, to investigate the effects of industry, in particular the oil industry, on the coastal environment of Greece. Two areas of the Greek coast were chosen for study, these areas being Thermaikos Gulf and the Saronic Gulf (see figure 1). For ease of interpretation the Saronic Gulf was subdivided into two areas, these being the Elevsis Gulf (map 2) in the north and the Kenkhreon Gulf (map 3) to the northwest.

Thermaikos Gulf receives outflow from the Axios, Loudhias and Alaikmon rivers. The rivers are utilised by several Greek industries as a convenient method of removing effluent which then empties into the Thermaikos Gulf. The Axios river originates in Yugoslavia and has a considerable pollutant loading before it crosses the border into Northern Greece (Vasilikiotis *et al* 1991). There are many domestic and industrial inputs into the gulf from the conurbation of Thessaloniki where there is an oil refinery which produces in excess of 20,000 barrels a day (Couper, A. 1983). Also there are fertilizer industries, steelworks, tanneries and a chlor-alkali plant.

The Elevsis Gulf is located in the north and Kenkhreon Gulf in the northwest of the Saronic Gulf (See maps 2 & 3). The Kenkhreon Gulf connects with the Gulf of Corinth and ultimately the Ionian Sea via a small canal at its western end. The Gulf of Elevsis is a shallow, semi-enclosed embayment which connects with the main body of the Saronic Gulf by means of two channels, one to the southwest and one to the southeast. It receives industrial pollutants from a number of sources. There are oil refineries (producing in excess of 100,000 barrels a day, Couper, A. 1983), shipyards, steelworks and several others. The sewage system of Athens, which contains both industrial and domestic effluent, discharges southeast of the Gulf. Inversion in the predominant clockwise circulation of the Gulf and wind driven currents often brings this polluted water into the Gulf (Scoullos 1986).

Oil is of environmental concern for several reasons, it's complicated nature results in a variety of detrimental effects on ecosystems. Oil is a mixture of fractions with differing volatility, molecular weight, toxicity and solubility in water. Oil from different sources will have different characteristics. There is a tendency for the relatively insoluble, high molecular weight fraction of the oil to end up in the sediments with the more soluble components, e.g. benzene, dispersed through the water column. The more volatile components tend to be released to the atmosphere. The amount of oil entering the sediment will therefore only be a proportion of the total released to the environment. The immediate physical effects of accidental oil spills on seabird and mammal populations are dramatic and well documented (Howarth & Marino 1991), however much oil enters the environment in a chronic manner by many routes. The majority of oil input into the marine environment comes from routine discharges associated with marine transportation and municipal and industrial wastes. Urban runoff and atmospheric fallout also are significant sources.

Many of the components of oil are extremely toxic and carcinogenic, particularly PAHs (polynuclear aromatic hydrocarbons). There are a variety of sub-lethal effects of oil-derived substances, including changes in physiology, reduction in growth and reproductive success, alteration of behaviour, and accumulation of hydrocarbons in tissues (Howarth & Marino 1991). Benthic organisms are adversly affected by oil in sediments at concentrations as low as 0.1 g/kg of sediment (Elmgren and Frithsen 1982). A more comprehensive description of the effects of oil in the marine environment is given by Howarth and Marino (1991).

Cadmium, chromium, lead and soluble copper compounds are included in the EEC list of priority pollutants. The toxicity of these metals is well documented (Brown & Kodama 1987) and this has prompted many investigations into their distribution in the environment (Bridges 1989, Nriagu 1990). Lead has been determined in the sediments of the Elevsis Gulf in a previous study by Scoullos (1986). Other workers have investigated heavy metals in other areas of the Greek coastal environment. The sediments of Thermaikos Gulf have been investigated by Chester and Voutsinou (1981) using a method which involved cold digestion of the sediments. The Axios river and Thessaloniki were identified in this study to be the major sources of Cd, Zn, Pb and Cu Pollution.

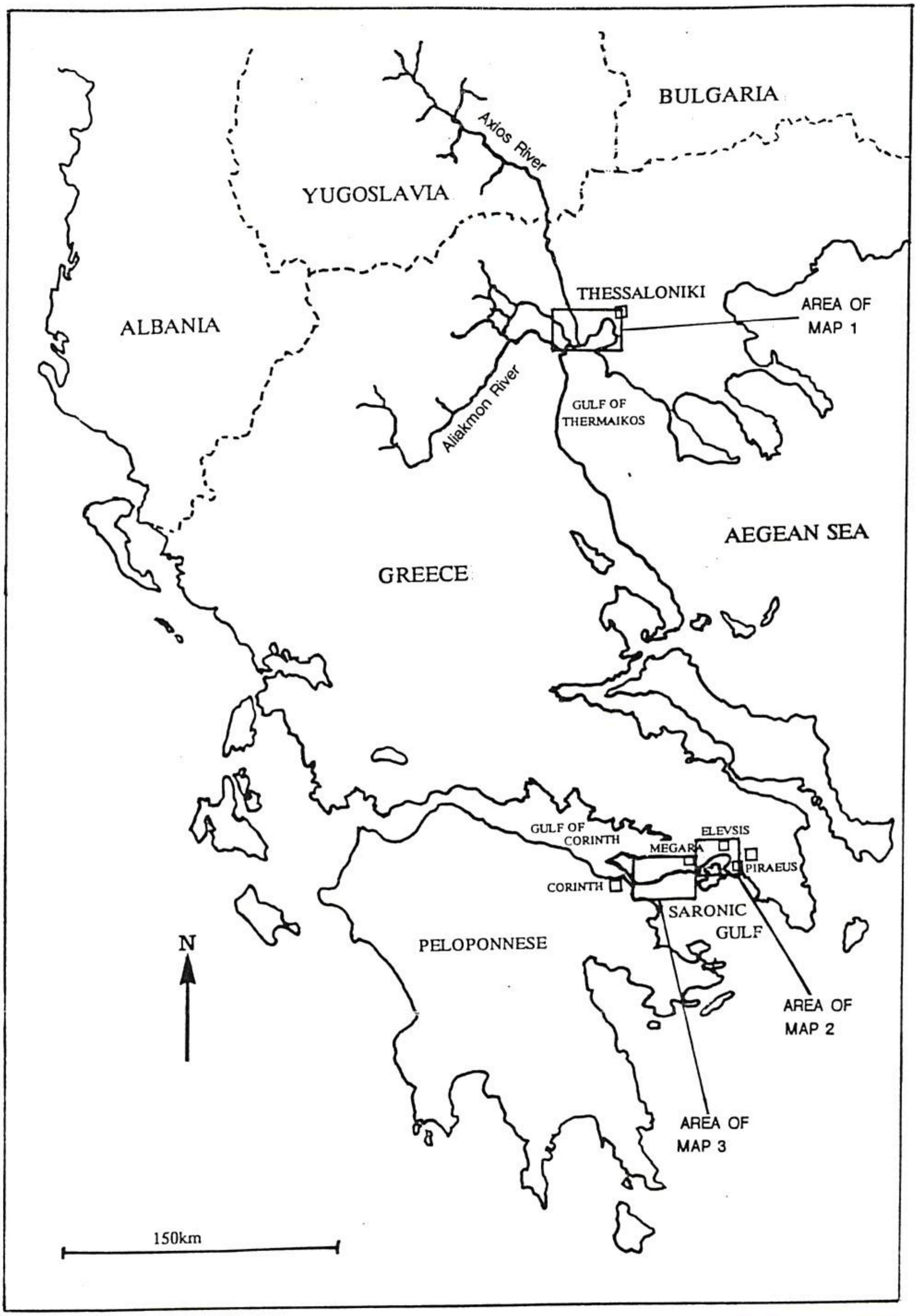
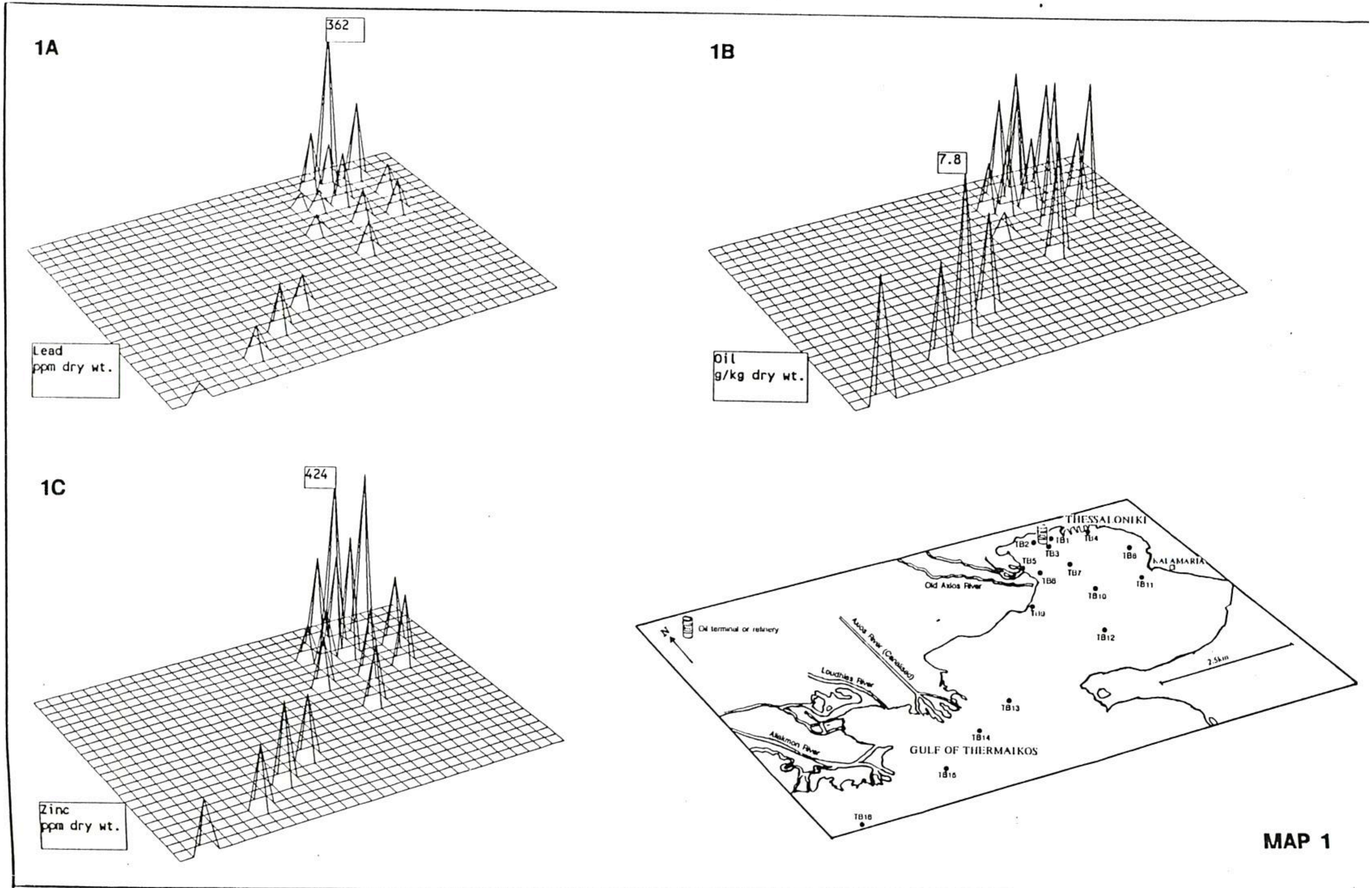


FIGURE 1: Location of sampling areas. The Saronic Gulf has been divided into two areas for simplicity (maps 2 and 3).



GULF OF THERMAIKOS. Concentrations of lead, zinc and oil. Note that the scales are different for each graph and that the north-south orientation of the map is altered for ease of interpretation. The highest concentration found in each sample set has been highlighted.

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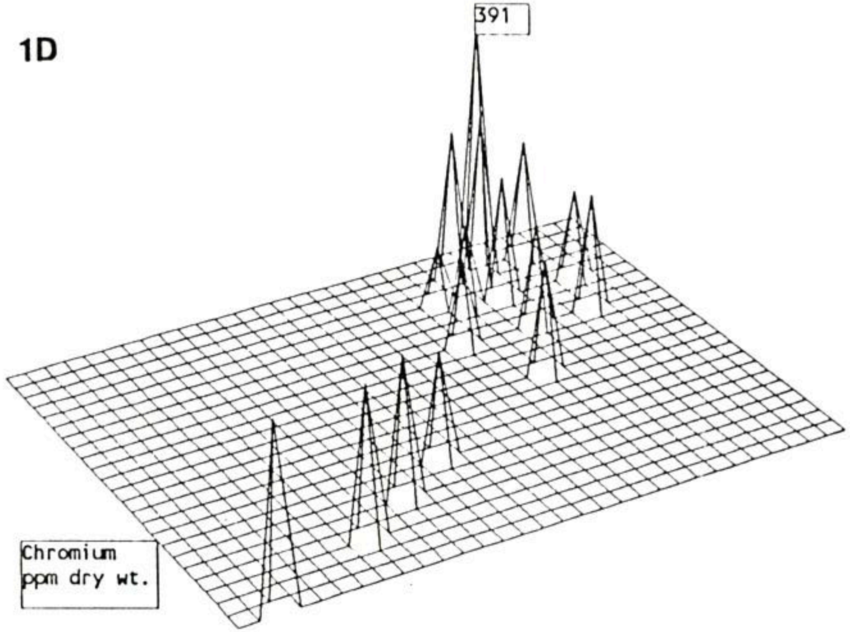

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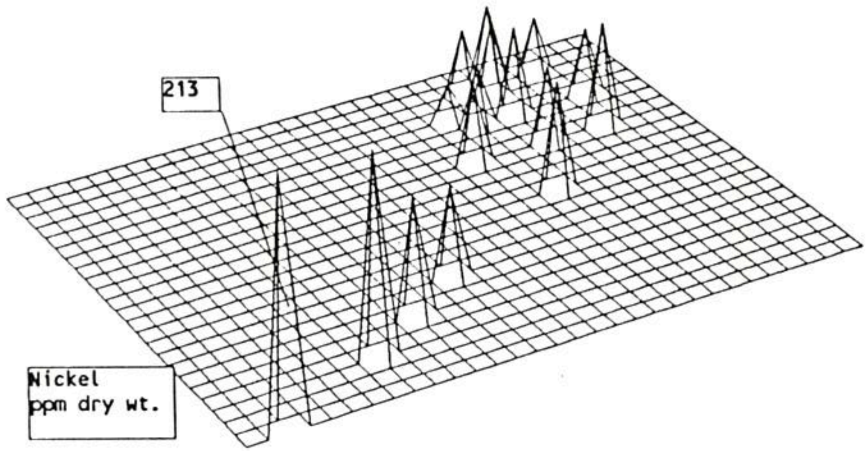

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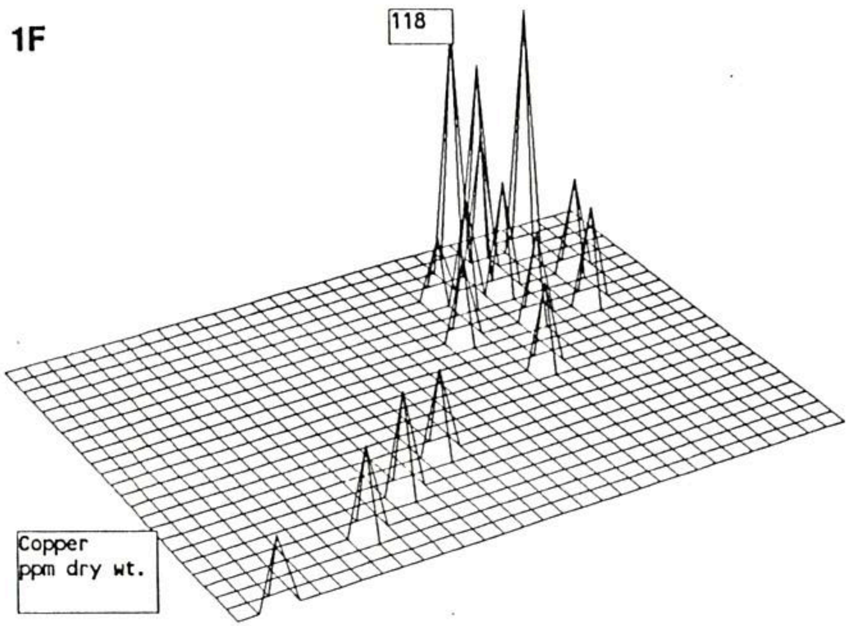
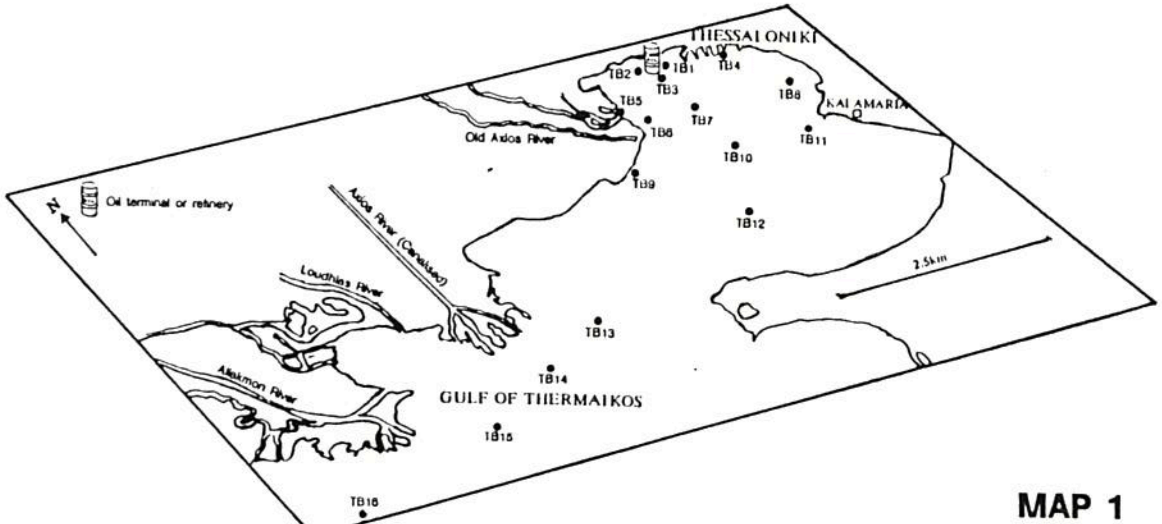
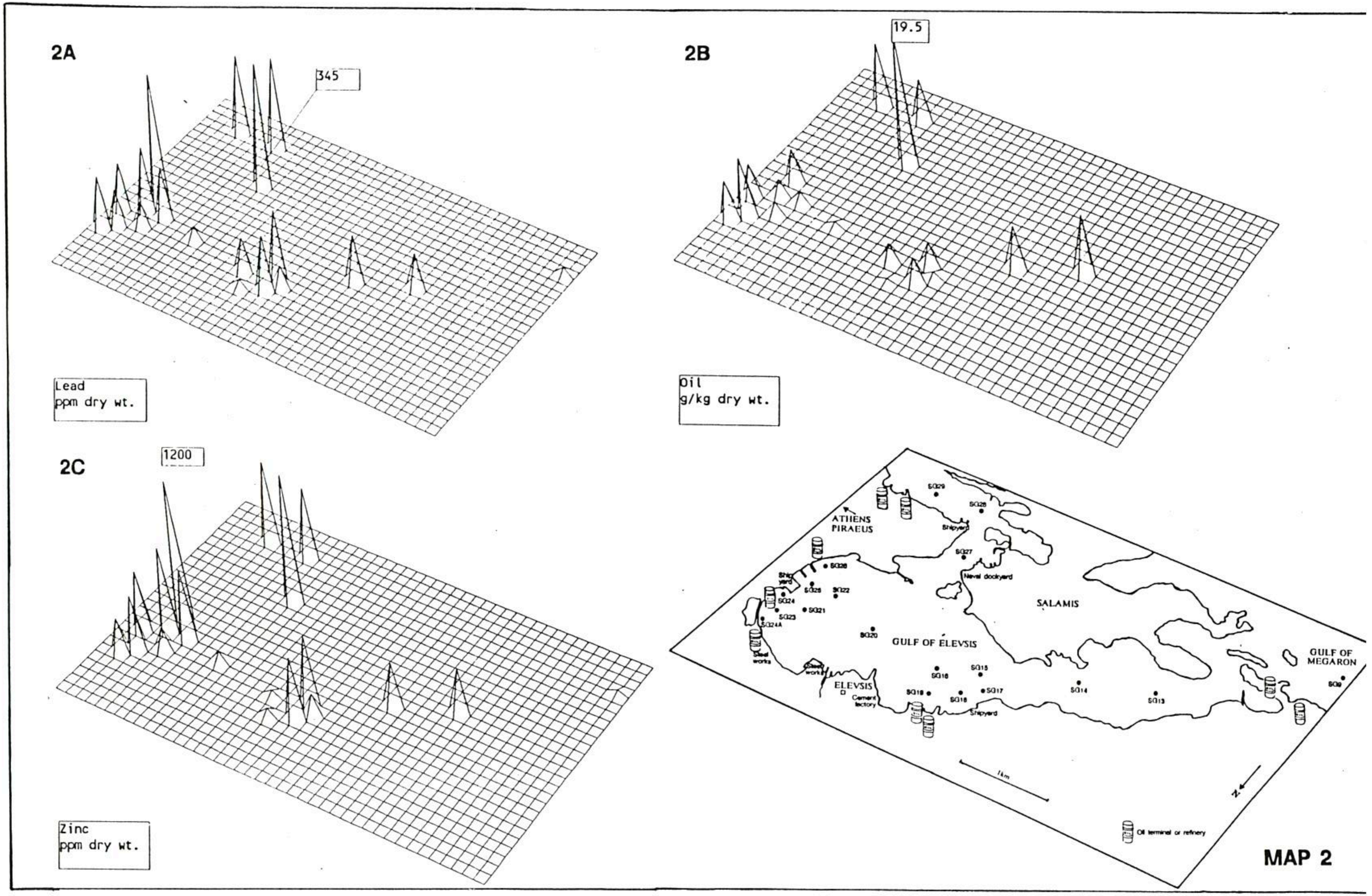


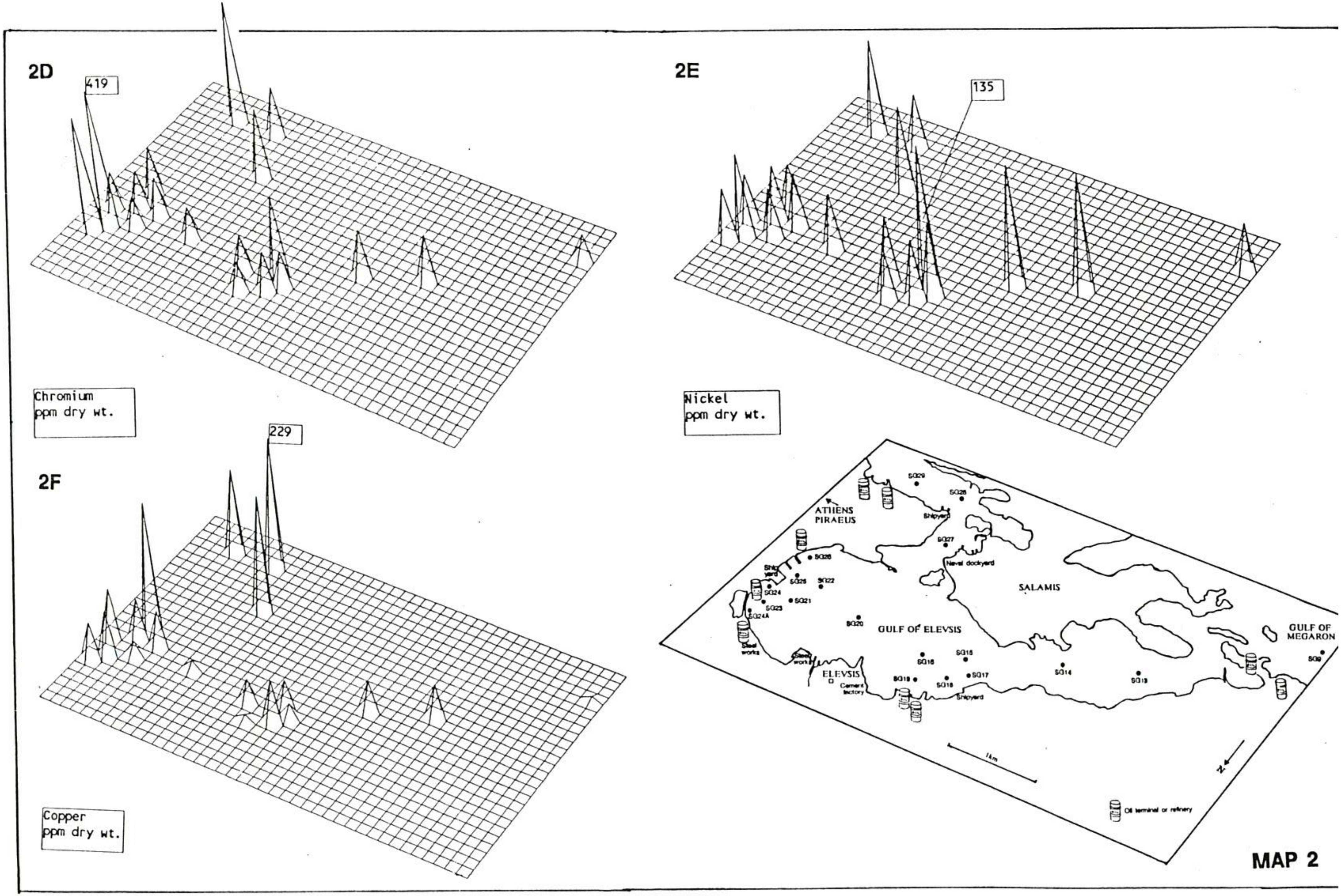


MAP 1

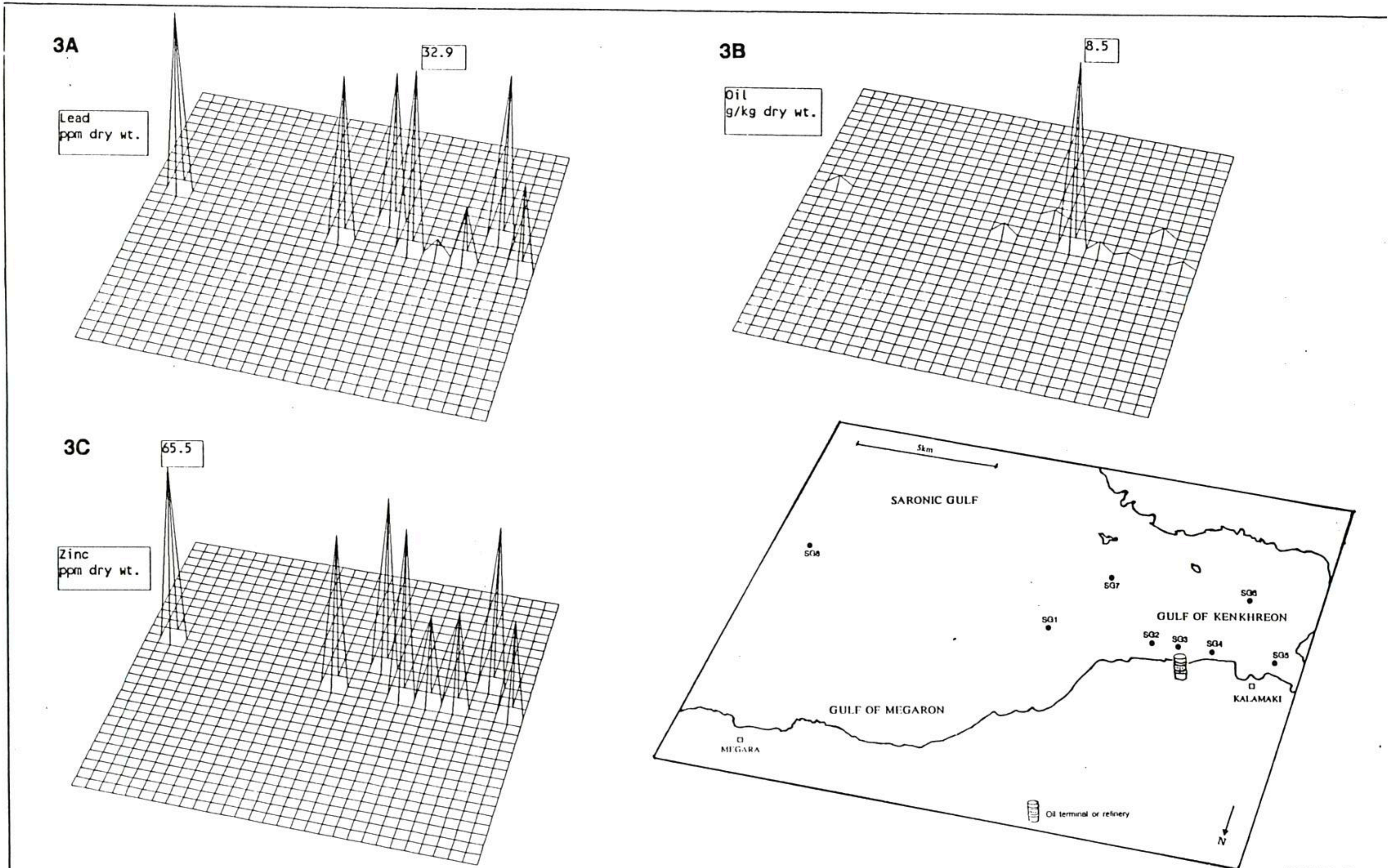
GULF OF THERMAIKOS. Concentrations of chromium, nickel and copper. Note that the scales are different for each graph and that the north-south orientation of the map is altered for ease of interpretation. The highest concentration found in each sample set has been highlighted.



GULF OF ELEVSIS. Concentrations of lead, zinc and oil. Note that the scales are different for each graph and that the north-south orientation of the map is altered for ease of interpretation. The highest concentration found in each sample set has been highlighted.

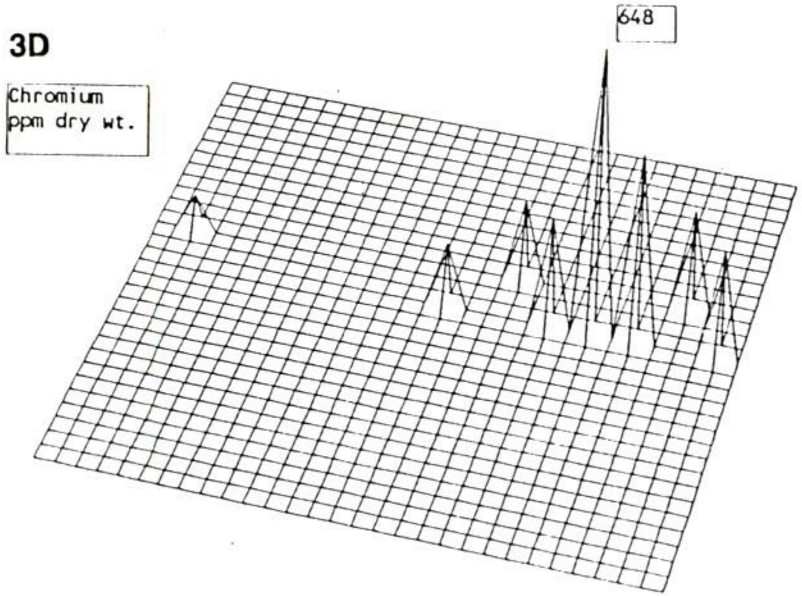


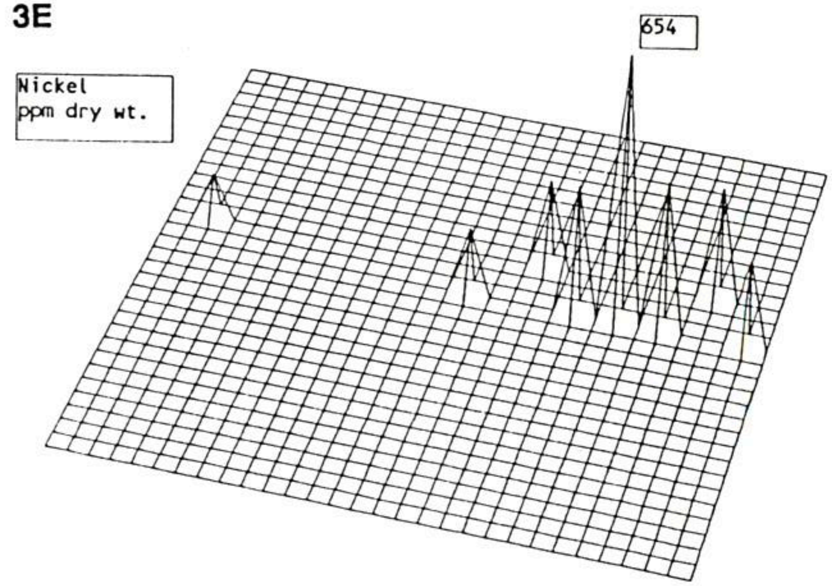
GULF OF ELEVSIS. Concentrations of chromium, nickel and copper. Note that the scales are different for each graph and that the north-south orientation of the map is altered for ease of interpretation. The highest concentration found in each sample set has been highlighted.

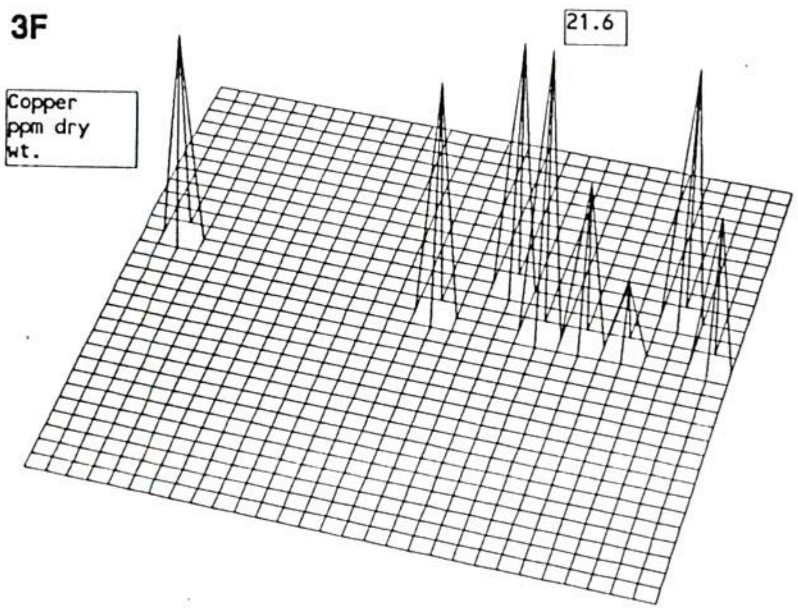


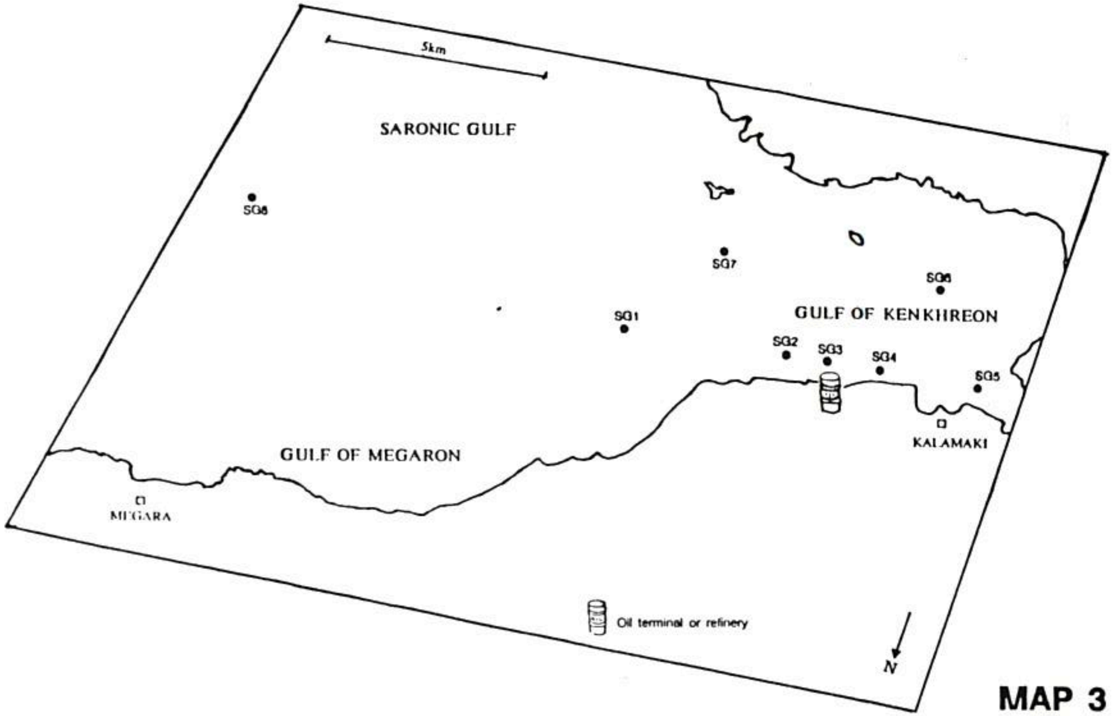
MAP 3

GULF OF KENKHREON. Concentrations of lead, zinc and oil. Note that the scales are different for each graph and that the north-south orientation of the map is altered for ease of interpretation. The highest concentration found in each sample set has been highlighted.









MAP 3

GULF OF KENKHREON. Concentrations of chromium, nickel and copper. Note that the scales are different for each graph and that the north-south orientation of the map is altered for ease of interpretation. The highest concentration found in each sample set has been highlighted.

SAMPLING AND ANALYSIS

Sediment samples were obtained using a Van Veen grab operated from the Greenpeace vessel MV Sirius in the spring of 1991. The location of these samples is shown in maps 1, 2 & 3. Samples from the Thessaloniki Bay area were labelled TB1-TB16 (map 1). Samples from the Saronic Gulf were labelled SG1-SG29 (see maps 2 & 3) but note that there are no samples SG10-SG12 due to sampling difficulties. Oil and heavy metals analyses were conducted at Queen Mary and Westfield College, University of London, U.K.

Oil (total hydrocarbon content)

50 mg of wet sediment and 10 g of 0.3 cm diameter glass balls were weighed into a 300 ml bottle. 25 ml of methanol was added and gently swirled by hand to dewater the sediment. The methanol-water mixture was decanted into a 250 ml beaker. Then 50 ml of 2:1 dichloromethane:methanol was added to the sediment which was then placed on a bottle roller for 16 hours. The extract was then decanted into the 250 ml beaker. This extraction step was repeated twice, first for 6 hours, then for 16 hours. All extracts were combined and filtered. The beaker and filter were washed twice with 10-20 ml of dichloromethane and the total filtrate was then gently swirled with 250 ml distilled water. After the phases separated, the distilled water was back-extracted with 10 ml dichloromethane and added to the cleaned organic phase. The solvent and water were removed by bubbling through nitrogen gas and the dry residue was weighed to obtain the final figure for total hydrocarbon content. For further information on this method consult Brown *et al* 1980. Two different standards were added to several samples to ascertain the oil-recovery.

Heavy metals

A range of seven metals was investigated, these being Cu, Cd, Cr, Pb, Ni, Zn & Ag. Sediments were oven-dried and their moisture content thereby determined. Preparation of the sample digests was by microwave digestion in a CEM MDS2000 sample preparation system. Approximately 1 gram of sample was weighed accurately into an acid washed teflon sample vessel and 20ml of 1:1 nitric acid:double distilled water added. The samples were microwaved at full power for one hour and pressure controlled to a maximum of 150psi. Duplicate samples were analysed for each sampling location and an average value thus obtained. Analysis was by Thermoelectron IL157 AAS calibrated against known standards and background corrected for all the metals except Cr.

RESULTS

The results are presented in the following table and graphically in mock 3-dimensional form (graphs 1a-f, 2a-f, & 3a-f). Only five of the seven metals are illustrated graphically, these metals being lead, copper, zinc, nickel and chromium. Silver and cadmium were not illustrated in this way because they were not detected in all the samples, these results will, however, be referred to in the text. The graphical representations were obtained using Wingz software. Concentrations of pollutant are illustrated relative to the highest value encountered in a given sample set, therefore the scale varies for each graph. The highest concentrations for each sample set are shown in boxes next to the sample location on the graph. The north-south orientation has been altered in the maps to aid interpretation of the graphical results. Care should be taken in interpreting these graphs because the data sets are limited, however they give a good overview and an indication of anthropogenic sources of these pollutants.

There are some limitations to the total hydrocarbon analysis method which should be kept in mind when interpreting the results. The method does not exclude natural hydrocarbons, although in practise this makes little difference because natural hydrocarbons contribute a small quantity in comparison to oil-derived hydrocarbons. The nitrogen blow-down at the end of the extraction procedure may remove some of the more volatile components of the oil (although, as mentioned in the introduction, this fraction is usually somewhat reduced by the time that oil reaches the sediment). Also the oil-recovery from the sediment will vary with the composition of the oil.

SAMPLE NUMBER	CADMIUM CONC. IN DRY SAMPLE (ppm)	LEAD CONC. IN DRY SAMPLE (ppm)	COPPER CONC. IN DRY SAMPLE (ppm)	NICKEL CONC. IN DRY SAMPLE (ppm)	ZINC CONC. IN DRY SAMPLE (ppm)	CHROME CONC. IN DRY SAMPLE (ppm)	SILVER CONC. IN DRY SAMPLE (ppm)	OIL CONC. IN DRY SAMPLE (g/kg)
TB1	4.29	362	91.0	63.1	424	391	<1.0	5.6
TB2	<3.0	140	118	57.7	239	239	<1.0	4.8
TB3	<3.0	119	68.5	65.7	250	270	<1.0	5.4
TB4	<3.0	186	117	51.1	440	192	2.36	5.0
TB5	<3.0	32.1	29.5	33.3	95.7	95.5	<1.0	2.4
TB6	<3.0	45.6	50.1	54.4	149	137	<1.0	3.5
TB7	<3.0	124	53.9	79.6	335	199	1.11	3.5
TB8	<3.0	65.2	45.3	60.1	190	144	2.13	3.2
TB9	<3.0	44.6	39.3	71.8	137	143	<1.0	1.2
TB10	<3.0	74.5	42.5	68.5	182	167	2.22	7.2
TB11	<3.0	76.0	47.0	93.4	195	188	<1.0	6.7
TB12	<3.0	70.4	40.8	97.4	163	182	<1.0	5.7
TB13	<3.0	69.4	39.4	78.8	173	170	<1.0	4.7
TB14	3.11	103	47.2	103	220	225	<1.0	7.8
TB15	<3.0	67.6	40.5	175	169	241	<1.0	4.6
TB16	<3.0	39.9	33.1	213	147	294	<1.0	6.1
SG1	<3.0	29.8	17.8	166	57.2	148	<1.0	0.5
SG2	<3.0	32.9	21.6	321	63.7	251	1.06	8.5
SG3	<3.0	3.0	12.3	654	33.1	648	<1.0	0.5
SG4	<3.0	10.0	5.3	368	36.3	433	<1.0	0.2
SG5	<3.0	16.1	11.6	217	36.7	252	<1.0	0.3
SG6	<3.0	30.2	19.2	281	60.4	236	<1.0	0.4
SG7	<3.0	27.3	18.8	220	64.4	184	<1.0	0.3
SG8	<3.0	32.7	15.7	110	65.5	130	<1.0	0.4
SG9	<3.0	24.1	8.38	46.4	41.9	81.9	<1.0	0.5
SG13	<3.0	98.8	60.7	114	402	143	<1.0	8.9
SG14	<3.0	128	54.4	118	393	146	<1.0	6.3
SG15	<3.0	185	49.9	135	617	247	<1.0	3.5
SG16	<3.0	92.2	41.7	61.7	300	107	1.02	2.6
SG17	<3.0	59.7	27.7	71.2	190	108	<1.0	1.0
SG18	<3.0	151	75.8	57.7	557	121	<1.0	3.7
SG19	<3.0	23.7	11.3	49.9	76.4	85.9	<1.0	0.4
SG20	<3.0	36.5	17.4	54.4	104	97.1	<1.0	0.8
SG21	<3.0	67.4	26.8	47.7	197	91.2	<1.0	1.9
SG22	<3.0	141	63	51.5	623	119	<1.0	1.7
SG23	<3.0	99.3	78.8	80.7	475	419	<1.0	8.5
SG24	<3.0	127	85.5	43.3	522	109	<1.0	4.7
SG24A	<3.0	143	62.7	50.0	304	349	<1.0	5.6
SG25	<3.0	177	69.2	52.0	741	115	<1.0	2.4
SG26	<3.0	341	212	39.2	1200	146	1.52	4.7
SG27	<3.0	345	201	80.9	1130	216	3.03	19.5
SG28	<3.0	254	229	51.7	631	153	2.13	6.2
SG29	3.32	220	146	91.8	730	382	3.32	9.4

TABLE 1: Heavy metals and oil in sediments. Samples TB1-TB16 are from the Gulf of Thermaikos, sample sites are illustrated in map 1 and results are illustrated graphically in graphs 1a-1f. Samples labelled 'SG' are from the Saronic Gulf, SG1-8 are from the Kenkhreon Gulf (map 3 and graphs 3a-3f) and SG9-SG29 are from the Elevsis Gulf (map 2 and graphs 2a-2f).

	Cd (ppm)	Pb (ppm)	Cu (ppm)	Ni (ppm)	Zn (ppm)	Cr (ppm)	Ag (ppm)	Oil g/kg
THERMAIKOS GULF This study	<3.0-4.29	32.1-362	29.5-118	51.1-213	95.7-440	95.5-391	<1.0-2.36	1.2-7.8
ELEVSIS GULF This study	<3.0-3.32	23.7-345	11.3-229	43.3-135	76.4-1200	85.9-419	<1.0-3.32	0.4-19.5
KENKHREON GULF This study	<3.0	3.0-32.9	5.3-21.6	46.4-654	33.1-65.5	81.9-648	<1.0-1.06	0.2-8.5
FORTH ESTUARY U.K. McIntyre 1988	-	-	-	-	-	-	-	0.33-4.15
HUMBER ESTUARY U.K.[a] Barr et al. 1990	-	36-187	19.2-90.5	15.6-61.7	96-361	42-175	-	-
ROTTERDAM[b] Nijssen 1988	3-21	80-320	39-203	24-66	256-1614	79-417	-	0.52-3.50
ELEVSIS GULF[c] Scoullos 1986	-	160-500	-	-	-	-	-	-
THERMAIKOS GULF[d] Chester & Voutsinou 1981	n.d.-3.0	13-228	4.0-37	35-160	23-299	31-107	-	-

TABLE 2: Comparative values from other studies.

[a] <90um particle fraction

[b] figures for dredged material, here the heavy metals have been extrapolated to a 50% fraction smaller than 16 um.

[c] <61um particle fraction

[d] <61um particle fraction and digestion in cold 0.5 N HCl

Thermaikos Gulf

oil

Oil concentrations are elevated throughout the gulf in comparison to other industrialised estuaries (see table 2). The harbour area and the Rivers Axios and Aliakmon are indicated as sources.

metals

The results indicate the harbour area of Thessaloniki and the canalised section of the River Axios to be sources of heavy metals pollution into the Gulf. The patterns of distribution for copper, lead, and zinc illustrate this clearly. Nickel and chromium concentrations are elevated in the region of the mouth of the Aliakmon River also, although the highest concentration of chromium is found in the region of Thessaloniki. In contrast the old River Axios inputs apparently unpolluted sediment as indicated by sample TB5 taken in the mouth of the River. Silver and cadmium were found in appreciable quantity in sediment sample SG1 near Thessoloniki. Cadmium was found at a concentration of 3.11ppm in sediment taken from the mouth of the River Axios (sample TB14). The overall level of metal pollution compares with that found in the highly polluted sediments of Rotterdam Harbour, and exceeds that reported for the Humber estuary (see table 2).

Elevsis Gulf

oil

Oil concentrations are elevated throughout the gulf in comparison to other industrialised estuaries (see table 2). It is evident from the graphical representation (see figure 2B) that there is some spatial variation in total oil content of sediment. Sample SG27 shows an exceptionally high level of total oil content of 19.5 g per 1 kg dry sediment. Other samples indicate a possible point source in the region of Piraeus, SG24a, SG23 and SG13 are also noteworthy.

metals

The sediments of the Elevsis Gulf exhibit elevated concentrations of all heavy metals in this study in comparison to the Megaron Gulf just to the west (sample SG9). The eastern channel of the gulf exhibits elevated concentrations of zinc, silver, cadmium, lead and copper. These compare with concentrations found in the highly polluted sediments of Rotterdam harbour And exceeds those founf in the Humber estuary (see Table 2). There are high concentrations of these metals (except cadmium)in sample SG26 also, which indicate a point source of these metals. A different pattern of distribution is indicated for chromium and nickel, it is obvious that there are several point sources releasing these metals into the Gulf. For example, samples SG24a and SG23 contain very high levels of chromium for which the only explanation can be localised anthropogenic input.

Kenkhreon Gulf

oil

Concentrations of total oil in sediment are lower here than in the Gulf of Elevsis with the exception of sample SG2. In general the total oil content compares with that found in sediments taken from the mouth of the Firth of Forth (McIntyre 1988). The Firth of Forth is an estuary in the U.K. which has the Grangemouth oil refinery situated at the landward end, sediments are most contaminated close to the refinery but the effect is less at the mouth of the estuary (see table 2). However the concentration found in sample SG2 exceeds that found even close to the refinery or in the highly-polluted sediments of Rotterdam harbour.

metals

Concentrations of nickel and chromium are much elevated in sediments close to the shore indicating a point source discharge of these metals. Detectable concentrations of silver were found in sample SG2. Concentrations of the other metals investigated were not high enough to discern an anthropogenic influence.

DISCUSSION

Thermaikos Gulf

The graphical representations of the results indicate that the canalised section of the Axios river is a source of both oil and heavy metals pollution. Thessaloniki harbour and the Aliakmon river are also implicated. Such a heavy input of pollutants is likely to be severely impacting the local ecosystem. The benthic fauna will be severely modified by oil pollution alone. It has been demonstrated, for example, that amphipod populations are severely perturbed by oil pollution (Sanders *et al* 1980). This is likely to have a "knock-on" effect on fish populations as amphipods contribute a large proportion to the diet of many demersal fish (Langton 1983). This is only one of a whole range of observed detrimental effects on fisheries, and wider ecosystems, caused by oil pollution. These effects are so many and varied as to be impossible to summarise in a document such as this, for further information see Howarth and Marino (1991).

As mentioned in the introduction, the concentration of oil found in the sediment will represent only a small proportion of the oil originally introduced into the environment. With such high concentrations as found in this study it is probable that elevated levels of hydrocarbons are dissolved in the seawater and are present in the atmosphere. Many of these more easily dissolved and volatile fractions are carcinogenic and probably present a health hazard to the inhabitants of this region.

The concentrations of heavy metals found in this study were higher than found by Chester and Voutsinou (1981) this was to be expected due to the difference in digestion procedures and so no conclusions about temporal change in these concentrations can be made. However the patterns of distribution of the metals studied are similar. Unfortunately the scope of this study was limited to the northern end of the Gulf. Sediments taken from the southern part of the gulf would be expected to exhibit a lowering trend (as shown by the work of Chester and Voutsinou 1981) if the only anthropogenic sources were those already identified above.

The elevated concentrations of nickel and chromium found in this study are a cause for concern. Many aspects of the ill-effect on human health caused by nickel in its various forms have been studied. Nickel and nickel compounds have been classified as human carcinogens by IARC. Nickel also causes contact dermatitis. Hexavalent chromium possesses carcinogenic properties. Possible sources of contamination in the Gulf are tanneries and metalworks.

Cadmium was found in samples near Thessaloniki at elevated levels, the presence of cadmium in the sediments at the levels found warrant further investigation. Cadmium does not occur freely in nature and there are no specific ores from which it is mined, it is of concern to human health and has been demonstrated to have several adverse effects.

From the results it is clear that the Gulf of Thermaikos is heavily polluted with oil and heavy metals. The sediments compare with those found in Rotterdam harbour. The majority of Rotterdam dredgings are considered too polluted to dump in the sea or to spread on agricultural land and therefore are treated as toxic waste by the Netherlands authorities (Nijssen 1988). The long term policy for these dredgings is to reduce pollutant loading of the sediments from source so that such polluted sediments are no longer produced. The implemention of such a policy in the Thermaikos Gulf should be considered urgently.

Saronic Gulf

It is clear from the results of this study that the Elevsis gulf is heavily polluted with heavy metals and oil. The sediments compare with those found in Rotterdam harbour except for oil which in some samples much exceeds these concentrations. There is considerable variation in the heavy metals content and distribution in the sediments sampled in the Saronic Gulf. It appears that there are several sources of anthropogenic enrichment of these metals. Scoullos (1986) reports higher concentrations for lead in sediments from the Elevsis Gulf than have been found in this study. This would be expected because his study quotes concentrations for total sediment whereas Scoullos reports the $<61^{m}$m fraction. It is likely that the domestic effluent from Athens is contributing directly to the high concentrations of pollutants found in samples SG28 and SG29. For instance a likely source of silver is from photographic processing, the effluent from which frequently is discharged into municipal sewage systems. The presence of silver is of concern because of its high toxicity, silver has been shown in laboratory experiments to be one of the most toxic elements to microorganisms and biochemical processes in the soil (Jones *et al* 1990). Sample SG27 has high concentrations of both lead and oil. Any of the following causes could explain this high concentration of oil, a large recent spill could have occurred or it is possible that this is caused by a leaking pipeline or chronic discharges from fuelling operations at the nearby naval dockyard.

The Kenkhreon Gulf sediments are, in general, less polluted than those of the Elevsis Gulf. There is a severe localised problem with chromium and nickel pollution however. Oil levels are elevated also, particularly in sample SG2. It is probable that the benthic community structure in the Elevsis and Kenkhreon Gulf has been severely modified by the impact of the oil pollution. Any fisheries in the area will be affected both directly by the oil pollution and indirectly through effects through the food chain (as mentioned in the discussion above).

From the results of this study it is clear that the Elevsis Gulf is suffering a high degree of environmental pollution due to insufficient regulation of many sewage and industrial inputs. The enclosed and shallow nature of the Gulf only serves to emphasize this problem. The Gulf gives industry a back door route to cheap and easy waste removal and provides no incentive for waste reduction.

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