

# Occurrence of Pesticides in Rain of the Axios River Basin, Greece

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The Axios River Basin is one of the most developed agricultural areas of Greece. Samples from a total of 205 rain events collected from eight sampling stations during 1997–1998 were analyzed for pesticide residues of which 186 events (90%) yielded at least one positive detection. Among 160 target pesticides and some major conversion products recovered by solid-phase extraction (SPE) and analyzed by GC–ITMS, 47 compounds were found in at least one rain event. The most frequently found pesticides included alachlor (49%), lindane (44%), parathion-methyl (38%), atrazine (30%), quintozone (28%), metolachlor (24%), prometryne (23%), and molinate (22%). Occurrences of diazinon, chlorpyrifos-ethyl, methidathion, ethofumesate, and parathion accounted for 14–17% of the samples analyzed. The remaining target analytes were present in less than the 10% of the analyzed samples. Concentrations of individual compound ranged from 0.002 to 6.82  $\mu\text{g/L}$ . Greater pesticide concentrations occurred during application seasons. The estimated annual deposition rates for the sum of the pesticides ranged from 51 to 395  $\mu\text{g/m}^2$  of soil surface.

## Introduction

During the last 30 years, an increasing number of researchers have well-documented the presence of pesticides in the atmosphere and rainfall as a result of drift during application, volatilization from treated surfaces, and wind erosion from soil.

Once pesticides enter the atmosphere, they can be transported often at considerable distances from their applications sites. This is especially true for the persistent organochlorine pesticides that have been detected even in remote regions, such as the North and South Poles (1, 2). Further studies especially during the past decade have demonstrated that atmospheric transport can be a problem, at least regionally, for pesticides from various chemical groups (3–6). Organophosphate insecticides and their oxon analogues were detected at an elevation of 1930 m in California Sierra Nevada Mountains, approximately 150 km from their nearest application sites (3), while triazines were found in rainwater at the island Helioland (North Sea), located 50 km off the coast (4).

More than 40 pesticides have been detected in precipitation in the United states (3, 7–10), Europe (4–6, 11–13), and Africa (14). Triazines and acetanilides were the most frequently found pesticides in these studies. Maximum reported concentrations are 40  $\mu\text{g/L}$  for atrazine (United states) (9) and 430  $\mu\text{g/L}$  for 2,4-D (South Africa) (14).

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TABLE 1. Description of Sampling Sites of the Axios River Basin

sampling site	elevation (m)	location of the sampler	main crops of the area
1	360	middle of a town	vines and orchards
2	70	open field	of apple, peach, and cherry trees
3	40	open field	cotton, corn, rice, sugar beets and vegetables
4	10	open field	wheat, oat and barley
5	10	open field	corn, wheat, oat, barley and vegetables
6	30	middle of a town	
7	280	middle of a town	
8	20	open field	

The basin of the Axios River (central Macedonia, northern Greece) (Figure 1) is one of the most developed agricultural areas of the country and receives annually a heavy load of numerous pesticides. In this area, the crop rotation schemes usually include high priced crops (rice, cotton, corn, sugar beets, vegetables, and orchards) and very rare crops of low pesticide input (alfalfa and wheat, oat, and barley). The occurrence of pesticide residues in most surface aquatic systems of the basin, even in remote areas of the highlands, triggered the investigation of the presence of pesticides in rainwater of the basin.

This study was initiated in June 1997 and continued until June 1998. The purpose of the present study (the first of this kind in Greece) was to monitor the occurrence of a wide range of pesticides in rainwater of the basin and to determine their temporal and spatial variations.

## Experimental Section

**Sample Collection.** The rain sampler consisted of a glass funnel (20 cm diameter) connected to a 2.5-L amber glass bottle. The sampler remained outdoors continuously for collecting both wet and dry deposition. The collection of rainwater was event-related. The collected rainwater was emptied into 1-L dark amber bottles and kept in ice until transferred to the laboratory. Samples were processed (extracted) as soon as delivered to the analytical laboratory.

**Sampling Sites.** Eight sampling sites (SS) were chosen to encircle the basin (Figure 1) and represent areas where the principle crops of the basin are produced. A brief description of the sampling sites is given in Table 1.

**Materials.** All solvents (ethyl acetate, acetone, and methanol) used in this study were of pesticide residue grade and were purchased from Merck (Darmstadt, Germany). Anhydrous sodium sulfate was of proanalysis grade and was also purchased from Merck. Lichrolut EN (a polymer of ethylvinylbenzene–divinylbenzene copolymer, 40–120  $\mu\text{m}$  particle size, 200 mg sorbent mass) SPE cartridges were purchased from Merck. Membrane (0.45  $\mu\text{m}$ ) and paper filters were purchased from Schleicher & Schuell (Dassel, Germany). Pesticide analytical standard materials were purchased from Promochem (Augsburg, Germany), Chem Servis (West Chester, PA), Riedel-de Haen (Seelze-Hannover, Germany), and Altech (Deerfield, IL). Analytical standards of atrazine, deisopropylatrazine, deethylatrazine, and metolachlor were donated by CIBA (Basel, Switzerland).

**Instrumentation.** A Tracker/Magnum ion trap mass spectrometer (ITMS) (Finnigan MAT, San Jose, CA) associated with a Varian (Varian Instruments, Sunnyvale, CA) model 3300 gas chromatograph was used for the analysis of pesticides and conversion products. An on-column septum-equipped programmable injector (SPI) was used on the gas

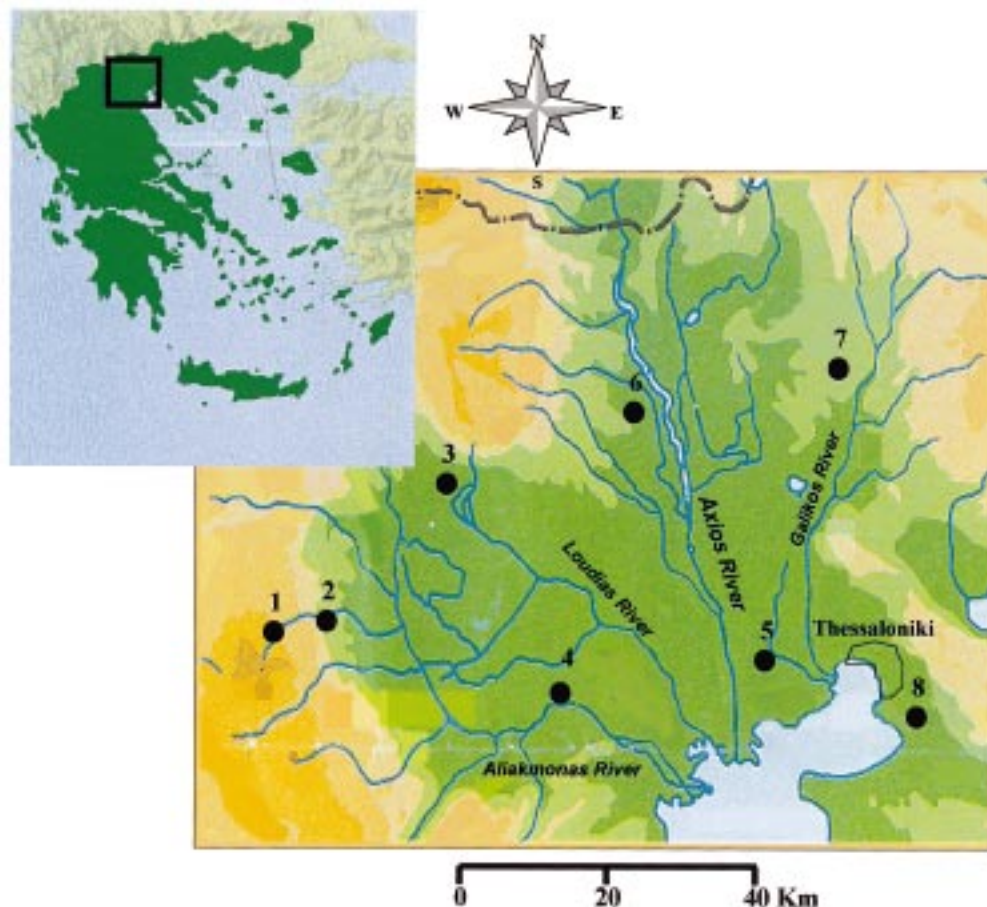


FIGURE 1. Geophysical map of the study area. Numbers 1–8 indicate the sampling sites; the geographical location of the study area on the map of Greece is indicated by the square.

chromatograph. The injector temperature was programmed from 55 °C (hold 0.5 min) to 200 °C at 145 °C/min (hold 15 min at 200 °C). A guard column (1.5 m × 0.25 mm i.d.) of deactivated fused silica (Alltech, Deerfield, IL) was inserted between the injector and the analytical column (30 m × 0.25 mm i.d. DB-1301 and 0.25 μm film thickness from J&W Scientific, Folsom, CA). The guard column was replaced periodically depending upon the type of samples analyzed. The oven was temperature programmed from 55 °C (hold 0.5 min at 55 °C) to 140 °C at 30 °C/min, from 140 to 190 °C at 3 °C/min (hold 2 min at 190 °C), from 190 to 220 °C at 4 °C/min (hold 1 min at 220 °C), and from 220 to 270 °C at 5.5 °C/min (hold 10 min at 270 °C). The total run time was 50 min. As carrier gas was used helium at 30 cm/s giving column head pressure of 7 psi. Injections of 2 μL were made by use of a model A200S autosampler. The ITMS was operated in the EI (electron impact) mode. Other operational parameters have been previously reported (15).

**Analysis.** For the analysis of pesticides and conversion products, samples were extracted by SPE on Lichrolut EN cartridges. Sorbed solutes were eluted with ethyl acetate, and the eluates were dried by passing through minicolumns containing granular anhydrous sodium sulfate. After being dried, eluates concentrated to small volumes were transferred into autosampler vials and concentrated to dryness by use of a nitrogen stream. The residue dissolved into ethyl acetate, the volume ranging from 1000 to 100 μL depending upon the size of the collected sample and maintaining a concentration ratio of 10 000, was analyzed by a GC–MS ion trap as previously described (15). The entire collected sample of a certain rain event was always processed using one SPE cartridge per maximum of 1 L of sample. Rain samples of

<100 mL were not considered, and thus rain events with <4 mm of rainfall were not included in this study.

In cases where sufficient dry deposition had occurred, the rainwater samples were filtered from paper filters before SPE extraction. Filtering of cloudy samples was necessary to avoid clogging of the SPE cartridge. The filters were extracted separately with 100 mL of acetone by shaking in glass vials overnight. Acetone extracts were dried over sodium sulfate, as described above, and extracts were combined with the respective SPE eluates. In preliminary studies, the extracts of the filtered material analyzed separately did not show the presence of detectable pesticide residues. However, it was considered appropriate to combine these extracts with the SPE eluates to avoid any probable constituent loss.

## Results and Discussion

A brief description of the selected sampling sites (SS) is given in Table 1. These SS are located on the periphery of the Axios River Basin (Figure 1) and the elevation of the respective areas ranging from 360 m (SS 1) to 10 m (SS 4 and 5). Three SS (1, 6, and 7) are located in residential areas, and the rest are located in open fields away from pesticide spray areas to avoid direct contamination of the sampling devices by spray drift.

A total of 205 rainwater samples, corresponding to equal number of rain events, was collected and analyzed for 160 compounds including pesticides and some of their major conversion products. Table 2 summarizes the collected data. In Table 2, the limits of detection (LODs) of the analytical method are also given. These LODs were calculated by processing 1 L of fortified samples with the final residue

**TABLE 2. Limits of Detections (LODs) of the Analytical Method, % of Rain Events (RE), among 205 RE, with Positive Findings and Minimum and Maximum Measured Concentrations, Mean Values of All Measured Concentrations, and Maximum Annual Deposition Rates ( $\mu\text{g}/\text{m}^2$ ) of 47 Target Analytes**

pesticides	LODs ( $\mu\text{g/L}$ )	% RE with positive findings	measured concn ( $\mu\text{g/L}$ )		mean concn ( $\mu\text{g/L}$ )	max annual deposition ( $\mu\text{g/m}^2$ )
			min	max		
Insecticides						
lindane	0.005	44	0.01	1.03	0.13	42.2
parathion-methyl	0.003	38	0.008	1.65	0.25	132.0
diazinon	0.005	17	0.008	0.21	0.07	10.8
chlorpyrifos-ethyl	0.010	16	0.03	0.2	0.08	19.2
methidathion	0.002	15	0.004	0.39	0.07	9.3
parathion-ethyl	0.002	14	0.004	0.22	0.07	7.5
phosmet	0.005	9	0.006	0.55	0.08	15.4
malathion	0.005	8	0.01	0.2	0.05	10.3
paraoxon-methyl	0.005	7	0.008	0.22	0.09	18.2
phosalone	0.005	6	0.01	0.43	0.07	14.0
carbofuran	0.005	3	0.02	1.01	0.31	10.1
malaaxon	0.010	3	0.02	0.1	0.04	5.2
fenoxycarb	0.005	3	0.005	0.07	0.02	0.3
azinphos-ethyl	0.010	2	0.02	0.2	0.07	1.4
azinphos-methyl	0.010	2	0.03	1.32	0.3	6.3
carbaryl	0.005	2	0.01	0.35	0.09	4.4
phorate sulfone	0.01	2	0.01	0.25	0.08	3.8
pirimicarb	0.005	2	0.08	0.42	0.21	6.1
dichlorvos	0.005	1	0.02	0.02	0.02	0.5
d-HCH	0.010	1	0.03	0.05	0.04	0.8
phorate sulfoxide	0.01	1	0.12	0.56	0.34	5.5
endosulfan a	0.050	1	0.07	0.09	0.08	1.8
endosulfan sulfate	0.050	0.5	0.08	0.08	0.08	2.1
pirimifos-methyl	0.005	0.5	0.007	0.007	0.007	0.3
Herbicides						
alachlor	0.005	49	0.007	2.2	0.14	34.3
atrazine	0.005	30	0.01	1.81	0.15	23.5
metolachlor	0.001	24	0.002	1.08	0.09	30.4
prometryne	0.001	23	0.003	2.9	0.28	30.5
molinate	0.005	22	0.03	6.82	0.56	110.5
ethofumesate	0.001	15	0.003	0.38	0.05	11.0
trifluraline	0.001	8	0.001	0.04	0.01	0.6
deethylatrazine	0.005	7	0.01	0.19	0.06	1.9
pendimethalin	0.005	6	0.004	0.07	0.02	2.1
EPTC	0.01	2	0.01	0.46	0.14	4.9
propanil	0.010	2	0.17	1.11	0.48	16.5
simazine	0.005	1	0.22	0.73	0.48	5.3
cyanazine	0.005	1	0.14	0.6	0.37	4.9
cycloate	0.005	1	0.004	0.004	0.004	0.1
deisopropylatrazine	0.050	0.5	0.1	0.1	0.1	0.7
propachlor	0.005	0.5	0.04	0.04	0.04	1.4
Fungicides						
quintozene	0.005	28	0.002	2.8	0.72	120.5
chlorothalonil	0.010	8	0.02	1	0.23	75.6
captan	0.005	2	0.009	0.11	0.03	3.4
metalaxyl	0.005	0.5	0.14	0.14	0.14	2.0
fenarimol	0.005	0.5	0.03	0.03	0.03	0.5
pyrazophos	0.005	0.5	0.01	0.01	0.01	0.4
fenpropimorf	0.005	0.5	0.005	0.005	0.005	0.2

dissolved in 100  $\mu\text{L}$  of ethyl acetate (concentration ratio 10 000) and analyzed by GC-ITMS (15). Thus in samples of rain events with volumes 100–<1000 mL, the respective LODs are in the range of 10–>1 higher. As mentioned in the Experimental Section, for samples >1000 mL, the final residue was dissolved in the appropriate volume (>100  $\mu\text{L}$ ) of ethyl acetate to maintain the 10 000 concentration ratio. Single and multiple detections of the target analytes occurred in 186 (90%) of the analyzed samples.

Forty-seven compounds were found at least in one rain event. Insecticides occurred more frequently than herbicides and fungicides; however, herbicides exhibited higher maximum concentrations (Table 2). Molinate, used exclusively in rice fields, yielded the highest concentrations. The concentration of molinate exceeded the 1  $\mu\text{g}/\text{L}$  level in seven rain events and reached the 6.82  $\mu\text{g}/\text{L}$  level once. These con-

centrations result from high rates of volatilization of molinate from field water surface and its vapor phase stability (16). Molinate has been observed in concentrations up to 1.72  $\mu\text{g}/\text{m}^3$  in air samples of California (16). Alachlor, lindane, parathion-methyl, atrazine, quintozone, metolachlor, prometryne, and molinate were the most frequently detected compounds. Similar concentrations with those found in the present study were reported by other investigators for atrazine (5–8, 10), alachlor (10), metolachlor (7, 10), deethylatrazine (6), and lindane (11, 13).

Seasonal trends were observed in the appearance of pesticides in rainwater. Sample data are presented in Figures 2–4 for SS 1, 5, and 7, respectively, representing the three crop subareas of the basin (Table 1). The highest concentrations for the herbicides of annual crops appeared in the period of March–June (Figures 3 and 4). Later detections were rare,



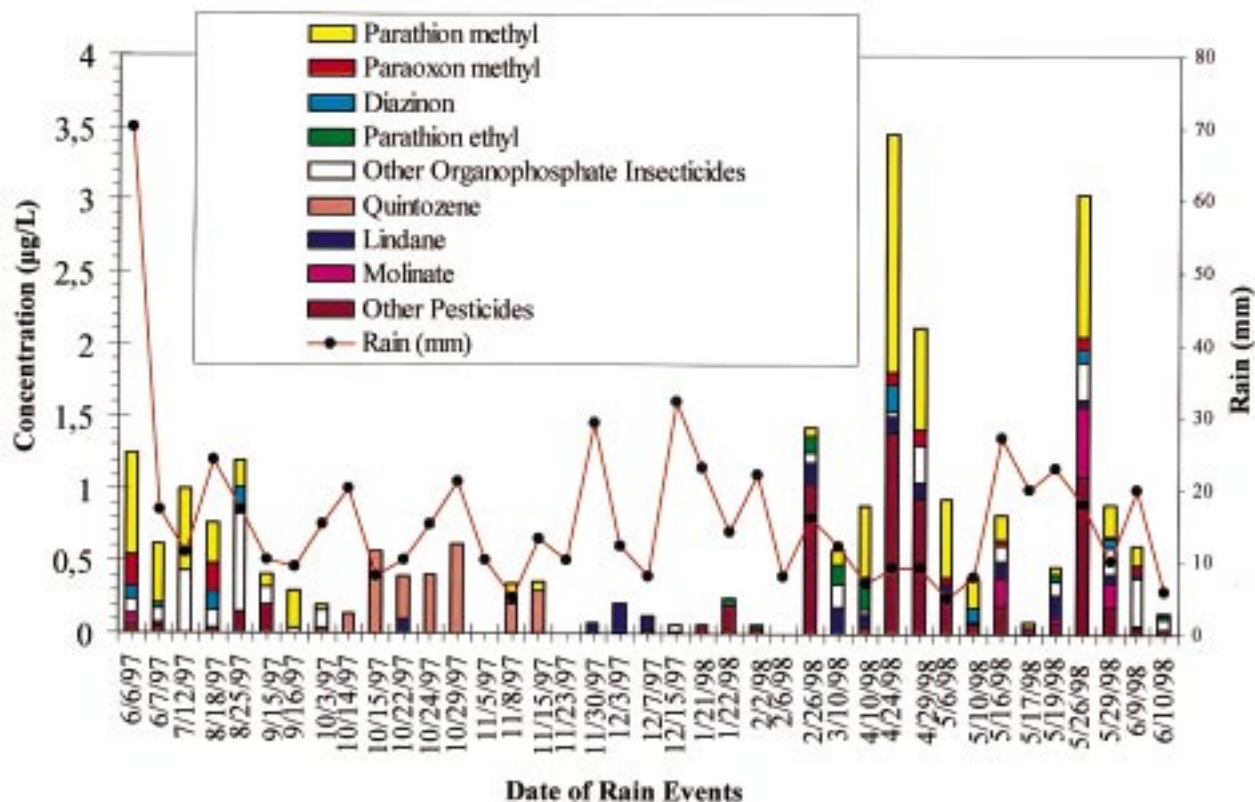


FIGURE 2. Pesticide concentrations ( $\mu\text{g/L}$ ) and respective rainfall (mm) per rain event at sampling site 1.

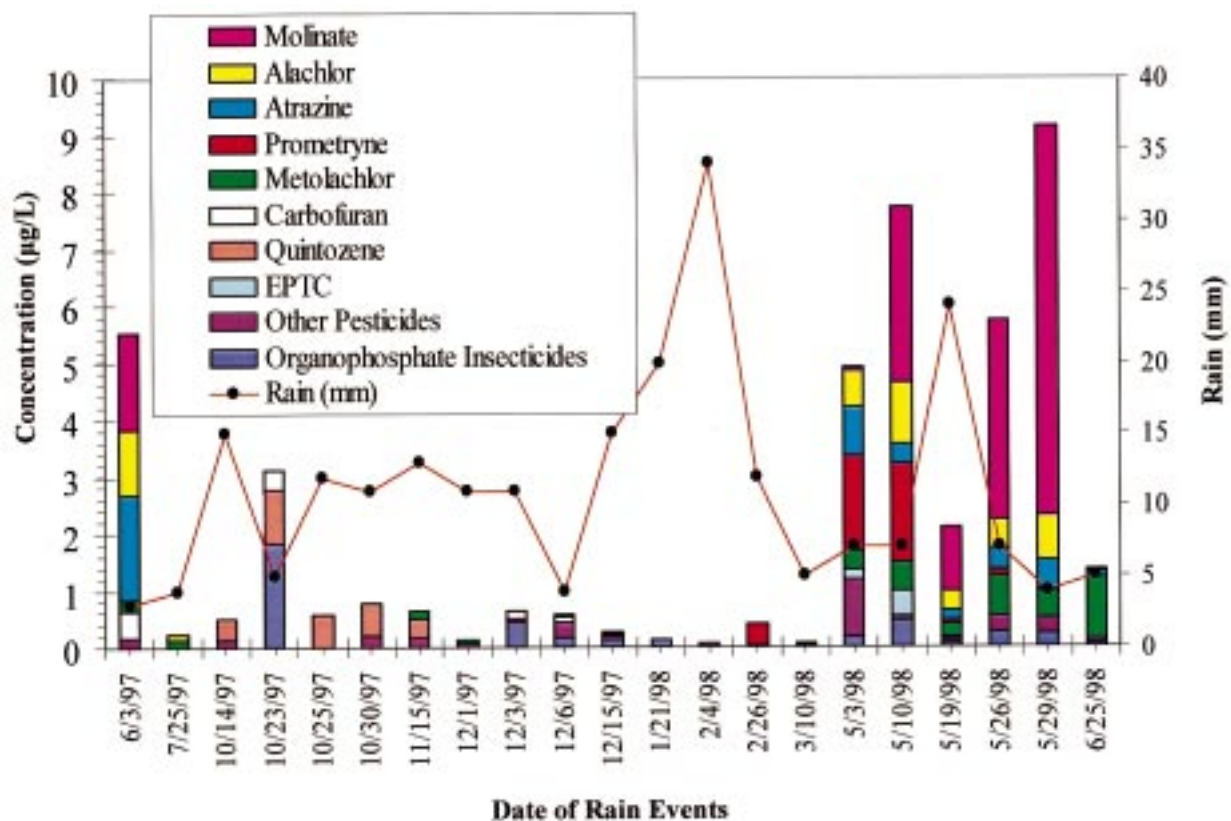


FIGURE 3. Pesticide concentrations ( $\mu\text{g/L}$ ) and respective rainfall (mm) per rain event at sampling site 5.

and except for alachlor, none of the other target pesticides were detected during the wintertime. Alachlor was also detected during winter months; however, the respective concentrations were relatively low. This seasonal distribution

in herbicide concentrations has been previously reported (6, 7, 9, 10, 12, 13).

At SS 1–3, insecticides were the predominant rainwater contaminants. Most insecticides also had peaks in concen-

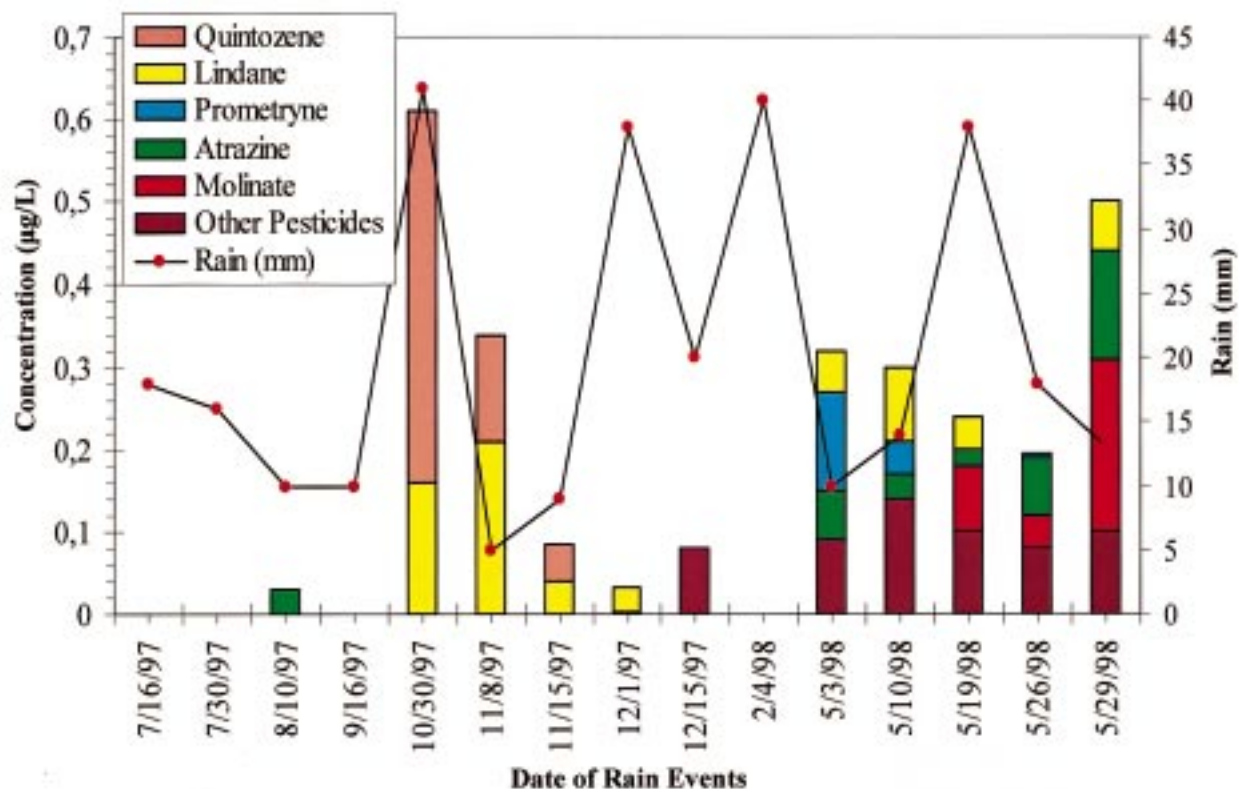


FIGURE 4. Pesticide concentrations ( $\mu\text{g/L}$ ) and respective rainfall (mm) per rain event at sampling site 7.

trations during the spring period (Figure 2); however, a few insecticides were also readily detected during other times of the year since these compounds are applied year-round. At SS 1 and 2, the concentration of parathion-methyl remained  $>0.1 \mu\text{g/L}$  from March to September. A seasonal appearance of quintozene was observed only during October and November (Figures 2–4), which corresponds with the period of its highest use as seed dressing for many annual crops.

The number of pesticide detections in samples collected during April–June was surprisingly high. Samples of this period contained an average of eight compounds each, while 50% of them had more than 10 detections. A maximum number of 20 compounds was detected in a single sample (SS 1).

Great differences in pesticide detections were seen among the eight sampling localities in relation to the different crops and pesticides used in each area. The organophosphate insecticides were found primarily and in higher concentrations at the western part of the basin (SS 1–3) where they are used extensively in orchards of this area. Concentrations of parathion-methyl reached  $1.65 \mu\text{g/L}$ , and concentrations of paraoxon-methyl reached  $0.22 \mu\text{g/L}$  at SS 1. In a similar pattern, herbicides used in annual crop production were detected in highest quantities in the middle part of the basin (SS 4–6), which is the area of their greatest use. Concentrations of molinate reached  $6.8 \mu\text{g/L}$ , atrazine reached  $1.8 \mu\text{g/L}$ , and concentrations of metolachlor amounted to  $1 \mu\text{g/L}$  at SS 5; while at SS 4, prometryne, alachlor, and ethofumesate had concentrations of 2.9, 2.2, and  $0.38 \mu\text{g/L}$ , respectively. SS 7 yielded the lowest proportion of detections, and this is in agreement with the lowest load of pesticides applied in this area. In Table 2, the maximum annual deposition rate ( $\mu\text{g/m}^2$ ) of each pesticide at the SS where its maximum concentration was recorded is also given. Among the pesticides reported in Table 2, parathion-methyl yielded the highest annual deposition rate ( $132 \mu\text{g/m}^2$ ) followed by the fungicide quintozene ( $120.5 \mu\text{g/m}^2$ ) and the herbicide molinate ( $110.5 \mu\text{g/m}^2$ ).

TABLE 3. Time Periods of Rainwater Collection, Annual Rainfall (mm), and Pesticide Deposition Rates ( $\mu\text{g/m}^2$ ) at the Different Sampling Sites of the Axios River Basin

sampling site	time periods of rainwater collections	annual rainfall (mm)	annual pesticide deposition ( $\mu\text{g/m}^2$ )
1	June 1997–June 1998	623	395
2	June 1997–June 1998	605	377
3	June 1997–June 1998	614	337
4	Sept 1997–June 1998	168	191
5	June 1997–June 1998	229	338
6	Oct 1997–June 1998	313	172
7	June 1997–June 1998	300	51
8	June 1997–June 1998	283	141

Although atrazine, alachlor, metolachlor, prometryne, and quintozene are not applied at the western part of the basin, they were detected at sites 1 and 2 and sometimes in high concentrations. Similarly molinate and ethofumesate were also found at all sampling localities, although they are used exclusively in the middle part of the basin. This strongly suggests that, in addition to vertical transport in the atmosphere, dispersion of pesticides in the atmosphere occurred, and this is in agreement with the variable winds of this area. Certainly, the possibility that sources of these pesticides are also located outside the boundaries of the Axios River Basin is not excluded; however, the present data are insufficient to support such hypothesis.

As has been reported by others (7, 9, 10) and also observed in this study, pesticides exhibited higher concentrations when the amount of rainfall was small. Nearly all the investigated pesticides reached their maximum concentrations at rain events that were less than 10 mm. However, the highest annual pesticide deposition rates were estimated for areas receiving the highest annual rainfall (Table 3). These estimated annual deposition rates ranged from 51 to  $395 \mu\text{g/m}^2$ . The SS 1–3 and 5 had the highest annual pesticide

deposition rates. Except for SS 5, SS 1–3 received also the highest rainfall levels.

Pesticide metabolites were occasionally detected in rainfall. Deethylatrazine (DEA) was found in 20% of the samples that contained atrazine. DEA was only detected during April–June, and the respective concentration levels ranged from 0.01 to 0.19  $\mu\text{g/L}$ . In contrast, deisopropylatrazine was found only in one sample at 0.1  $\mu\text{g/L}$ . The median ratio of DEA to atrazine concentration was 0.4. These results agree with those reported by others (4, 6, 10) and suggest that DEA is a very common metabolite of atrazine in rainwater, while deisopropylatrazine has little importance. DEA in rainwater may result from the photocatalyzed degradation of atrazine in the atmosphere (10).

Paraoxon-methyl and malaoxon were detected in 19 and 37%, respectively, of the samples that contained the parents compounds. The median ratio of oxon to thion forms of parathion-methyl was 0.25, while for malathion the respective ratio was 1.11. Oxidation of phosphorothioate insecticides has been reported (3, 16–18) as an important reaction occurring in the atmosphere, leading to the formation of the more toxic and reactive oxons. In a few samples, phorate sulfone and phorate sulfoxide were also detected without the presence of phorate itself. Because no published data were found on this matter, it is uncertain if these compounds were the result of the atmospheric oxidation of phorate or if they originated, through volatilization, from soil surfaces.

The most frequently found pesticides and in higher concentrations (alachlor, lindane, parathion-methyl, atrazine, quintozone, metolachlor, prometryne, and molinate) are among those, except quintozone, used most widely in the basin of the Axios River. However, all these pesticides, except molinate, are relatively nonvolatile compounds with vapor pressure  $<7.0$  mPa (20  $^{\circ}\text{C}$ ). Also these compounds, except quintozone, have Henry's law constants ( $H$ )  $<0.20$  Pa  $\text{m}^3/\text{mol}$  (19). The  $H$  value for quintozone is 4.43 Pa  $\text{m}^3/\text{mol}$  (calculated from literature available data on vapor pressure and water solubility of quintozone). Thus, the presence of most of these pesticides in rainwater of the Axios River Basin was not expected on the basis of these physicochemical properties. It is apparent that the environmental stability of these compounds in both the wet and gas phases is the determining factor for their occurrence in rainwater since pesticides used as widely as those mentioned above and having more favorable vapor pressure and  $H$  values (i.e., trifluralin and pendimethalin) were found less frequently and at much lower concentrations due to their well-known photoinstability.

## Conclusions

A variety of pesticides, including parent compounds and major conversion products, are present in rainwater of both agricultural and residential areas of the Axios River Basin. Presently, the environmental impact of pesticides found in

rainwater is difficult to assess. It was reported in the Introduction that this investigation was triggered by previous findings that pesticide residues are present in most aquatic systems of the basin, even in remote areas of the highlands (unpublished data); in fact, the latter aquatic systems are used as drinking water sources. Therefore, what is most certain at the present is that rainwater of this area, due to the presence of pesticides at concentrations higher than 0.1  $\mu\text{g/L}$  in most rain events of the year, does not comply with the drinking water quality standards of the European Union (EU) Directive 83/98 (20), which is of great concern.

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