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Heavy Metal Concentrations in Ten Species of Fishes Caught in Sinop Coastal Waters of the Black Sea, Turkey

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Abstract

In the present study, the heavy metal concentrations (zinc, copper, cadmium and lead) have been measured in dorsal muscle tissue of ten species (*Trachurus mediterraneus*, *Sprattus sprattus sprattus*, *Mullus surmelatus*, *Sarda sarda*, *Mugil cephalus*, *Scorpaena porcus*, *Sparus aurata, Umbrina cirrosa*, *Spicara maena* and *Solea solea*) from Sinop coast of the Black Sea, Turkey during fish season in 2010. Significant differences in metal concentrations were found between the species (P<0.05). In general, it was found that the levels of heavy metals studied were lower than the maximum permissible limit of the food regulations of the Ministry of Agriculture, Fisheries and Food (MAFF), the Turkish Food Codex and Commission Regulation (EC). Cd showed the lowest concentrations in all fish samples followed by Pb. The results of the present study were compared with the other studies and discussed.

Keywords: Black Sea, fishes, zinc, copper, cadmium, lead.

Türkiye' de, Karadeniz'in Sinop Kıyılarında On Farklı Balık Türünde Ağır Metal Konsantrasyonları

Özet

Mevcut bu çalışmada 2010 yılı avcılık sezonunda Karadeniz'in Sinop kıyılarından örneklenen on balık türünün (*Trachurus mediterraneus*, *Sprattus sprattus sprattus*, *Mullus surmelatus*, *Sarda sarda*, *Mugil cephalus*, *Scorpaena porcus*, *Sparus aurata*, *Umbrina cirrosa*, *Spicara maena* ve *Solea solea*) sırt kas dokularındaki ağır metal (çinko, bakır, kurşun ve kadmiyum) konsantrasyonları belirlenmiştir. Metal konsantrasyonlarında türler arası önemli farklılıklar bulunmuştur (P<0,05). Genel olarak çalışılan ağır metal düzeyleri Tarım, Balıkçılık ve Gıda Bakanlığı (MAFF), Türk Gıda Kodeksi Tebliği, Avrupa Birliği Komisyon Tüzüğünce belirlenen gıda maddelerindeki bulaşanların maksimum limitlerinden daha düşük bulunmuştur. Kadmiyum tüm balık örneklerinde en düşük değerde bulunurken bunu kurşun izlemiştir. Çalışma sonuçları diğer çalışmalarla karşılaştırılarak tartışılmıştır.

Anahtar Kelimeler: Karadeniz, balık, çinko, bakır, kurşun, kadmiyum.

Introduction

The contamination of the Black Sea waters, sediments and organisms with a wide range of pollutants has become a matter of great concern over the last few decades (Secrieru and Secrieru, 2002; Ergül *et al*., 2008; Bat *et al*., 2009; Boran and Altınok, 2010). Heavy metals are natural trace components of the marine environment, but their levels have increased due to domestic, industrial, mining and agricultural activities (Bakan and Büyükgüngör, 2000; Altas and Büyükgüngör, 2007). Discharge of heavy metals into river or any marine environment can change both marine species diversity and ecosystems, due to their toxicity and accumulative behavior (Bat, 2005; Bakan and Böke Özkoç, 2007; Bat *et al*., 2009). Marine organisms such as fish accumulate heavy metals to concentrations many times higher than present in water or sediment (Bryan, 1976; Phillips and Rainbow, 1994; Bat *et al*., 2009; Boran and Altınok, 2010). Thus, heavy metals acquired through the food chain as a result of pollution are potential chemical hazards, threatening consumers. At low levels, some heavy metals such as copper and zinc are essential for enzymatic activity and many biological processes. However, some heavy metals, such as cadmium and lead which may be introduced into the aquatic environment from anthropogenic activities have no known essential role in living organisms, and are toxic at even low concentrations. The essential metals also become toxic at high concentrations (Bryan, 1976). Once in the marine environment these heavy metals can be concentrated in fish tissues.

Thus the aim of the present study is to:

1. Determine and compare the concentrations of Zn, Cu, Pb and Cd, in edible parts (dorsal muscle tissue) of ten selected common coastal Black Sea fish species,

2. Describe differences of the concentrations of these heavy metals between species,

3. Compare with the guidelines set down by the Ministry of Agriculture, Fisheries and Food (MAFF), the Turkish Food Codex, Commission Regulation (EC) for the safe consumption limits of fish and the other studies.

Materials and Methods

Most of the fishing industry in Turkey is based on the Black Sea waters. A total of 399656 tons of sea fish were landed by Turkish fish vessels in 2010. The amount of fishing carried out in 2010 in the Black Sea consisted of 76% of the total Turkish catch (Turkish Statistical Institute, 2011). Catches of fish from Turkish waters in 2010 are given in Table 1 (Turkish Statistical Institute, 2011). These data show that these species of fish are economically important for human consumption.

The fishes were sampled in 2010, by trap-nets, gill-nets, bottom trawl, long line fishing, hand line fishing and were purchased from the fish market at Sinop port during fishing season from the Black Sea. Figure 1 shows fish sampling area.

Selected ten fish species are; Mediterranean horse mackerel (*Trachurus mediterraneus* Steindachner, 1868), sprat (*Sprattus sprattus sprattus* Linnaeus, 1758), stripped mullet (*Mullus surmelatus* Linnaeus, 1758), Atlantic bonito (*Sarda sarda* Bloch, 1793), flathead mullet (*Mugil cephalus* Linnaeus, 1758), black scorpion fish (*Scorpaena porcus* Linnaeus, 1758), sea bream (*Sparus aurata* Linnaeus, 1758), Shi drum (croaker) (*Umbrina cirrosa* Linnaeus, 1758), picarel (*Spicara maena* Linnaeus, 1758) and common sole (*Solea solea* Linnaeus, 1758), out of which in total 258 adult specimens were investigated.

Sampled individuals from each species taken randomly were rinsed in clean sea water and then filleted. The fillets from each fish were thoroughly chopped and mixed and a subsample of about 60 g taken. All prepared fish samples were stored deep frozen at -21°C until their analysis. The fish samples were thawed and then approximately 20 g of samples

Table 1. Catches of fishes from Turkish waters in 2010 (in tons) (Turkish Statistical Institute, 2011)

Species	Total	Eastern Black Sea	Western Black Sea
Mediterranean horse mackerel	14,392	7,968	2,879
Sprat	57,023	56,739	100
Stripped mullet	4,455	1,064	2,309
Atlantic bonito	9,401	3,408	2,914
Flathead mullet	3,119	851	366
Black scorpion fish	254	55	13
Sea bream	1,164	No record	No record
Shi drum (Croaker)	41	6	-
Picarel	1,243	20	37
Common sole	1,166	No record	No record

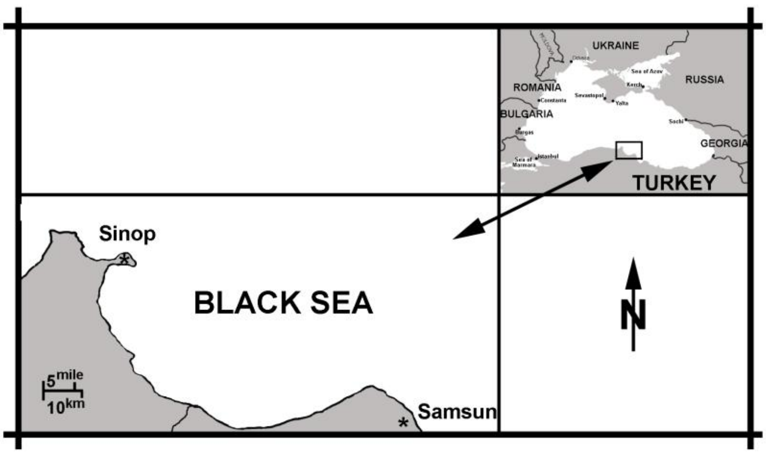

Figure 1. Fish sampling area from Sinop coasts of the Black Sea, Turkey.

were introduced in acid cleaned jars and digested with hot concentrated nitric acid to obtain release of heavy metals. All organic materials in each sample were completely digested. The digests were allowed to cool, filtered through a 0.45 µm Millipore membrane filter, transferred to 50 ml volumetric flasks and made up to mark with 1% nitric acid and diluted with double distilled water to 25 ml. The digestion of each sample was made in triplicate and in all experiences three blanks were also performed in order to check for possible contamination. The digests were kept in plastic bottles and later all determinations were made using Atomic Absorption Spectrophotometer (AAS) (modified from Bernhard, 1976). The values were used to plot a standard curve. The standards and blank were treated in the same way as the real samples to minimize matrix interferences during analysis. Metal contents were expressed as µg g^{-1} wet weight. Working calibration standards of all metals were prepared by serial dilution of concentrated stock solutions (Merck, Germany) of 1000 mg/ l. These and blank solutions were also analysed in the same way as for the digested samples.

Statistical Analysis

Statistical analysis of data was carried out using Statistica 7.0 statistical package program. A one-way analysis of variance (ANOVA) was performed, followed by Duncan comparisons for the source of statistically significant differences of metal concentrations between species. Differences in mean values were accepted as being statistically significant if P<0.05 (Zar, 1984).

Results and Discussions

The data obtained for heavy metal contents in fish (mg/kg wet wt.) are presented in Figure 2. The metal concentrations decrease in the order Zn>Cu>Pb>Cd. The ranges of essential metals Zn and Cu in fish muscles were 5.95-45.35 and 0.77-7.77 mg/kg wet wt., respectively. The Zn, Cu, Pb and Cd concentrations in tissues displayed species differences (P<0.05). The Zn concentration was the highest in sprat (45.35 mg/kg wet wt.). Mean concentrations of Zn found in all fish samples studied are all less than 50 mg/kg wet wt., well below the guideline level (MAFF 1995; Anonymous 1995). Cu concentrations in all fish samples were low (maximum Cu concentration was 7.77 mg/kg wet wt. in sprat) and quite below the guideline level of 20 ppm (MAFF, 1995; Anonymous, 1995).

The ranges of non-essential metals Pb and Cd were 0.03-0.28 and 0.02-0.09 mg/kg wet wt., respectively. The muscle concentration of non-essential element Pb in all fish samples was below the detection limit. Turkish Food Codex (Anonymous, 2008) and Commission Regulation (EC) (Anonymous, 2006) indicate that maximum level is 0.30 mg/kg wet wt. for Pb. The lead and cadmium concentrations were the highest in lipid-rich pelagic

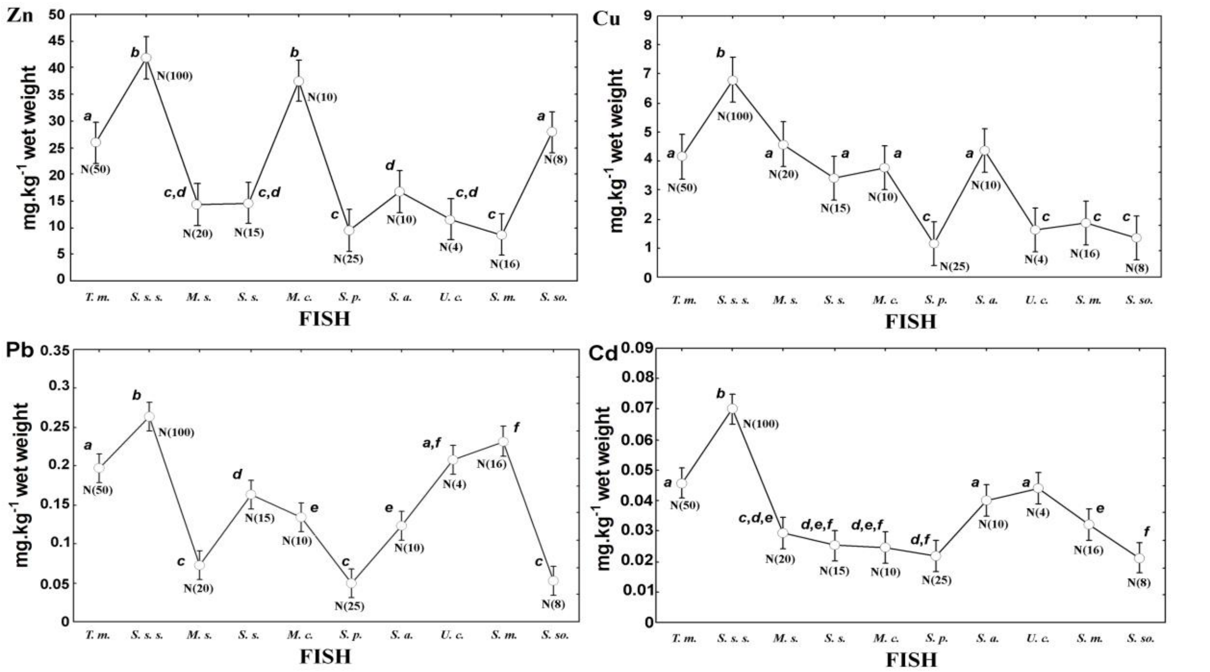

Figure 2. The means with standard deviations (vertical line) of Zn, Cu, Pb and Cd concentrations (µg/g wet wt.) in the dorsal muscle tissues of ten species of fishes from Sinop coastal waters of the Black Sea during fishing season in 2010.
T.m. = Trachurus mediterraneus, S.s.s. = Sprattus sprattus sprattus, M.s. = Mullus surmelatus, S.s. = Sarda sarda, M.c. = Mugil cephalus, S.p.= Scorpaena porcus, Sa = Sparus aurata, U.c. = Umbrina cirrosa, S.m. = Spicara maena, S.so. = Solea solea.
a, b, c, d, e, f = The same letters beside the vertical bars in each graph indicate the values are not significantly different (P>0.05).
N = Number of fish samples

fish *S. sprattus sprattus*. The maximum concentations of Pb and Cd were 0.28 and 0.09 mg/kg wet wt., respectively, Cd concentration was, however, well below the proposed maximum in the food safety regulations (<0.2 mg/kg wet wt.) (MAFF, 1995).

To protect consumers of marine foodstuffs, the EU set a maximum limit for Cd of 0.05 mg/kg wet wt. in fishery products (Anonymous, 2006). The muscle concentration of non-essential element Cd in all fish samples except sprat was below the detection limit. The mean concentration of Cd in sprat was 0.07 mg/kg wet wt. For physiological reasons, certain species accumulate Cd more readily than others and for these species a higher acceptable limit applies (0.10 mg/kg wet weight for bonito (*S. sarda*), common two-banded seabream (*Diplodus vulgaris*), eel (*Anguilla anguilla*), grey mullet (*Mugil labrosus labrosus*), horse mackerel or scad (*Trachurus* sp.), louvar or luvar (*Luvarus imperialis*), mackerel (*Scomber* species), sardine (*Sardina pilchardus*), sardinops (*Sardinops* sp.), tuna (*Thunnus* sp., *Euthynnus* sp.s, *Katsuwonus pelamis*) and wedge sole (*Dicologoglossa cuneata*); 0.20 mg/kg wet weight for bullet tuna (*Auxis species*); 0.30 mg/kg wet weight for anchovy (*Engraulis* sp.), swordfish (*Xiphias gladius*) (Anonymous, 2006). Although there was no criteria particularly for sprat, its biology similar to anchovy.

Differences in metal concentrations related to diet and feeding habits of benthic and pelagic fish species (Bustamente *et al.*, 2003). They show that benthic fish generally accumulate higher concentrations of heavy metals than pelagic fish. Whereas, Topping (1973) suggested that mainly plankton feeding fish contain much higher concentrations of some heavy metals than bottom feeding fish. This is agreed with the present study. Sprat is zooplanctonivorous fish and has high metabolic rate. According to their ecology and food habits, sprat seems suitable as tools for descriptions of environmental conditions of coastal waters. It is also known that metal concentrations in fish tissues are related to the pollution status of the regions. Sprat is migrating between the open sea and inshore areas. Yılmaz (2003) found that concentrations of heavy metals were higher in fish skin than in muscles tissues. The reason for high Cd concentrations in sprat could be due to the metal complexion in skin with the mucus that is impossible to be removed completely from sprat the tissue before the analysis. Indeed it should be reported that for small fish the skin may be an important site for the uptake of metals due to their high surface area to body ratio.

Statistical analysis of metal concentrations showed a significant ($P<0.05$) difference between species (Figure 2).

The heavy metal levels in muscle tissues of similar fish species from the Black Sea coast of Turkey have been investigated by several researchers (Table 2). When the metal concentrations were compared among the Black Sea coasts, Zn and Cu concentrations were found to be highest in *Engraulis encrasicolus* of Bartin coast (Türkmen *et al.*, 2008) and *Mullus barbatus* of Trabzon coasts (Topcuoglu *et al.*, 1990), respectively.

As the concentrations of zinc are high at Bartin and other coasts of the Black Sea, it is possible that these high concentrations are due to naturally occurring processes. Zn is an abundant element and is

Table 2. Heavy metal concentrations (µg metal g^{-1} wet wt.) in similar fish species from the Black Sea coasts of Turkey

Fish Species	Area	Zn	Cu	Pb	Cd	References
Trachurus trachurus	Igneada	-	0.36-0.68	-	-	Ünsal *et al.*, 1993
Trachurus trachurus	Inebolu	-	1.24-2.8	0.02-0.06	-	Ünsal *et al.*, 1993
Trachurus trachurus	Sakarya	-	0.06-0.24	0.27-0.66	-	Ünsal *et al.*, 1993
Trachurus trachurus	Sinop	3.28±0.66	0.79±0.06	0.74±0.21	0.028±0.002	Bat *et al.*, 1996
*Trachurus trachurus**	Samsun	12.05±2.30	1.52±0.35	0.85±0.16	0.47±0.10	Tüzen, 2003
*Trachurus trachurus**	Samsun, Sinop, Terme, Fatsa, Ordu	27.70±1.00	1.79±0.12	0.60±0.07	0.012±0.002	Nisbet *et al.*, 2010
Trachurus mediterraneus	Sinop	17.89-32.38	2.22-6.21	0.17-0.23	0.043-0.048	Present study
Engraulis encrasicolus	Trabzon	10.8±1.29	0.88±0.10	0.12±0.03	0.03±0.01	Türkmen *et al.*, 2008
Engraulis encrasicolus	Sinop	10.6±0.88	1.12±0.16	0.27±0.05	0.02±0.00	Türkmen *et al.*, 2008
Engraulis encrasicolus	Bartin	45.6±22.1	8.58±2.15	0.87±0.40	0.06±0.02	Türkmen *et al.*, 2008
*Engraulis encrasicolus**	Samsun, Sinop, Terme, Fatsa, Ordu	26.25±1.67	2.73±0.21	0.70±0.07	0.035±0.005	Nisbet *et al.*, 2010
Sprattus sprattus sprattus	Sinop	38.34-45.35	5.72-7.77	0.24-0.28	0.05-0.09	Present study
*Mullus barbatus**	Trabzon	11.5±3.5	9.10±5.9	6.86±0.26	<0.1	Topcuoglu *et al.*, 1990
Mullus barbatus	Sinop	2.42±0.27	0.76±0.07	0.28±0.06	0.023±0.002	Bat *et al.*, 1996
*Mullus barbatus**	Samsun	-	-	0.0815±0.003	<0.02	Das *et al.*, 2009
*Mullus barbatus**	Sinop	-	-	0.0515±0.0005	<0.02	Das *et al.*, 2009
Mullus barbatus	Sinop	9.90	8.968	0.424	0.076	Bat *et al.*, 2006
*Mullus barbatus**	BS	4.3	0.01	0.077	0.017	Dalman *et al.*, 2006
*Mullus barbatus**	Sinop	1.424-63.290	0.380-2.714	-	-	Turk Culha *et al.*, 2007
*Mullus barbatus**	Samsun, Sinop, Terme, Fatsa, Ordu	23.71±0.71	3.14±0.31	0.92±0.12	0.020±0.002	Nisbet *et al.*, 2010
*Mullus surmelutus**	Sinop	28.0±9.0	4.20±1.8	<0.5	0.42±0.09	Topcuoglu *et al.*, 1990

- : not measured; *: expressed in µg metal g^{-1} dry wt.
BS: Black Sea (modified from Bat *et al.*, 2009 and Boran and Altınok, 2010).

Table 2. (continued)

Fish Species	Area	Zn	Cu	Pb	Cd	References
Mullus surmelutus	Sinop	10.41-19.71	3.78-5.39	0.05-0.10	0.025-0.035	Present study
Sarda sarda	Samsun	11.20±1.44	1.28±0.14	0.22±0.04	0.09±0.02	Tüzen, 2003
*Sarda sarda**	Samsun, Sinop, Terme, Fatsa, Ordu	19.55±1.20	1.74±0.18	0.90±0.11	0.025±0.005	Nisbet *et al*., 2010
Sarda sarda	BS	21	1.90	0.28	0.35	Durali *et al*., 2010
Sarda sarda	BS	64.9	1.43	0.61	0.13	Tüzen, 2003
Sarda sarda	BS	48.7	0.84	0.76	0.90	Uluozlu *et al*., 2007
Sarda sarda	Sinop	12.75-17.56	2.75-4.12	0.13-0.19	0.023-0.028	Present study
Mugil cephalus	BS	86.2	2.14	0.68	0.35	Tüzen, 2003
Mugil cephalus	BS	40.2	1.26	0.61	0.45	Uluozlu *et al*., 2007
Mugil cephalus	Sinop	30.88-42-65	2.86-4.61	0.09-0.19	0.02-0.03	Present study
Scorpaena porcus	Sinop	7.44-12.3	0.88-1.70	0.03-0.07	0.020-0.023	Present study
Sparus aurata	Sinop	10.72-22.34	3.48-5.21	0.11-0.14	0.038-0.042	Present study
Umbrina cirrosa	Sinop	8.94-14.75	0.91-2.64	0.20-0.22	0.041-0.047	Present study
*Spicara smaris**	Samsun, Sinop, Terme, Fatsa, Ordu	24.35±1.96	0.35±0.10	0.67±0.10	0.67±0.10	Nisbet *et al*., 2010
*Spicara smaris**	Sinop	6.234-57.743	0.610-4.161	--	--	Turk Culha *et al*., 2007
Spicara smaris	Trabzon	12.2±2.63	0.83±0.10	0.15±0.04	0.02±0.00	Turkmen *et al*., 2008
Spicara maena	Sinop	5.88-11.12	1.11-2.75	0.22-0.24	0.028-0.036	Present study
*Psetta maxima**	Samsun, Sinop, Terme, Fatsa, Ordu	24.83±1.71	2.13±0.21	0.73±0.21	0.73±0.21	Nisbet *et al*., 2010
Psetta maxima	BS	45.2	0.75	0.28	0.10	Tüzen, 2003
Solea solea	Sinop	18.63-32.38	0.77-2.10	0.03-0.08	0.020-0.023	Present study

- : not measured; *: expressed in µg metal g^{-1} dry wt.
BS: Black Sea (modified from Bat *et al*., 2009 and Boran and Altınok, 2010).

found in most rocks and sediment. Studies on copper were quite variable throughout the catchment and seem to be significantly different between sites. Cu concentrations in fish were high in Trabzon coast, suggesting a possible point source of copper from these catchment. Although Cu is essential element required by all living organisms, it is considered as toxic to aquatic organisms at high concentrations.

Pb concentrations were found to be the highest in *Mullus barbatus* of Trabzon coasts (Topcuoglu *et al*., 1990) and it was followed by *S. sarda* (Nisbet *et al*., 2010). Cd concentrations were found to be the highest in *S. sarda* (Uluozlu *et al*., 2007) and it was followed by *Psetta maxima* and *Spicara smaris* (Nisbet *et al*., 2010).

From this analysis, it is quite apparent that Sinop coastal waters of the Black Sea are not generally polluted by heavy metals. There are some areas where elevated concentrations may occur near industrial hot spots (Table 2) and it will be important to complete a more detailed survey of coastal sites.

Sinop coast is a relatively unpolluted marine environment, since almost no industry and only small settlements exist in the surrounding region. Studies on heavy metal concentrations in marine organisms have provided an opportunity to observe the mechanisms of bioaccumulation of heavy metals, which is an important component in assessing the effects of pollution on marine ecosystems.

Conclusions

Based on the analyses of fish samples, heavy metal concentrations in fish from Sinop coastal waters of the Black Sea are low. All fish samples tested were well within the limits set by the Commission Regulation (EC) (Anonymous, 2006) and Turkish Food Codex (Anonymous 2008) for metals except Cd in sprat. For Cd, levels were well below the guideline level (MAFF 1995; Anonymous 1995). Fish is an important food resource for human consumption and a major component of the marine ecosystem, thus assessment of the heavy metal effects is particularly important. Therefore studies on the presence of heavy metals in marine fish have contributed to the accumulation of new data on their levels in species of marine organisms with commercial significance, including implications for estimating the risk of consumer exposure to metals and thus to make a more valid conclusion further experimentation would be required. Besides, studies on heavy metal in aquatic organisms should be conducted over a longer period before making any decision.

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