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A Preliminary Study on the Metal Content of Mussels, *Mytilus galloprovincialis* (Lmk.) in the Eastern Black Sea

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Abstract: The seasonal variation in the mercury, copper and lead concentrations of mussels, *Mytilus galloprovincialis* inhabiting the eastern Black Sea coast of Turkey has been studied.

The distribution of mercury concentrations between the sampling stations was more or less homogenous, while copper and lead concentrations decreased in the middle but increased in the eastern and western parts of the studied region.

The averages of four sets of samples obtained at different periods indicated the highest Hg concentrations to be present in Samsun. Similarly the highest Cu concentrations were measured in mussels from Rize and Hopa and the highest Pb concentration, in mussels from Sinop and Samsun.

Regardless of the sampling locations, the highest average Hg and Pb concentrations were obtained in June, and the highest Cu concentrations, in December. A significant difference in concentrations was observed between sampling periods.

Key Words: Mussel; Metal content; Eastern Black Sea

Doğu Karadeniz'de Midyelerin, *Mytilus galloprovincialis*, (Lmk.) Metal Miktarı Üzerine Bir Araştırma

Özet: Türkiye'nin Doğu Karadeniz kıyısında yaşayan midyelerde, *Mytilus galloprovincialis*, civa, bakır ve kurşun konsantrasyonlarının mevsimsel değişimi incelendi.

Adı geçen bölgede civa konsantrasyonları, örnekleme istasyonları arasında az çok homojen olarak dağılmasına karşın, bakır ve kurşun konsantrasyonları orta kısımda azalmış, doğu ve batı kısımda ise artmıştır. Dört ayrı zamanda alınan örneklerin ortalamasına göre en yüksek Hg konsantrasyonu Samsun bölgesinde ölçülmüştür. Yine aynı ortalamalara göre en yüksek Cu konsantrasyonu Rize bölgesinde, kurşun konsantrasyonu ise Sinop ve Samsun bölgesinde bulunmuştur.

Örnekleme bölgeleri dikkate alınmadan yapılan değerlendirmeye göre ise ortalama en yüksek Hg ve Pb konsantrasyonları Haziran ayında, Cu konsantrasyonu ise Aralık ayında ölçülmüştür. Örnekleme zamanlarına göre konsantrasyonlar arasında önemli farklar gözlenmiştir.

Anahtar Kelimeler: Midye; Metal miktarı; Doğu Karadeniz.

Introduction

The eastern Black Sea coast of Turkey between longitudes 35° 00" and 41° 45", ise more populated and therefore more industrialized than the western coast. The number of major Turkish industries in the Black Sea region amounts to 236 (1) and most of these are situated along the eastern coast. This region is also rich in ore deposits, especially in copper and lead ores (2). On the other hand, it has been suggested that the fresh water discharged along the eastern coast by three major rivers, Kızılırmak, Yeşilırmak and Çoruh, contributes to the pollution of the sea.

Because of their capacity to accumulate metals from the environment, bivalves have been widely used as qualitative biological indicators of metal pollution (3-6). Of the bivalves, mussels now receive considerable attention as subjects of pollution studies. They have been proposed as suitable indicators for monitoring trace metal pollution in the marine environment (7-14).

The genus *Mytilus* offers most of the requisite features of a bioindicator: world-wide distribution, non-migrant species with long survival, reasonable size, easy sampling, abundance, tolerance to brackish water, and the ability to concentrate numerous contaminants (5, 15-18).

Mytilus galloprovincialis is abundant along the Black Sea coast of Turkey (19). Although this species is widely used for monitoring trace metal pollution in the Mediterranean Sea (15, 20-23), there have been few studies of its metal content in the Black Sea environment (24-26).

The aim of this study is to evaluate the degree of metal contamination of the eastern Black Sea using the indicator species, *Mytilus galloprovincialis*.

Materials and Methods

Mussels, *M. galloprovincialis*, were collected from six stations (Figure 1) along the eastern Black Sea coast in June, August, October and December of 1991 according to the procedure described in UNEP/FAO/IAEA, (27).

As the concentration of most heavy metals in mussels is influenced by the size of these organisms (28-29), efforts have been made to collect mussels of the same size (4.7±0.8 cm). After collection, all samples were put into plastic bags and frozen at -20 °C until analysis. Frozen samples were first thawed, removed from their shells and rinsed with distilled water. The soft parts of at least three mussels were digested in a teflon cup with concentrated nitric acid using a stainless stell block. After digestion, mercury was analyzed by cold vapor Atomic Absorption Spectrophotometry (Varian model AA-6) according to the method described in UNEP/FAO/IAEA/IOC, (30). Copper and lead were analyzed by a GBC Atomic Absorption Spectrophotometer (Model 902). International standards from CBR (Community Bureau of Reference) and blanks were included in each set of samples to check the precision and accuracy of the analysis.

Results

Figures 2a, b and c show the variations of mercury, copper and lead in the soft parts of *M. galloprovincialis* collected in June, August, October and December from the six sampling areas.

Mercury

Mercury concentrations fluctuated both with location and with season (Figure 2a). The average mercury concentrations in the soft parts of mussels from the Sinop location ranged from 6 to 20 ng g^{-1} although the mussels were always roughly the same size. The high values were obtained in June and October and the low values in August and December. The mercury content of the samples taken at Samsun station showed a decrease from 170 ng g^{-1} in June to 10 ng g^{-1} in December (Figure 2a). This value of 170 ng g^{-1} was the highest mercury concentration obtained throughout the sampling period. At the Trabzon station, mercury concentrations in the mussels remained at nearly the same level (~20 ng g^{-1}) throughout the sampling periods except for in December when a level of 46 ng g^{-1} was measured. A similar pattern to Trabzon was observed in the seasonal variation of the mercury contents of mussels from Sürmene (Figure 2a); concentrations ranged from 10 ng g^{-1} in June to 25 ng g^{-1} in December. Whilst the mussels sampled in October and December from Rize contained quite low concentrations, mercury could not be detected from those collected in June. At the easternmost station, Hopa, the mercury concentrations showed fluctuations during sampling periods, the lowest value (2.67 ng g^{-1}) being obtained in August and the highest value (43.67 ng g^{-1}), in December.

Copper

Of the three metals studied, copper had the highest values in the soft part of *M. galloprovincialis* in most sampling periods. The concentrations increased towards the eastern part of he studied region (Figure 2b). Seasonal fluctuations were observed in the copper concentrations of mussels from Sinop and Samsun. At Sinop station, the highest value of 3.18 µg g^{-1} was obtained in August, followed by 2.17 and 1.12 µg g^{-1} in December and June, respectively. The lowest concentration of the four sampling periods was found to be 0.16 µg g^{-1} in October. In Samsun, despite the very low copper concentration (0.09 µg g^{-1}) found in October, for other sampling periods concentrations varied between 1.79 and 2.77 µg g^{-1}. Compared with the other sampling locations, relatively low concentrations were measured in Trabzon. Copper concentrations in Sürmene samples also fluctuated between 0.12 µg g^{-1} (in October) and 3.39 µg g^{-1} (in August). The mean copper concentrations measured in Sürmene were higher than those found in Trabzon.

The highest copper concentration and the highest seasonal variations were both observed in mussels from Rize. The concentrations varied between 0.25 and 13.33 µg g^{-1}. At Hopa station, which is the easternmost station of the studied area, copper values showed a step-wise increase from June to December (Figure 2b). Following Rize samples, the highest copper concentrations were obtained from this location.

Lead

In contrast to copper, the lead content of the mussels decreased towards the eastern part of the studied region until Hopa station where an increase in lead concentration was observed (Figure 2c). Samples taken from the Sinop area in June contained the highest lead concentrations (2.58 µg g^{-1}) compared with all other sampling areas and sampling periods. At Samsun station, considerable levels were also obtained in June and August (1.37 and 1.32 µg g^{-1}, respectively) while those measured in October and December were found to be less than half these values. Despite the high lead accumulations encountered in Sinop and Samsun, relatively low concentrations were obtained in samples from Trabzon, Sürmene and Rize. Seasonal variations occurred in the lead content of mussels from Hopa.

Discussion

The difference in metal concentrations observed in sampling locations is obviously due to an increase and/or decrease in the input of pollutants from different sources. In addition, the fluctuations observed during the sampling periods resulted both from exogenous and endogenous factors (7, 31-32). The average mercury concentrations in the soft parts of mussels from the Sinop location changed from 6 to 20 ng g^{-1}, although the mussels were more or less of the same size. Phillips (7) studied the net uptake of zinc, cadmium, lead and copper by *Mytilus edulis* and found that in mussels of similar size (by shell length), the wet weight varied with season and that concentrations of metals were inversely proportional to these weight variations. According to this author, seasonal weight variations are related to the annual reproductive cycle of the mussels. Likewise, Simpson (33) pointed out that seasonal differences in heavy metal concentrations in samples of *Mytilus edulis* and other marine invertebrates may simply be a function of the seasonality of the reproductive cycle. The results obtained by Feyzioğlu (34) showed June and October to be pre-spawning preiods for *M. galloprovincialis* on the south-eastern Black Sea coast. This may be a reason for the high mercury concentrations during these sampling periods. The value of 170 ng g^{-1} obtained in Samsun samples was the highest mercury concentration obtained throughout the whole sampling period. This concentration probably resulted from freshwater input. According to meteorological data (35), the rainfall in the Samsun region reached its highest level in May and June and the flows of the Kızılırmak and Yeşilırmak rivers which discharge into the sea on both sides of Samsun city (Figure 1) also reached their maximum in May and June (36). On the other hand, some authors (37-38) have indicated that rivers are the most important sources of mercury in the Mediterranean. Similarly Amiard et al., (18) have pointed out that changes in land drainage leading to changes of metal inputs into coastal areas may result in fluctuations in the metal concentrations in living organisms. Salinity changes caused by these rivers may also be a reason for changes in the mercury concentrations of mussels, as has been observed for the other meatls (7). The highest mercury concentration obtained in

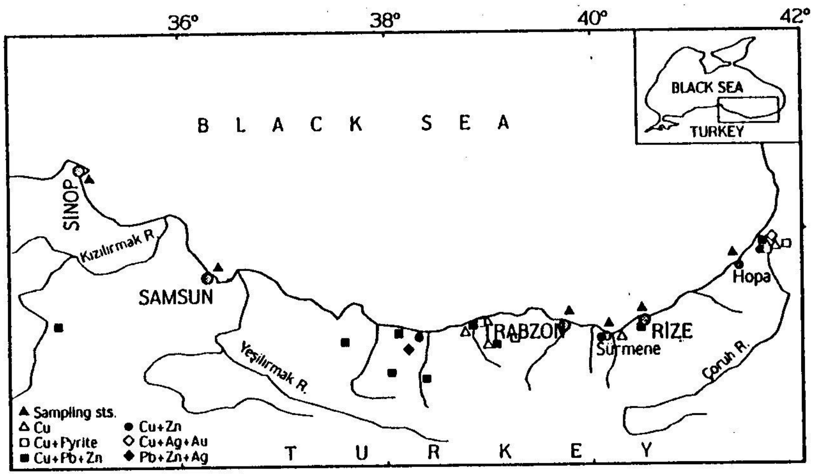

Figure 1. Sampling Stations and Localities of Economic Mineral Deposits (modified from Yücesoy & Ergin, 1992)

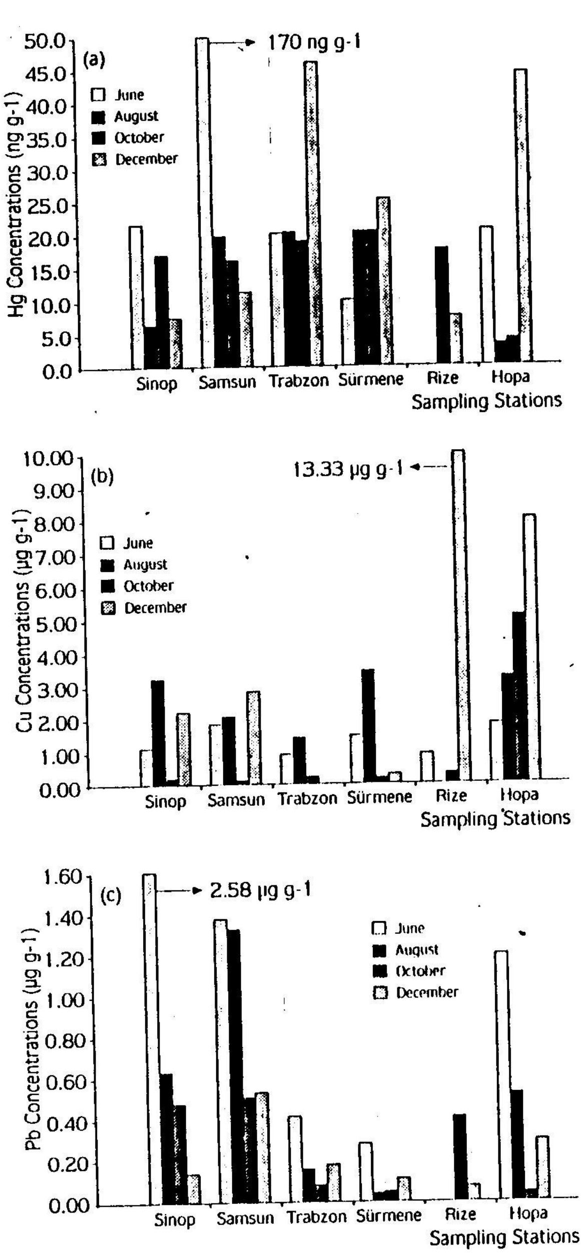

Figure 2. Variations in the Concentrations of Mercury (a), copper (b) and Lead (c) in the Soft parts of *M. galloprovincialis* with Space and Time.

June could also have been caused by a sudden pulse of mercury entering the sampling area at the time of sampling. Leonzio et al., (1981, cf. 39) have observed great variation in mercury concentration of *M. galloprovincialis* within a distance of 92 m from a source of mercury. The steady levels of mercury, observed at Trabzon station in June, August and October lead to the suggestion that a constant input of mercury from industrial and urban sources occurs in this area during most of the year. The high concentration measured in December may be due either to high rainfall in October and November or to low phytoplankton abundance at this location (34, 40) since higher phytoplankton productivity has been associated with low metal concentrations (41). The possible reason for the similarity in the temporal variation of the mercury contents of mussels between Trabzon and Sürmene is that these two cities are close to each other and have the same climatic conditions; however Trabzon is more populated; and industrialized than Sürmene. Accordingly, the slightly higher values obtained from Trabzon mussels presumably result from Trabzon's larger population. At the Hopa station, the lowest value was obtained in August and the highest value in December. With the exception of April, the annual rainfall is always high in this region compared with the western stations of the studied area. Both June and December follow the rainy spring and autumn seasons, respectively. Thus a direct run-off of mercury could be expected due to drainage especially from agricultural areas since this metal is also found in pesticides (42).

The results discussed above show that the highest mercury concentrations were found in late spring or early summer at the two western stations, Sinop and Samsun, while the highest concentrations were observed in late autumn or early winter at the eastern stations of the study area. Omitting the mercury concentration in June obtained from Samsun, the average mercury concentrations in *Mytilus galloprovincialis* collected from six locations throughout the sampling period were lower than or similar to those found by various other authors (39, 43) in different parts of the Mediterranean. The highest mercury value acquired on one occasion only in the Samsun location is nearly at the same level as those values measured in polluted areas of the Tyrrhenian Sea (43).

With the exception of the Hopa location, copper concentrations are similar to those found in *M. galloprovincialis* from the Gulf of La Spezia, Italy (44), Saronicos Gulf, Liguraian and Adriatic Seas (43) and in *M. edulis* from Port Phillip Bay, Australia (8). Because the copper concentrations obtained from Black Sea mussels (25-26) are given on a dry weight basis, no comparison could be made. The concentrations increased towards the eastern part of the studied region (Figure 2b). Seasonal fluctuations were observed in the copper concentrations of mussels from Sinop and Samsun. The reasons for these inconsistancies are not easy to explain. Phillips (7) has suggested that such seasonal fluctuations may be due to the variation in the wet mass of mussels with season. Such fluctuations may be related to phytoplankton productivity and availability of food (41), and to the reproductive cycle of the mussels (33). Unfortunately, detailed studies of neither phytoplankton productivity along the southeastern Black Sea coasts nor of the reproductive cycle of *M. galloprovincialis* in the study area yet exist. A contribution to the copper pollution could also be expected from the copper refinery in Samsun. Relatively low concentrations were measured in Trabzon and so the mean copper concentrations obtained in Sürmene were higher than those found in Trabzon although these two cities are close to each other. There is a copper refinery and some copper mines around Sürmene city (Figure 1), and the higher values probably resulted from the drainage of these enterprises.

The highest copper concentration was measured in Rize. This enormous peak observed in December presumably originated from an instantaneous or accidental run-off of copper-containing effluents from the Çayeli copper refinery and also from copper mines which are located all around this city. Fowler and Oregioni (15) have suggested that peak metal concentrations result from high precipitation and increased run-off bringing particulate metals to the sea. Following the Rize samples, the next highest copper concentrations were obtained from the Hopa location. This region is very rich in copper mines (Figure 1). The metal from these mines is processed in copper refineries occurring in the same area. They obviously constitute the main sources resposible for the high copper concentrations in the mussels from this location. Inputs from the Çoruh river and from some small creeks in this area are additional reasons for the elevated copper concentrations. Yücesoy and Ergin (45) studied the heavy-metal geochemistry of surface sediments from the southern Black Sea shelf and upper slope and found the highest copper content in the sediments from the Hopa region.

The high lead content of mussels from the Sinop area remained within the same ranges as those found in *Mytilus galloprovincialis* from the Adriatic Sea (43). The Sinop area is far from the impact of any rivers. However, in 1991 when the samples were collected, the rainfall maintained a high level throughout the winter and spring seasons (35). It is likely, therefore, that the very high concentrations obtained in June originated from industrial and/or domestic effluents which were discharged deliberately or carried by torrents of the ground water into the sea. Phillips (16) studied the lead, iron and manganese content of *M. edulis* in Scandinavian waters and has suggested that the majority of lead entering the coastal waters of Sweden is derived from storm-water. Likewise, Chow et al. (9) obtained a correlation between lead content of *Mytilus* and human activity along the coastal zone. In addition, used lead batteries were occasionally encountered on the Sinop coast. A contribution to pollution should also be expected from these materials. Additional metal inputs would also be derived from natural geological sources since zinc and lead are commonly found associated with economically important deposits on land (Gümüş, 1979, cf. 45). For example, sediments from the southeastern Black Sea showed significant lead contribution from the mineral deposits and mining activities which are genetically related to the volcanic massive Zn-Pb-Cu sulfides formed in the Cretaceous to Tertiary times (45). The high lead concentrations obtained in June and August in the Samsun samples were found to be less than or at the same level as those reported for mussels from different parts of the Mediterranean Sea (43). As was mentioned previously, this area receives very important freshwater input from the Kızılırmak and Yeşilırmak rivers, draining a large area where some important lead deposits occur. Seasonal variations in the metal content of mussels can be attributed to the flow regime of these rivers. Amiard et al. (18) have suggested that changes in land drainage leading to changes in the metal inputs into coastal areas may result in fluctuations in the metal concentrations occurring in living organisms. Furthermore, Samsun is the most populated city of the eastern Black Sea region and has a heavy traffic flow both in the city and in the harbour. Leaded petrol could be another explanation for the high lead levels in the sea water and thus in mussels from this location. Despite the high lead concentrations encountered in Sinop and Samsun, relatively low concentrations were obtained in samples from Trabzon, Sürmene and Rize. The factors controlling the availability of sediment-bound lead to the estuarine bivalve, *Scobicularia plana*, were studied and considerable variations in lead levels between estuaries were observed (4). It was suggested that one of the reasons for these variations is the difference between the characteristics of the sediments such as size, organic carbon content etc. The relationships were observed between the clay, silt, sand, $CaCO_3$ and organic carbon in the sediments and their heavy metal concentrations (45). The slightly high lead concentration obtained from the Hopa samples in June resulted presumably from freshwater runoff throughout the winter and spring periods. Elevated concentrations obtained in August and December may be of industrial and/or urban origin.

Conclusion

The following conclusions can be deduced from the results discussed above:

- Regardless of sampling location, the highest average Hg, Cu and Pb concentrations were obtained in June, December and again in June, respectively.

- With the exception of quite high mercury and copper concentrations obtained in the Samsun and Rize locations, respectively, the highest average mercury value was recorded in Trabzon samples and the highest coper concentration was found in the Hopa location. Both Sinop and Samsun samples contained the highest lead concentrations.

- The concentrations of the three metals measured in mussels throughout the sampling period were below the maximum permissible levels accepted by different countries (39, 46).

- The distribution of mercury concentrations among sampling stations was more or less homogenous, while the distribution of copper and lead decreased in the central but increased in the eastern and western parts of the study region.

- Several investigators have observed a significant correlation between heavy metal concentrations in mussels and phytoplankton productivity (41) in the surrounding region and the reproductive cycle of the mussels (7, 33). Unfortunately, no detailed study has yet been carried out either on the biology of *M. galloprovincialis* or on phytoplankton production in the study area.

- The difference in concentrations between sampling locations was insignificant, while a significant difference ($P<0.01$) was observed between the sampling periods.

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