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Research Article

Determination of Heavy Metals and Pesticides in Different Types of Fish Samples Collected from Four Different Locations of Aegean and Marmara Sea

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Heavy metals and pollutants cause serious damage to the ecological environment and accumulate in marine species in the seas. These pollutants and heavy metals accumulating in living species are a serious source of danger for human health. For this purpose, in this study, heavy metal (lead, mercury, copper, zinc, arsenic, cadmium, silver, manganese, and nickel) and pesticide (p-p′-DDE, α-BHC, endosulfan, endosulfan sulfate, endrin, aldrin, heptachlor, heptachlor epoxide, methoxychlor, p-p′-DDD, p-p′-DDT, β-BHC, cypermethrin, and dieldrin) analyses of four different fish species (*Pomatomus saltatrix*, *Dicentrarchus labrax*, *Mugil cephalus*, and *Sparus aurata*) collected from the Aegean and Marmara seas were carried out by gas chromatography-mass spectroscopy and using an inductively coupled plasma-mass spectrometer. We observed serious and remarkable arsenic, lead, and cadmium concentrations in the muscle meat of fish sample. p-p′-DDE and endosulfan were determined in every fish sample of each region. Heptachlor concentration was determined as 0.0598 μg/g in *Dicentrarchus labrax* sample from Marmara Sea, which is nearly nine thousand times more than the maximum allowable concentration of environmental quality standards biota of heptachlor listed in 2013/39/EU. The results show an indication of the significant health risks associated with the consumption of these contaminated fish in the Aegean and Marmara seas. In the Turkish food codex and in the 2013/39/EU directive, some heavy metals that do not have the maximum allowable concentration limits should be urgently indicated.

1. Introduction

Today, with an ever-rapidly increasing human population also increases their consumption needs. To make life more standardized, and to increase production and productivity, application of widespread and unconscious use of chemicals cause direct environmental damage, as air, soil, and water pollution, degrading plant and animal health and existence [1, 2]. Among these chemicals, pesticides are used extensively in the fields of agriculture, public health, environmental health, and veterinary medicine [3]. Pesticides are chemical substances used to prevent, control, or reduce harmful biological organisms such as insects, plant pathogens, weeds, molluscs, birds, mammals, fish, nematodes, and microorganisms that are harmful to human and animal food sources and also to the ecosystems that inhabit them [4]. These chemicals, which are useful when applied at the required dosages and durations, may affect the nontarget organisms as a result of their careless and intensive use, while polluting the soil and aquatic ecosystem. Pesticides are transported by surface streams, evaporated into the atmosphere, going through the absorption/desorption process, infiltrated from the soil or by taking plants into the plant, or degraded by the chemical, as microbial and photodegradation directly from one ecosystem to another. The transport of pesticides into aquatic ecosystems is direct, via spray application to the water sources or by being mixed into the groundwater. This phenomenon threatens the life of the environment [5].

Chemical properties of pesticides such as water solubility, vapor pressure, soil tendency, and resistance to time of disintegration, determine the amount of pesticides transported from one medium to another. Most pesticides are of

organic molecular structure and are hydrophobic in nature; they are bound to suspended particles on the water surface or accumulate in the sediment layer at the bottom of lakes and seas. Therefore, benthic invertebrates, bottom fish, and other vertebrates fed in the sediment environment are at greater risk of exposure to these contaminants than pelagic creatures; it is reported that pesticides can be found in the concentrations that can cause ecotoxicological effects in aquatic ecosystems although the bioaccumulation properties of the modern pesticides used in the present day are low. Pesticides are taken through organism by aquatic organisms such as epidermis, gill epithelium, and digestive system. Each of these chemical substances passes through a number of biological membranes until they are assimilated in the body [6]. The structures of these biological membranes and toxic compounds are the most important factors controlling the access to the organism. For example, small molecule and fat-soluble substances are taken by passive diffusion that does not require energy use to the organism.

The accumulation of compounds in the body by passive diffusion varies depending on factors, such as the animal's metabolic rate, nutritional status, size, age, and temperature of ambient water. Wastewater is the result of the production and subsequent use of heavy metals and compounds and contains high concentrations of toxic heavy metals and compounds [7–9]. Copper, zinc, nickel, and manganese from these heavy metals are dangerous in high concentrations. Mercury, cadmium, lead, and arsenic are toxic even at very low concentrations [10, 11]. These pollutants affect the air, water, soil, and living creatures. If heavy metals are absorbed above the permissible limit values, they cause serious health problems in humans [12]. Heavy metals cause serious health problems, such as loss of growth, cancer, organ damage, damage to the nervous system, and even death in very high amounts [8, 13]. Exposure to some metals, such as mercury and lead, causes autoimmunity diseases that attack their own cells in the immune system. This may lead to conditions such as rheumatic calcification, kidney disorders, circulatory and nervous system problems, and damage to the fetal brain [14]. Heavy metals are toxic when they exceed the concentration limit. In contrast to this general representation, heavy metals do not act in live bodies only depending on their concentration. The effect of heavy metals depends on the species of the organism and the nature of the metal ion (solubility value, chemical structure, redox and complex formation ability, the shape of the body, the frequency of the environment, the local pH value, etc.) [15–18]. For this reason, the maximum concentration limit values of the nutrients and foodstuffs grown in the drinking water, soil, and sea are determined because they are regularly consumed [19–23].

2. Materials and Methods

2.1. Sample Collection. Figure 1 presents general view of sampling area. Figures 2–5 present sample collection of different fish species from Aegean Sea (38°71′N, 26°61′E), Bandırma Coast of Marmara Sea (40°53′N, 28°25′E), Istanbul coast of Black Sea (41°21′N, 29°34′E), and Duzce Coast of Black Sea (41°19′N, 31°13′E), respectively.

2.2. Samplings. Fish samples were provided from fishermen at Bandırma, Izmir, Istanbul, and Duzce and transferred to our laboratory in polyethylene boxes at −4°C. We obtained three samples for each of fish species. The samples were washed with distilled water and placed in −80°C freezer until extraction process. Muscle tissues and internal organs of fish samples were collected separately. 15 g of muscle tissues of fish samples was shredded by mixer and added to 15 mL Acetic acid-Acetonitrile mixture solution (1%). The samples were extracted according to AOAC QuECHERS, AOAC 2007.01 method [24]. After that, the extraction solution of each fish sample was filtered from a dispersive clean-up tube, C18 (Octadecyl-modified silica, Macherey-Nagel Chromabond), to remove the fatty acids, sugar, and organic acids from the sample solution.

2.3. Instruments and Chemicals. Gas chromatography-mass spectroscopy (GC-MS), Thermo TRACE™ 1300 ISQ LT Single Quadrupole Mass Spectrometer (Thermo Fisher, Inc., Waltham, MA, USA), and an inductively coupled plasma-mass spectrometer (ICP-MS), NexION 2000 B Perkin Elmer, were used for the determination of pesticides and elements. CEM MARS-6 closed vessel microwave digestion system was used for preparation method of fish samples for ICP-MS analysis. Analytical grade chemicals and organic solvents were purchased from Sigma Aldrich. ICP-MS standard solutions were obtained from Perkin Elmer. Certificated levels of standard solutions are Pb: 999 $\mu g \cdot mL^{-1} \pm 5\,\mu g/mL$, Hg: 998 $\mu g \cdot mL^{-1} \pm 5\,\mu g/mL$, Cu: 999 $\mu g \cdot mL^{-1} \pm 5\,\mu g/mL$, Zn: 998 $\mu g \cdot mL^{-1} \pm 5\,\mu g/mL$, As: 1003 $\mu g \cdot mL^{-1} \pm 5\,\mu g/mL$, Cd: 999 $\mu g \cdot mL^{-1} \pm 5\,\mu g/mL$, Ag: 998 $\mu g \cdot mL^{-1} \pm 5\,\mu g/mL$, Mn: 1002 $\mu g \cdot mL^{-1} \pm 5\,\mu g/mL$ and Ni: 999 $\mu g \cdot mL^{-1} \pm 5\,\mu g/mL$. Bi, In, Sc, and Ge were used as internal standards for ICP-MS analysis. All pestanal® analytical standards (p-p′-DDE, α-BHC, endosulfan sulfate, endrin, heptachlor, heptachlor epoxide, methoxychlor, p-p′-DDD, p-p′-DDT, β-BHC, aldrin, cypermethrin, dieldrin, and endosulfan) were purchased from Sigma Aldrich. All laboratory equipment was washed with HNO_3 solution (1/6, v/v) before use.

2.4. GC-MS Analyses. We used TG-5MS (30 m × 0.25 × 2.5 μm) column for the separation of pesticides. The inlet temperature was 250°C. The following gradient temperature method was applied. The initial oven temperature was 60°C and was kept for 1 min. The oven temperature was increased to 150°C at a rate of 10°C per minute with 1 min hold time and then it was increased to 250°C at a rate of 10°C per minute kept for 1 min. Finally, the oven temperature was increased to 350°C at a rate of 20°C/min with 3 min hold time. The split flow was 45 mL/min. The carrier gas flow was 1.5 mL/min. The carrier gas was Helium with 99.995% purity. Retention times and recovery % values of the target analytes are presented in Table 1. After the mass detection of each compound with four fragment ions, we obtained the calibration curve and equations for each compound.

Figure 1: General view of sampling area.

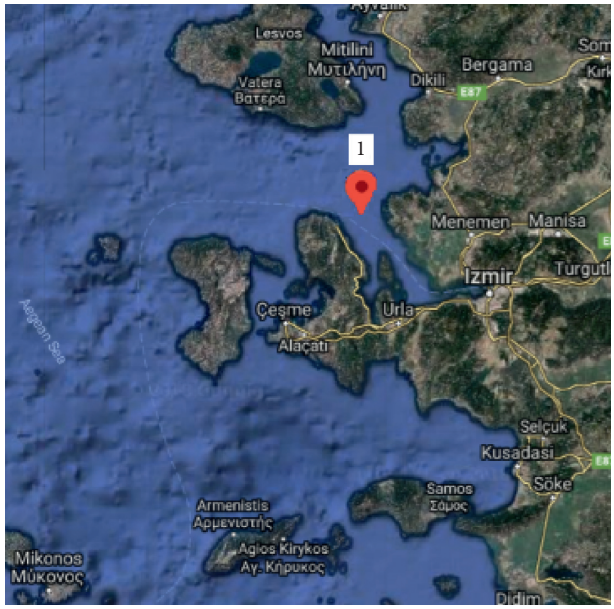

Figure 2: Sample collection from Aegean Sea, Izmir (38°71′N, 26°61′E).

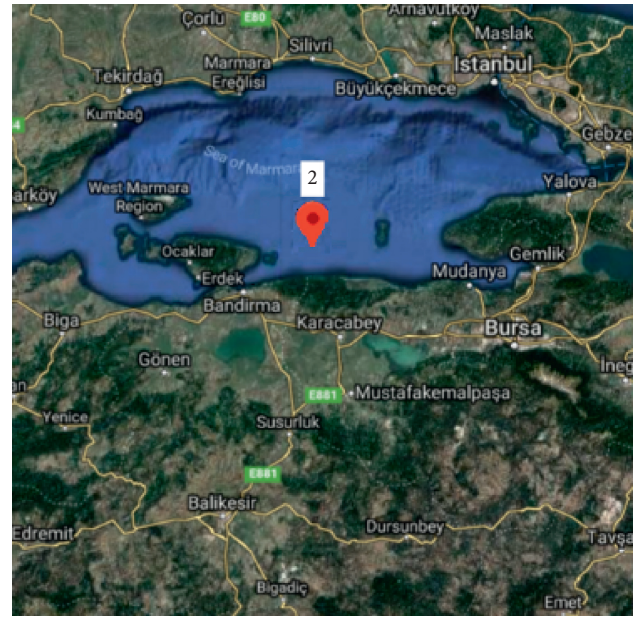

Figure 3: Sample collection from Bandırma Coast of Marmara Sea (40°53′N, 28°25′E).

2.5. ICP-MS Analyses. We had used solubilization process for muscle tissues and internal organs of each fish samples by microwave digestion system before determination of the samples by ICP-MS. ICP-MS parameters for determination of target heavy metals in fish samples are Rf power 1300 W, gas flow rate 1.5 mL/min, coolant gas 15 mL/min, auxiliary gas mL/min, nebulizer gas 0.65 mL/min, sample flow rate 1.5 mL/min, flush time 20 sec, and read time 3 seconds, respectively. The concentration of calibration solutions were 1 μg/kg, 5 μg/kg, 10 μg/kg, 30 μg/kg, 500 μg/kg, and 1000 μg/kg, respectively. Au standard was spiked to the blank and standard solutions for the determination of Hg. Limits of detections of target heavy metals were Pb: 0.04 ngL^{-1}, Hg: 1 ngL^{-1}, Cu: 0.2 ngL^{-1}, Zn: 0.7 ngL^{-1}, As: 0.4 ngL^{-1}, Cd: 0.07 ngL^{-1}, Ag: 0.09 ngL^{-1}, Mn: 0.1 ngL^{-1}, and Ni: 0.2 ngL^{-1} (accuracy of the results was applied using different statistical methods at 95% confidence interval, significance, $p < 0.05$).

3. Results and Discussion

3.1. Heavy Metal Analyses by ICP-MS. Domestic, industrial, and ship wastes pose a threat to aquatic organisms and ecological environment as a result of the evacuation of the boats to the cove by the tons of dead fish, port operations, and the discharge of the products resulting from mining activities into the sea. Wastewater is the result of the production and subsequent use of heavy metals and compounds

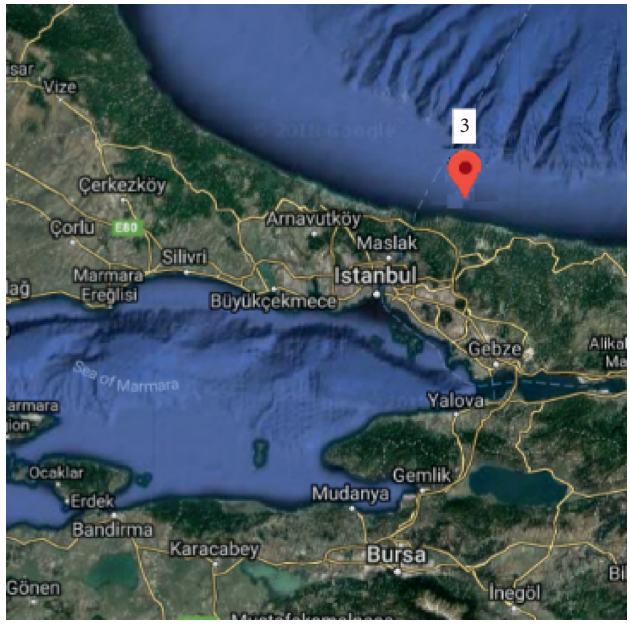

Figure 4: Sampling collection from Istanbul coast of Black Sea (41°21′N, 29°34′E).

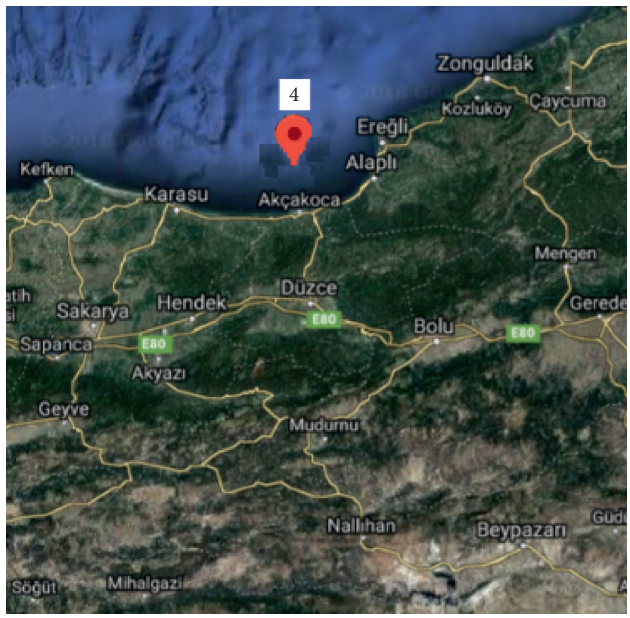

Figure 5: Sample collection from Duzce Coast of Black Sea (41°19′N, 31°13′E).

and contains high concentrations of toxic heavy metals and compounds. The maximum levels for Hg, Pb, Cd, and Sn in foodstuffs are presented in Commission Regulation (EC) No. 1881/2006 [25], which indicates the maximum levels for chemical pollutants in the muscle meat of fish. Table 2, Figures 6–9, and Table 3 present the heavy metal residues (As, Mn, Hg, Ag, Cd, Ni, Zn, Cu, and Pb) (μg/g) in muscle tissues and internal organs of fish samples for four different fish species (*Pomatomus saltatrix*, *Dicentrarchus labrax*, *Mugil cephalus*, and *Sparus aurata*) from four different cost regions of Turkey.

Table 1: Retention time and recovery (%) values of each target pesticide.

Pesticide	Recovery % ($n = 3$), mean ± SD	RT (min)
α-BHC	86.16 ± 0.98	15.41
β-BHC	94.06 ± 1.56	16.13
Heptachlor	86.30 ± 0.70	17.75
Aldrin	101.56 ± 0.59	18.46
Heptachlor epoxide	99.82 ± 0.71	19.28
Endosulfan	85.03 ± 1.80	19.98
p-p′-DDE	95.21 ± 0.88	20.41
Dieldrin	92.39 ± 1.65	20.48
Endrin	96.46 ± 0.99	20.91
p-p′-DDT	102.11 ± 0.90	21.19
p-p′-DDD	98.64 ± 0.76	21.27
Endosulfan-sulfate	96.50 ± 1.21	21.95
Methoxychlor	105.63 ± 1.98	23.12
Cypermethrin	95.55 ± 1.32	25.26

In Table 2, the highest arsenic concentration (1.18 μg/g) was observed in *Pomatomus saltatrix* from Istanbul. The second highest As concentration was observed in *Pomatomus saltatrix* from Bandırma. Unfortunately, the maximum level of arsenic has not been determined yet by the European Commission (EC No. 78/2005) nor the Turkish Food Codex (TFC) [26, 27]. When we checked the As concentration for internal organs of every fish sample except *Sparus aurata* from Bandırma, it can be clearly seen that there was a really serious As concentration in each of the fish species from four different regions (Table 3). So it is urgently necessary to determine maximum limit for arsenic, by both the EC and TFC. Arsenic-containing pesticides (insecticides, ink, and textile dyes) are used unconsciously to increase the amount of arsenic in nature by hundreds of times. Today, the use of arsenic dyes and pesticides is prohibited. In addition, arsenic causes many diseases such as skin cancer, circulatory disorder, heart failure, chronic poisoning, extreme fatigue, and cancer. Arsenic first accumulates by binding to thiol groups in the liver and then in keratin-rich tissues [28]. Tuzen determined the amount of arsenic in meat of the fish from the Black Sea region of Turkey in the range of 0.11–0.32 μg/g in 2009 [10]. When we compared As concentration of Tuzen's research with Table 2, it is observed that there has been a significant increase in the amount of arsenic in the last decade in Turkey. Increasing of As amount is alarming and should be controlled immediately.

The main routes of transmission of lead are contaminated drinking water, vegetable and animal foods (especially fish), lead-coated containers, water pipes, cosmetics, insecticides, dyes, cigarettes, and gasoline. The concentration of lead in muscle tissues of fish samples varies between 2.38 and 0.10 μg/g in Table 2. The highest Pb residue (2.38 μg/g) was observed in *Sparus aurata* sample from Izmir (Aegean Sea). The amount of Pb concentration is over the maximum level by EC and TFC. In Commission Regulations No. 1881/2006, the maximum level (ML) of Pb was established as 0.3 μg/g for muscle meat of fish. The amount of Pb residues was presented in Table 3 for internal organs of fish samples. It does not state any ML for internal organs of fish. However,

Table 2: Concentration of Pb, Hg, Cu, Zn, As, Cd, Mn, and Ni residues (μg/g) in muscle tissues of fish samples.

Fish species		Pb	Hg	Cu	Zn	As	Cd	Mn	Ni
Izmir	*Pomatomus saltatrix*	0.34 ± 0.06	—	—	64.94 ± 9.09	—	5.15 ± 0.27	2.22 ± 0.15	1.85 ± 0.03
	RSD	2.4			14.5		5.2	7.6	2.7
	Dicentrarchus labrax	0.11 ± 0.008	—	8.09 ± 1.13	59.25 ± 7.70	0.68 ± 0.07	0.24 ± 0.01	6.79 ± 0.13	11.25 ± 0.56
	RSD	8.1		14.5	13.3	11.2	5.9	2.4	5.3
	Mugil cephalus	2.34 ± 0.04	—	—	50.14 ± 7.01	—	5.40 ± 0.25	4.16 ± 0.29	1.66 ± 0.03
	RSD	2.4			14.5		5.2	7.6	2.7
	Sparus aurata	2.38 ± 0.14	—	3.39 ± 0.20	67.09 ± 9.39	—	4.96 ± 0.24	4.71 ± 0.84	4.03 ± 0.68
	RSD	6.9		6.9	14.2		5.2	18.2	17.4
Bandırma	*Pomatomus saltatrix*	0.09 ± 0.004	0.13 ± 0.006	7.10 ± 0.92	39.15 ± 5.87	1.00 ± 0.16	—	4.80 ± 0.91	4.30 ± 0.08
	RSD	5.2	5.4	13.8	15.6	16		19.9	2.5
	Dicentrarchus labrax	0.14 ± 0.007	—	7.74 ± 1.08	80.29 ± 14.45	0.75 ± 0.03	—	6.47 ± 0.77	5.07 ± 0.50
	RSD	5.2		14.3	18.7	4.4		12	10.9
	Mugil cephalus	0.39 ± 0.07	—	—	32.07 ± 5.13	—	4.98 ± 0.49	6.91 ± 0.48	1.23 ± 0.11
	RSD	2.2			16.6		10.9	7.4	9.4
	Sparus aurata	0.44 ± 0.03	—	0.49 ± 0.05	24.20 ± 0.72	—	0.20 ± 0.008	2.92 ± 0.23	1.49 ± 0.07
	RSD	7.9		11.8	3.6		4.5	8.1	5.7
Istanbul	*Pomatomus saltatrix*	0.12 ± 0.008	—	4.59 ± 0.82	49.48 ± 7.61	1.18 ± 0.20	—	3.83 ± 0.53	6.33 ± 0.58
	RSD	7.7		18	15.4	17		14.2	9.3
	Dicentrarchus labrax	0.12 ± 0.004	—	7.96 ± 0.31	64.29 ± 5.14	0.70 ± 0.02	—	6.57 ± 0.13	7.87 ± 0.07
	RSD	4.2		4.3	8.7	3.4		2.8	1.9
	Mugil cephalus	0.39 ± 0.01	—	0	52.08 ± 3.12	—	5.18 ± 0.05	3.71 ± 0.33	1.58 ± 0.11
	RSD	3.2		0	6.6		1.9	9.4	7.2
	Sparus aurata	0.36 ± 0.007	—	8.43 ± 0.59	49.45 ± 2.96	—	1.27 ± 0.003	3.33 ± 0.03	3.60 ± 0.18
	RSD	2.6		7.3	6.8		3.2	1.6	5.6
Duzce	*Pomatomus saltatrix*	0.10 ± 0.004	—	9.46 ± 1.51	67.55 ± 2.70	—	—	7.00 ± 0.84	3.71 ± 0.40
	RSD	4.4		16	4.7			12.8	11.9
	Dicentrarchus labrax	0.13 ± 0.007	—	7.82 ± 0.54	72.00 ± 2.16	0.72 ± 0.02	0.18 ± 0.01	6.67 ± 0.20	8.19 ± 0.32
	RSD	6.5		7.5	3.9	4.2	7.6	3.6	4.6
	Mugil cephalus	0.26 ± 0.07	—	—	44.94 ± 1.79	—	5.04 ± 0.20	4.36 ± 0.26	1.70 ± 0.05
	RSD	3.0			4.5		4.9	6.9	3.3
	Sparus aurata	0.16 ± 0.004	—	10.13 ± 0.81	38.05 ± 2.66	—	0.17 ± 0.001	3.00 ± 0.3	2.74 ± 0.19
	RSD	3.6		8.4	7.5		1.9	10.8	7.4

the high amount of Pb must be taken under control because there is no possibility of being expelled from the body since the half-life is 20 years. The other similar studies were reported by the amount of Pb as 0.22–0.85 μg/g at the middle of the Black Sea, and 0.33–0.93 μg/g, 1.41–3.92 μg/g, 0.38–5.20 μg/g, and 0.83–3.71 μg/g in fish sample at Aegean Sea and Black Sea in fish samples.

Cadmium is a harmful element in terms of human health, although it has benefits in the production of many products. It is one of the most dangerous heavy metal pollutants in the ecosystem. It is among the most toxic metals for living organisms. The compounds are also highly toxic. Cadmium used in batteries shows carcinogenic effects. It is not an essential element for biological functions. Cadmium is linked to lung diseases, prostate cancer, tissue damage, central nervous system and immune system problems, anemia, diarrhea, chronic problems, destruction of the adrenal gland, abdominal pains, vomiting, bone disorders, reproductive system problems, infertility, prostate problems, psychological problems, damage to the brain and spinal cord, and DNA damage as a result of cancer triggers [13, 17, 19]. The ML of Cd is 0.05 μg/g in EC and TFC. In Table 2, a high concentration of Cd in muscle meat of *Mugil cephalus* samples (5.40, 4.98, 5.18, and 5.04 μg/g) are especially remarkable from each of the four different regions. This phenomenon is attributed to the fact that *Mugil cephalus* likes dirty and turbid waters, because it has an ability to live in dirty water while other fish species do not. The other fish species—*Pomatomus saltatrix*, *Dicentrarchus labrax,* and *Sparus aurata*—have also high Cd amounts that are over ML of EC and TFC. The amount of Cd in internal organs of fish samples is under ML except *Dicentrarchus labrax* (1.50 μg/g) from Bandırma and *Pomatomus saltatrix* (0.42 μg/g, 0.13 μg/g) from Istanbul and Duzce, respectively

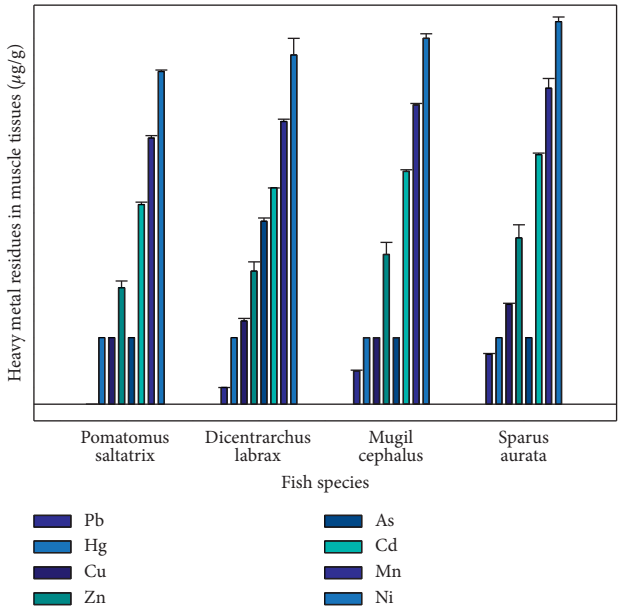

Figure 6: Concentration of Pb, Hg, Cu, Zn, As, Cd, Mn, and Ni residues (μg/g) in muscle tissues of fish samples from Izmir, Aegean Sea.

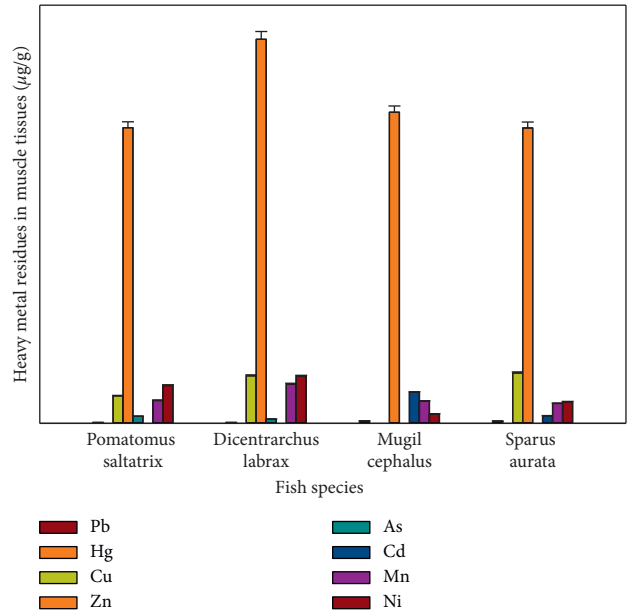

Figure 8: Concentration of Pb, Hg, Cu, Zn, As, Cd, Mn, and Ni residues (μg/g) in muscle tissues of fish samples from Istanbul, Cost of Black Sea.

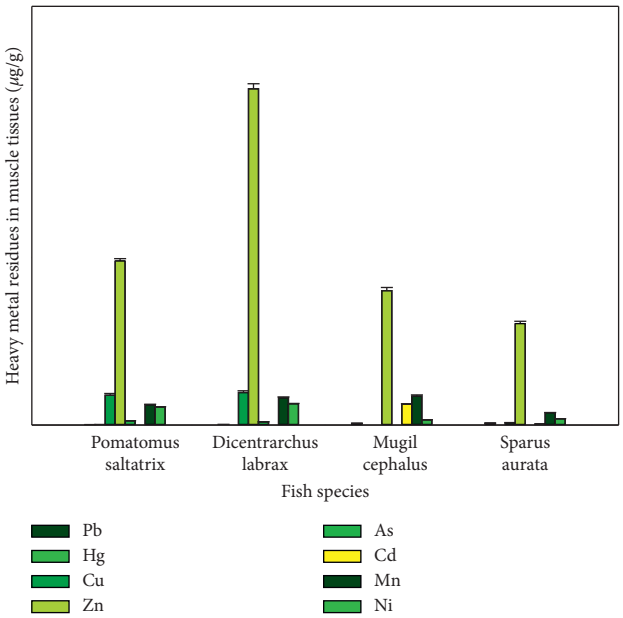

Figure 7: Concentration of Pb, Hg, Cu, Zn, As, Cd, Mn, and Ni residues (μg/g) in muscle tissues of fish samples from Bandırma Cost of Marmara Sea.

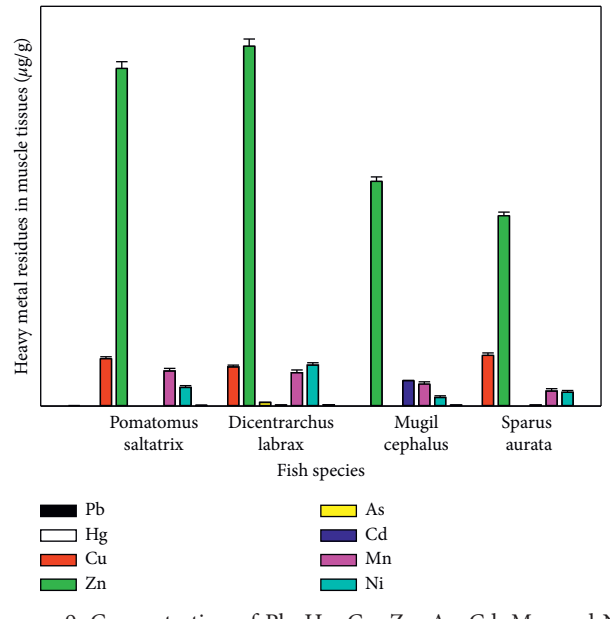

Figure 9: Concentration of Pb, Hg, Cu, Zn, As, Cd, Mn, and Ni residues (μg/g) in muscle tissues of fish samples from Duzce, Cost of Black Sea.

(Table 2). The other researchers reported the concentration of Cd in fish samples as 0.02–0.24 μg/g at Black Sea, 0.02–0.35 μg/g at Marmara Sea, and 0.13–0.47 μg/g at Marmara (Istanbul), Aegean, and Mediterranean Seas. When we compared our results with the other results, increasing concentration of Cd should be taken into serious consideration.

According to the TFC, the maximum Cu concentration allowed in the meat of fish was determined as 20 μg/g from the sea and lakes. The ML of Cu is listed as 30 μg/g and 20 μg/g at the Food and Agriculture Organization (FAO) and WHO, respectively [29, 30]. In this study, Cu concentration is determined in muscle meat of fish samples under the ML of FAO and WHO (Table 2). But when we analyzed the internal organs of fish samples (Table 3), it can be clearly seen that Cu concentration is over ML in many samples. Specifically, there were above-limit Cu amounts in the samples from Bandırma, Istanbul, and Duzce. This could be due to the fact that these cities are within the industrial

Table 3: Concentration of Pb, Cu, Zn, As, Cd, Ag, Mn, and Ni residues (μg/g) in internal organs of fish samples.

Fish species		Pb	Cu	Zn	As	Cd	Ag	Mn	Ni
Izmir	*Pomatomus saltatrix*	0.13 ± 0.005	12.30 ± 1.59	70.66 ± 12.71	1.76 ± 0.08	—	—	7.42 ± 0.37	8.23 ± 0.24
	RSD	4.7	13	18.1	5.3			5.9	3.2
	Dicentrarchus labrax	0.09 ± 0.002	35.33 ± 0.70	66.28 ± 6.62	1.80 ± 0.10	—	—	11.51 ± 1.03	4.44 ± 0.26
	RSD	3.2	2.8	10.9	6.6			9.2	6.2
	Mugil cephalus	0.10 ± 0.007	31.17 ± 0.31	187.10 ± 7.48	2.39 ± 0.04	—	—	15.24 ± 1.21	13.33 ± 0.39
	RSD	7.6	1.7	4.7	2.8			8.5	3.4
	Sparus aurata	0.08 ± 0.007	22.31 ± 2.23	60.01 ± 8.40	1.99 ± 0.17	—	—	7.25 ± 0.72	5.05 ± 0.55
	RSD	9.5	10.8	14.4	9.8			10.3	11.4
Bandırma	*Pomatomus saltatrix*	0.09 ± 0.006	13.89 ± 1.80	65.43 ± 7.5	5.20 ± 0.1	—	—	4.74 ± 0.04	1.26 ± 0.08
	RSD	7.5	13.8	12.5	2.6			1.4	7.7
	Dicentrarchus labrax	0.08 ± 0.002	66.70 ± 2.00	88.51 ± 6.19	0.49 ± 0.02	1.50 ± 0.09	—	4.52 ± 0.31	4.90 ± 024
	RSD	3.1	3.6	7.5	5.1	6.4		7.2	5.6
	Mugil cephalus	0.16 ± 0.006	68.40 ± 1.36	177.10 ± 10.62	5.02 ± 0.25	—	1.02 ± 0.06	6.42 ± 0.06	18.81 ± 2.82
	RSD	4.6	2.8	6	5.9		6.1	1.7	15.9
	Sparus aurata	0.06 ± 0.001	11.12 ± 1.44	48.21 ± 1.44	—	—	—	6.83 ± 0.20	4.79 ± 0.43
	RSD	2.2	13.2	3.2				3.2	9.2
Istanbul	*Pomatomus saltatrix*	0.15 ± 0.004	8.63 ± 0.69	60.95 ± 3.65	2.32 ± 0.04	0.42 ± 0.01	—	2.87 ± 0.08	1.44 ± 0.05
	RSD	3.6	8.2	6.1	2.1	4.2		3.6	4.7
	Dicentrarchus labrax	0.06 ± 0.003	50.56 ± 1.51	76.34 ± 6.87	1.20 ± 0.007	—	—	6.18 ± 0.43	4.66 ± 0.23
	RSD	6.5	3.6	9.9	6.4			7.2	5.7
	Mugil cephalus	0.14 ± 0.008	54.58 ± 1.09	158.10 ± 11.06	3.69 ± 0.29	—	—	10.56 ± 0.52	15.63 ± 0.78
	RSD	6.7	2.7	7.5	8.2			5.9	5.8
	Sparus aurata	0.07 ± 0.005	18.46 ± 1.47	59.32 ± 2.37	1.66 ± 0.13	—	—	5.55 ± 0.38	4.67 ± 0.42
	RSD	8.0	8.8	4.4	8.9			7.3	9.4
Duzce	*Pomatomus saltatrix*	0.11 ± 0.005	10.38 ± 0.31	88.69 ± 14.19	4.37 ± 0.30	0.13 ± 0.01	—	7.30 ± 1.02	5.73 ± 0.28
	RSD	5.8	3.6	16.2	7.1	9.4		14.3	5.3
	Dicentrarchus labrax	0.07 ± 0.003	44.98 ± 01.34	79.46 ± 7.15	1.69 ± 0.10	—	—	7.68 ± 0.46	4.81 ± 0.24
	RSD	5.9	3.6	9.9	6.9			6.9	5.0
	Mugil cephalus	0.13 ± 0.007	70.89 ± 1.41	164.80 ± 4.94	4.24 ± 0.12	—	—	8.56 ± 0.59	16.60 ± 0.49
	RSD	6.5	2.0	3.6	3.4			7.9	3.9
	Sparus aurata	0.09 ± 0.003	7.45 ± 0.74	76.3 ± 8.39	1.37 ± 0.02	—	—	6.19 ± 1.05	3.47 ± 0.20
	RSD	4.4	10.1	11.2	2.4			17.1	6.1

center of Turkey. Unfortunately, there is no report of any ML for Cu at EC.

Taking in large amounts of zinc is highly toxic to the human body. It can cause nausea, vomiting and diarrhea, restlessness, sweating and tremor, and cholesterol instability. It also causes weakening of the immune system by reducing the enzymes in the body and tumor formation in excessive use. A wide range of security leads to excessive intake of zinc toxicity [8, 13]. The ML of zinc in fish samples was reported to be 50 μg/g, 30 μg/g, and 50 μg/g at TFC, FAO, and MAFF, respectively [26, 30, 31]. The highest concentration of Zn was analyzed as 80.29 μg/g in muscle meat of *Dicentrarchus labrax* from Bandırma (Table 2). In Table 3, the highest Zn concentration (187 μg/g) was observed in internal organs of *Mugil cephalus* samples from Izmir (Aegean Sea). *Mugil cephalus* has the highest concentration when we compared to all regions. Other researchers reported Zn amount as 9.5–22.9 μg/g from the Black Sea, 16.10–31.40 μg/g from the Mediterranean, and 63.90–73.80 μg/g from Marmara.

Taking high amounts of manganese has toxic effects on mammals. In addition, spasms can cause conditions such as tremors and mental disorders. According to recent studies, the manganese content of drinking water increased while children's memory and cognitive abilities decreased. In another study, it was determined that children who were born as a result of taking manganese from pregnant women had behavioral disorders [14]. Despite these toxic effects of Mn, there is no report of any ML at neither the FAO nor the TFC. The highest Mn concentrations were observed in muscle meat of *Pomatomus saltatrix* (7.00 μg/g) from Duzce and internal organs of *Mugil cephalus* (15.24 μg/g) from Izmir (Tables 2 and 3). In other studies, Mn concentration was reported as 2.76–9.10 μg/g from the Black Sea, 0.18–2.78 μg/g from the Aegean Sea, and 0.10–0.99 μg/g from the Mediterranean Sea.

Nickel is absorbed by the respiratory tract and its absorption through the skin is slow. It is distributed in the kidney, pituitary, lung, skin, adrenal, ovary, and testes. It induces the synthesis of metallothionine. Its excretion mainly occurs in the urine [14]. The highest Ni concentration was observed in muscle meat of *Dicentrarchus labrax* (11.25 μg/g) from Izmir (Table 2). It is difficult to compare the amounts of Ni in our study of muscle meat and internal organs of fish samples with EC or TFC because there is no report of any ML for Ni at EC No. 1881/2006 commission or in the TFC. Considering all of this toxicity of nickel, there should be a reported permissible upper limit of Ni in the EC and TFC.

Silver is an element that cannot be easily removed from the human body and is constantly stored in the body. Prolonged exposure to silver, silver powder, and even other chemical compounds with silver increases the amount of silver in the body. First of all, it changes color without being noticed, when it is exposed to the silver light that accumulates in the organs, under the skin, and in different parts of the body [9]. As a result, the person exposed to silver gains a purple or blue color. This situation is known as Argyria-Arjiri in medical science. In the present study, we analyzed the Ag amount in muscle meat and internal organs of fish samples. We did not observe any Ag concentration in muscle meat of the samples (Table 2). We only observed Ag concentration (1.02 μg/g) in internal organs of the *Mugil cephalus* sample from Bandırma (Table 3).

The rate of mercury has increased 3 times in the environment by increasing usage of industry. There is a small amount of mercury in seafood. However, some game fish contain higher levels of mercury and, when eating these fishes regularly, they may cause accumulation of mercury in the body [17, 28]. Therefore, it is important to determine Hg residues in sea food. We only had determined 0.13 μg/g Hg in muscle tissues of *Pomatomus saltatrix* from Bandırma which is under the ML. But the mercury concentration of sea water samples had a serious issue in CF region. We did not determinate any Hg residues in the internal organs of fishes.

3.2. Analysis of Some Organochlorine Pesticides by GC/MS. Endosulfan, cypermethrin, heptachlor, and endrin were stated in the list of priority substances in the field of water policy in directive 2013/39/EU [32]. Endosulfan, endrin, heptachlor, p-p′-DDD, p-p′-DDT, dieldrin, and cypermethrin were listed in Annex I environmental quality standards for priority substances and certain other pollutants in directive 2013/39/EU. This type of pesticide is used for industrial and agricultural purposes and causes environmental pollution. The pollution in the seas has increased significantly in the recent years with the increase of the population in Turkey. For this purpose, we studied the residues of these pesticides and also α-BHC, β-BHC, p-p′-DDE, and methoxychlor that appears predominantly in the fish liver, in *Pomatomus saltatrix*, *Dicentrarchus labrax*, *Mugil cephalus*, and *Sparus aurata* from four different regions of Turkey. Pesticides resulted in biological accumulation in the fish species. Table 4 presents the residues of some of pesticides in the muscle meat of *Pomatomus saltatrix*, *Dicentrarchus labrax*, *Mugil cephalus*, and *Sparus aurata*. We observed remarkable α-BHC residues in *Mugil cephalus* and *Sparus aurata* samples from each region (Table 4).

Endosulfan sulfate is a type of pesticide commonly found. It was defined that endosulfan is the most common species of pesticides in nutrients, solids, and water in Europe. It was only presented as MAC—environmental quality standards (EQS) of endosulfan for inland surface (0.01 μg/L) and other surface (0.004 μg/L) waters. There is no defined MAC EQS biota that relates to fish in 2013/39/EU [32]. In Table 4, the highest endosulfan sulfate concentration was observed as 0.0214 μg/g in *Mugil cephalus* from Izmir and Istanbul. It was observed in every fish sample of each region except *Pomatomus saltatrix* from Duzce. Directive 2006/77/EC presented maximum content of endosulfan and its derivatives as 0.005 μg/g feeding stuff for fish. The highest obtained value is over the maximum content according to the directive 2006/77/EC [33].

Endrin residues were observed in every fish sample. *Mugil cephalus* samples had the highest concentration of endrin (0.2957 μg/g) from Istanbul. When we compared the endrin residues of the samples, it was listed as *Mugil cephalus* > *Sparus aurata* > *Dicentrarchus labrax* > *Pomatomus saltatrix*. Unfortunately, there is no stated MAC for EQS biota in 2013/39/EU although the usage of it is prohibited in all European countries. Only directive 2006/77/EC mentioned maximum level of endrin as 0.01 μg/g for all feedstuffs.

Heptachlor is one of the pesticides that disrupt the endocrine system. Congenital deformities in children, cancer, especially hormonal cancers, delay in sexual development, and delay in the development of the nervous system are possible effects that may be related to the endocrine system in humans due to heptachlor. The MAC EQS biota of heptachlor is defined as 6.7×10^{-6} μg/g in 2013/39/EU. In the present study, the lowest (0.0059 μg/g) and the highest (0.0598 μg/g) heptachlor residues were observed in the *Dicentrarchus labrax* sample from Istanbul and from Bandırma, respectively (Table 4). The highest heptachlor concentration is nearly nine thousand times more than MAC EQS biota of heptachlor listed in 2013/39/EU. This is a serious issue that should be taken under control, urgently.

Organic pollutants such as organochlorine pesticides (OCPs) in food products tend to accumulate in biological organisms because they are not easily degradable in the environment and are lipophilic. Methoxychlor is a predominantly observed pesticide species, especially in fish liver. Methoxychlor residues were observed in all regions and fish species samples (Table 4). Amounts of methoxychlor residues are higher in Duzce (0.0511 μg/g in *Pomatomus saltatrix*, 0.0178 μg/g in *Dicentrarchus labrax*, 0.0149 μg/g in *Mugil cephalus*, and 0.0157 μg/g in *Sparus aurata*) than the other regions. There is no maximum allowable limit for methoxychlor in neither 2006/77/EC, 2013/39/EU, nor TFC.

Particularly, organic chlorinated insecticides are stored in human fat tissues. This storage is limited not only to

Table 4: The residues of some of pesticides (μg/g) in the muscle meat of *Pomatomus saltatrix*, *Dicentrarchus labrax*, *Mugil cephalus*, and *Sparus aurata*.

Pesticide species	Izmir (mean ± SD)				Bandırma (mean ± SD)				Istanbul (mean ± SD)				Duzce (mean ± SD)			
	Pomatomus saltatrix	*Dicentrarchus labrax*	*Mugil cephalus*	*Sparus aurata*	*Pomatomus saltatrix*	*Dicentrarchus labrax*	*Mugil cephalus*	*Sparus aurata*	*Pomatomus saltatrix*	*Dicentrarchus labrax*	*Mugil cephalus*	*Sparus aurata*	*Pomatomus saltatrix*	*Dicentrarchus labrax*	*Mugil cephalus*	*Sparus aurata*
p-p′-DDE	0.0453 ± 0.000054	0.0207 ± 0.00095	0.1019 ± 0.0028	0.0089 ± 0.00032	0.0356 ± 0.0018	0.0819 ± 0.0024	0.1650 ± 0.0051	0.0065 ± 0.00023	0.0148 ± 0.0011	0.0509 ± 0.0025	0.1001 ± 0.0064	0.0036 ± 0.00021	0.0133 ± 0.00022	0.0305 ± 0.00061	0.1023 ± 0.00018	0.0029 ± 0.000084
RSD	1.2	4.6	2.8	3.7	5.1	3.0	3.1	3.6	7.5	5.1	6.4	6.1	1.7	2.0	1.8	2.9
α- BHC	—	—	0.0155 ± 0.000961	0.0401 ± 0.0028	—	—	0.0028 ± 0.0001204	0.0562 ± 0.0029	0.0026 ± 0.00024	—	0.0083 ± 0.00036	0.0456 ± 0.0044	0.0061 ± 0.00026	—	0.0045 ± 0.000504	0.0024 ± 0.000057
RSD	—	—	6.2	7.1	—	—	4.3	5.3	9.5	—	4.4	9.8	4.4	—	11.2	2.4
Endosulfan-sulfate	0.0110 ± 0.00035	0.0013 ± 0.000036	0.0214 ± 0.00040	0.0054 ± 0.00035	0.0093 ± 0.00035	0.0037 ± 0.00019	0.0196 ± 0.00072	0.0086 ± 0.00030	0.0115 ± 0.00039	0.0042 ± 0.00032	0.0214 ± 0.001	0.0046 ± 0.00026	—	0.0080 ± 0.00053	0.0146 ± 0.00027	0.0045 ± 0.00009
RSD	3.2	2.8	1.9	6.6	3.8	5.2	3.7	3.5	3.4	7.8	5.0	5.7	—	6.7	1.9	2.0
Endrin	0.0312 ± 0.0015	0.0644 ± 0.0032	0.2847 ± 0.027	0.0684 ± 0.00095	0.0231 ± 0.001	0.0598 ± 0.0057	0.2893 ± 0.011	0.0710 ± 0.0012	0.0420 ± 0.0005	0.0620 ± 0.0014	0.2957 ± 0.01	0.0750 ± 0.0045	0.0329 ± 0.0025	0.0549 ± 0.0051	0.1794 ± 0.0075	0.0976 ± 0.0039
RSD	4.9	5.0	9.7	1.4	4.7	9.6	4.0	1.8	1.2	2.4	3.7	6.0	7.8	9.3	4.2	4.0
Heptachlor	—	0.0084 ± 0.00043	0.0460 ± 0.0041	0.0094 ± 0.00015	—	0.0598 ± 0.0055	0.0081 ± 0.00039	0.0077 ± 0.00013	0.0082 ± 0.000098	0.0059 ± 0.00013	0.0340 ± 0.0016	0.0074 ± 0.00019	—	—	—	—
RSD	—	5.2	9.0	1.6	—	9.2	4.9	1.8	1.2	2.3	4.8	2.6	—	—	—	—
Methoxychlor	0.0222 ± 0.00066	0.0210 ± 0.00054	0.0137 ± 0.001	0.0123 ± 0.001	0.0326 ± 0.0023	0.0288 ± 0.00057	0.0155 ± 0.0012	0.0120 ± 0.00042	0.0226 ± 0.00085	0.0189 ± 0.0016	0.0119 ± 0.00059	0.0094 ± 0.00049	0.0511 ± 0.003	0.0178 ± 0.0015	0.0149 ± 0.00028	0.0157 ± 0.00037
RSD	3.0	2.6	7.5	8.6	7.3	2.0	7.9	3.5	3.8	8.7	5.0	5.3	6.0	8.6	1.9	2.4
p-p′-DDD	0.00036 ± 0.000018	0.0294 ± 0.0019	0.0059 ± 0.00042	0.0717 ± 0.0012	0.00078 ± 0.000056	0.0125 ± 0.0011	0.0017 ± 0.000081	0.0519 ± 0.0021	0.00045 ± 0.000028	0.0122 ± 0.00097	0.0068 ± 0.00033	0.0426 ± 0.0039	0.0197 ± 0.00061	0.0136 ± 0.0013	0.0092 ± 0.00052	0.1027 ± 0.0029
RSD	5.0	6.5	7.2	1.8	7.3	9.4	4.8	4.2	6.4	8.0	4.9	9.2	3.1	9.8	5.7	2.9
p-p′-DDT	0.00058 ± 0.000026	0.0019 ± 0.00017	0.0574 ± 0.0027	0.0972 ± 0.0019	0.0065 ± 0.00024	0.4324 ± 0.036	0.0046 ± 0.00033	0.0814 ± 0.0020	0.0142 ± 0.00069	0.0070 ± 0.00063	0.0036 ± 0.00014	0.0079 ± 0.00012	0.0278 ± 0.00041	0.0049 ± 0.00012	0.0080 ± 0.00039	0.1359 ± 0.0013
RSD	4.5	9.0	4.8	2.0	3.7	8.4	7.3	2.5	4.9	9.0	4.1	1.6	1.5	2.5	4.9	1.0
Cypermethrin	0.0053 ± 0.00015	0.0075 ± 0.000082	0.0519 ± 0.0012	0.0018 ± 0.00052	0.0050 ± 0.00021	0.0038 ± 0.00011	0.0082 ± 0.00029	0.0049 ± 0.00011	0.1143 ± 0.009	0.049 ± 0.0039	—	0.0069 ± 0.00018	0.0080 ± 0.0006	0.0092 ± 0.00052	0.0060 ± 0.00058	0.0093 ± 0.00062
RSD	4.5	1.1	2.5	2.9	4.3	2.9	3.6	2.4	7.9	8.0	—	2.7	7.5	5.7	9.7	6.7

adipose tissue but also within the fat material in the organs. This is a strong protection mechanism for mammals. The toxic substance disappears rapidly from the circulation and undergoes metabolism more slowly, and the toxic concentration is prevented from going into rapidly sensitive organs [34]. In a study by Cingi and Dökmeci, it was found that DDT, lindane, and dieldrin were mostly found in fat tissue and then in the brain and blood [35]. In addition, the excretion of dieldrin was found to be less and slower than the others. In a study conducted by Kelle, DDT, heptachlor, and BHC residues were detected in all of the human fat tissue samples and the detected values were found to be higher in the fat tissue samples of older people compared to the younger people. DDT also accumulates in the breasts of women, especially in breast milk, and can cause serious health problems in newborn babies [36]. In the present study, we observed p-p′-DDD residues in every fish sample from each region. The lowest and highest p-p′-DDD concentration were 0.00036 μg/g in the *Pomatomus saltatrix* sample from Izmir and 0.1027 μg/g in *Sparus aurata* sample from Duzce (Table 4). The concentration of p-p′-DDD can be listed as *Sparus aurata* > *Dicentrarchus labrax* > *Mugil cephalus* > *Pomatomus saltatrix* according to the fish species. When tested, p-p′-DDE was determined in every fish sample of each region. The highest concentration of p-p′-DDE was observed in the *Mugil cephalus* samples (0.1650 μg/g) especially from the Bandırma region. When we compared the residues of p-p′-DDE regionally, it is listed as Bandırma > Duzce > Izmir > Istanbul.

In general, p-p′-DDT, p-p′-DDE, and p-p′-DDD concentrations were determined in all fish species and regions. Maximum p-p′-DDT, p-p′-DDE, and p-p′-DDD concentrations are 0.4324 μg/g (*Dicentrarchus labrax* from Bandırma), 0.1650 μg/g (*Mugil cephalus* from Bandırma), and 0.1027 μg/g (*Sparus aurata* from Duzce), respectively. There is no MAC EQS biota for p-p′-DDT, p-p′-DDE, and p-p′-DDD in 2013/39/EU but Directive 2006/77/EC listed the maximum level of p-p′-DDT, p-p′-DDE, and p-p′-DDD as 0.05 μg/g for all feedstuffs. Obtained values are over the ML according to Directive 2006/77/EC. When we compare between the residues of p-p′-DDT, p-p′-DDE, and p-p′-DDD, residue of p-p′-DDT is higher than others. DDE is caused by the disintegration of DDT in the living body. In many animals, it emerges as a product of the body's effort to purify itself from DDT. DDD is caused by the metabolism of DDT in some organisms. In fact, p, p′-DDE, o, p′-DDE, p, p′-DDD, and p, p′-DDD are the disintegration products of DDT. Therefore, DDT concentration is higher than the others and this is the usual result and the high concentration of DDT shows that the pollution in that ecosystem is based on the old times.

The MAC of cypermethrin (CYP) is 6×10^{-4} μg/L and 6×10^{-5} μg/L for inland surface and other surface waters in 2013/39/EU. There is no stated MAC for EQS biota for fish. We obtained cypermethrin residues in every species of samples from each region, except *Mugil cephalus* from Istanbul (Table 4). The obtained results are significant and quite high. The *Pomatomus saltatrix* sample had 0.1143 μg/g cypermethrin concentration from Istanbul. The high concentration of CYP is alarming and must be under control.

β-BHC, aldrin, dieldrin, heptachlor epoxide, and endosulfan were studied in every fish sample (*Pomatomus saltatrix*, *Dicentrarchus labrax*, *Mugil cephalus*, and *Sparus aurata*) from each region and we did not determine any residues in the samples.

4. Conclusion

In the present study, we carried out the analysis of some heavy metals and pesticides that create a danger to the ecological system and human health in four different fish samples obtained from the Aegean and Marmara Seas. We tried to draw attention to heavy metals and pesticide accumulations in the fish living in the Aegean and Marmara Seas. We investigate the residues of As, Mn, Hg, Ag, Cd, Ni, Zn, Cu, and Pb in muscle meat and internal organs of fish samples. The maximum levels for Hg, Pb, Cd, and Sn in foodstuffs are revealed in Commission Regulation 1881/2006 EC, which is the framework for EU legislation setting maximum levels for chemical contaminants in food. Unfortunately, there are no maximum limits for arsenic, manganese, nickel, copper, and zinc levels in food at the EU level; these heavy metals do not fall within the scope of this Regulation.

Directive 98/83/EC regulates the concentration of Cd, Cr, Cu, Pb, Hg, and Ni on the quality of water intended for human consumption. Considering the obtained results in this study, especially pertaining to arsenic concentration, it is necessary to make an arrangement to determine the maximum concentration limits in food/fish for As, Mn, Ni, Cu, and Zn, which are not currently regulated in directive 1881/2006 EC, in the near future. The MAC EQS biota of heptachlor is defined as 6.7×10^{-6} μg/g in 2013/39/EU. The highest heptachlor concentration is nearly nine thousand times more than the MAC EQS biota of heptachlor listed in 2013/39/EU. This is a serious issue that should be taken under control urgently.

Endosulfan, cypermethrin, heptachlor, and endrin were stated in the list of priority substances in the field of water policy in directive 2013/39/EU and endosulfan, endrin, heptachlor, p-p′-DDD, p-p′-DDT, dieldrin, and cypermethrin were listed in Annex I in directive 2013/39/EU. We observed and determined remarkable residues for α-BHC, p-p′-DDD, p-p′-DDT, p-p′-DDE, endosulfan sulfate, endrin, heptachlor, methoxychlor, and CYP in the muscle meat of *Pomatomus saltatrix*, *Dicentrarchus labrax*, *Mugil cephalus*, and *Sparus aurata* from four different regions of Turkey.

Among these pesticides analyzed in this study, only heptachlor has MAC EQS biota in 2013/39/EU. When we consider residues of pesticides determined in this study, the obtained results are significant and quite high and should be taken under control urgently.

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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Supplementary Materials

Determination of some heavy metals and pesticides in different types of fish samples. (*Supplementary Materials*)

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