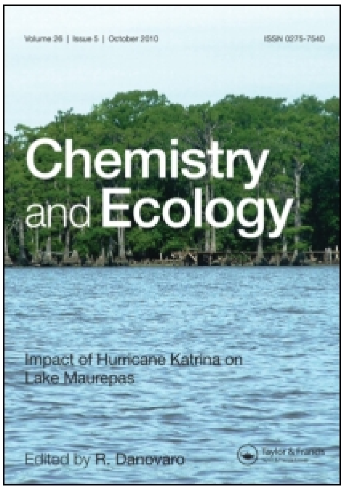

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DDT in fish from the Bulgarian region of the Black Sea

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In spite of a worldwide reduction in the utilization of organochlorine pesticides (OCPs), they are still a problem for the aquatic environment and human health. The Black Sea is still being polluted with persistent chemicals, including OCPs. Aquatic organisms (sprat, scad, bluefish, shad, belted bonito, goby, and black mussel) with different feeding behaviours were sampled on a seasonal basis from the Bulgarian region of the Black Sea, and the concentrations of 13 OCP residues were determined. Although many of the OCPs were not detected in the samples, in all samples 1,1,1-trichloro-2,2-bis(4-chlorophenyl) ethane (DDT) was present mainly in the form of its metabolites 1,1-dichloro-2,2-bis(4-chlorophenyl) ethane (DDD) and 1,1-dichloro-2,2-bis(4-chlorophenyl) ethylene (DDE). Only about 12% of the total DDT was present as the parent compound pp-DDT, which suggests that it was not being used recently in the region. The total DDT concentrations were generally below 150 $\mu g\, kg^{-1}$ fresh weight, but higher levels—up to 354 $\mu g\, kg^{-1}$ fresh weight—were also measured for fish species with a high fat content. Between-species differences were observed, even when the concentrations were presented on a fat-level basis. DDT concentrations did not show any significant changes over the 2-yr sampling period. Fish sampled in the northern areas of the Bulgarian Black Sea coast seemed to contain higher DDT levels than those from the southern areas, suggesting a major (historical) influence of the Danube River. For permanent monitoring purposes, the utility of Black Sea gobies and scad should be considered.

Keywords: Organochlorine pesticides; DDT; Fish; Black Sea

1. Introduction

Organochlorine pesticides (OCPs) have been widely used in the past, but due to their toxicity, stability, and bioaccumulation, especially in the aquatic food web, they can still be a current problem for human health through fish consumption. Many of these compounds, like 1,1,1-trichloro-2,2-bis(4-chlorophenyl) ethane (DDT), have endocrine-disrupting activities and are possible carcinogens for humans [1–3].

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The Black Sea is the world's largest natural anoxic water basin below 180 m in depth. It is a closed sea with a very high degree of isolation from the world's oceans, but it receives freshwater inputs from some of the largest rivers in Europe: the Danube, the Dniester, and the Dnieper [4]. For this reason, the Black Sea is considered one of the most polluted seas, and the increasing concentrations of nutrients in recent years have led to a higher degree of eutrophication. The fishery yield has declined dramatically, and the tourism industry has also suffered from serious pollution of the Black Sea.

The biogeochemistry of DDT in Black Sea was studied in the sediments [5] and waters [6]. Both studies concluded that in spite of the regulations, there has possibly been recent use of DDT in the region. The Danube and the rivers from the Russian Federation [7] and Turkey [8] have been discharging large quantities of this pesticide in the Black Sea. The pesticide DDT and its metabolites 1,1-dichloro-2,2-bis(4-chlorophenyl) ethane (DDD) and 1,1-dichloro-2,2-bis(4-chlorophenyl) ethylene (DDE) were the main organohalogen contaminants recently measured in sediments and biota samples from the Danube delta [9]. However, apart from one study of DDT in the harbour porpoise [10], to our knowledge there have been no investigations of the pollution of fish from the Black Sea with OCPs, like DDT. The aim of this study was to investigate the current levels of pollution with DDT and other OCPs and the seasonal/geographical variation of their concentrations in different aquatic organisms sampled from the Bulgarian Black Sea coast.

2. Materials and methods

2.1 *Sampling*

After studying the fish market and fishing practices in Bulgaria, several of the most commercial fish species were selected for this study. These species were sprat (*Sprattus sprattus*), scad (*Trochurus mediterranens*), bluefish (*Pomatomus saltatrix*), shad (*Alosa pontica*), and belted bonito (S*arda sarda*). Black sea gobies (*Mesogobius batrachocephalus*, *Neogobius melanostomus*, *Neogobius ratan*) were also selected because they are non-migratory species and feed mainly on benthic organisms. Three black mussel samples (*Mytilus galloprovincialis lam*) were used to compare the DDT concentrations with those found in fish.

In order to study the seasonal variations, most of the species were sampled twice a year for a period of 2 yr. The bluefish samples were collected only in the autumn. The study area is presented in figure 1. With this sampling strategy, it was possible to cover the entire Bulgarian Black Sea coast and to include three important fishing regions: north (near Cape Kaliakra); Varna Bay and south (Bourgas, Nessebar, Rezovo).

2.2 *Analysis*

The analytical method for determination of residues of OCP has been described in detail elsewhere [11]. The fish and black mussels were frozen (−18 °C) after sampling. Each sample was about 5 kg, and half was kept frozen as a control. In the laboratory, the whole bodies of the fish and the black mussels (except the shells) were homogenized to pool samples of about 2 kg each, and subsamples of 10 g were taken from them for extraction. The OCPs were extracted with methylenechloride/hexane (3/1 v/v) in a Soxhlet apparatus; the extracts were evaporated to allow the determination of lipid content and purified by column chromatography with Florisil (Carlo Erba). The OCPs were determined by capillary gas chromatography with an electron-capture detector. Thirteen OCPs were measured: aldrin, dieldrin, α-, β-, and

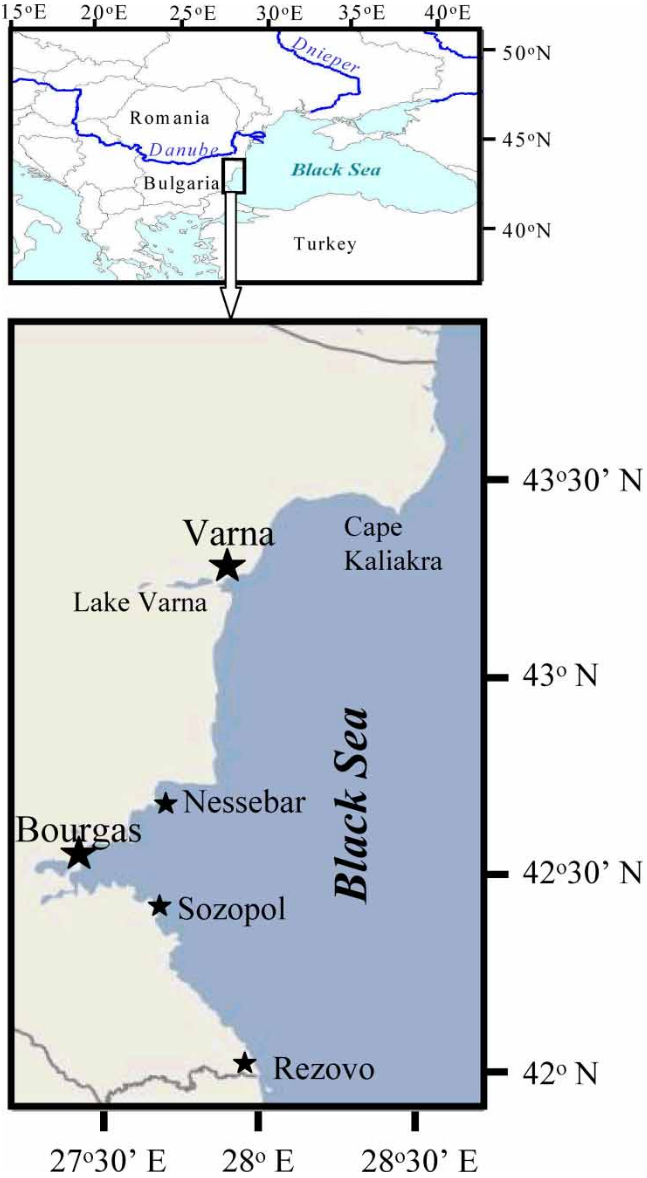

Figure 1. Map of the Bulgarian Black Sea coast with the main regions of sampling.

γ-hexachlorcyclohexane isomers (HCH), heptachlor, heptachlor epoxide, endrin, pp-DDT, op-DDT, pp-DDD, op-DDD, and pp-DDE. The recoveries were determined by analysing fish samples, spiked with a mixed standard (Supelco, Bellefonte, PA) and were between 76.4% and 108.1%. The method detection limit for α-, β-, and γ-HCH, heptachlor, heptachlorepoxide, aldrin, dieldrin, and pp-DDE was $0.2\,\mu\text{g kg}^{-1}$ fresh weight. For endrine, it was $0.3\,\mu\text{g kg}^{-1}$. For op-DDD, pp-DDD, and pp-DDT, it was $0.4\,\mu\text{g kg}^{-1}$, and for op-DDT $0.5\,\mu\text{g kg}^{-1}$.

The statistical analysis of the data was based on the comparison of average values or assessing the significance of correlation by a t-test [12].

3. Results and discussion

Most of the OCPs studied were not detected in the fish samples. Some of them, like HCH isomers, were occasionally detected and will be the subject of a future investigation. However, pp-DDE was present in all samples with most of them also containing pp-DDD and the parent compound pp-DDT. The sum of concentrations for these three compounds was reported here as total DDT ($\mathrm{DDT_{TOT}}$).

3.1 *Levels of pollution*

The average values, standard deviations, and ranges for the lipid content and the concentrations of pp-DDT, pp-DDE, and pp-DDD are summarized in table 1, on the basis of both fresh weight and fat levels. The concentrations of DDT and its metabolites in fish from the Black Sea were similar to those found in the case of fish from other polluted marine environments [13, 14]. Fish from the Black Sea were more polluted with DDT compared with fish from the Adriatic Sea [15, 16] and North Sea [17]. The concentrations of $\mathrm{DDT_{TOT}}$ in fish from the Black Sea were higher than the reference value of 14 $\mu\mathrm{g\,kg^{-1}}$ fresh weight recommended for protection of aquatic life [18]. The possible influence on the health of large predators should be a subject of future investigation.

3.2 *DDT compositions*

The concentrations based on fresh weight of pp-DDE, pp-DDD, and pp-DDT are presented in figure 2 as a function of $\mathrm{DDT_{TOT}}$ for all samples. There are good linear dependencies ($p = 0.01$) irrespective of the species showing similar DDT composition of all Black Sea fish and the mussel samples studied. The main form is pp-DDE, accounting for 57 % of $\mathrm{DDT_{TOT}}$, followed by pp-DDD (31%) and the parent insecticide pp-DDT (12%), showing the lack of significant new input of pp-DDT from the Bulgarian coastline. These values observed were similar to those found in the fish from the Bering Sea, the Gulf of Alaska, and the Sea of Japan [19], and different for fish from the Salton Sea [20] and the China Sea [13] where more recent input of the parent insecticide was noticed. The distribution of DDT and its metabolites in harbour porpoise from the Black Sea, sampled in 1993 [10], showed 46% pp-DDE, 34% pp-DDD, 16% pp-DDT, and 4% op-DDT. These values were similar to those found in the current study but showed a lower DDE/DDT ratio. This difference may reflect the decomposition of the parent insecticide in the decade between 1993 and 2003 with a half-life for pp-DDT of more than 10 yr.

3.3 *DDT and lipid content*

In figure 3, the $\mathrm{DDT_{TOT}}$ concentration is represented as a function of lipid content for the Black Sea fish and mussels studied. Even if, for the individual species, there was often no relationship, the points for all samples followed a statistically significant non-linear increase in the $\mathrm{DDT_{TOT}}$ concentrations with fat content ($R^2 = 0.60$, $p = 0.01$). As shown in other studies, the variation of $\mathrm{DDT_{TOT}}$ concentration in marine species can be quite complex and shows, in some cases, a linear relationship with lipid content [21] and for other cases no relationship at all [22].

Table 1. Summary of the results (average values ± standard deviations and ranges) for the concentrations of DDT and its metabolites DDD and DDE for fish and black mussel species from the Bulgarian part of the Black Sea.

	Lipids (%)	pp-DDE ($\mu g\,kg^{-1}$ f.w.)	pp-DDD ($\mu g\,kg^{-1}$ f.w.)	pp-DDT ($\mu g\,kg^{-1}$ f.w.)	pp-DDE ($\mu g\,kg^{-1}$ fat)	pp-DDD ($\mu g\,kg^{-1}$ fat)	pp-DDT ($\mu g\,kg^{-1}$ fat)
Black mussel	3.0 ± 0.3	6.3 ± 1.0	2.4 ± 1.9	0.9 ± 1.2	213 ± 26	85 ± 68	30 ± 39
($n = 3$)	(*2.6–3.3*)	(*5.3–7.3*)	(*<0.4–3.6*)	(*<0.4–2.3*)	(*192–242*)	(*<15–131*)	(*<15–75*)
Bluefish	16.1 ± 4.6	52.5 ± 18.3	24.8 ± 9.7	10.7 ± 4.5	328 ± 93	156 ± 52	68 ± 27
($n = 6$)	(*10.6–23.5*)	(*23.0–78.1*)	(*9.9–36.7*)	(*5.8–17.6*)	(*218–440*)	(*93–213*)	(*34–99*)
Shad	23.5 ± 3.8	119.4 ± 51.0	66.3 ± 22.1	24.7 ± 10.8	518 ± 239	290 ± 121	107 ± 49
($n = 11$)	(*19.2–30.8*)	(*54.3–225.2*)	(*30.4–108.5*)	(*8.7–40.8*)	(*283–927*)	(*158–566*)	(*36–189*)
Belted bonito	12.9	21	11.5	6.1	150	84	44
($n = 2$)	(*7.4–18.3*)	(*8.7–33.3*)	(*5.3–17.8*)	(*2.6–9.7*)	(*118–182*)	(*71–97*)	(*35–53*)
Goby	4.6 ± 2.5	20.2 ± 13.5	7.2 ± 4.7	3.0 ± 3.0	653 ± 885	212 ± 200	70 ± 87
($n = 12$)	(*1.4–9.2*)	(*5.2–45.7*)	(*2.0–17.5*)	(*<0.4–10.9*)	(*115–3310*)	(*37–699*)	(*<15–319*)
Scad	11.2 ± 7.6	27.1 ± 22.8	17.9 ± 23.2	5.4 ± 5.7	319 ± 309	170 ± 170	68 ± 62
($n = 9$)	(*2.4–21.5*)	(*2.9–61.9*)	(*<0.4–67.9*)	(*<0.4–18.6*)	(*61–855*)	(*<15–489*)	(*<15–165*)
Sprat	9.4 ± 4.5	30.5 ± 6.5	17.9 ± 3.8	6.8 ± 1.8	472 ± 462	272 ± 243	105 ± 104
($n = 13$)	(*1.7–16.0*)	(*20.6–40.5*)	(*13.3–24.7*)	(*4.1–9.9*)	(*198–1911*)	(*103–1005*)	(*36–429*)

Note: The concentrations are presented independently from the sampling sites and periods. The data are given both on a fresh weight and on a fat-level basis. In cases where the substance is not detected, half of the detection limit is used in the calculation.

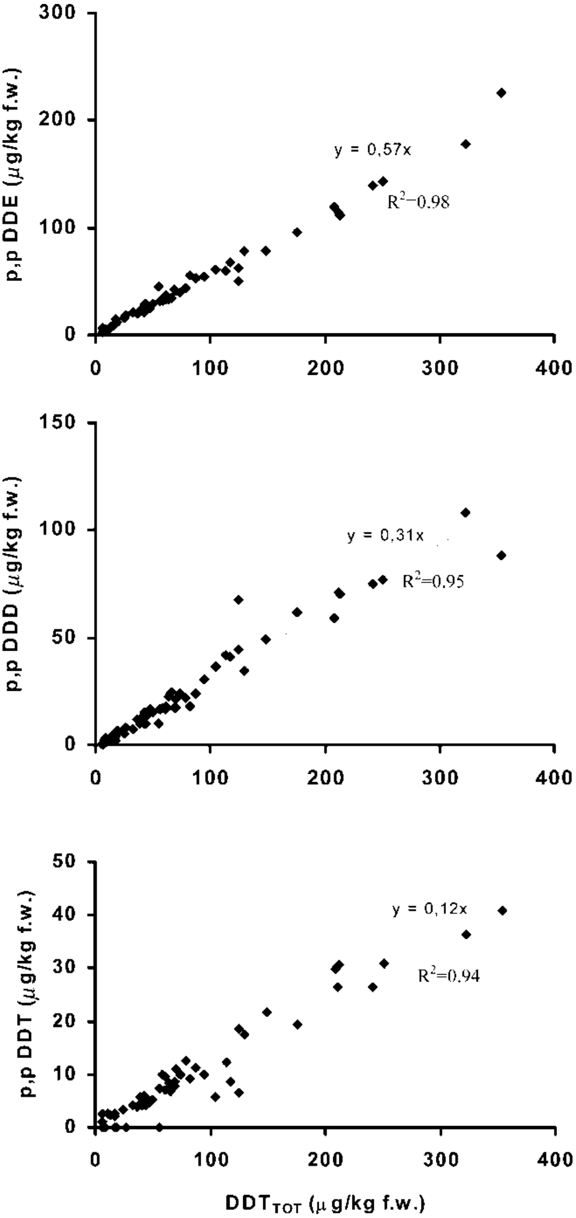

Figure 2. Concentrations of DDT and its metabolites DDE and DDD as a function of DDT_{TOT} for fish and mussels from Bulgarian part of the Black Sea (all samples).

3.4 *Temporal variations*

In table 2, the temporal variations of DDT_{TOT} concentrations were expressed on a fat-level basis for those fish species for which seasonal data are available (bluefish, shad, goby, scad, and sprat). There were no statistically significant ($p = 0.05$) seasonal differences of the average DDT_{TOT} concentrations for each species which could possibly be explained by the large standard deviations and the limited data. The average concentration of DDT_{TOT} in all of these fish for the first year of the study (autumn 2003 and spring 2004, $n = 26$) was 751 ±

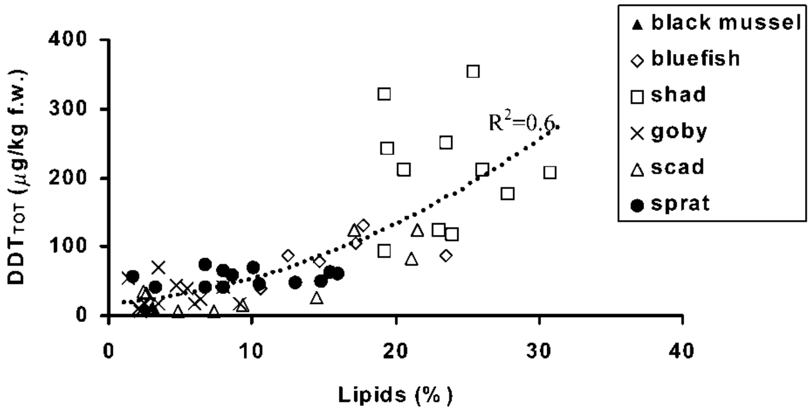

Figure 3. Total DDT concentrations vs lipid content in fish and mussels from Bulgarian part of the Black Sea.

Table 2. Seasonal dynamics of DDT$_{TOT}$ concentrations ($\mu g\,kg^{-1}$ fat) for fish from the Bulgarian part of the Black Sea.

	Autumn 2003	Spring 2004	Autumn 2004	Summer 2005
Bluefish	448 ± 136 (*367–605*), $n = 3$	–	657 ± 104 (*538–732*), $n = 3$	–
Shad	985 ± 458 (*492–1396*), $n = 3$	952 ± 634 (*542–1682*), $n = 3$	778 ± 269 (*492–1027*), $n = 3$	959 (*677–1242*), $n = 2$
Goby	487 ± 280 (*191–748*), $n = 3$	431 (*347–515*), $n = 2$	867 ± 810 (*299–2048*), $n = 4$	1811 ± 1923 (*488–4017*), $n = 3$
Scad	352 ± 248 (86–578), $n = 3$	833 (159–1509), $n = 2$	461 (190–732), $n = 2$	686 (124–1248), $n = 2$
Sprat	*798 ± 386* (*378–1305*), $n = 4$	*1374 ± 1707* (*362–3345*), $n = 3$	*808 ± 261* (*632–1107*), $n = 3$	*432 ± 98* (*337–533*), $n = 3$

Note: The data are presented as average values ± standard deviations, and ranges and the number of samples (n) are also given.

675 $\mu g\,kg^{-1}$ fat, and for the second year (autumn 2004 and summer 2005, $n = 25$) it was 845 ± 775 $\mu g\,kg^{-1}$ fat. For the 2-yr period studied, there was no observable difference in the DDT$_{TOT}$ concentration ($p = 0.1$) which was in accordance with the persistence of DDT and its metabolites in the environment. However, much higher concentrations were measured in fish from the Danube delta about 30 yr ago [23] showing the decline of DDT levels in fish over longer time periods. For this 2-yr study, the temporal change can be excluded from further investigation and between-species differences and geographical variations will be considered irrespective of the sampling time.

3.5 *Between-species differences*

There were up to two orders of magnitude between-species concentration differences of DDT$_{TOT}$ on a fresh weight basis (see table 1 and figure 3). The mussel samples presented much lower concentrations of DDT and metabolites compared with fish, mostly due to their low lipid content. Similar concentrations were measured in the same mussel species from the Turkish coast of the Black Sea [24]. On the other hand, oily fish, like shad, showed a much higher DDT$_{TOT}$ concentration, attaining 354 $\mu g\,kg^{-1}$ fresh weight.

A better comparison between species may be obtained by expressing the DDT concentrations on a fat level basis. Even if more uniform, the results on this basis still revealed some between-species differences in DDT concentration (table 1) due to the different position of the organisms in the aquatic food web. For example, the DDT concentrations, expressed on a fat basis, were significantly lower (one-sided t-test) for mussels compared with bluefish ($p = 0.05$), goby ($p = 0.05$), sprat ($p = 0.05$), and shad ($p = 0.02$). Shad also showed significantly higher DDT_{TOT} levels on a fat basis compared with scad ($p = 0.05$) and bluefish ($p = 0.01$).

3.6 *Geographic variation*

In figure 4, the geographical variations were presented for DDT_{TOT} concentrations on a fat-level basis for goby, scad, and shad. The results for goby and scad showed that the northern coast was generally more polluted with DDT than the southern coast, possibly due to historical discharges from the Danube into the Black Sea [7]. Point sources of DDT are also possible. For example, the highest concentrations were measured in samples from the Varna Bay area. It is known from the literature that local sources, like urban centres, could pollute coastal waters [13, 25]. Most of the other species studied did not present any trend in the geographical variations and could not be used to assess the sources of pollution. The utility of the Black Sea gobies and scad for biomonitoring purposes should be considered as indicator species for a future evaluation of the Black Sea pollution with DDT.

3.7 *DDT intake by Black Sea fish consumption*

According to the data of the National Statistics Institute, the fish consumption in Bulgaria is low—between 2.1 and 4.6 kg yr^{-1} with the corresponding probability distribution for it

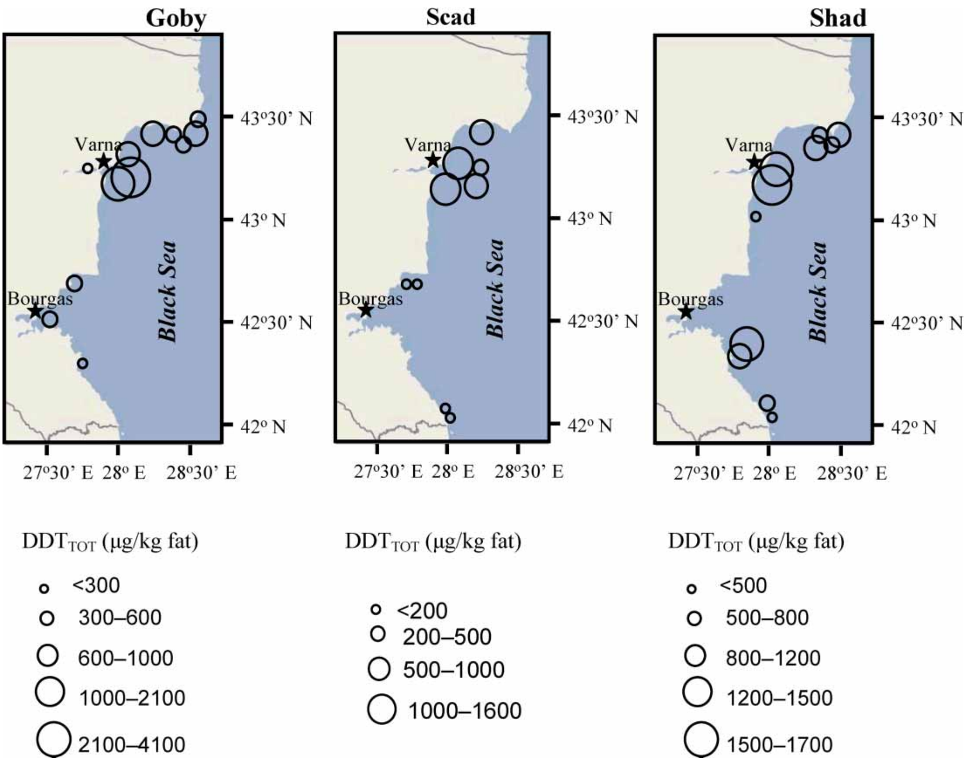

Figure 4. Regional distribution of DDT_{TOT} for goby, scad, and shad.

denoted as P_{fish} [26]. The values for the fish intake (kg yr^{-1}) and the corresponding P_{fish} (%) were as follows: 2.1 (9.9%), 2.4 (11.3%), 2.7 (15.2%), 3.0 (17.7%), 3.3 (17.0%), 3.6 (12.8%), 3.9 (6.7%), 4.2 (2.1%), and 4.7 (7.4%). On the other hand, from the measured DDT_{TOT} concentrations, found for all fish samples, the corresponding probability distribution function P_{conc} could be estimated. For DDT_{TOT} concentrations of less than 30 $\mu\text{g kg}^{-1}$ fresh weight, the corresponding P_{conc} value was 26.8%; for the range 30–90, 90–180, and 180–360 $\mu\text{g kg}^{-1}$, the corresponding values were 46.4%, 14.3%, and 12.5%, respectively. The DDT intake from fish consumption was estimated by multiplying the quantity of fish eaten annually by Bulgarians [26] and the DDT_{TOT} concentrations found in this study. For each DDT intake value, the corresponding probability of the intake $P_{\text{DDT}} = P_{\text{fish}} P_{\text{conc}}$ can be estimated.

The DDT_{TOT} intake was less than 0.5 mg yr^{-1} for 86% of the population and between 0.5 and 1.2 mg yr^{-1} for 14% of the population, and did not exceed the EPA reference value [2]. However, the calculated value took into account only raw fish from the Black Sea and was probably overestimated. The lack of consumption data for different fish species also did not permit more precise calculations. A future study of fish found in the market (not only from the Black Sea) and the eventual decrease in DDT concentration upon cooking [27] is necessary in order to determine the real DDT intake. Special emphasis should be given to human subgroups with increased fish consumption. However, almost certainly, DDT from Black Sea fish does not represent a significant danger to human health due mostly to the low fish consumption rates of the Bulgarian population.

4. Conclusions

This study of DDT levels in different aquatic organisms from the Black Sea provides useful information both for assessing human health and ecological risks of pesticide pollution. For a future evaluation of the Black Sea pollution with DDT, the utility of goby and scad for biomonitoring purposes should be considered.

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