Fate of pesticides in soil in a coastal lagoon area and associated water quality impacts

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Abstract Pesticides used on agricultural lands are among the significant diffuse sources of pollutants. They are poisons and can be particularly dangerous when misused and/or applied without care. Although certain characteristics of pesticides are well known, their final characteristics after they reach a waterbody are extremely difficult to estimate. Regarding the soil and water environment, it is necessary to conduct both a field study and laboratory analysis. Such a study has been performed in the catchment area of Dalyan Lagoon, Turkey, joining the Mediterranean Sea, that was selected as the pilot region. The input of pesticides, transport in soil and transfer to the water environment together with their probable impacts on water quality are determined by conducting detailed field surveys and water residue analyses. Within the scope of the study, the behavior and fate of pesticides both in soil and water are investigated in detail. Water residue experiments are conducted on four sets of water samples representing the seasons of the year 1999–2000 for the pre-selected 6 pesticides. Two sets of water samples (surface and bottom) are taken from the 16 stations along the lagoon channel and two lakes within the catchment area. The results and findings show the significance of the use of pesticides as they exist in the water environment around and/or slightly above the limits stated in the national current regulations except dichlorvos which presents very high values compared to its tolerance limit. Of particular interest, the results are used to enlighten the farmers and the public and increase awareness of pesticides as contaminants in valuable coastal waters.

Keywords Agricultural pollution; Dalyan Lagoon; pesticides; trend; water quality; water residue analyses

Introduction

Even though Turkey has the largest agricultural area in Europe, she uses the least quantity of pesticides per hectare (Tanik *et al.*, 1998). However, one of the problems that the country faces at the beginning of the new century is the careless practice and/or misuse of pesticides causing probable detrimental effects on water resources. In this paper, the fate of pesticides in soil and associated water quality impacts will be investigated in a selected catchment area of Dalyan Lagoon at the southwest of Turkey ending in the Mediterranean Sea. The area is approximately 130 km^2, out of which 23% is devoted to agriculture, and is one of the sensitive coastal areas of the country where the typical Mediterranean crops; cotton, citrus fruits, wheat, corn and peas are grown and horticulture is practised. Main features of the lagoon and its catchment area are given in Tanik *et al.* (2001). Current annual pesticide consumption and distribution in terms of crops, frequency and type of application are determined through field surveys. The fate of pesticides in soil, considering the main mechanisms like persistence and mobility, is evaluated together with their physical and chemical characteristics to find out how much is lost via surface runoff or by leaching. To predict in a general sense how a pesticide behaves in the environment, many intrinsic properties like soil characteristics and climatic conditions, besides pesticide properties, help to make a rough estimation of its transportation route (Hutson and Roberts, 1994; Rào *et al.*, 1999). Pesticides used in the area are evaluated and classified according to their chemical class, structure, form, vapor pressure, solubility in water, DT_{50} (time necessary for the first 50% of pesticide to dissipate), GUS (groundwater ubiquity score), KO_C (organic carbon partition coefficient), stability, toxicological classification based on WHO and EPA criteria, etc. (Guvensoy, 2000; Un, 2000; Tanik *et al.*, 2000).

Pesticide use in the area is approximately 12 kg-lt/ha which is quite high compared to the overall annual consumption of Turkey (1.25 kg-lt/ha) and of Megacity Istanbul (3.5–4 kg-lt/ha) (Zeren, 1997; Tanik *et al.*, 1999). Based on the properties of pesticides, they are classified in three groups according to pollution potentials and to their transportation route; those reaching the water media through surface runoff, those through leaching and those in a transient condition. Tables 1, 2 and 3 summarize the main soil and water reactions of these classified pesticides together with their annual consumption, half-lives (Extoxnet, 1996; BCPC, 1998) and toxicity class according to WHO standards for oral LD_{50} (mg/kg of male mice) (Ware, 1994).

Only 6 typical pesticides are selected for further water residue analysis to discuss their water quality impacts. Deltamethrin, diazinon and endosulfan belong to Group 1, where endosulfan is one of the most used pesticides in the area, diazinon is known with its long half-life in water (HL_W) and deltamethrin has unknown HL_W. Dichlorvos and metalaxyl are representatives of Group 2, where dichlorvos has high toxicity and is publicly known to be consumed like aspirin in the country and has no substitutes, although the officially stated consumption value is quite low, and metalaxyl has comparably higher HL_W. Methidahion belongs to Group 3 with high toxicity and unknown HL_W.

Experimental study

Sampling stations

Pesticide residue analyses were conducted on water samples taken along the channel and from the two lakes (Alagol and Sulungur) situated on both sides of the channel, to detect the impact of selected pesticides on water quality. 16 stations were selected in the lagoon system whose locations are shown in Figure 1. Such a selection depended on various factors, summarized as follows. The stations along the main channel and its branches are needed to detect the spatial variations (Station nos 1, 2, 15, 6, 7, 13 and 5). Stations constituting the boundary conditions for the system are nos: 0 and 14. Stations representing the lake systems are 3 and 4 for Alagol and 8, 9, 10 11, 12 for Sulungur Lake. Residual pesticide analyses were performed seasonally where two sets of samples (surface and bottom) were taken from each station as horizontal and vertical salinity gradients were observed throughout the cruises. Surface water samples were collected from 0.5 m depth. Bottom water sampling depths were chosen according to the depth of the stations and vertical salinity gradients. The cruises took place in: April 1999 (Spring), August 1999 (Summer), November 1999 (Autumn) and March 2000 (Late Winter) further mentioned as Set 1, Set 2, Set 3 and Set 4 for evaluating seasonal variations. Out of the six selected pesticides, four had appeared (endosulfan, deltamethrin, dichlorvos and diazinon) in the aquatic system.

Experimental method

The pesticide residue analyses in water samples were conducted at the Laboratory of Mersin University, Department of Environmental Engineering. The AOAC Official Method 985.22 "Organochlorine and Organophosphorus Pesticide Residues" method and a Hewlett Packard 6890 GC instrument with an HP autosampler were used during the experiments. Samples were injected in the splitless mode into a HP5 capillary column coated with 5% phenyl methyl siloxan (30 m × 0.32 mm × 0.25 μm). N_2 was used as the carrier gas. The injection temperature was 240°C. The column was programmed from 100°C to 215°C at 5°C/min with NPD and μECD detectors at 270°C and 290°C, respectively.

Table 1 Properties of pesticides likely to appear in surface water – Group 1

Pesticide name	Consumption (kg/year)	Toxicity class[1]	HL_S (days) *	HL_S (days) **	HL_S (days) ***	Soil reactions *	HL_W (days) *	Water reactions *
Abamectin	425	IV	8hr-1d			microbial degradation, rapid photodegradation	4	rapid degradation, did not hydrolyze, photodegradation
Captan	420.8	List 5	1–10		3		15	rapid degradation
Copper Sulfate	32917.5	II				adsorption to organic material on soil		persistent, binds to water particles and sediment
Chlorfluazuaran	667	IV						
Chlorpyrifos	2551.5	II	60–120	28	30	degradation by UV, chemical hydrolysis	60–120	hydrolysis, volatilization, photolysis
Deltamethrin	448	II	7–15			degradation by microbes		uptake by plants, evaporation, rapidly adsorbed
Diazinon	170	II	14–28	30	40	degradation by microbes	180	degradation
Endosulfan	3577	II	50	42	120	degradation by bacteria and fungi	21	degradation
Furathiocarb	600	Ib						
Glyphosate	2867	III	47			degradation by microbes, adsorption to soil	12–70	microbial degradation, adsorbed to suspended organisms
Iprodione	94.9	List 5	14			degradation by soil microbes		rapid degradation under aerobic conditions
Lufenuron	267.5	III						
Mancozeb	306.2	List 5	1–7	15		rapid degradation	1–2	degradation
Maneb	277.1	List 5	12–36	15		breakdown in aerobic and anaerobic conditions	1hr	degradation
PCNB	8362	List 5	21d–1yr			evaporation, biotransformation, breakdown by soil bacteria	2–5	volatilization, unchanged by sunlight, stable, adsorbed by biota
Procymidone	94.8	List 5			120			
Tebuconazele	37.4							insoluble, adsorbed to particles and particulate
Trifluralin	2417		45–180			degradation by soil microbes, decomposed by UV volatilization		
Tetradifon	1021.5	List 5			60			
Thiocylam	562.5	II						
Hydrogen-Oxalate								
Thiram	45	III	15			degradation by microbes, photodegradation		hydrolysis and photodegradation

HL_S: Half-life in soil, HL_W: Half-life in water, [1] (Ware, 1994)
* (Extoxnet, 1996), ** (Unlu *et al.*, 1997), *** (Rao *et al.*, 1999)

Table 2 Properties of pesticides likely to appear in groundwater – Group 2

Pesticide name	Consumption (kg/year)	Toxicity class[1]	HL_S (days) *	HL_S (days) **	HL_S (days) ***	Soil reactions *	HL_W (days) *	Water reactions *
2–4 D	83	III	<7	15		degradation by microbes	>7	microbial degradation
Cadusafos	960	III						
Carbosulfan	2683	II		290				
Carboxin	596	List 5	3			rapid degradation	7	oxidation, microbial degradation
Dazomet	3110	III						
Dichlorvos	210	Ib	7			hydrolysis, biodegradation	4 hydrol. 57–400 volatiliz.	biodegradation, hydrolysis, volatilization
Formothion	883	II	1–14					
Metalaxyl	202.8	III	70		21		>28	
Monocrotophos	219	Ib	<7			biodegradation		
Propineb	420.8	List 5						

Table 3 Properties of pesticides likely to appear both in groundwater and surface water (transient condition) – Group 3

Pesticide name	Consumption (kg/year)	Toxicity class[1]	HL_S (days) *	HL_S (days) **	HL_S (days) ****	Soil reactions *	HL_W (days) *	Water reactions *
Bromopropylate	530.35	III		481				
Cypermethrin	235	II	4–56			degradation by microbes		
Fenamiphos	205	Ia	50			breakdown in aerobic conditions	4 hr	rapid degradation, stable in neutral water under dark conditions
Methidathion	285.5	Ib	5–23		3–18	degradation by soil microbes		

HL_S: Half-life on soil, HL_W: Half-life in water, [1] (Ware, 1994)
* (Extoxnet, 1996), ** (Unlu *et al.*, 1997), *** (Rao *et al.*, 1999), **** (BCPC, 1998)

Results and discussion

The results of the experiments indicated that two of the selected pesticides (methidathion and metalaxyl) were not detectable in the water samples ($<10^{-4}$ µg/L). Methidathion has low mobility in soil and rapidly degrades by chemical, photolytic and biological processes with a representative half-life of 7 days in soil.

None of the methidathion and its breakdown products have been detected in any groundwater resources (Extoxnet, 1996). In this study, methidathion appeared only in the first set of samples (April 1999) and was detected in two stations, one at the entrance of the channel (Station 0) and the other in the middle of the reed beds (Station 15) at very low concentrations. This is probably due to its short half-life in soil and water, relatively low amount of application and application frequency, which is once a year. Metalaxyl was not detected in any of the sets and stations. Its typical half-life in moist soils is about 70 days; however, increased sunlight may increase the rate of breakdown in the soil. It is poorly sorbed by soils and highly soluble in water. Exposure to sunlight reduces the half-life to 1 week (Extoxnet, 1996). As the pilot region bears typical Mediterranean climatic conditions, half-life may be low and thus it cannot be detectable. The data on the other four selected pesticides are given in Tables 4 and 5 and results are discussed separately, in turn. Values are to be evaluated according to the current Turkish Aquatic Products Regulation (TAPR, 1995) where 51 common pesticides used in Turkey are listed with their acceptable concentrations in the water environment. The tolerable limits for 3 of the pesticides; endosulfan, dichlorvos and diazinon have been stated in the current legislation as 0.2 µg/L, 0.07 µg/L, and 0.9 µg/L.

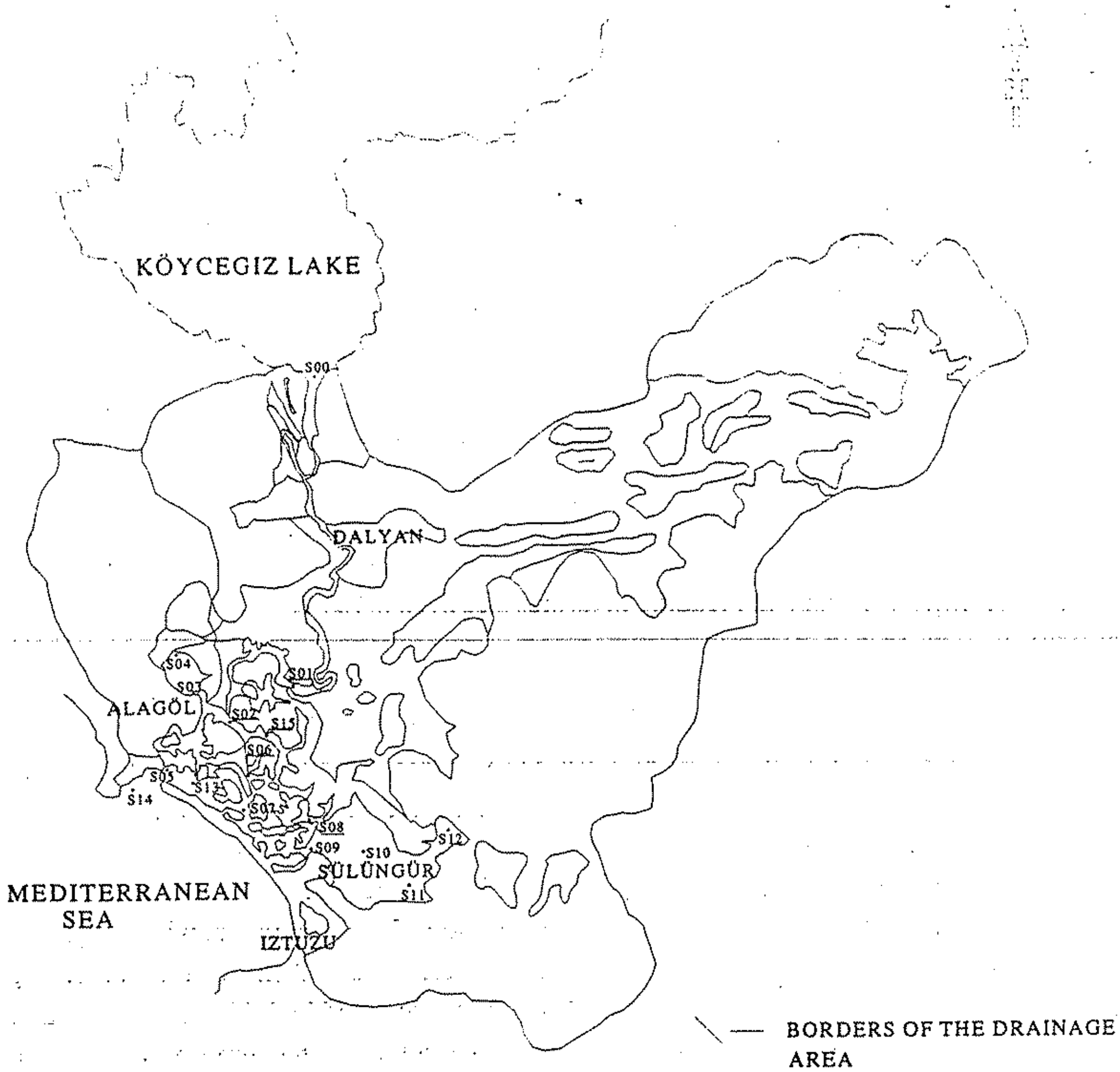

Figure 1 Location of sampling stations at Dalyan Lagoon catchment area

Deltamethrin

It is used in horticulture in an area of 94.3 ha with an annual consumption of 448 kg. It is applied once a year between June and September, preferably in June/July. Thus, water residue of deltamethrin was not detected in the first set of experiments representing spring, indicating that it completely disappeared in water. This may be the reason why a tolerance value for it was excluded in the current legislation on pesticides. In the second set of experiments representing summer, the residue values were higher compared to the following two sets as expected. Such higher values performed a decreasing trend in the third set and almost became insignificant in the final set. Summer values were higher as deltamethrin was applied in this period and the probable water reactions such as uptake by plants, evaporation and adsorption had not been accelerated. Effects of reactions were observed in the following sets. Generally, higher surface values were detected in the second set. At the entrance to the lagoon (Station 0), the higher surface value was due to transport of deltamethrin from Koycegiz Lake. At the outlet of the lagoon (Station 14), the residue was somehow diluted and relatively lower values were detected. In the third set, bottom values were much higher which may be explained by the low water solubility of deltamethrin and increase in sorption. It is obvious that sorption to sediment was significant. It is also interesting to note that values at stations near the reed beds (Stations 13 and 5) were relatively low, supporting the occurrence of the reaction, which is uptake by plants.

Diazinon

It is used for protection of cotton in an area of 1787.7 ha with an annual consumption value of 170 kg. It is applied once or twice a year between May and September. The tolerance value is 0.9 μg/L according to TAPR (1995). Diazinon residues appeared in all the four sets of experiments. In general, it was observed that part of the residue reached to the lagoon system from Koycegiz Lake. In the two lakes, none of the residues exceeded the limit value. In the second set, only at three of the stations (Stations 0, 7 and 11) were residue values almost at the limit value. With higher values than the first set, it is obvious that agricultural activities accelerated between April and August and most preferably, diazinon was applied only once in May–August period. The values of the third set were almost lower than the second set proving the information that the application took place between May–August and as diazinon has a very long half-life in water (~6 months) it was further detected in the other sets. The general trend in the third set designates lower surface but higher bottom values correlating with the fact that accumulation in sediment occurred due to low solubility and fairly strong adsorptive capacity of diazinon. In the fourth set, even lower values varying between 0.25–0.45 μg/L were observed. During winter, as precipitation increases, the water is diluted. Furthermore, the decrease in temperature affects the biodegradation rate, which extends its half-life leading to its presence even in Set 4.

Endosulfan

It is used in cultivation of cotton and horticulture in an area of 1,882 ha, with an annual consumption of 3,577 kg. It is one of the highly used pesticides in the area and is applied twice a year except in the December–March period. The tolerance value is 0.2 μg/L according to TAPR (1995). In the first set representing spring, generally no residues were observed correlating with the characteristics of endosulfan that it is strongly adsorptive. The values of this set were all under the tolerance value except Station 15-bottom value, which is just in the reed bed area. In the second set, higher values were observed showing a decreasing trend along the channel except for Stations 6 and 7 near the reed bed. However, no major fluctuations were observed. The results of the third set indicated that application of endosulfan continued during summer and with precipitation in autumn, residues reached the

Table 4 Results of the pesticide experiments – Deltamethrin and Diazinon residues (values in μg/L)

	Station	Deltamethrin								Diazinon							
		Set 1		Set 2		Set 3		Set 4		Set 1		Set 2		Set 3		Set 4	
		Surface	Bottom	Surface	Bottom	Surface	Bottom	Surface	Bottom	Surface	Bottom	Surface	Bottom	Surface	Bottom	Surface	Bottom
Alagol Lake	4	N.D.	N.D.	2.421	2.637	2.135	1.993	0.737	1.108	N.D.	0.180	0.692	0.754	0.671	0.586	0.234	0.368
	3	N.D.	N.D.	1.851	2.421	2.847	2.136	N.D.	1.972	0.380	0.380	0.577	0.700	0.847	0.613	0.278	0.440
Sülüngür Lake	8	N.D.	N.D.	1.851	1.567	N.D.	2.991	0.885	N.D.	0.340	0.320	0.530	0.526	0.868	0.856	0.263	0.297
	9	N.D.	N.D.	2.137	1.425	2.137	N.D.	N.D.	N.D.	N.D.	N.D.	0.686	0.429	0.658	0.821	0.472	0.368
	10	N.D.	N.D.	2.563	1.995	2.140	2.278	N.D.	N.D.	N.D.	N.D.	0.779	0.683	0.700	0.660	0.274	0.321
	11	N.D.	N.D.	2.848	2.281	2.847	2.993	N.D.	N.D.	0.270	0.820	0.906	0.658	0.858	0.891	N.D.	0.638
	12	N.D.	N.D.	2.848	2.136	N.D.	2.847	0.999	1.290	N.D.	N.D.	0.826	0.613	0.840	0.818	0.457	0.380
Lagoon Channel	0	N.D.	N.D.	2.849	2.281	2.420	2.859	1.105	N.D.	N.D.	0.240	0.936	0.653	0.715	0.820	0.345	0.446
	1	N.D.	N.D.	2.065	1.994	1.708	2.847	1.480	N.D.	0.440	N.D.	0.659	0.606	0.511	0.848	0.497	0.296
	2	N.D.	N.D.	2.136	1.853	1.566	2.847	1.179	1.308	N.D.	N.D.	0.638	0.622	0.555	0.848	0.347	0.425
	15	N.D.	N.D.	2.422	N.D.	2.847	1.423	N.D.	N.D.	N.D.	N.D.	0.689	0.856	N.D.	0.436	0.325	0.369
	6	N.D.	N.D.	2.849	2.279	N.D.	1.281	0.665	N.D.	0.280	N.D.	0.828	0.732	0.554	0.000	0.238	0.423
	7	N.D.	N.D.	2.848	3.134	2.848	2.848	1.365	0.667	N.D.	N.D.	N.D.	0.904	0.837	0.832	0.379	0.261
	13	N.D.	N.D.	N.D.	N.D.	0.248	1.423	N.D.	0.740	0.330	0.160	N.D.	N.D.	N.D.	0.427	0.221	0.211
	5	N.D.	N.D.	2.564	1.925	1.996	2.135	1.105	N.D.	N.D.	0.440	0.737	0.630	0.601	0.611	0.330	0.249
	14	N.D.	N.D.	2.136	1.994	1.994	2.852	N.D.	1.255	0.330	0.260	0.689	0.606	0.607	0.867	0.336	0.396

N.D. Not Detected

Table 5 Results of the pesticide experiments – Endosulfan and Dichlorvos residues (values in µg/L)

	Station	Endosulfan								Dichlorvos							
		Set 1		Set 2		Set 3		Set 4		Set 1		Set 2		Set 3		Set 4	
		Surface	Bottom	Surface	Bottom	Surface	Bottom	Surface	Bottom	Surface	Bottom	Surface	Bottom	Surface	Bottom	Surface	Bottom
Alagol Lake	4	N.D.	0.051	0.189	0.176	0.161	0.161	0.056	0.103	1.100	0.750	2.527	2.526	2.151	1.962	1.053	1.489
	3	N.D.	N.D.	0.141	0.185	0.214	0.160	0.073	0.109	1.620	1.620	1.807	2.463	2.960	N.D.	5.457	1.825
Sulungur Lake	8	0.097	0.097	0.142	0.120	0.213	0.224	0.067	0.073	1.450	1.410	1.796	1.514	N.D.	2.985	N.D.	1.320
	9	N.D.	N.D.	0.162	0.108	0.161	0.214	0.122	0.089	2.340	1.380	N.D.	1.382	2.043	N.D.	2.236	1.748
	10	N.D.	0.108	0.199	0.151	0.162	N.D.	0.073	0.094	0.790	1.570	3.041	1.909	2.242	3.531	1.386	1.349
	11	0.077	0.189	0.221	0.171	0.213	N.D.	N.D.	0.139	1.140	2.770	2.768	2.156	N.D.	N.D.	N.D.	N.D.
	12	N.D.	0.056	0.215	0.161	0.214	N.D.	0.142	0.097	1.870	0.780	2.754	2.230	N.D.	N.D.	1.386	3.704
Lagoon Channel	0	N.D.	0.054	0.213	0.172	0.184	0.161	0.842	0.118	1.130	0.980	2.773	2.249	N.D.	N.D.	1.540	2.438
	1	N.D.	N.D.	0.157	0.151	0.128	N.D.	0.117	0.777	N.D.	1.100	1.963	2.154	N.D.	N.D.	4.567	N.D.
	2	N.D.	N.D.	0.161	0.140	0.118	0.008	0.788	0.132	1.690	1.530	2.283	1.756	1.825	N.D.	1.659	7.500
	15	0.164	0.214	0.183	N.D.	0.213	N.D.	0.081	N.D.	2.210	2.320	2.344	3.530	N.D.	N.D.	1.480	5.907
	6	N.D.	0.082	0.215	0.198	0.133	0.096	0.050	N.D.	1.220	1.840	N.D.	2.231	N.D.	1.351	0.927	0.054
	7	N.D.	0.198	0.215	0.245	0.214	0.216	0.108	0.057	2.870	3.140	2.714	3.486	N.D.	N.D.	1.868	0.952
	13	N.D.	0.100	N.D.	N.D.	N.D.	0.107	0.056	0.056	1.450	0.700	2.308	2.069	2.725	N.D.	1.318	N.D.
	5	N.D.	0.130	0.198	0.147	0.150	0.215	0.084	N.D.	N.D.	1.850	N.D.	1.864	N.D.	N.D.	1.531	1.210
	14	N.D.	0.150	0.162	0.151	0.149	0.216	0.084	0.103	2.140	1.130	2.135	2.099	N.D.	N.D.	1.513	1.726

N.D. Not Detected

lagoon slightly exceeding the limit at many stations. No shock changes were detected. In general, channel values were relatively lower than those detected at the two lakes, which may be explained by the prevailing hydraulic conditions that are not evaluated in this study. In the final set, much lower values were seen compared to the third set with the exceptional values of Stations 0, 1 and 2, that can only be explained by heavy precipitation and runoff carrying the residuals from Koycegiz Lake. Excluding these higher values, others ranged between 0.005–0.14 μg/L, which lie well below the limit. Similarly, in the lakes, bottom values were higher due to high adsorptive capacity of endosulfan.

Dichlorvos

It is used in cultivation of cotton and horticulture again like endosulfan in an area of 1882 ha, with an annual consumption of 210 kg. It is applied once or twice a year between June and November. Dichlorvos appeared in all the four sets of experiments with very high values. In the first set representing spring, it existed in almost all samples, both surface and bottom, with high values exceeding the national tolerance limit 0.07 μg/L. Higher values were also detected in surface samples of both lakes and along the channel. Similar high concentrations continued to prevail in the second set reflecting the application period of dichlorvos. According to the information gathered from the local authorities, heavy irrigation is practised in cotton cultivation for almost three months a year from June to August. Therefore, dichlorvos enters the lagoon system in very high amounts through leaching to groundwater. In the third set, dichlorvos was not detected at more than half of the stations, but where detected, still exceeded the limit, ranging between 1.5–3 μg/L. As dichlorvos has no significant rapidly occurring reaction in the water environment, it stays in solution form and does not adsorb to sediment according to Extoxnet (1996) which fully supports the findings. Additionally, it is known that as water solubility of a chemical is high, no volatilization occurs. In the final set, the values were comparatively lower than Set 3, but within a range well above the limit. To sum up, dichlorvos was observed in Set 2 and 3 due to intensive leaching following irrigation in summer and heavy precipitation in autumn. In other samples, the values of residues were comparatively lower due to slow rate of reactions and long half-life in water.

Conclusions

A detailed field survey on the use of pesticides in the catchment area of Dalyan Lagoon is followed by a literature-based determination of fate of pesticides both in soil and in water. Besides, external factors like climatic conditions and soil properties are investigated. Water residue analyses of the six selected pesticides are conducted to strengthen the expected impacts on water quality. Out of the six pesticides, four appeared in the four sets of samples gathered seasonally. The fate of a pesticide in the aquatic environment was found to be significantly influenced by abiotic factors like adsorption, volatilization, hydrolysis and photolysis and by biotic factors like degradation. To assess the fate of a pesticide in the environment, models become more valuable and such systems can focus on specific questions about fate under existing conditions. Since extrapolation of the data from the natural environment is limited to certain qualitative aspects, the verification of the fate of a pesticide and associated water quality impacts requires appropriate monitoring studies, as was experienced in this study.

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