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Abstract

Lead pollution and its sources have been investigated in the south-eastern and south-western Black Sea. Surficial sediments and mussels were collected in different seasons of the year from the south-eastern and south-western Black Sea and analysed for their lead contents.

In the south-eastern Black Sea sediments from the easternmost and from the central stations contained the highest lead concentrations. Sediments contained 8 to 10 times the lead concentrations of sediments from the south-west. Samples taken from rivers, streams and effluents had especially high lead concentrations. The highest lead concentrations were found in sediments collected in October and December.

Lead concentrations in mussels from the south-eastern coast were comparatively lower compared to those from the south-western coast. The highest average values occurred in December and in October.

In the south-western Black Sea, the highest lead concentrations in sediments were obtained in those from Inebolu, followed by those from around Zonguldak. The concentrations increased from the middle to the west of the southern Black Sea coast, possibly due to the effect of the Danube River. Sediments taken in September had the highest lead concentrations, followed by samples taken in December.

Very high lead concentrations (> 10 $\mu g\, g^{-1}$) were observed in mussels from those stations where the sediments also contained high concentrations. Mussels had their highest lead concentrations in January and April.

Keywords: Black Sea, Pollution, Lead, Sediment, Mussel.

Introduction

The Black Sea, with a surface area of approximately 4.2 x 10^5 km^2, a volume of 5.3 x 10^5 km^3 and a maximum depth of ~2300 m, constitutes one of the world's largest inland basins (OGUZ *et al.,* 1992). The sea borders several different cities and countries. It is an important area for fishing and aquaculture. It receives an enormous quantity of pollutant-containing sedimentary material, being fed from all sides by several large rivers and many streams, each of which possesses a large drainage area. According to ZAITSEV (1992) 4500 tons of lead reach the Black Sea annually through this river network. TUNCER *et al.* (1998) reported the total annual flux of lead from the Turkish coast to be around 60 tons, 42 of which entered through rivers and 18 tons from domestic and industrial discharges. The atmosphere constitutes another source of lead to the Black Sea (HACISALIHOĞLU *et al.,* 1992; KUBILAY *et al.,* 1995)

One notes that the Black Sea region, especially in the south-east, is rich in mines and ores from these mines are processed in smelters situated along the south-eastern coast. These activities result in a large input of Pb-bearing material into the Sea.

Sediments, being reservoirs for most pollutants including heavy metals, have been used to indicate the magnitude and distribution of lead contamination of the aquatic environment. They have been the subject of previous investigations (LARSEN & JENSEN, 1989 ; JONES & TURKI, 1997).

Mussels have been recommended by several authors (PHILLIPS, 1976a,b ; ÜNSAL, 1978, 1982, 1984 ; ÜNSAL & BEŞIKTEPE, 1994) as indicator organisms for monitoring trace metal pollution because of their ability to filter large quantities of water and so accumulate large amounts of pollutants from the environment. SCHULZ-BALDES (1974) and PHILLIPS (1976 a, b, 1978) suggested the mussel, *Mytilus edulis,* being as an ideal indicator of lead in the marine environment. *Mytilus galloprovincialis,* which is the most abundant mussel species along the shore of the Black Sea (MULTU, 1990), was chosen for this study.

The present investigation aims to determine the magnitude, distribution and sources of lead pollution along the Turkish coast of the Black Sea.

Materials and Methods

Surface sediments were collected in 1993 from the south-eastern and in 1995 from the south-western Black Sea coast of Turkey with an Ekman grab sampler.

A total of 31 stations (14 sources and 4 controls in the east and 9 sources and 4 controls in the west) were sampled. The locations of these stations are shown in Fig.1 A & B. However, fine sediment could not be found at all stations. At stations situated in rivers, streams and industrial discharge channels, samples were taken from three locations: from a calm area, close to the point of discharge and from an offshore location situated 50-100 m from the discharge. Samples were collected seasonally as far as possible, they were sub-sampled immediately, placed in pre-cleaned, acid-washed plastic jars, taken to the laboratory and stored at -20 °C until analysis.

Whenever they were observed, mussels were also collected from the same stations as the surface sediments. Mussels were placed in plastic bags, taken to the laboratory and stored at - 20 °C until analysis. As the concentration of most heavy metals in mussels is influenced by their size (BOYDEN, 1977) efforts were made to collect mussels of uniform size (4-5 cm).

Sediments were wet-sieved through a 100μm mesh size plastic screen and the fraction <100 μm was taken for analysis since metals, including Pb, are mainly absorbed by small particles (SHIMKUS, 1991 ; YÜCESOY & ERGIN, 1992 ; PULS *et al.,* 1997). Approximately 1 g of the wet sediment taken for analysis was digested in a teflon cup with concentrated HNO_3 at 140 °C for 8 hours.

The soft flesh of the mussels was excised for analyses. At least three individuals taken from each sampling point were pooled to obtain a composite sample. Three 0.5 g sub-samples from each composite were taken and digested in the same way as sediments.

Total lead concentrations in sediments and mussels were determined by a GBC flameless Atomic Absorption Spectrophotometer (Model 902) according to the procedures described by UNEP/FAO/ IAEA/IOC (1984). Both International Standards from the CBR (Community Bureau of Reference) and blanks were included in each set of samples to check the precision and accuracy of the analyses. Because results in the literature are usually given on a dry weight basis, the dry weights of sediments and mussels from each station were determined. The dry weight/wet weight ratios averaged 0.729 and 0.725 for sediments from the south-eastern and south-western Black

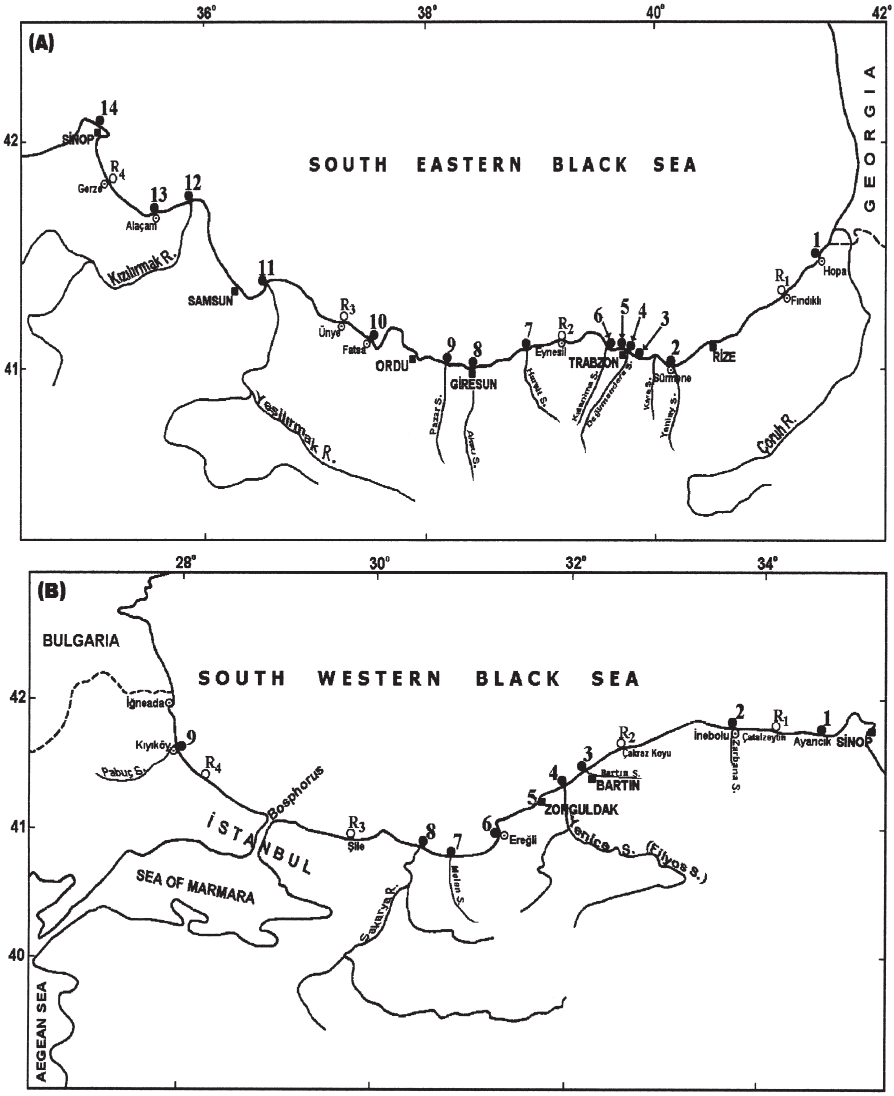

Fig. 1: Sampling stations in the south-eastern (A) and south-western (B) Black Sea (R: Reference stations).

Sea respectively and were 0.218 and 0.182 for mussels collected from the same regions.

Results

Sediments

The spatial distribution of total lead concentrations in surficial sediments from the south-eastern Black Sea is presented in Figure 2A. The values shown in this figure are the averages of four seasonal samplings at each station. Figure 2A shows a remarkable variation in lead contamination, minimum and maximum average lead concentrations of 3.06 and 617.8 μg g^{-1} wet weight being observed. Stations 1 and 2 located, along the easternmost coast of the southern Black Sea (Fig. 1A), were the most consistently polluted. All the samples (that is from source, inshore and offshore) from these two stations contained very high lead concentrations (310.7-565.5 μg g^{-1}). The lead concentrations in the samples, taken from Harşit, Aksu and Pazar streams around Giresun (Sts. 7, 8 and 9 respectively) and from their discharge points, ranged from 9.10 to 617.75 μg g^{-1} (Fig. 2A). These results demonstrate the contribution of terrigenous sources to the lead pollution of the south-eastern Black Sea.

Table 1 shows the concentration factors (CF) for the surface sediments of all sampling stations. These are calculated according to HORNUNG & KRUMGALZ (1984) as the ratio of the average metal concentration measured at a given station to the average control stations' value. The extremely high (up to 84.2) contamination factors obtained for each of the three sampling points at Sts. 1,2,7,8 and 9 (Table 1) confirm the high levels of the lead pollution at these stations and therefore in the eastern Black Sea.

In contrast to the high lead pollution observed in sediments from the south-east, the samples from the south-western coast contained very low average lead concentrations (Fig. 2B). The highest recorded average values, 20.8 and 42 μg g^{-1}, were observed in the samples offshore from Sts. 2 and 8 respectively. The highest CF obtained in this part of the Black Sea was 7.73 (Table 1), 10 times lower than the CFs found for the eastern part. Although the lead concentrations in the sediments sampled

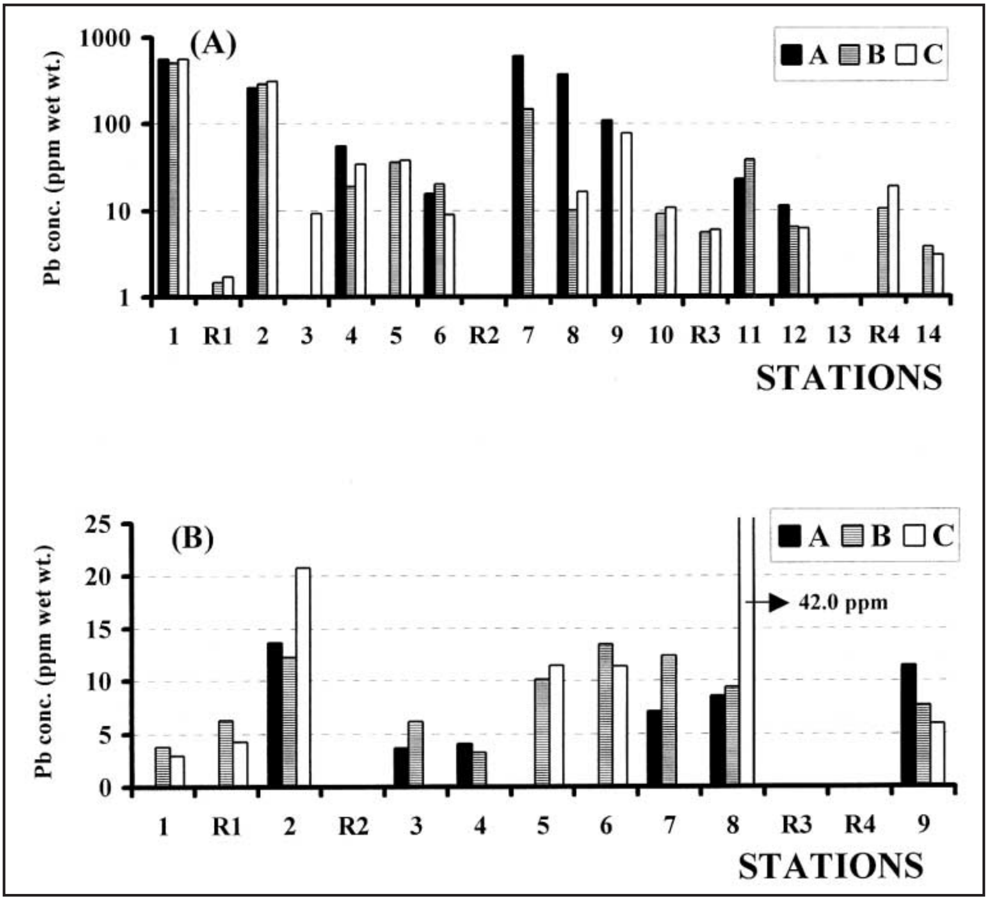

Fig. 2: Lead concentrations in sediments from the south-east (A) and south-west (B) of the Black Sea (Average of four sampling periods). A: Samples from rivers, streams and/or wastes ; B: Samples from discharge points or shore C: Samples from offshore ; R: Reference stations.

from the rivers, streams and effluents were moderately high at Sts. 2 and 8 (Fig. 2B), they were, however, much lower than concentrations found at the analogous source stations in the south-east. The lead pollution of the south-western Black Sea by these sources was, therefore, less pronounced compared to the eastern Black Sea.

With respect to seasonal variation, the peak value was obtained in October (363.2 μg g^{-1}) and the lowest one in April (28.5 μg g^{-1}) in the south-eastern Black Sea. The concentrations were also significantly higher in December and July compared to April but lower than those of October (Fig.3A).

In the south-west, the average peak lead concentrations were observed in September and December (11.91 and 10.73μg g^{-1} respectively). The concentrations in January (6.2 μg g^{-1}) and April (6.9 μg g^{-1}) were about half those observed in September (Fig. 3B)

Mussels

Lead concentrations in the mussels inhabiting 10 source and four reference stations along the south-eastern Black Sea coast are presented in Fig. 4. These concentrations are, as in the case of sediments, the average values over four sampling periods. Because the mussel species used in this study *(Mytilus galloprovincialis)* occurs only in marine environments, samples could be, and were, taken only from the coast and offshore. Stations 10 and 14, situated in Fatsa and Sinop respectively (Fig. 1A) had the highest Pb concentrations (both 1.7 μg g^{-1}). Mussels from other sources contained the same (or at times lower) lead concentrations as those at reference stations (0.04 to 0.3 μg g^{-1} ; Figure 4A). The mussels taken from coastal areas contained less lead than those from offshore stations except at St.14 where both coastal and offshore samples had elevated lead concentrations.

Figure 4B shows the average lead concentrations in the mussels sampled from different locations along the western Black Sea coast. With the exception of Stations 2 and 9, where lead concentrations were around 0.1 μg g^{-1}, mussels throughout the south-western coast possessed significantly higher lead contents (>10μg g^{-1} at Sts. 1, 3 and 5) than mussels from the south-east (1.7 μg g^{-1}). Elevated concentrations persisted all the year round in Zonguldak harbour. However the high average concentrations at the other stations resulted from exceptionally high values measured in one or two sampling periods. Samples from reference stations also possessed

Table 1
Concentration factors (CF) for the surface sediments from the south-eastern and south-western Black Sea.

S.E. Black Sea		S.W. Black Sea	
Stations	Concentration Factors	Stations	Concentration Factors
1A	76.72	1B	0.69
1B	68.87	1C	0.54
1C	77.04	2A	2.52
2A	35.99	2B	2.26
2B	40.00	2C	3.83
2C	42.33	3A	0.67
3C	1.27	3B	1.13
4A	7.59	4A	0.76
4B	2.59	4B	0.59
4C	4.75	5B	1.87
5C	5.26	6B	2.48
6A	2.14	6C	2.10
6B	2.74	7A	1.32
6C	1.23	7B	1.29
7A	84.16	8A	1.58
7B	20.11	8B	1.73
8A	51.58	8C	7.73
8B	1.37	9A	2.11
8C	2.25	9B	1.42
9A	14.90	9C	1.09
10B	1.24		
10C	1.48		
11A	3.13		
11B	5.28		
12A	1.54		
12B	0.88		
12C	0.88		
14B	0.51		
14C	0.42		

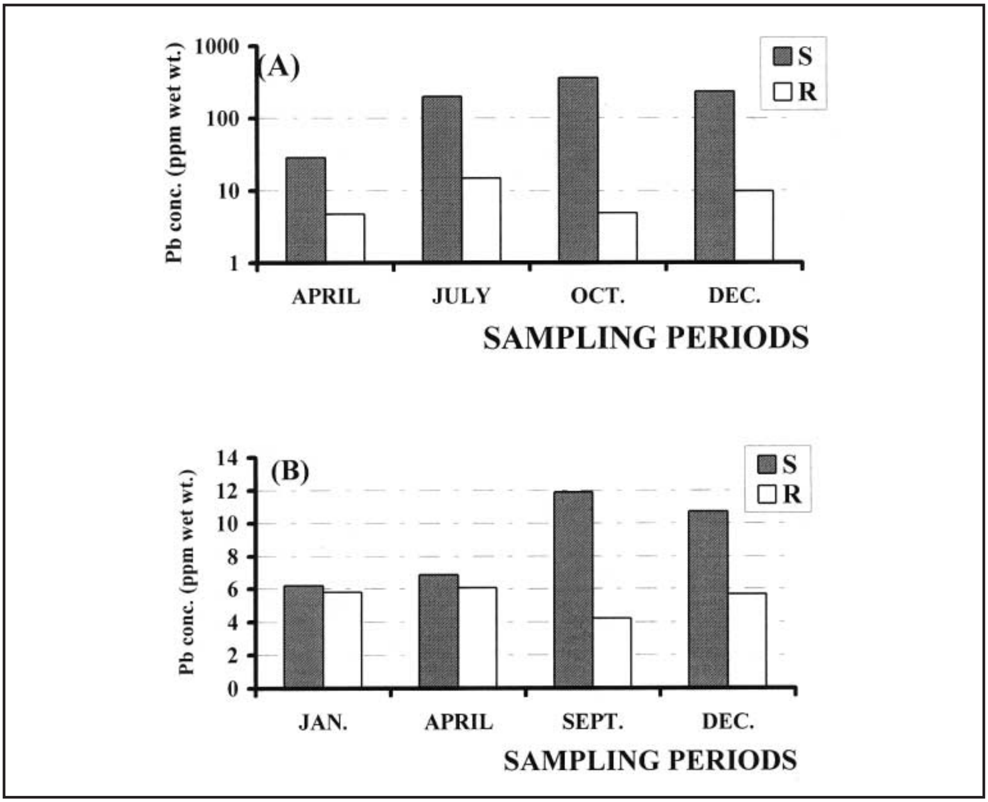

Fig. 3: Seasonal variations of lead concentrations in sediments from the south-eastern (A) and south-western (B) Black Sea (Average of all stations). (S: Source ; R: Reference stations).

quite high Pb concentrations (5.4 μg g^{-1}; Fig. 4B).

Average seasonal lead concentrations observed in the mussels sampled at all stations are shown in Figs. 5A and 5B. The seasonal variations in lead concentrations in mussels from the south-western and south-eastern Black Seas were different; in the south-east, the highest average values were observed in December (0.6 μg g^{-1}) and October (0.5 μg g^{-1}) whilst the lowest average occurred with 0.1 μg g^{-1} in July (Fig. 5A).

In April lead concentrations in mussels were low in those sampled from eastern coasts, but peaked in those from western coasts (0.2 μg g^{-1} in the east; 30.3 μg g^{-1} in the west) (Fig. 5B). In the south-west, the average concentration was also quite high (24.1μg g^{-1}) in mussels collected in January, they decreased, however, to 1.3 and 0.04 μg g^{-1} in December and September respectively (Fig. 5B).

Discussion

In the south-eastern coast of the Black Sea, stations located in Hopa and Sürmene were the most consistently polluted areas (Figs.1A and 2A). These two stations are situated at the discharge points of the Hopa and Sürmene Copper Industries. The samples taken from the industrial effluents contained very high lead concentrations, indicative of the industries' contribution to the lead pollution. Furthermore, the Çoruh River discharging into the Black Sea to the north of Hopa carries the wastes of the Murgul Copper Industry. Thus it appears that the local copper industries are causing serious lead pollution, and are

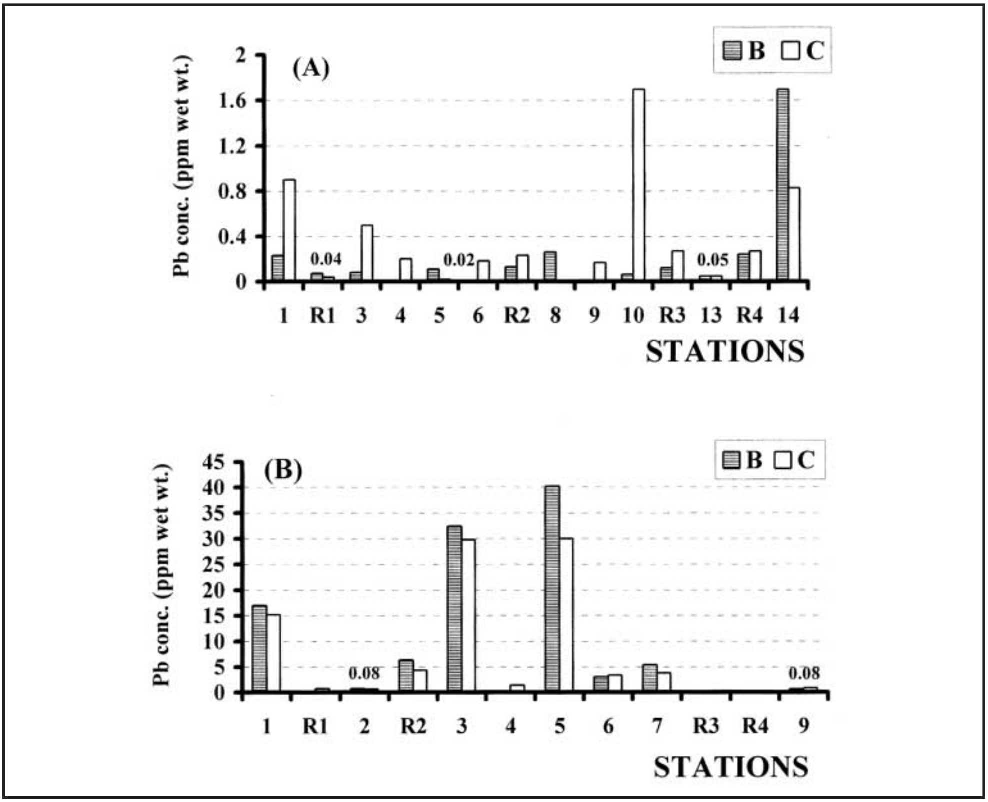

Fig. 4: Lead concentrations in mussels from the south-east (A) and south-west (B) of the Black Sea (Average of four sampling periods). B: Samples from discharge points or shore ; C: Samples from offshore R: Reference stations.

perhaps the largest source of lead contamination in the region. The role of the industry was discussed in the distribution of heavy metals (including lead) in sediments of the Venice lagoon where the lead concentrations changed between 21-929 $\mu g\ g^{-1}$ (FRIGNANI *et al.* 1999). Moreover, sediments from the south-eastern coast may well carry lead load transported both from the Rim current and the Caucasian coast by semi-permanent currents and/or eddies. Elevated lead concentrations, found at stations around Giresun, almost certainly originated from the local lead mines and smelters. The data obtained from these stations agree well with those reported by YÜCESOY & ERGİN (1992) who also found the highest lead concentrations to be present in sediments from this region. The Kızılırmak and Yeşilırmak rivers are also potential regional sources of lead pollution, the input through these rivers being found to be 8500 and 5800 kg y^{-1} respectively (TUNCEL *et al.,* 1993).

In the south-western Black Sea, the Pb levels in sediments were, with the exception of St. 2, considerably lower compared to the south-east (Fig. 2B). Station 2, where the highest concentrations were observed, is located in the mouth of Zarbana stream, which receives wastes from a copper mine in Küre which are probably the main source of the lead in the sediments. Part of the lead concentrations observed in sediments in Zonguldak and to its west may have originated in dust from Istanbul. KUBILAY *et al.* (1995) found the average lead concentration in the airborne material collected at and around the city of Istanbul to

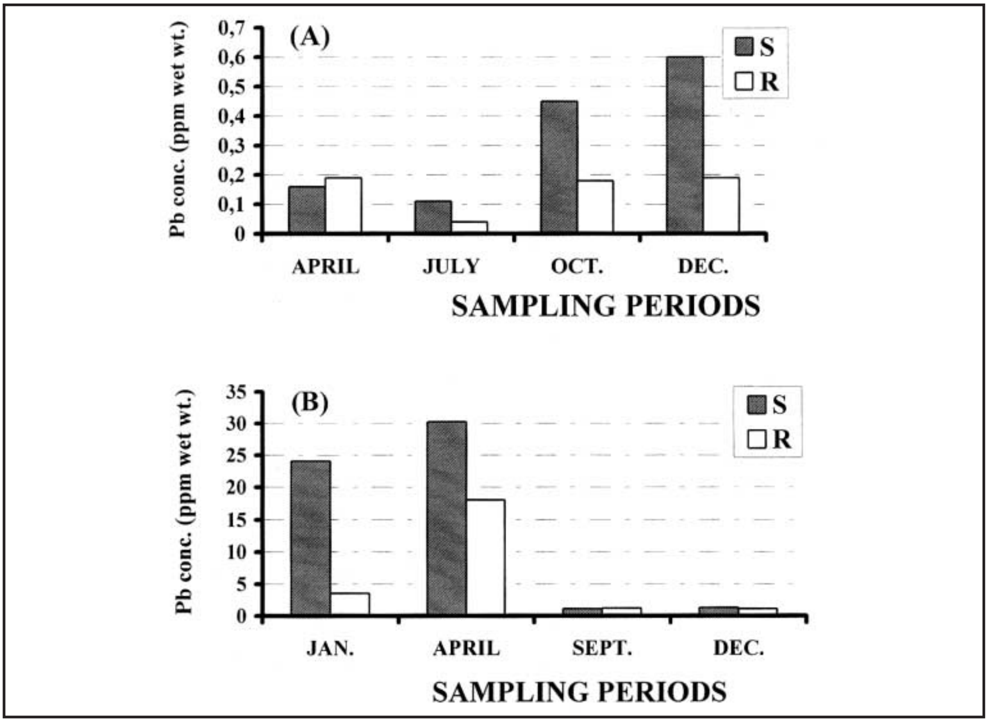

Fig. 5: Seasonal variations of lead concentrations in mussels from the south-eastern (A) and south-western (B) Black Sea (Average of all stations). (S: Source ; R: Reference stations).

be 270 ng m^{-3} compared with 19 ng m-3 generally observed over the Black Sea. Similarly, HACISALIHOĞLU *et al.* (1992) estimated the total input of atmospheric lead to the south-western and south-eastern Black Sea to be 2400 and 1500 tons per year respectively.

A notable amount of lead must have been transported in particulate form or on the particles contained in rivers' and/or stream's waters. HARALDSSON & WESTERLUND (1988) reported the percentage of particulate lead in waters of the Black Sea to be 42%. TUNCEL *et al.* (1993) calculated the total lead flux from the rivers and streams in the region to be 21603 kg y^{-1}. Rivers and streams are, therefore, the main sources of lead in the western Black Sea coast of Turkey as was indicated also by UJEVIC *et al.* (1999) for the Adriatic Sea.

The station, located in the extreme west of the region (St. 9), has been affected by both the Pabuç stream and to a large extent by the Danube river. Significantly high lead concentrations were measured in sediments from this stream. The contribution of the Danube river to the lead pollution in the Black Sea has been studied by several authors (ZAITSEV, 1992 ; KUBILAY *et al.,* 1995 ; TUNCER *et al.,* 1998).

The highest average lead concentrations in sediments from the south-eastern Black Sea were measured in October, followed by December and July (Fig. 3C). The flows of the main rivers, Kızılırmak and Yeşilırmak and streams decreased from July to December (ÜNSAL *et al.,* 1995). Suspended particles carried by these sources and containing lead absorbed on their surface, therefore settled down to the bottom and generated high concentrations in the sediments.

In the south-western Black Sea, the highest annual average was observed in September, followed by December (Fig. 3D). In this region, the rainfall and the river flow are at their lowest levels in July and August (ÜNSAL *et al.,* 1997). During this calm period, particles containing lead, were, therefore, deposited on the bottom and contributed to the high concentrations observed in September. On the other hand, the September maxima could be attributed to phytoplankton. PULS *et al.,* (1997) suggested phytoplankton transfers 21 tons of Pb per year to the sediments of the German Bight. Usually two phytoplankton blooms occur in the Black Sea; in Spring and Autumn (SOROKIN, 1983). However unusual temporal variations have been observed during the last decade (YILMAZ *et al.,* 1998).

The highest lead concentrations in mussels from the south-eastern Black Sea were found at Sts. 10 and 14 (Fig.4A) although sediments from the same stations possessed low concentrations of lead (Fig 2A). There was no correlation between lead concentrations in sediments and in mussels at these stations. This is consistent with previous studies (HORNUNG, 1989; HORNUNG *et al.,* 1989) that have shown no relationship between the lead concentrations in mollusc bivalvia, gastropods and in sediments. However, at St. 1, high concentrations in mussels and sediments coincided and it is assumed that the high lead content of mussels at this station is related not only to the high concentrations in sediments but also to other sources such as phytoplankton.

The mussels from the south-western Black Sea contained more Pb than those from the south-east (Fig 4A and 4B). The highest concentrations were observed in mussels collected from Sts. 1, 3 and 5. The station situated in Zonguldak harbour (St. 5) was found to be the most polluted. As was indicated above, heavy boat traffic occurs in this harbour. FOWLER & OREGIONI (1976) explained the extremely high levels of lead in mussels from TOULON & MARSEILLE (France) as a consequence of the local pollution due to the boat traffic and industry in these ports. Lead concentrations in mussels increased towards the west. As was suggested for sediments, lead pollution in the south-west of the Black Sea originated not only from local sources but also from the west e.g, the Danube river. It is emphasised that about 75% of pollutants entering the Black Sea are contributed by this river (VORONTSOV & KOLTSOV, 1991). The annual load of Pb from the Danube river is 4500 tons (ZAITSEV, 1992; TUNCER *et al.,* 1998).

The highest lead concentrations in mussels along the south-eastern coast were found in December, followed by October (Fig. 5A), whilst in the south-west, the highest concentrations were observed in April, followed by January (Fig. 5B). The parameters governing the seasonal uptake of metals by mussels appear to include spawning, body weight, salinity and water temperature (PHILLIPS, 1976a, 1978; SIMSPON, 1979; OREN *et al.,* 1980). Many workers have observed a decrease in body-weight and an increase in metal concentration after spawning (PHILLIPS, 1976a; SIMSPON, 1979; PHILLIPS, 1980). In the present study, the increase observed in the average lead concentrations in October and December coincided with an increase in the weight: size ratio. A similar observation was recorded by BRYAN & UYSAL (1978) who found an increase in lead concentration with increasing dry weight of whole soft body parts of the bivalve, *Scrobicularia plana,* from the Tamar Estuary in England.

PHILLIPS (1978) reported concentrations of lead in mussels to be highest in mussels taken from low-salinity waters on the Scandinavian coasts. The Black Sea receives fresh water from heavy rainfall, from major rivers such as the Danube, Sakarya, Filyos, Kızılırmak, Yeşilırmak and to some extent the Çoruh and also from several small and large streams occurring along the Turkish coast. These inputs obviously result in fluctuations in salinity, which may be expected to differ along the south-east and south-west coasts (ÜNSAL *et al.,* 1995). Therefore, peaks in lead concentrations may appear in different months of the year in mussels from these two regions of the Black Sea (Figs. 4A and 4B).

The annual variation of temperature is also different in the south-east and south-western Black Sea (ÜNSAL *et al.,* 1997). BRYAN (1973) suggested the effect of temperature on the rate of excretion by mussels might produce seasonal changes.

Phytoplankton abundance has been suggested as another factor, which influences the seasonality of metals in mussels. According to BRYAN (1973) the period of highest phytoplankton productivity is correlated with low metal concentrations in scallop species. Conceivably, (i) greater availability of food increases the metabolic rate of the animal and increases the excretion of waste products; (ii) during times of high productivity, the amount of metal per phytoplankton cell may be lower than at other times since the amount available in the water is limited. As a result, the amounts of metal available from the food may be lower than at less productive times.

Conclusions

Sediments taken from rivers, streams and effluents along the south-eastern Black Sea coast contained high lead (up to 617.8 $\mu g\ g^{-1}$) concentrations. These concentrations were often higher than those measured immediately downstream on the shore and in the sea.

Two stations in Hopa and Sürmene and three stations around Giresun were found to be the most polluted, the highest lead concentrations were observed in autumn and early winter.

Mussels collected from the south-eastern coastal waters contained comparatively low concentrations of lead (0.01-1.70 $\mu g\ g^{-1}$) confirming the lead pollution in this region to be in the form of particles which either fail to reach the mussels or are not assimilated.

Despite the significant sources of lead documented along the west and north-west coasts of the Black Sea, low concentrations of lead (2.9-42.0 $\mu g\ g^{-1}$) were generally observed in sediments from the south-western coast. Significant concentrations of lead (0.08-40.2 $\mu g\ g^{-1}$) were observed, however, in mussels inhabiting south-western coastal waters. These concentrations were higher than those observed in similar mussels from the south-east.

The highest seasonal averages were obtained in October and December for sediments and mussels from the south-east and in September and April for those from the south-west.

The low levels in sediments of the south-western Black Sea lead to the suggestion that, significant amounts of this metal, adsorbed on suspended particles and/or contained in phytoplankton (also in zooplankton), are transported by surface currents to the offshore and/or to the Marmara and Aegean Seas.

Acknowledgments

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