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The status of pesticide pollution in surface waters (rivers and lakes) of Greece. Part I. Review on occurrence and levels

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Information on pesticide pollution of surface waters in Greece is reviewed.

Abstract

This review evaluates and summarizes the results of long-term research projects, monitoring programs and published papers concerning the pollution of surface waters (rivers and lakes) of Greece by pesticides. Pesticide classes mostly detected involve herbicides used extensively in corn, cotton and rice production, organophosphorus insecticides as well as the banned organochlorines insecticides due to their persistence in the aquatic environment. The compounds most frequently detected were atrazine, simazine, alachlor, metolachlor and trifluralin of the herbicides, diazinon, parathion methyl of the insecticides and lindane, endosulfan and aldrin of the organochlorine pesticides. Rivers were found to be more polluted than lakes. The detected concentrations of most pesticides follow a seasonal variation, with maximum values occurring during the late spring and summer period followed by a decrease during winter. Nationwide, in many cases the reported concentrations ranged in low ppb levels. However, elevated concentrations were recorded in areas of high pesticide use and intense agricultural practices. Generally, similar trends and levels of pesticides were found in Greek rivers compared to pesticide contamination in other European rivers. Monitoring of the Greek water resources for pesticide residues must continue, especially in agricultural regions, because the nationwide patterns of pesticide use are constantly changing. Moreover, emphasis should be placed on degradation products not sufficiently studied so far.

Keywords: Occurrence; Pesticide residues; Freshwaters; Greece

1. Introduction

Worldwide pesticide usage has increased dramatically during the last two decades, coinciding with changes in farming practices and the increasingly intensive agriculture. This widespread use of pesticides for agricultural and non-agricultural purposes has resulted in the presence of their residues in various environmental matrices. Pesticide contamination of surface waters has been well documented worldwide and constitute a major issue that gives rise to concerns at local, regional, national and global scales (Planas et al., 1997; USGS, 1999; Huber et al., 2000; Cerejeira et al., 2003).

Pesticide residues reach the aquatic environment through direct run-off, leaching, careless disposal of empty containers, equipment washing, etc. Although significant advances have been made in controlling point-source pollution, little progress has been accomplished in the area of nonpoint-source pollution of surface waters. This is because of the seasonality, inherent variability and multiplicity of origins of nonpoint-source pollution (Albanis et al., 1998; Pereira and Hostettler, 1993).

Pesticides are primarily moved from agricultural fields to surface waters in surface run-off (Richards and Baker, 1993). The amount lost from fields and transported to surface waters depends on several factors, including soil

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characteristics, topography, weather, agricultural practices, and chemical and environmental properties of individual pesticides (Wagenet, 1987; Leonard, 1990). The combined effect of these factors on the temporal and spatial magnitude of pesticide concentrations and fluxes in large integrating river systems is largely unknown (Larson et al., 1995). Chemical reactions and physical displacements influence the persistence of the chemicals in the soil, but with different environmental implications. Chemicals, which are sufficiently resistant to degradation and are adequately soluble to be transported in water, may reach the water bodies in significant amounts (Wauchope, 1978; Wagenet, 1987).

The trends in pesticide consumption used in Greece during the period 1986–1998 are given in Fig. 1A (FAO, 2004). The relative contribution of the various chemical families to the total pesticide use is given in Fig. 1B. Organophosphorus compounds, triazines and dithiocarbamates represent the main chemical categories of pesticides used in Greece. Other categories such as dinitroanilines, carbamates and amides follow with percentages less than 5% of the total used pesticides. Organochlorine insecticides such us DDT, endrin, dieldrin, aldrin, heptachlor, heptachlor epoxide and technical mixtures of

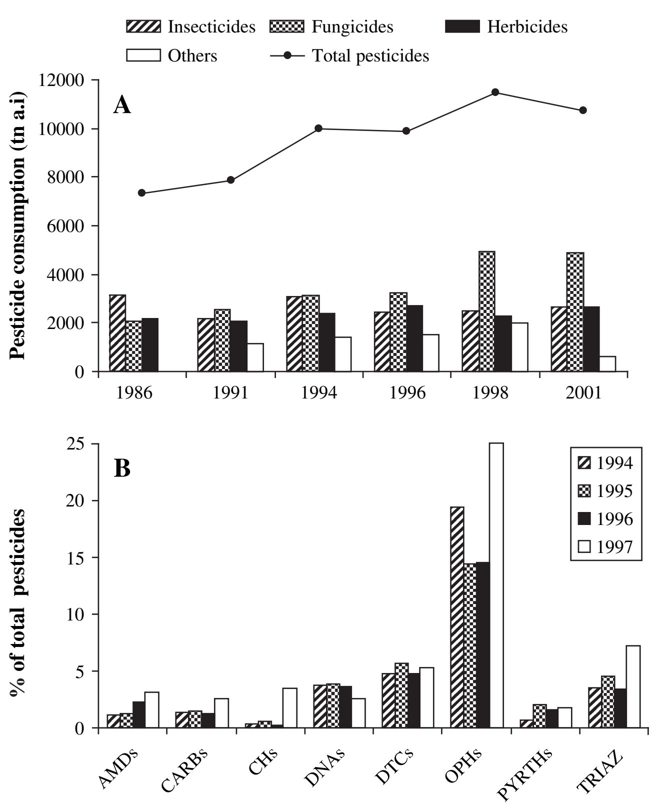

Fig. 1. The trends in the consumption of pesticides in Greece (FAO, 2004). (A) During the period 1986–2001. Insecticides: acaricides, molluscicides, nematocides and mineral oils; fungicides: bactericides and seed treatments; herbicides: defoliants and dessicants. Total pesticides may include other pesticides such as growth regulators and rodenticides. (B) The relative contribution of the different chemical families to the total pesticides used. AMDs, amides; CARBs, carbamates; CHs, chlorinated hydrocarbons; DNAs, dinitroanilines; DTCs, dithiocarbamates; OPHs, organophosphates; PYRTHs, pyrethroids; TRIAZ, triazines.

BHCs and HCHs were extensively used in Greece until their ban in 1972 (Albanis et al., 1994) and still persist in the aquatic environment (Albanis et al., 1998).

Although the physicochemical properties and the pollution pattern by nutrients and metals of the Greek lakes and rivers have already been reviewed (Skoulikidis et al., 1998), the contamination of surface waters by pesticides has not been performed so far. The aim of the present review is to compile all available data on pesticide pollution of Greek surface freshwaters, to discuss the seasonal patterns and their occurrence and finally to compare them with pesticide levels in other European surface waters.

2. Occurrence and environmental levels

It should be noted that the literature available data reviewed herein concerning river water pollution by pesticides, include several principal rivers of Greece that drain major agricultural areas; however, there is a lack of data for some other important rivers mainly in central and Southern Greece (e.g. Sperheios). In addition, though the existing data cover the last two decades only few concern annual monitoring surveys that include all the categories of pesticides. The rivers that were monitored in a systematic way are Aliakmon, Axios, Loudias, Louros, Arachthos and Kalamas. The above river deltas are regions protected by international conventions as they constitute important aquatic ecosystems.

The water bodies of Greece that have been monitored for pesticide residues are shown in Fig. 2. Generally the pesticides that are most commonly detected are those that are widely applied, have low K_{oc} values and high environmental persistence. Table 1 lists selected commonly occurring pesticides in surface waters of Greece as well as their use, physicochemical properties and run-off indices. We can distinguish two main groups of pesticides according to their occurrence and the concentration range detected. The first group consists of pesticides that were occasionally seen in surface waters of Greece. The compounds in this group have one or more of the following characteristics: low application rates (metribuzin, azinphos-methyl), use only in limited geographical areas (molinate, propanil), short soil lifetimes (malathion, parathion, EPTC, propanil), short aquatic lifetimes (malathion, propanil), lower run-off hazard (propanil, malathion, EPTC). The second group includes those compounds that are usually found in Greek surface waters and exhibit seasonally elevated concentrations (atrazine, simazine, alachlor, metolachlor, diazinon, trifluralin, parathion methyl, lindane). The compounds of this group have higher application rates and widespread use and (or) higher run-off hazard, with little loss within the riverine network or the lake (longer aquatic half-lives).

Another way to separate the detected pesticides is by type and application method, i.e. surface-applied herbicides, incorporated herbicides, and insecticides. As is shown in Table 1, surface-applied herbicides such as atrazine, simazine, alachlor and metolachlor have greater load as percent of use (LAPU) mean values compared to the incorporated herbicides and

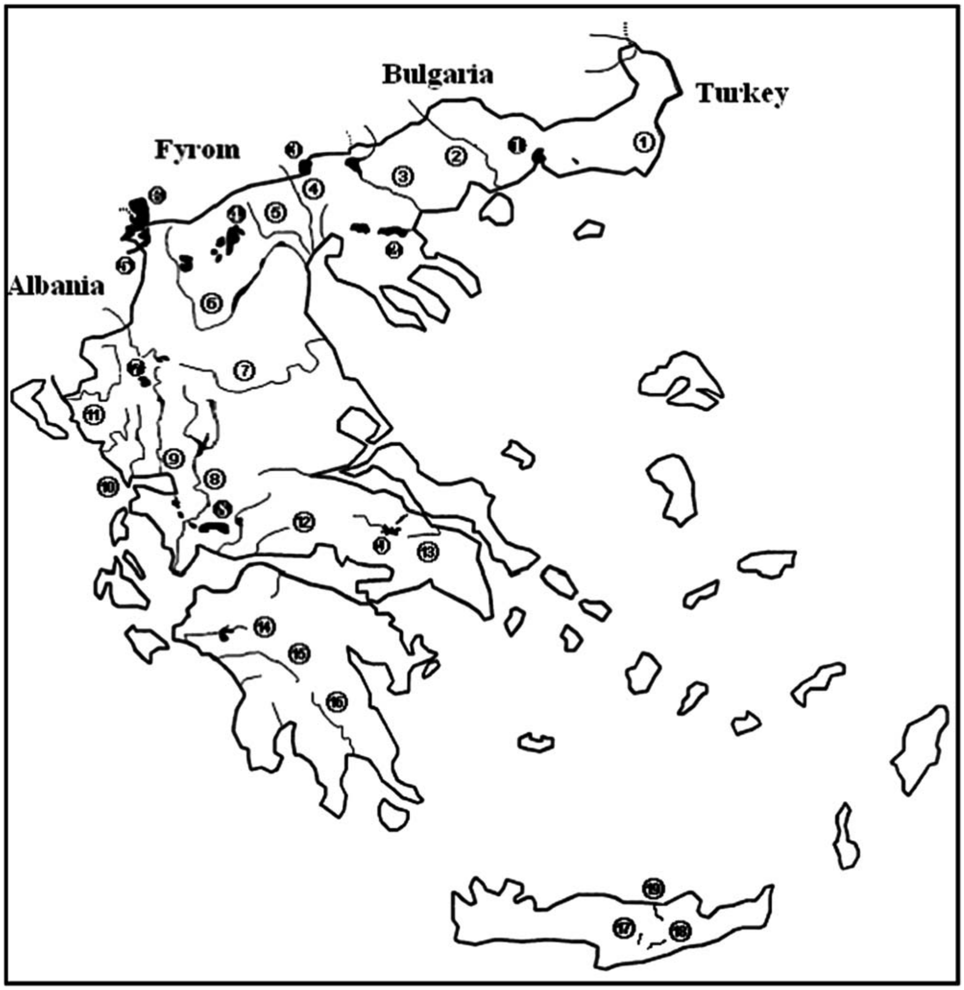

Fig. 2. The Greek rivers and lakes that have been monitored for pesticide residues. Rivers (numbered black on white background): 1, Evros; 2, Nestos; 3, Strymonas; 4, Axios; 5, Loudias; 6, Aliakmonas; 7, Pinios; 8, Acheloos; 9, Arachthos; 10, Louros; 11, Kalamas; 12, Mornos; 13, Asopos; 14, Pinios Peloponnese; 15, Alphios; 16, Evrotas; 17, Honos; 18, Havgas; 19, Aposelemis. Lakes (numbered white on black background): 1, Vistonida; 2, Volvi; 3, Doirani; 4, Vegoritida; 5, Mikri Prespa; 6, Megali Prespa; 7, Pamvotida; 8, Trichonida; 9, Iliki.

insecticides, so they are more frequently detected in the surface waters (Capel et al., 2001).

In the following paragraphs the pesticide occurrence and levels is discussed based on the chemical families and the mode of action. The occurrence of organochlorine insecticides in rivers and lakes of Greece is shown in Tables 2–5. Among organochlorines, only endosulfan is in use for applications mainly in vineyards, orchards and vegetable farming. Lindane was used in Greece in some cultivations and seed treatment (Albanis, 1997) until June 2002, when it was phased out according to EC Directive2000/801. Concentrations of organochlorines are generally very low with the exception of hexachlorocyclohexane isomers and aldrin, which show the highest levels especially in rivers from Northern Greece. For these rivers (Evros, Axios) the higher concentrations of organochlorines were generally detected in the samples from the border sampling stations, denoting probable transboundary pollution from the neighbouring countries as is stated in the corresponding monitoring studies (Angelidis and Albanis, 1996; Papadopoulou-Mourkidou, 2002; Golfinopoulos et al., 2003). Especially in the Axios river, lindane was detected in 100% of the samples collected from the points located at the entrance of the river to Greek territory, showing clear transboundary pollution. This observation is justified also from the fact that lindane manufacturing is active in Skopja (Papadopoulou-Mourkidou, 2002). Other sources of organochlorine pesticides residues in Greek surface waters might be wet and dry deposition. It is known that persistent organochlorine compounds undergo atmospheric long-range transport before wet and dry deposition (Cleemann et al., 1995; van Pul et al., 1998; Beyer et al., 2000). Thus, organochlorine pollution is not only a matter of residual concentrations in the region that have been applied but could be due to long-range transport from neighbouring regions or countries. In conclusion, water pollution due to organochlorine pesticides shows that they still persist in the aquatic environment of Greece.

Regarding herbicides, atrazine, simazine, metolachlor and alachlor were the most frequently detected compounds in waters for most of the rivers (Tables 6–8) (Albanis, 1991, 1992; Samara et al., 1994; Albanis and Hela, 1998; Papadopoulou-Mourkidou, 2002), followed by trifluralin, molinate, prometryne and propanil. The highest concentrations of

Table 1
Agricultural use, physicochemical properties, and estimated run-off capacity of the most frequently detected pesticides in Greek freshwaters

Pesticide (use[a])	MAT[b]	MAR[c] (g a.i./ha)	$DT_{50\ s}$ (days[d])	$DT_{50\ w}$ group[e]	K_H[f]	K_{oc}[g]	Major loss process[h]	LAPU[i]	RRPI[j]
Alachlor (H)	sur	3500	15	E	6.2E-3	170	T	0.27	113
Atrazine (H)	sur	8000	60	G	2.9E-4	100	T	0.66	17
Azinphos-methyl (I)	fol	1900	10	C	3.2E-3	1000	T	0.32	100
Carbofuran (I)	inc	2200	50	C	5.1E-4	22	T	0.32	4
Diazinon (I)	fol	1500	40	E	6.7E-2	1000	T	0.085	25
EPTC (H)	inc	7500	6	C	1.02	200	T	0.034	333
Lindane (I)	sur	2500	400	G	0.13	1100	V	0.00045	2
Malathion (I)	fol	3000	1	B	2.3E-3	1800	T	0.045	555
Methyl parathion (I)	fol	3500	5	D	2.1E-2	5100	T	0.009	39
Metolachlor (H)	sur	3500	90	E	9E-4	200	T	0.5	22
Metribuzin (H)	sur	1000	40	F	2.4E-4	60	T	0.40	15
Molinate (H)	pad	5000	21	C	0.746	190	T	4.9	90
Parathion (I)	fol	3500	14	D	1.2E-2	5000	T	0.19	14
Propanil (H)	sur	5000	1	A	3.6E-3	149	T	6.4	>1000
Simazine (H)	sur	2500	60	D	2.9E-6	130	T	0.52	21
Trifluralin (H)	inc	1300	60	E	9.5E-3	8000	V	0.054	2

[a] H, herbicide; I, insecticide.
[b] MAT, major application type; Sur, soil surface applied; inc, soil incorporated; fol, foliar applied; pad, added to rice paddy.
[c] MAR, maximum application rates (g active ingredient/ha); from Fielding et al., 1991.
[d] $DT_{50\ s}$, half life in soil from Wauchope et al., 1992.
[e] $DT_{50\ w}$, half life in water: Group A: ~0.5–1 day; B: ~1–4 days; C: ~4–12 days; D: ~12–40 days; E: ~40–120 days; F: ~120–420 days; G: ~420–1200 days; from Mackay et al., 1997.
[f] Henry's law constant (K_H, Pa m^3 mol^{-1}); from Tomlin, 1995.
[g] Organic-carbon normalized water-solid distribution coefficient (K_{oc}); from Wauchope et al., 1992.
[h] T, transformation; V, volatilization; from Capel et al., 2001.
[i] LAPU, Load as percent of use (%) for pesticides for small watersheds (<100,000 ha).
[j] RRPI, relative run-off potential index, from Hornsby and Augustijn-Beckers, 1991 (smaller RRPI values indicate greater run-off hazard).

alachlor and atrazine were measured in the River Loudias at 9300 ng/L and 5900 ng/L, respectively in 1988–1989, as well as for prometryne in the Aliakmon river at 6100 ng/L in 1988–1989 and propanil in the Axios river at 20,600 ng/L in 1993–1994. Among triazines the compound with the highest frequency of detection was atrazine followed by simazine and prometryne. It must be noted that atrazine and simazine were withdrawn in Greece in September 2004 (2004/248/EC and 2004/247/EC). Water pollution by triazines and chloroacetanilides was the highest in the estuarine areas, showing that many of these compounds are transported over significant distances from their application sites (Albanis et al., 1998).

Concerning insecticides, diazinon, methyl parathion and parathion were the compounds detected in most Greek rivers followed by fenthion, carbofuran and malathion (Table 9). Higher levels of insecticides were recorded for the River Axios, up to 2000 ng/L for malathion, parathion and pyrazophos, 362 ng/L for parathion methyl and 7300 ng/L for carbofuran. Diazinon, ethion and disulfoton were found in higher levels (up to 775, 30 and 70 ng/L, respectively) in River Kalamas while fenthion's maximum concentration (230 ng/L) was observed in the Evrotas river. Parathion and parathion methyl were in use before 2003 and their authorization was revoked according to Directives 2001/520/EC and 2003/166/EC. Thus, residues of these compounds were not recorded in the most recent studies.

The reported data in the literature for the occurrence of herbicides as well as organophosphorus and carbamate insecticides in Greek lakes are given in Table 9. The data refer to Lakes Pamvotida, Iliki, Marathonas and Mornos. The last three are important water-supplying reservoirs for the city of Athens. The concentrations of the detected pesticides were higher in Lake Pamvotida compared to other lakes. Pesticide levels in Greek lakes were also presented in recent studies (Lekkas et al., 2003, 2004) but they were not included in the table as they report a range of concentrations for all lakes monitored, not discussing them separately. Data of these studies are reviewed in subsequent paragraphs.

Captafol, captan, chlorothalonil, folpet, metalaxyl, flutriafol and vinclozolin were the fungicides monitored in riverine waters of Greece (Loudias, Axios, Aliakmon, Nestos, Kalamas, Evrotas and Louros rivers). The results from the reported studies (Albanis and Hela, 1995; Albanis et al., 1995b; Angelidis et al., 1996; Readman et al., 1997; Lambropoulou et al., 2002; Papadopoulou-Mourkidou et al., 2004b) show sporadic run-off of few compounds, mainly captan, chlorothalonil and folpet, in the adjacent water bodies. Captan was found in Loudias, Axios, Nestos, Evrotas and Louros rivers with maximum concentrations of 24, 40, 80, 260 and 32 ng/L, respectively. Folpet was found only in Loudias river at concentration levels up to 50 ng/L, chlorothalonil (max. 7 μg/L) and flutriafol (0.5 μg/L) were detected in limited number of samples in the Axios river. The results indicate that fungicides do not generally threaten contamination of freshwater and estuarine environments, probably due to their low persistence (Readman et al., 1997).

Table 2
Levels (ng/L) of organochlorine insecticides (BHC, HCHs, DDT and related compounds) residues in water samples from various Greek rivers at different sampling periods

River	Sampling period	BHC	HCHs	4,4′-DDE	4,4′-DDD	4,4′-DDT	Methoxychlor	Dicofol	Refs.
Aliakmon	12/1990–09/1992	–[a]	n.d.–130[c]	n.d.	n.d.	n.d.	–	–	Albanis et al., 1995b
	05/1996–04/1997	–	n.d.[c]	n.d.	n.d.	n.d.	–	–	Albanis et al., 1998
	During 2000	2.78	6.35[c]	2.17	0.50	n.d.	–	–	Kamarianos et al., 2002[e]
Loudias	05/1996–04/1997	–	n.d.–22[c]	n.d.–15	n.d.	n.d.	–	–	Albanis et al., 1998
	During 2000	1.00	1.80[c]	1.60	1.10	n.d.	–	–	Kamarianos et al., 2002[e]
Axios	1993–1994	–	n.d.–310[c]	n.d.	n.d.–5	n.d.–27	–	–	Patsias and Papadopoulou-Mourkidou, 2002
	During 1994	–	n.d.–80[c]	n.d.	n.d.	n.d.	n.d.	n.d.	Papadopoulou-Mourkidou, 1996
	11/1996	–	490[c]	n.d.	n.d.	n.d.	–	–	Miliadis and Malatou, 1997
	06/1996–06/1998	n.d.[b]	n.d.–233[d]	n.d–25	n.d.	n.d.–35	n.d.	–	Golfinopoulos et al., 2003
	1997–1998	–	n.d.–132[c]	n.d.	n.d.	n.d.–1000	–	–	Papadopoulou-Mourkidou, 2002
	During 2000	3.53	2.93[c]	3.20	0.50	n.d.	–	–	Kamarianos et al., 2002[e]
Strimonas	06/1996–06/1998	n.d.	n.d.–59[d]	n.d.–31	n.d.	n.d.	n.d.	–	Golfinopoulos et al., 2003
	During 2000		4.90[c]	1.00	0.55	n.d.	–	–	Kamarianos et al., 2002[e]
Nestos	06/1996–06/1998	n.d.–14	n.d.–68[d]	n.d.–64	n.d.	n.d.	n.d.	–	Golfinopoulos et al., 2003
	During 2000	6.05	6.68[c]	5.50	1.15	n.d.	–	–	Kamarianos et al., 2002[e]
Evros	08/1992–07/1993	–	n.d.–34[c]	n.d.	n.d.–56	n.d.–10	–	–	Angelidis and Albanis, 1996
	06/1996–06/1998	n.d–131	n.d.–189[d]	n.d.–9	n.d.	n.d.	n.d.	–	Golfinopoulos et al., 2003
	During 2000	2.65	12.50[c]	6.00	1.60	n.d.	–	–	Kamarianos et al., 2002
Kalamas	09/1984–09/1985	–	n.d.–5.6[c]	–	–	–	–	–	Albanis et al., 1986a
Arachthos	03/1992–02/1993	–	n.d.	n.d.–11	n.d.	n.d.	–	–	Albanis et al., 1995a
Mornos	12/1992–02/1993	–	n.d.–5[d]	1	n.d.	n.d.	–	–	Tsipi and Hiskia, 1996
Louros	03/1992–02/1993	–	n.d.–16[c]	n.d.	n.d.	n.d.	–	–	Albanis et al., 1995a
Evrotas	08/1991–08/1992	–	–	–	–	–	–	n.d.–80	Angelidis et al., 1996
Honos	11/1995–06/1996	–	2–6[d]	1–10	n.d.	2	n.d.	–	Vassilakis et al., 1998
Havgas	11/1995–06/1996	–	2–26[d]	1	n.d.	2–4	n.d.	–	Vassilakis et al., 1998
Aposelemis	11/1995–06/1996	–	5–171[d]	1–37	n.d.	2–3	n.d.	–	Vassilakis et al., 1998

[a] –, not monitored.
[b] n.d., not detected.
[c] Refers only to lindane (γ-HCH).
[d] Data shown the sum of α-, β-, γ-, δ- hexachlorocyclohexane isomers.
[e] Mean values were reported in this study.

The pesticide occurrence corresponds to specific applications on cultivations from the regions into the drain basin of the rivers. Examples concerning rivers Axios, Loudias and Aliakmon in Northern Greece, Louros, Arachthos and Kalamas in Western Greece and the Evrotas river in Southern Greece follow. The rivers Axios (300 km long, 65 m^3/s mean flow rate), Loudias (38 km long, 20 m^3/s mean flow rate) and Aliakmon (310 km long, 42 m^3/s mean flow rate) drain 93,500 ha of agricultural land, which supports mainly cotton, corn, rice, fruit trees (mainly peach and apples), sugar beet and vegetables while rare crops include alfalfa and grains (Albanis et al., 1998; Papadopoulou-Mourkidou et al., 2004a). The southern part of the Axios river basin is considered one of the main rice cultivation regions in Greece, where rice is cultivated under submerged conditions. The contamination of water bodies is considered higher in the areas where rice is cultivated under flooded conditions because irrigation increases the likelihood of transport of pesticides via run-off to surface water. Molinate and propanil were detected mainly in these rivers and, especially in the Axios river, propanil was the most frequently detected pesticide (Papadopoulou-Mourkidou et al., 2004b). Atrazine, simazine, prometryne, alachlor and terbufos were regularly detected at concentrations exceeding 0.1 μg/L (Papadopoulou-Mourkidou et al., 2004b).

In River Axios, for which a complete screening agrochemical pollution survey was conducted in 1993–1994 and 1997–1998 periods (Papadopoulou-Mourkidou, 2002), several other pesticides were detected apart from these reported in Tables 6–8 and Table 10 but with a low frequency of detection (≤2%) and in low concentrations. The levels ranged from 0.001 μg/L for ethofumesate to 0.464 μg/L for bromopropylate in the 1997–1998 period, and from 0.002 μg/L for desmetryne to 0.132 μg/L for ethofumesate in the 1993–1994 period. The detected compounds in the 1997–1998 period include mevinphos, furalaxyl, cyanofos, cycloate, carbophenothion ethyl, terbumeton, atraton, coumaphos and napropamide whereas fluometuron, carbosulfan, desmetryne, methidation, pirimiphos methyl and *cis*-permethrin were detected in the 1993–1994 period. It must be noted also that the Axios river, originating from FYROM and discharging into the North Aegean Sea, is already highly polluted with pesticides when entering the Greek/FYROM boarder due to significant point and diffuse pollution sources located in the FYROM territory (Papadopoulou-Mourkidou et al., 2004a).

In north-western Greece, Louros (80 km long, 19.4 m^3/s mean flow rate) and Arachthos (110 km long, 61 m^3/s mean flow rate) rivers drain 74,700 ha of agricultural land. The rivers approach the sea through lowlands, which support intense agriculture including citrus fruits, olives, corn, alfalfa and cotton (Albanis et al., 1995a). The Kalamas river (96 km long, 43 m^3/s), with a few tributaries, an agricultural area of ca.

Table 3
Levels (ng/L) of organochlorine insecticide (cyclodienes) residues in water samples from various Greek rivers at different sampling periods

River	Sampling period	Aldrin	Dieldrin	Endrin	Endrin ketone	Endrin aldehyde	Isodrin	α,β-Endosulfan[a]	Endosulfan sulphate	Heptachlor	Heptachlor epoxide	Refs.
Aliakmon	12/1990–09/1992	n.d.[b]	n.d.	n.d.	–[c]	–	–	–	–	n.d.	n.d.	Albanis et al., 1995b
	05/1996–04/1997	n.d.	n.d.	n.d.	–	–	–	–	–	n.d.	n.d.	Albanis et al., 1998
	During 2000	n.d.	0.98	0.80	–	–	–	–	–	n.d.	n.d.	Kamarianos et al., 2002[d]
Loudias	05/1996–04/1997	n.d.	n.d.	n.d.	–	–	–	–	–	n.d.–8	n.d.	Albanis et al., 1998
	During 2000	n.d.	0.90	0.80	–	–	–	–	–	n.d.	0.80	Kamarianos et al., 2002[d]
Axios	1993–1994	n.d.	n.d.	n.d.	–	–	–	n.d.–1741	–	n.d.–20	n.d.–5	Papadopoulou-Mourkidou, 2002
	06/1996–06/1998	n.d.–34	n.d.–17	n.d.	n.d.	n.d.–40	n.d.	n.d.	n.d–58	n.d.–20	n.d.	Golfinopoulos et al., 2003
	1997–1998	n.d.	n.d.	n.d.	–	–	–	2	–	n.d.	n.d.	Papadopoulou-Mourkidou, 2002
	During 2000	n.d.	1.55	n.d.	–	–	–	–	–	n.d.	n.d.	Kamarianos et al., 2002[d]
Strimonas	06/1996–06/1998	n.d.–101	n.d.	n.d.	n.d.	n.d.–80	n.d.	n.d.–22	n.d.–23	n.d.	n.d–24	Golfinopoulos et al., 2003
	During 2000	n.d.	1.40	0.85	–	–	–	–	–	n.d.	0.70	Kamarianos et al., 2002[d]
Nestos	06/1996–06/1998	n.d.–22	n.d.–15	n.d.	n.d.	n.d.	n.d.	n.d.	n.d–12	n.d.	n.d.–9	Golfinopoulos et al., 2003
	During 2000	n.d.	2.00	0.80	–	–	–	–	–	n.d.	n.d.	Kamarianos et al., 2002[d]
Evros	08/1992–07/1993	n.d.	n.d.	n.d.	–	–	–	n.d.	–	–	–	Angelidis and Albanis, 1996
	06/1996–06/1998	n.d.–34	n.d.	n.d.	n.d.–25	n.d.–40	n.d.	n.d–20	n.d.–19	n.d.–20	n.d.	Golfinopoulos et al., 2003
	During 2000	n.d.	2.20	n.d.	–	–	–	–	–	–	0.95	Kamarianos et al., 2002[d]
Kalamas	09/1984–09/1985	–	n.d.	–	–	–	–	–	–	–	–	Albanis et al., 1986a
Arachthos	03/1992–02/1993	n.d.	n.d.	n.d.	–	–	–	–	–	n.d.	n.d.	Albanis et al., 1995a
Mornos	12/1992–02/1993	1	2	n.d.	–	n.d.	–	3	4	n.d.	n.d.	Tsipi and Hiskia, 1996
Louros	03/1992–02/1993	n.d.	n.d.	n.d.	–	–	–	–	–	n.d.	n.d.	Albanis et al., 1995a
Evrotas	08/1991–08/1992	–	–	–	–	–	–	n.d.–190	–	–	–	Angelidis et al., 1996
Honos	11/1995–06/1996	1–4	1–10	1–2	–	n.d.	–	n.d.	n.d.	1–20	1–13	Vassilakis et al., 1998
Havgas	11/1995–06/1996	n.d.	1	n.d.	–	n.d.	–	n.d.	1–2	1–50	1–6	Vassilakis et al., 1998
Aposelemis	11/1995–06/1996	1–24	1–15	1–6	–	n.d.	–	n.d.	1–4	1–51	n.d.	Vassilakis et al., 1998

[a] Sum of the two isomers.
[b] n.d., not detected.
[c] –, not monitored.
[d] Mean values were reported in this study.

Table 4
Levels (ng/L) of organochlorine insecticides (BHCs, HCHs, DDT and related compounds) residues in water samples from various Greek lakes at different sampling periods

Lake	Sampling period	BHC	HCHs	4,4′-DDE	4,4′-DDD	4,4′-DDT	Methoxychlor	Dicofol	Refs.
Trichonida	10/1996	–[a]	n.d.[b]	n.d.	n.d.	n.d.	n.d.	30	Miliadis and Malatou, 1997
Iliki	12/1992–02/1993	–	n.d.–5[d]	3	2	n.d.	–	–	Tsipi and Hiskia, 1996
	10/1996–12/1996	–	100–200[c]	n.d.	n.d.	n.d.	–	–	Miliadis and Malatou, 1997
Marathonas	12/1992–02/1993	–	n.d.–5[d]	2	n.d.	n.d.	–	–	Tsipi and Hiskia, 1996
Pamvotida	09/1984–09/1985	–	n.d.–38[c]	–	–	–	–	–	Albanis et al., 1986a
Volvi	06/1996–06/1998	n.d.–24	n.d.–84[d]	n.d.	n.d.–112	n.d.	n.d.	–	Golfinopoulos et al., 2003
Vistonida	06/1996–06/1998	n.d.–33	n.d.–66[d]	n.d.	n.d.–18	n.d.	n.d.–56	–	Golfinopoulos et al., 2003
Vergoritida	06/1996–06/1998	n.d.–15	n.d.–421[d]	n.d.–9	n.d.	n.d.	n.d.	–	Golfinopoulos et al., 2003
L. Prespa	06/1996–06/1998	n.d.	n.d.–232[d]	n.d.	n.d.	n.d.	n.d.	–	Golfinopoulos et al., 2003
S. Prespa	06/1996–06/1998	n.d.–440	n.d.–95[d]	n.d.–13	n.d.–12	n.d.	n.d.	–	Golfinopoulos et al., 2003

[a] –, not monitored.
[b] n.d., not detected.
[c] Refers to lindane (γ-HCH).
[d] Data shown is the sum of α-, β-, γ-, δ-, hexachlorocyclohexane isomers.

45,000 ha, and a mean annual flow-rate of 54 m^3/s, discharges into the Ionian Sea. Major cultivation includes maize, sorghum, cereals, alfalfa, vegetables, potatoes, citrus fruits and olives (Lambropoulou et al., 2002). The main herbicides (atrazine, simazine, alachlor, metolachlor, trifluralin) and insecticides (diazinon, parathion methyl, carbofuran) detected correspond to applications in the previous cultivations. Evrotas river (82 km long, 15 m^3/s mean flow rate), in the Southern part of Greece, drains ca. 30,000 ha of agricultural land in which the main crops are oranges and olives with ploughed cultivations of vine trees and vegetables (Angelidis et al., 1996). Thus, in the Evrotas river mostly insecticide residues of several compounds, especially organophosphates, are detected although in lower levels than herbicides.

The Evros (530 km long of which 204 in the Greek territory, 50 m^3/s mean flow rate) is a multinational river of the Balkan peninsula, with a catchment area of 53,000 km^2 divided between Bulgaria (66.2%), Turkey (27.5%) and Greece (6.3%). Agrochemical pollution sources include the run-off from intensively cultivated plains of Plovdiv (in Bulgaria) and Andrianoupolis (in Turkey). In Greece, the river receives the run-off from the greater part of the 140,000 ha which constitute the cultivated land of the prefecture of Evros and main crops include cotton, wheat, beet and corn (Angelidis and Albanis, 1996). The detected pesticides include mainly atrazine, simazine, alachlor, trifluralin and diazinon (Angelidis and Albanis, 1996; Papastergiou and Papadopoulou-Mourkidou, 2001) as well as organochlorine pesticides probably due to transboundary pollution (Angelidis and Albanis, 1996; Golfinopoulos et al., 2003).

The existing data show that Axios, Aliakmon and Loudias are the rivers most polluted by pesticide residues based both on the number of the detected compounds and the maximum detected concentrations. The pesticide pollution is mainly characterized by herbicidal compounds rather than by insecticides.

Data on pesticide occurrence in rivers such as Asopos, Alphios and Pinios of the Peloponnese and in the lakes Doirani and Volvi were first recorded in recent studies (Lekkas et al., 2003, 2004) but the levels are not referred to herein as they did not concern individual pesticide concentrations but ranges for all rivers and lakes monitored. The results of a nationwide monitoring survey showed that the most commonly encountered organochlorine insecticides in Greek surface waters are α-endosulfan, β-endosulfan and endosulfan sulphate (up to 43 ng/L, 23 ng/L and 28 ng/L, respectively). This trend is not depicted in previous studies (Tables 2–5). As regards organophosphorus insecticides the most frequently detected compounds are fenthion, azinphos methyl, azinphos ethyl, parathion ethyl and coumaphos. Other organophosphorus pesticides include fenitrothion, parathion methyl, dimethoate, demethon-S-methyl, methamidophos, mevinphos and malathion. The concentrations of the measured insecticides were comparable to those reported already in Tables 8 and 9. In the rivers, maximum concentrations of 158 ng/L for triazophos and 96 ng/L for disulfoton were found while in the lakes maximum concentrations of 131 ng/L and 78 ng/L were measured for demeton-S-methyl and methamidophos, respectively. Concerning herbicides, atrazine (up to 500 ng/L for lakes and 330 ng/L for rivers) is the compound with the highest frequency of detection followed by simazine (up to 32 ng/L for lakes and 640 ng/L for rivers) and prometryne (up to 10 ng/L for lakes and 780 ng/L for rivers), in agreement with the previous studies reported in Tables 6–8. Terbuthylazine was detected in rivers Evros and Axios (up to 37 ng/L) and in lake Pamvotida (up to 30 ng/L). Finally, the presence of several phenylurea herbicides, such as linuron, monolinuron, chlorotoluron and metobromuron, is reported in the same studies for certain rivers and lakes. Higher concentrations were recorded for metobromuron (up to 144 ng/L) in rivers and chlorotoluron (up to 98 ng/L) in lakes. Diuron was present only in Lake Vistonida (up to 85 ng/L) and this could be due to the replacement of this compound by other phenylurea herbicides.

Comparing these results and the concentrations reported in Tables 6–9, we can conclude that the patterns of agrochemical use are constantly changing as the popularity of existing pesticides rise and fall and as new compounds are introduced into farming and others are withdrawn.

The entrance of pesticides into freshwaters through wet and dry deposition was revealed also in monitoring studies in the Aliakmon and Loudias river basins (plain of Imathia) during

Table 5
Levels (ng/L) of organochlorine insecticide (cyclodienes) residues in water samples from various Greek lakes at different sampling periods

Lake	Sampling period	Aldrin	Dieldrin	Endrin	Endrin ketone	Endrin aldehyde	Isodrin	α,β-Endosulfan[a]	Endosulfan sulphate	Heptachlor	Heptachlor epoxide	Refs.
Iliki	12/1992–02/1993	1	4	1	–[b]	2	–	10	n.d.[c]	1	n.d.	Tsipi and Hiskia, 1996
Marathonas	12/1992–02/1993	1	3	n.d.	–	n.d.	–	3	n.d.	n.d.	n.d.	Tsipi and Hiskia, 1996
Pamvotida	09/1984–09/1985	–	n.d.–1.2	–	–	–	–	–	–	–	–	Albanis et al., 1986a
Volvi	06/1996–06/1998	n.d.–23	n.d.–31	n.d.–20	n.d.	n.d.–64	n.d.	n.d.–39	n.d.–49	n.d.	n.d.	Golfinopoulos et al., 2003
Vistonida	06/1996–06/1998	n.d.–200	n.d.	n.d.	n.d.	n.d.	n.d.–57	n.d.	n.d.–40	n.d.	n.d.	Golfinopoulos et al., 2003
Vergoritida	06/1996–06/1998	n.d.–72	n.d.	n.d.	n.d.	n.d.–26	n.d.	n.d.–12	n.d.–39	n.d.–20	n.d.–18	Golfinopoulos et al., 2003
L. Prespa	06/1996–06/1998	n.d.–36	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.–19	n.d.	n.d.	Golfinopoulos et al., 2003
S. Prespa	06/1996–06/1998	n.d.–95	n.d.–14	n.d.–21	n.d.	n.d.–26	n.d.	n.d.–37	n.d.–37	n.d.–10	n.d.–12	Golfinopoulos et al., 2003

[a] Sum of the two isomers (β-endosulfan was not detected).
[b] –, not monitored.
[c] n.d., not detected.

1996–1997 by Albanis et al. (1998) and the Axios river basin during 1997–1998 by Charizopoulos and Papadopoulou-Mourkidou (1999). For the central part of Imathia plain the higher concentrations of pesticides detected in rainfall waters were 0.007 μg/L for atrazine, 0.028 μg/L for DEA, 0.005 μg/L for simazine, 0.004 μg/L for carbofuran, 0.017 μg/L for diazinon, 0.004 μg/L for parathion and 0.006 μg/L for parathion methyl (Albanis et al., 1998). In the Axios river basin the most frequently found pesticides included alachlor (49%), parathion methyl (38%), atrazine (30%), quintozene (28%), metolachlor (24%), prometryne (23%) and molinate (22%). Occurrence of diazinon, chlorpyrifos-ethyl, methidathion, ethofumesate and parathion accounted for 14–17% of the samples analysed. Many other pesticides, including some of the most detected in Greek freshwaters (i.e. carbofuran, malathion, prometryne, EPTC, propanil, propachlor, trifluralin), were present in less than 10% of the samples. The concentrations of individual compounds ranged from 0.002 to 6.82 μg/L (Charizopoulos and Papadopoulou-Mourkidou, 1999). The presence of triazine herbicides and other pesticides in the rainwater was also reported for water bodies in European countries (Dubus et al., 2000). These results show that atmospheric transport and deposition of pesticides in neighbouring water bodies from their application sites can be a problem, at least regionally.

It must be noted that most of the commonly detected pesticides, i.e. alachlor, atrazine, simazine, trifluralin, diuron, lindane and endosulfan, in the surface waters of Greece are included in the priority list of the water framework directive (2000/60/EC) showing the need for monitoring and establishment of environmental quality standards for these compounds.

3. Seasonal patterns

Critical factors for the time elapse between the period of pesticide application in cultivation and their occurrence in rivers include the characteristics of the catchment (size, climatological regime, type of soil or landscape) as well as the chemical and physical properties of the pesticides (Capel et al., 2001). The size of the drainage basin affects the pesticide concentration profile, and Larson and co-workers showed that in large rivers (i.e. Missouri and Mississippi) the integrating effects of the many tributaries result in elevated pesticide concentrations that spread out over the summer months. In rivers with relatively small drainage basins (50,000–150,000 km^2) pesticide concentrations increased abruptly and the periods of elevated concentrations were relatively short—about one month—as pesticides were transported in run-off from local spring rains in the relatively small area (Larson et al., 1995). Although the drainage basins of the Greek rivers are small and short periods of increased pesticide concentrations would be expected, more outspread pesticide concentration profiles are observed. This is probably due to delayed leaching from soil as a result of dry weather conditions, which is reflected by the low mean annual discharges (15–65 m^3/s). As is stated in the corresponding studies, the major inputs of the compounds into the rivers occurred in

Table 6
Levels (ng/L) of s-triazine herbicide residues in water samples from various Greek rivers at different sampling periods

River	Sampling period	Atrazine	DEA	Simazine	Prometryne	Terbuthylazine	Metribuzin	Refs.
Aliakmon	03/1988–02/1989	n.d.–200	–[c]	n.d.–100	n.d.–6100	–	–	Albanis, 1991, 1992
	During 1991[a]	n.d.–700	–	n.d.–300	n.d.–500	–	n.d.–1100	Readman et al., 1993
	12/1990–09/1992	n.d.–2160	–	n.d.–340	n.d.	–	–	Albanis et al., 1995b
	05/1996–04/1997	n.d.–48	n.d.[d]–235	n.d.–27	n.d.		–	Albanis et al., 1998
Loudias	03/1988–02/1989	n.d.–5900	–	n.d.–50	n.d.–3000	–	–	Albanis, 1991, 1992
	During 1991[a]	n.d.–700	–	n.d.–300	n.d.–500	–	n.d.–1100	Readman et al., 1993
	05/1996–04/1997	n.d.–310	n.d.–407	n.d.–125	n.d.	–	–	Albanis et al., 1998
Axios	03/1988–02/1989	n.d.–3300	–	n.d.–50	n.d.–3100	–	–	Albanis, 1991, 1992
	During 1991[a]	n.d.–700	–	n.d.–300	n.d.–500	–	n.d.–1100	Readman et al., 1993
	1993–1994	n.d.–1230	n.d.–7	n.d.–3180	n.d.–550	n.d.–1	n.d.	Papadopoulou-Mourkidou, 2002
	06/1996	600	–	–	n.d.	–	–	Miliadis and Malatou, 1997
	1997–1998	n.d.–1000	n.d.–124	n.d.–214	n.d.–131	n.d.–13	n.d.	Papadopoulou-Mourkidou, 2002
Pinios	10/1996	n.d.	–	–	800	–	–	Miliadis and Malatou, 1997
Nestos	08/1993 and 06/1994	n.d.	–	120/–	n.d.	–	–	Albanis and Hela, 1995
Evros	08/1992–07/1993	n.d.–630	–	n.d.–320	n.d.	–	–	Angelidis and Albanis, 1996
Kalamas	09/1984–09/1985	2–84	–	n.d.–15	–	–	–	Albanis et al., 1986a
	09/1998–09/1999	n.d.–3870	n.d.–90	n.d.–490	n.d.	n.d.	–	Albanis et al., 2004
	01/2000–12/2000	20–230	30–90	n.d.	n.d.	10–20	–	Lambropoulou et al., 2002
Arachthos	During 1991[b]	n.d.–260	–	n.d.–400	n.d.	–	n.d.	Readman et al., 1993
	03/1992–02/1993	n.d.–240	–	n.d.–110	n.d.	–	n.d.	Albanis et al., 1995a
	09/1998–09/1999	n.d.–22	n.d.	n.d.–98	n.d.	n.d.	–	Albanis et al., 2004
Louros	During 1991[b]	n.d.–260	–	n.d.–400	n.d.	–	n.d.	Readman et al., 1993
	03/1992–02/1993	n.d.–4100	–	n.d.–1450	n.d.	–	n.d.	Albanis et al., 1995a
	08/1993 and 06/1994	n.d.	–	135; 70	n.d.	–	–	Albanis and Hela, 1995
	01/1995–08/1996	5–195	n.d.–215	n.d.–177	n.d.	–	–	Albanis and Hela, 1998
	09/1998–09/1999	n.d.–204	n.d.–128	n.d.–222	n.d.	n.d.	–	Albanis et al., 2004
Evrotas	08/1991–08/1992	30–1160	–	30–290	n.d.	–	n.d.	Angelidis et al., 1996
Honos	11/1995–06/1996	10–30	–	10–36	n.d.	n.d.	40–46	Vassilakis et al., 1998
Havgas	11/1995–061996	10–56	–	10–39	n.d.	n.d.	n.d.	Vassilakis et al., 1998
Aposelemis	11/1995–06/1996	10–40	–	10–28	n.d.	n.d.	n.d.	Vassilakis et al., 1998

[a] Data were grouped for the three rivers (Axios, Loudias and Aliakmon) in the corresponding reference.
[b] Data were grouped for the two rivers (Arachthos and Louros) in the corresponding reference.
[c] –, not monitored.
[d] n.d., not detected.

the summer period (May–August) after their application in agricultural fields, as a result of their surface run-off and the lower flow rates of the rivers (Albanis, 1991; Albanis et al., 1995b, 1998; Angelidis and Albanis, 1996; Albanis and Hela, 1998). In some cases a lower peak is observed in late September–October due to the rainfall events after the dry summer period (Albanis et al., 1998; Lambropoulou et al., 2002). Low concentrations were observed during the winter months because of dilution effects due to high-rainfall events and the increased degradation of pesticides after their application. Thus, pesticides were flushed to the surface water systems as pulses in response to late spring and early summer rainfall, as reported also elsewhere (Thurman et al., 1991). This trend is also followed for the occurrence of pesticides in Spanish (Planas et al., 1997) and Italian (Arno river) (Griffini et al., 1997) surface waters. The highest percentages of samples containing pesticides from Spanish surface waters were found during the months of April (55%), May (33%), June (44%) and July (33%), while for November and December the lowest presence of these compounds were observed. Similarly, the maximum concentrations of pesticides were observed each year in May and June during 1992–1995 in the case of the Arno river.

The character of the landscape in combination with the type of cultivation in the catchment area may well affect the temporal variations in riverine concentrations of pesticides. For example, for the relatively large basin of the River Rhone, the concentration of triazines display a short peal from late April until late June with relatively constant concentrations during the rest of the year (Steen, 2002), due to the fact that herbicides are used in vineyards situated on mountain slopes that promote rapid run-off. For the River Ebro, elevated herbicide concentrations were found to continue long after the application period, i.e. from May through December (Gascon et al., 1998). A similar pattern was found for the river Scheldt with peak discharge in August of 1996 (Steen et al., 2001).

As far as the metabolites and especially for DEA, early rainfall followed by a dry summer will result in an early peak concentration in surface waters. A wet summer will delay the maximum concentrations of metabolites and increase their run-off into surface water, occasionally resulting in a slight separation of the parent atrazine maximum concentrations,

Table 7
Levels (ng/L) of various commonly used herbicides in water samples from various Greek rivers at different sampling periods

River	Sampling period	Alachlor	Metolachlor	Molinate	Propanil	Trifluralin	2,4-D	MCPA	EPTC	Bentazon	Mecoprop	Diuron	Refs.
Aliakmon	03/1988–02/1989	n.d.–5500	100[c]	740[c]	280[c]	n.d.–950	n.d.–650	n.d.–4850	40[c]	–[d]	–	120[c]	Albanis, 1991, 1992
	12/1990–09/1992	n.d.–210	n.d.–340	n.d.	n.d.[e]	n.d.–550	n.d.–120	n.d.–1560	n.d.–120	–	–	n.d.	Albanis et al., 1995b
	During 1991[a]	n.d.–1300	n.d.–500	n.d.–900	–	n.d.–460	n.d.–1200	n.d.–800	–	n.d.	–	n.d.–700	Readman et al., 1993
	05/1996–04/1997	n.d.–23	n.d.	n.d.–112	n.d.	n.d.	–	–	–	–	–	–	Albanis et al., 1998
Loudias	03/1988–02/1989	n.d.–9300	500[c]	750[c]	420[c]	n.d.–500	n.d.–900	n.d.–3800	250[c]	–	–	100[c]	Albanis, 1991, 1992
	05/1996–04/1997	n.d.–265	n.d.–558	n.d.–320	n.d.–340	n.d.–16	–	–	–	–	–	–	Albanis et al., 1998
	During 1991[a]	n.d.–1300	n.d.–500	n.d.–900	–	n.d.–460	n.d.–1200	n.d.–800	–	n.d.	–	n.d.–700	Readman et al., 1993
Axios	03/1988–02/1989	n.d.–8500	600[c]	30[c]	25[c]	n.d.–350	n.d.–400	n.d.	310[c]	–	–	100[c]	Albanis, 1991, 1992
	During 1991[a]	n.d.–1300	n.d.–500	n.d.–900	–	n.d.–460	n.d.–1200	n.d.–800	–	n.d.	–	n.d.–700	Readman et al., 1993
	1993–1994	n.d.–260	n.d.–520	n.d.–300	n.d.–20600	n.d.–152	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	Papadopoulou-Mourkidou, 2002
	1997–1998	n.d.–31	n.d.–1000	n.d.–768	n.d.–85	n.d.–312	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	Papadopoulou-Mourkidou, 2002
Evros	08/1992–07/1993	n.d.–370	n.d.	–	–	n.d.–21	–	n.d.	–	n.d.	–	–	Angelidis and Albanis, 1996
Kalamas	01/2000–12/2000	40–130	n.d.	–	–	20–300	–	–	40–120	–	–	–	Lambropoulou et al., 2002
	09/1998–09/1999	n.d.–939	n.d.	n.d.	n.d.	n.d.–325	–	–	n.d.–1851	–	–	–	Albanis et al., 2004
Arachthos	03/1992–02/1993	n.d.–350	n.d.–60	–	–	n.d.–90	n.d.	n.d.	–	–	–	n.d.–260	Albanis et al., 1995a
	During 1991[b]	n.d.–1400	n.d.–800	n.d.	–	n.d.–360	n.d.–500	n.d.–900	–	n.d.	–	n.d.–600	Readman et al., 1993
	09/1998–9/1999	n.d.	n.d.	n.d.	n.d.	n.d.–15	–	–	n.d.–120	–	–	–	Albanis et al., 2004
Louros	08/1993/06/1994	n.d.	n.d.	55/50	n.d.	n.d.	–	n.d.	n.d.	–	–	–	Albanis and Hela, 1995
	03/1992–02/1993	n.d.–1650	n.d.–1120	n.d.	n.d.	n.d.–360	n.d.	n.d.	–	–	–	n.d.–140	Albanis et al., 1995a
	01/1995–08/1996	n.d.–39	n.d.–257	n.d.–36	n.d.–50	n.d.–206	–	–	–	–	–	–	Albanis and Hela, 1998
	During 1991[b]	n.d.–1400	n.d.–800	n.d.	–	n.d.–360	n.d.–500	n.d.–900	–	n.d.	–	n.d.–600	Readman et al., 1993
	09/1998–09/1999	n.d.–1026	n.d.	n.d.	n.d.	n.d.–201	–	–	n.d.–897	–	–	–	Albanis et al., 2004
Evrotas	08/1991–08/1992	n.d.–570	n.d.	–	–	–	–	–	–	–	–	n.d.	Angelidis et al., 1996
Honos	11/1995–06/1996	–	–	–	–	–	40–194	10–52	–	n.d.	10–29	–	Vassilakis et al., 1998
Havgas	11/1995–06/1996	–	–	–	–	–	n.d.	n.d.	–	10–24	10–24	–	Vassilakis et al., 1998
Aposelemis	11/1995–06/1996	–	–	–	–	–	40–300	10–170	–	n.d.	n.d.	–	Vassilakis et al., 1998

[a] Data were grouped for the three rivers (Axios, Loudias and Aliakmon) in the corresponding reference.
[b] Data were grouped for the two rivers (Arachthos and Louros) in the corresponding reference.
[c] Mean values.
[d] –, not monitored.
[e] n.d., not detected.

Table 8
Levels (ng/L) of s-triazine herbicide, organophosphorus and carbamate insecticide residues in water samples from various Greek lakes at different sampling periods

s-Triazines

Lake	Sampling period	Atrazine	DEA	Simazine	Prometryne	Terbuthylazine	Aminotriazole	Refs.
Pamvotida	09/1984–09/1985	n.d.–390	–[a]	n.d.–80	–	–	n.d.–14	Albanis et al., 1986a
	08/1993 and 06/1994	140/85	n.d.[b]	n.d.	n.d.	n.d.	n.d.	Albanis and Hela, 1995
	04/1998–04/1999	n.d.–792	n.d.–120	n.d.–28	n.d.	n.d.	–	Albanis et al., 2004
Iliki	12/1992–02/1993	n.d.	–	n.d.	–	n.d.	–	Tsipi and Hiskia, 1996
Marathonas	12/1992–02/1993	40[c]	–	40[c]	–	15	–	Tsipi and Hiskia, 1996
Mornos	12/1992–02/1993	45[c]	–	45[c]	–	n.d.	–	Tsipi and Hiskia, 1996

Organophosphorus and carbamate insecticides

Lake	Sampling period	Parathion M.	Diazinon	Azinphos M.	Carbofuran	Carbaryl	Malathion	Ethion	Refs.
Pamvotida	09/1984–09/1985	n.d.–12	n.d.–57	n.d.–16	n.d.–42	n.d.–38	–	–	Albanis et al., 1986b
	04/1998–04/1999	n.d.	n.d.–2105	–	n.d.–158	–	n.d.–1227	n.d.–993	Albanis et al., 2004
Mornos	05/1996	n.d.	n.d.	100	–	–	n.d.	–	Miliadis and Malatou, 1997

[a] –, not monitored.
[b] n.d., not detected.
[c] Data reported as the sum of atrazine and simazine.

giving a 'second flush' of triazine metabolites to surface waters (Thurman et al., 1994).

In general, results revealed that highest concentrations correspond to agricultural applications with episodic peaks correlating with meteorological and hydrological events such as rainstorms and flooding.

Similar trends and temporal variations were also observed in lakes. The only difference is that residues were detected during a longer period as a result of the lower water flushing and renewal time as compared to rivers. Seasonal trends with a concentration increase of some s-triazines in the epilimnia were observed in several lakes of Switzerland around June, indicating also fast response of the aquatic environment to new applications of these herbicides in spring (Buser, 1990). Concentrations for atrazine, terbuthylazine, simazine and prometryne varied from non-detectable to 20 ng/L, with atrazine showing the highest concentration followed by terbuthylazine.

4. Comparison with European surface waters

Within Europe, the contamination of freshwaters by pesticides follows comparable concentration levels and patterns as recorded in most countries including Italy, Spain, France, Portugal, United Kingdom, Germany and Netherlands. Table 10 shows herbicide concentration data in major rivers from Mediterranean and European countries. We selected data mostly from the Mediterranean region in order have comparable climatic conditions, cultivations and type of landscape with those reported for Greece. It is apparent that the five most commonly encountered compounds in European freshwaters were atrazine, simazine, metolachlor, alachlor and molinate. s-Triazine herbicides are widely applied herbicides in Europe for pre- and post-emergence weed control among various crops as well as in non-agricultural purposes. Levels of simazine are generally lower than those reported for atrazine, a fact also observed in Greek waters. Low levels of atrazine in the River Po compared with other areas probably arise owing to the banning of this compound in Italy for agricultural use. The occurrence of high levels of terbuthylazine and/or metolachlor might be related to the use of these herbicides as substitutes for atrazine, as all three compounds have overlapping herbicidal properties (Tomlin, 1995). In some studies acetamide herbicides alachlor and metolachlor (which are also used to control grasses and weeds in a broad range of crops) were also detected at levels comparable to those of the triazines (Table 10). Molinate was detected in River Po in higher concentrations than in Greek surface waters, while propanil and trifluralin concentrations in European surface waters are low-level but scarce.

Concerning insecticide concentrations in European freshwaters, mainly organophosphates and organochlorines insecticides have been detected. Diazinon was detected in surface waters of Spain and in River Scheldt (Netherlands) at concentrations up to 530 and 87 ng/L, respectively (Planas et al., 1997; Steen et al., 2001). Parathion methyl was detected in the Segre (Spain), Elbe and Rhine rivers (Germany) at concentrations up to 270, 332 and 40 ng/L, respectively (Planas et al., 1997; Gotz et al., 1998), and fenitrothion was detected in the Segre (Spain) and Humber rivers (UK) at concentrations up to 490 and 270 ng/L, respectively (Planas et al., 1997; House et al., 1997). Finally, malathion was detected only in River Arno (Italy), at 170 ng/L (Griffini et al., 1997), ethion in the Segre river basin (Spain) at 10 ng/L (Planas et al., 1997), and dimethoate in the Elbe and Rhine rivers (Germany) at 3210 and 50 ng/L, respectively (Gotz et al., 1998). Similar trends were found in Greece with the exception of River Axios in which higher amounts were reported for parathion, malathion and carbofuran.

Among organochlorine insecticides, lindane was the most frequently detected compound. It was detected in the Ebro river delta (Spain) at concentrations up to 2100 ng/L, in the Tejo and Sado rivers (Portugal) up to 14 and 86 ng/L, respectively, in the Humber and Thames rivers (UK) up to 97 and 13 ng/L, respectively, and in the Elbe and Rhine (Germany) rivers up to 29 and 30 ng/L, respectively. Other organochlorine insecticides include α-endosulfan that was detected in rivers Segre, Tejo and Sado and Elbe at 10, 4 and 181 and 5 ng/L, respectively (Aguilar et al., 1997; House et al., 1997; Power et al., 1999;

Table 9
Levels (ng/L) of various commonly used insecticides in water samples from various Greek rivers at different sampling periods

River	Sampling period	Malathion	Fenthion	Diazinon	Parathion methyl	Parathion ethyl	Ethion	Disulfoton	Demeton-S-methyl	Pyrazophos	Carbofuran	Phosalone	Refs.
Aliakmon	12/1990–09/1992	–[a]	n.d.–110	n.d.–160	n.d.–210	n.d.	–	–	–	–	n.d.	–	Albanis et al., 1995b
	05/1996–04/1997	n.d.[b]	n.d.	n.d.	n.d.	n.d.	n.d.	–	–	–	n.d.	–	Albanis et al., 1998
Loudias	05/1996–04/1997	n.d.–18	n.d.	n.d.–28	n.d.–183	n.d.	n.d.	–	–	–	n.d.	–	Albanis et al., 1998
Axios	1993–1994	n.d.–2000	n.d.–5	n.d.–90	n.d.–250	n.d.–290	n.d.	–	–	–	n.d.–7300	–	Papadopoulou-Mourkidou, 2002
	11/1996	n.d.	–	n.d.	n.d.	n.d.	–	–	600	n.d.	–	n.d.	Miliadis and Malatou, 1997
	1997–1998	n.d.–1000	n.d.	n.d.–102	n.d.–362	n.d.–2000	n.d.	–	–	n.d.–2000	–	–	Papadopoulou-Mourkidou, 2002
Pinios	10/1996	100	–	n.d.	n.d.	n.d.	–	–	n.d.	n.d.	–	n.d.	Miliadis and Malatou, 1997
Evros	08/1992–07/1993	–	n.d.	n.d.–210	n.d.–120	n.d.	–	–	–	–	–	–	Angelidis and Albanis, 1996
	11/1996	n.d.	–	10	n.d.	n.d.	–	–	n.d.	n.d.	–	n.d.	Miliadis and Malatou, 1997
Kokinos	05/1996	n.d.	–	n.d.	n.d.	n.d.	–	–	n.d.	100	–	100	Miliadis and Malatou, 1997
Kalamas	09/1984–09/1985	–	–	n.d.–52	n.d.–32	–	–	–	–	–	n.d.–14	–	Albanis et al., 1986b
	09/1998–09/1999	n.d.	n.d.	n.d.–775	n.d.–271	n.d.–40	n.d.	–	–	–	n.d.–160	–	Albanis et al., 2004
	01/2000–12/2000	–	10–30	40–250	50–90	20–40	10–30	10–70	–	–	30–150	–	Lambropoulou et al., 2002
Arachthos	09/1998–09/1999	n.d.	n.d.	n.d.–57	n.d.	n.d	n.d	–	–	–	n.d.–553	–	Albanis et al., 2004
Louros	08/1993 and 06/1994	n.d.	–	n.d.	n.d.	n.d.	n.d.	–	–	–	n.d.	–	Albanis and Hela, 1995
	01/1995–08/1996	–	–	–	n.d.–5	–	–	–	–	–	n.d.–15	–	Albanis and Hela, 1998
	09/1998–09/1999	n.d.	n.d.	n.d.–234	n.d.–70	n.d.	n.d.	–	–	–	n.d.–111	–	Albanis et al., 2004
Evrotas	08/1991–08/1992	n.d.	n.d.–230	n.d.–90	n.d.–30	n.d.	–	–	–	n.d.	n.d.–10	–	Angelidis et al., 1996

[a] –, not monitored.
[b] n.d., not detected.

Table 10
Levels (ng/L) of various commonly used herbicides in water samples from various Mediterranean and European rivers at different sampling periods

River	Sampling period	Atrazine	Terbuthylazine	Simazine	Alachlor	Metolachlor	Molinate	Propanil	Trifluralin	Diuron	Refs.
Spain											
Ebro river	03/1991–06/1991	17–190	–[b]	28–138	<1–206	32–132	<1–38	–	<1	–	Readman et al., 1993
	03/1995–06/1996	22–118	–	18–51	4–14	n.d.–4	–	–	–	–	Gascon et al., 1998
	04/1998–07/1998	180–240	–	50–1060	–	n.d.–200	n.d.–290	–	–	–	Aguilar et al., 1999
Llobregat river	During 2000	<25–29	n.d.[c]	<25–84	n.d.	n.d.	–	–	n.d.	–	Quintana et al., 2001
Surface waters	02/1993–12/1993	10		10	n.d.	330–3000	60–880	10	–	–	Planas et al., 1997
Segre river basin	05/1995–06/1995	10–90	10	3–100	10	10–80	10–140	n.d.	n.d.	–	Planas et al., 1997
Ter, Llobregat river	05/1995–06/1995	20–30	n.d.	n.d.	n.d.	n.d.	10–80	n.d.	n.d.	–	Planas et al., 1997
Guarena, Almar rivers	06/1998–12/1998	n.d.–140	–	–	90–160	–	–	–	–	n.d.	Garabias-Martinez et al., 2000
France											
Rhone river	04/1991–11/1991	40–291	–	22–372	–	–	–	1–3	–	–	Readman et al., 1993
Loire river	During 1995[a]	251	19	91	n.d.	14	n.d.	–	n.d.	traces	Steen, 2002
Marne river	1992–1993	60–1300	10–1540	5–840	–	–	–	–	–	n.d.–1027	Garmouma et al., 1998
Italy											
Po river	03/1991–07/1991	21–118	4–149	6–81	<30–106	<30–605	<3–1750	<3	–	–	Readman et al., 1993
	05/1988–06/1991	20–688	n.d.–320	n.d–180	11–213	–	n.d.–3158	–	–	–	Brambilla et al., 1993; Galassi et al., 1992
Arno river	1992–1995[a]	60–160	200–2270	100–300	20–440	160–3680	n.d.	n.d.	n.d.	–	Griffini et al., 1997
Tuscany surface water	–	n.d.–50	n.d.–500	n.d.–620	n.d.	n.d.–250	n.d.	n.d.	n.d.–30	–	Sbrilli et al., 2005
Portugal											
Tejo	1990–1993	80–630	n.d.	100–294	n.d	n.d.	42–580	n.d.	–	–	Cerejeira et al., 2003
Guadiana	1998–1999	n.d.–170	n.d.	n.d.–310	n.d.	n.d.	n.d.	n.d.	–	–	Cerejeira et al., 2003
Sado	1998–1999	n.d.	n.d.	n.d.	n.d.	n.d.	<20–4800	n.d.	–	–	Cerejeira et al., 2003
UK											
Humber river	04/1994–041995	40–540	–	20–140	–	–	–	–	–	50–8700	House et al., 1997
Thames river	1988–1997	38–120	–	40–167	–	–	–	–	–	–	Power et al., 1999
Netherlands											
Scheldt river	1996–02/1998	60–750	–	50–570	2–100	25–1000	–	–	–	n.d–1350	Steen et al., 2001; Steen, 2002
Germany											
Elbe river	1992–1993	13–347	2–52	7–331	–	–	–	–	–	–	Gotz et al., 1998
Rhine river	1992–1993[a]	130	420	110	–	–	–	–	–	–	Gotz et al., 1998

[a] Maximum values.
[b] –, not monitored.
[c] n.d., not detected.

Gotz et al., 1998; Cerejeira et al., 2003) and aldrin that was detected in the Ebro delta and river Sado at 1500 ng/L and 179 ng/L, respectively (Aguilar et al., 1997; Cerejeira et al., 2003). Aldrin was found at lower levels in surface waters of Greece.

Fungicides were not generally present at high concentrations in European surface waters and usually the detected levels were below detection limits. Only sporadic run-off of certain fungicides (e.g. captafol, captan, carbendazim, dicloran and folpet) was also reported in estuaries of major Mediterranean rivers. This is in accordance with the reported concentrations in Greece. Low concentrations of captafol and folpet were detected in the Po river mouth (<1–14 ng/L) while relatively high concentrations (<2–474 ng/L) of dicloran were recorded in samples from the Rhone river. High concentrations of carbentazim were found in Ebro delta rice fields (up to 200 μg/L) (Readman et al., 1997).

Finally, in a limited mention for the United States, the most commonly encountered compounds include also atrazine, simazine, alachlor and metolachlor from herbicides and diazinon, malathion from insecticides (USGS, 1999). Generally, acetamide and triazine herbicides are widely used to control grasses and weeds in a broad range of crops, and were detected at comparable levels.

5. Conclusions

Pesticides that belong to s-triazines and amide herbicides as well as organophosphorus insecticides were more frequently detected in Greek surface waters. Similar trends with exceptions owing to local scale applications or site characteristics were found between Greek and other European surface freshwaters. Organochlorines pesticides still persist in the aquatic environment of Greece. Monitoring focused on certain key regions should be continued to better understand the effects of these mitigation methods. Time profiles are influenced by a number of factors, which cannot always be predicted. Thus, monitoring of pesticides should be performed over a period of time and with a frequency that allows all seasonal events to be accounted for.

The countrywide patterns of chemical use are constantly changing as the popularity of existing herbicides rises and falls and as new compounds are introduced into farming. The research should be completed for pesticide degradation products commonly found in surface waters. Only a few degradation products were examined in the previous nationwide investigations.

With sufficient continued monitoring, the overall trend towards reduced pesticide loading into aquatic systems caused by the combinations of use rate reduction, use restrictions and alterations of agricultural management practices may become discernible.

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