

Radioactive Pollution in Freshwater Ecosystems from Macedonia, Greece

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Received: 29 March 1995/Revised: 29 June 1995

Abstract. The levels of radioactive contamination by artificial radiocesium (Cs-137, Cs-134) as well as the concentrations of natural radioisotopes (K-40, Ra-226, Ra-228, Th-228) in sediments and aquatic plants were evaluated in selected aquatic biotopes in Macedonia, Greece. Radionuclide analysis was performed in Marinelli beakers by a gamma spectrometry system with an Hp-Ge detector of 20% relative efficiency. Cs-137 concentration activities are higher in sediment than in plant material; in general, roots showed greater Cs-137 concentration than leaves, while stems showed the lowest concentration. Cs-137 is significantly higher at 95% confidence level in the river Axios than in the rivers Strymon and Aliakmon. Leaves, especially young ones, show K-40 activities higher than roots or stems. From the studied watersystems, the river Axios and Prespa Lake are more contaminated from Cs-137 and Cs-134. K-40 activity is significantly higher at the 95% confidence level in the river Axios than in the rivers Aliakmon and Pinios. The K-40 activity is also high, especially in Prespa Lake. High activities of Ra-226, Ra-228, Th-228, and K-40 were found in Polyphytos Lake, presumably as a result of the operation of the coal power plants in this area.

Traces of radioactivity are normally found in all types of water. The concentration and composition of these radioactive constituents vary from place to place, depending essentially on the radiochemical composition of the soil and rock through which the raw water may have passed (Chouroulinkov and Jaylet 1989). The occurrence of radioactive elements in natural phosphate rocks was established by Strutt (1906). Uranium, thorium, and potassium as "parent" radionuclides are the most common ones in nature. Indium-115 is also a parent radionuclide, but it is rare. The half-lives involved have permitted these elements to survive through the geologic ages.

The development of atomic energy plants and the use of radioisotopes in biomedical research considerably increased the production of radioactive chemicals. The greatest amount of radioactive wastes results from fuel processing plants. A small plant of this type releases annually between 500 and 1500 m³ of water containing tritium, strontium-90, cesium-137, ruthenium-106, cerium-144, and iodine-131. According to prediction values (Ramade 1981), in Western Europe by the year

2000, the amount of wastes would have increased so that their activity would be 30-fold higher compared to that of 1980. The nuclear industry is not always in a position to dispose of radioactive wastes, since it can neither eliminate nor transform radiation. One can only protect man and the environment via a decrease in the density of the flow of radiation, using dilution or screens.

Cesium isotopes produced by nuclear weapon tests (Cs-137), especially during the 1960s, and the Chernobyl accident of 1986 (Cs-134, Cs-137) are firmly bound to clay mineral particles in river or lake sediments (Robbins *et al.* 1992; Kaminski 1991) as a result of the selective irreversible binding sites available at micaceous clay minerals like illite (Comans *et al.* 1991).

Cesium is less strongly bound by organic soils (de Preter 1990). From the sediment active zone, cesium is lost to the hypolimnion by recycling or remobilization. This is attributed to ion-exchange displacement of cesium from sediment by cations such as ammonium, iron, or magnesium under anaerobic conditions (Evans *et al.* 1983; Comans *et al.* 1989). Furthermore, a seasonal cycling both of dissolved cesium radionuclides and ammonium ions was observed and was attributed to the redissolution of cesium radionuclides unspecifically bound in the sediment by ion exchange with ammonium ions (Lindner *et al.* 1993; Wunderer *et al.* 1993; Alberts *et al.* 1979; Evans *et al.* 1983; Comans *et al.* 1989; Davison *et al.* 1993).

This paper presents the results of radiocesium introduced mainly from the Chernobyl accident as well as natural radionuclides in sediment and aquatic plants collected from lakes and rivers of Macedonia, Greece.

Studied Area—Sampling Stations

Rivers

River Strymon: The river Strymon originates from Bulgaria and flows into the Strymonikos gulf. Along the Greek part of the river, alluvial deposits in valleys, amphibolites, and gneisses are present (Bornovas and Rondogianni-Tsiambau 1983).

River Axios: The river Axios originates from former Yugoslavia and after flowing 220 km within this area and 80 km within Greece, it flows into the Thermaikos gulf. The main

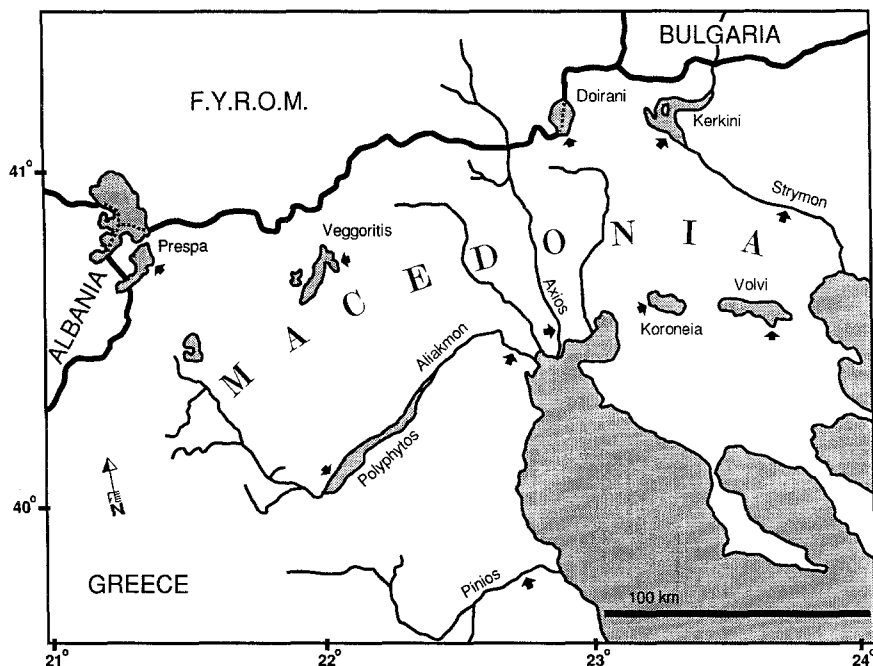


Fig. 1. Map of Macedonia (Northern Greece) showing the studied rivers and lakes. Arrows point to sampling stations

geological formations that the river runs through are igneous rocks, alluvial deposits, and ophiolites in general.

River Aliakmon: The river Aliakmon is the longest river in Greece (350 km). The river originates from W. Macedonia and flows into the Thermaikos gulf. The basement of the river consists mainly of alluvial deposits, a few ophiolites, gneisses, shists, and amphibolites.

River Pinios: The Pinios river, together with its tributaries, is the unique receiver of Thessalia plain, a 9.747 km² hydrological basin. The river flows into the mouth of Thermaikos gulf. Alluvial deposits and gneisses are present in the river's basement.

Lakes

Volvi: Lake Volvi, a warm monomictic lake, has a surface area of 68.6 km² and a mean depth of 13.5 m. The basement of the lake consists of alluvial deposits, amphibolites, gneisses, igneous rocks, and shists with marble intercalations (Bornovas and Rondogianni-Tsiambau 1983).

Koroneia: The lake has a surface area of 42 km² and a maximum depth of 8.5 m. The basement of the lake is analogous to Lake Volvi.

Polyphytos: It is a reservoir created on the Aliakmon river in 1975 for cooling requirements of thermal electricity generation plants and hydroelectricity generation. The reservoir's surface area is 70 km², and the maximum depth is 91 m. The basement of the reservoir consists of lacustrine, terrestrial deposits, and igneous rocks.

Veggortitis: The lake has a surface area of 59 km² and a maximum depth of 46 m. The lake is surrounded by alluvial and lacustrine deposits.

Prespa: The lake has a surface area of 266 km² and a maximum depth of 55 m. The basement of the lake consists of crystalline limestones, alluvial deposits, carbonate, and plutonic and igneous rocks.

Doirani: It is a shallow lake with a maximum depth of 8 m and a surface area of 52 km². The surrounding formation consists of alluvial deposits, volcano-sedimentary rocks, and quartzites.

Kerkini: It is a reservoir that was made after constructing a dam and impounding the Strymon river in 1932 for flood control, irrigation, and fisheries. The reservoir's surface ranges to 76 km² and the maximum depth is 6.5 m. The basement of the reservoir consists mainly of alluvial valley deposits.

Analysis Procedures and Instrumentation

The sampling sites are shown in Figure 1. Samples were collected during the summer and autumn of 1993. The plant materials were initially cleaned with fresh water and then with distilled water, and air-dried at room temperature to constant weight. Drying of the sampled material is important as it protects the plant material from microbial decomposition and ensures a constant reference weight in contrast to fresh weight, which is difficult to quantify (Markert 1993). For the most uniform possible distribution of elements in the sample, the material was pulverized in a Mulinex mill. After homogenizing, the samples were stored in polyethylene containers at room temperature in a desiccator. Sediment samples were collected from a 10 cm water depth using a shovel. Three replicates from each location were taken. After air-drying at room temperature, the sediment samples were passed through a 2 mm sieve. 200–300 g of each sample were measured in a standard geometric (Marinelli) beaker using a gamma spectrometer with a 4000 channel analyser and an Hp-Ge detector of 20% relative efficiency. The counting time for all samples was 20,000 s. The gamma spectrometer was calibrated using standard reference sources, the overall efficiency being about 12% for the Marinelli system. For statistical analysis of the data, Pearson's correlation coefficient and paired T-test were used. The measurements were carried out at the Environmental Radioactivity Laboratory of the N.C.S.R. "Democritus," Athens.

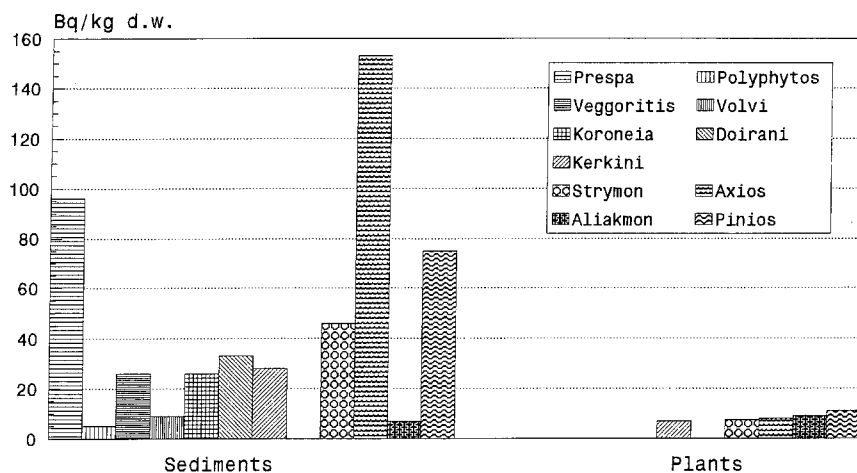


Fig. 2. Cs-137 concentrations in lake and river sediments and plants. Mean standard error (MSE) ± 2.0 Bq/kg for sediment and ± 2.7 Bq/kg for plant samples

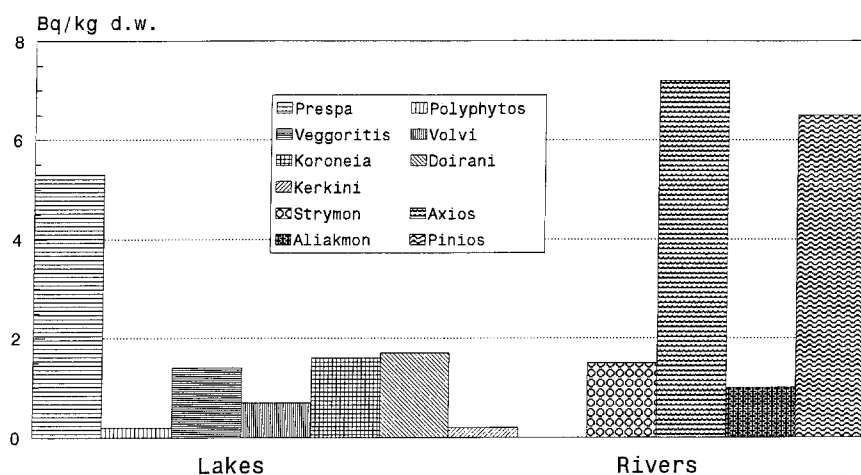


Fig. 3. Cs-134 concentrations in lake and river sediments. MSE ± 0.9 Bq/kg

Results

The cesium-137 and -134 content in lake and river sediments is shown in Figures 2 and 3, respectively. The highest concentration of radionuclides (152.7 Bq/kg for Cs-137 and 7.2 Bq/kg for Cs-134) occurred in the sediment of the river Axios. Among the studied lakes, Prespa seems to be most contaminated, both with Cs-137 (96.2 Bq/kg) and Cs-134 (5.29 Bq/kg). The Cs-137 content in aquatic plants is also shown for comparison in Figure 2. The detailed values of Cs-137 for each collected plant species are illustrated in Table 1. Results of paired t-tests for Cs-137 and K-40 are presented in Table 2.

The Ra-228 and Ra-226 concentration in sediments is shown in Figure 4 and Figure 5, respectively. Among the rivers, Axios shows the highest concentrations in both radionuclides (Ra-228 62.0 Bq/kg, Ra-226 43.3 Bq/kg), followed by Pinios (Ra-228 57.8 Bq/kg, Ra-226 34.3 Bq/kg) and Aliakmon (Ra-228 54.1 Bq/kg, Ra-226 30.7 Bq/kg). Among the lakes, the highest concentrations were observed in Polyphytos Lake (Ra-228 72.6 Bq/kg, Ra-226 49.6 Bq/kg).

K-40 content, both in sediment and aquatic plants (mean value), is shown in Figure 6. Plants show higher K-40 concentrations than sediments, the highest values of which are observed in Prespa (1942.6 Bq/kg) and Volvi (1298.6 Bq/kg) lakes. The detailed values for each plant species are illustrated in Table 1.

The Th-228 content in lake and river sediments is shown in Figure 7. The two lakes Polyphytos and Prespa show the highest Th-228 concentrations (44.1 Bq/kg and 39.0 Bq/kg, respectively).

Discussion

Cs

It is obvious from Figure 2 that Cs-137 activities are higher in sediment than in plant material. Pearson's correlation coefficient between 11 plant samples and sediments showed no significant correlation. The highest correlation coefficient value for Cs-137 was 0.897 between the sediment and *Ceratophyllum demersum* (leaf). Sediments of high clay content can effectively immobilize cesium by chemical binding. In this case, the sediment acts like a sink for Cs and in time, very little of the nuclide is available for biological incorporation. On the other hand, the activity once taken up by plants will be reduced not only by physical decay of the respective radionuclides, but also by environmental removal processes (Keppel 1966; Ertel *et al.* 1989). This field loss is influenced by various factors (Miller and Hoffman 1983). In general, roots showed higher Cs-137 concentration than leaves, while stems showed the lowest con-

Table 1. K-40 and Cs-137 activity in various waterplant species from Macedonia, Greece

	K-40	Cs-137
Kerkini Lake		
<i>Nymphaea alba</i> (rhizom)	320.5 ± 22	4.0 ± 2.1
<i>Nymphaea alba</i> (leaf)	536.9 ± 65	b.d.l.
<i>Paspalum pasalodes</i> (leaf)	574.9 ± 57	4.2 ± 1.3
<i>Paspalum pasalodes</i> (root)	363.0 ± 29	16.0 ± 7.0
<i>Phragmites australis</i> (root)	31.7 ± 18	19.1 ± 6.1
<i>Phragmites australis</i> (young stem)	1310.0 ± 70	b.d.l.
<i>Phragmites australis</i> (rhizom)	824.5 ± 50	b.d.l.
<i>Phragmites australis</i> (old leaf)	452.0 ± 34	b.d.l.
<i>Phragmites australis</i> (young leaf)	1242.0 ± 64	b.d.l.
<i>Potamogeton gramineus</i> (leaf)	536.5 ± 60	6.5 ± 4.9
<i>Potamogeton pectinatus</i> (leaf)	615.5 ± 58	7.0 ± 5.7
<i>Ranunculus sardous</i> (leaf)	497.8 ± 44	7.7 ± 3.6
<i>Rumex crispus</i> (root)	543.3 ± 29	3.4 ± 1.3
<i>Rumex crispus</i> (leaf)	1565.7 ± 32	2.5 ± 1.6
<i>Trapa natans</i> (root)	66.2 ± 32	7.2 ± 5.0
<i>Trapa natans</i> (stem)	852.1 ± 48	b.d.l.
<i>Trapa natans</i> (leaf)	960.6 ± 51	6.5 ± 4.8
Strymon River		
<i>Ceratophyllum demersum</i> (leaf)	1192.9 ± 50	7.0 ± 4.9
<i>Ceratophyllum demersum</i> (stem)	958.8 ± 76	b.d.l.
<i>Cladophora</i> sp.	1011.0 ± 64	13.6 ± 5.0
<i>Cyperus longus</i> (root)	266.4 ± 36	24.5 ± 3.1
<i>Cyperus longus</i> (stem)	390.0 ± 20	2.2 ± 1.3
<i>Cyperus longus</i> (leaf)	509.9 ± 45	b.d.l.
<i>Hydrocharis morsus</i> (root)	738.1 ± 63	10.0 ± 5.3
<i>Hydrocharis morsus</i> (leaf)	831.9 ± 45	b.d.l.
<i>Lemna polyrrhyza</i>	1241.2 ± 70	8.1 ± 6.9
<i>Myriophyllum</i> sp. (root)	354.7 ± 20	2.4 ± 1.0
<i>Myriophyllum</i> sp. (leaf)	657.4 ± 63	b.d.l.
<i>Paspalum pasalodes</i> (root)	313.5 ± 66	16.2 ± 7.8
<i>Paspalum pasalodes</i> (stem)	302.5 ± 50	2.2 ± 1.3
<i>Paspalum pasalodes</i> (leaf)	635.3 ± 76	b.d.l.
<i>Potamogeton crispus</i> (root)	284.3 ± 55	12.8 ± 5.8
<i>Potamogeton crispus</i> (leaf)	1107.0 ± 72	b.d.l.
<i>Potamogeton nodosus</i> (stem)	938.2 ± 72	b.d.l.
<i>Potamogeton nodosus</i> (leaf)	1521.6 ± 99	b.d.l.
<i>Spyrogyra</i> sp.	277.8 ± 33	6.5 ± 3.7
<i>Trapa natans</i> (root)	459.7 ± 45	7.0 ± 3.6
<i>Trapa natans</i> (leaf)	980.8 ± 78	3.5 ± 2.0
Axios River		
<i>Ceratophyllum demersum</i> (stem)	835.3 ± 67	8.0 ± 4.0
<i>Ceratophyllum demersum</i> (leaf)	1452.0 ± 79	15.2 ± 5.5
<i>Cladophora</i> sp.	1051.2 ± 46	8.4 ± 3.9
<i>Cyperus longus</i> (root)	366.4 ± 36	28.5 ± 5.1
<i>Cyperus longus</i> (stem)	360.0 ± 20	2.2 ± 1.3
<i>Cyperus longus</i> (leaf)	709.9 ± 45	4.3 ± 1.1
<i>Myriophyllum</i> sp. (leaf)	658.2 ± 41	5.7 ± 3.5
<i>Paspalum pasalodes</i> (root)	452.2 ± 35	20.5 ± 6.0
<i>Paspalum pasalodes</i> (stem)	325.9 ± 30	b.d.l.
<i>Paspalum pasalodes</i> (leaf)	675.8 ± 43	5.1 ± 2.5
<i>Potamogeton nodosus</i> (stem)	935.0 ± 68	4.0 ± 2.3
<i>Potamogeton nodosus</i> (leaf)	1130.9 ± 76	8.2 ± 2.8
<i>Potamogeton pectinatus</i> (leaf)	1020.0 ± 68	12.5 ± 4.5
Aliakmon River		
<i>Ceratophyllum demersum</i> (leaf)	1356.7 ± 65	8.7 ± 5.8
<i>Ceratophyllum</i> sp. (leaf)	905.7 ± 61	12.3 ± 5.5
<i>Cladophora</i> sp.	882.6 ± 70	b.d.l.
<i>Cyperus longus</i> (root)	300.5 ± 45	8.1 ± 4.1
<i>Cyperus longus</i> (stem)	432.0 ± 65	b.d.l.
<i>Cyperus longus</i> (leaf)	623.5 ± 61	2.2 ± 1.3
<i>Hydrodictyon</i> sp.	652.0 ± 81	7.2 ± 4.5
<i>Myriophyllum</i> sp. (leaf)	550.2 ± 40	b.d.l.
<i>Paspalum pasalodes</i> (root)	392.7 ± 28	8.2 ± 3.1
<i>Paspalum pasalodes</i> (stem)	298.8 ± 30	b.d.l.
<i>Paspalum pasalodes</i> (leaf)	565.0 ± 48	5.2 ± 4.2
<i>Potamogeton crispus</i> (leaf)	666.8 ± 28	5.1 ± 3.0
<i>Potamogeton nodosus</i> (stem)	835.3 ± 63	b.d.l.
<i>Potamogeton nodosus</i> (leaf)	690.2 ± 56	9.9 ± 5.3
<i>Potamogeton pectinatus</i> (leaf)	727.2 ± 31	6.7 ± 3.1
Pinios River		
<i>Ceratophyllum demersum</i> (leaf)	1025.0 ± 52	10.5 ± 5.1
<i>Ceratophyllum</i> sp. (leaf)	484.6 ± 43	4.4 ± 4.2
<i>Cladophora</i> sp.	983.5 ± 62	7.2 ± 3.1
<i>Cyperus longus</i> (root)	285.1 ± 49	12.7 ± 4.5
<i>Cyperus longus</i> (stem)	392.7 ± 53	b.d.l.
<i>Cyperus longus</i> (leaf)	532.4 ± 68	5.3 ± 2.1
<i>Myriophyllum</i> sp.	508.7 ± 58	25.5 ± 3.9
<i>Paspalum pasalodes</i> (root)	402.5 ± 45	12.3 ± 4.5
<i>Paspalum pasalodes</i> (stem)	310.2 ± 29	b.d.l.
<i>Paspalum pasalodes</i> (leaf)	623.1 ± 42	b.d.l.
<i>Potamogeton nodosus</i> (stem)	825.2 ± 50	b.d.l.
<i>Potamogeton nodosus</i> (leaf)	576.7 ± 75	b.d.l.

b.d.l.: Below detection limits

Table 2. Result of significance of difference at 95% confidence level for K-40 and Cs-137 from four rivers, according to paired t-test

	K-40			Cs-137		
	Axios	Aliakmon	Pinios	Axios	Aliakmon	Pinios
Strymon	0.730	0.908	1.341	2.635	0.902	0.249
Axios	(-)	2.953	2.837	(+)	2.790	0.980
Aliakmon		(+)	1.223		(+)	1.058
Pinios			(-)			(-)

(-) significantly not different
(+) significantly different

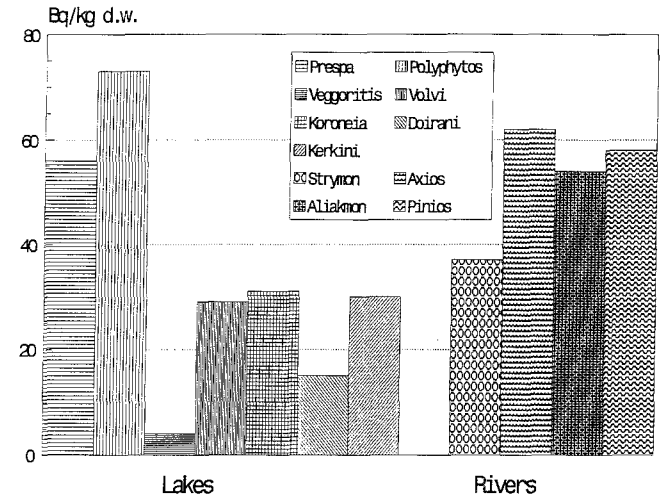


Fig. 4. Ra-228 concentrations in lake and river sediments. MSE ± 5.3 Bq/kg

centration, with a single exception of *Phragmites australis*. Mineeva *et al.* (1990), using *Lamium maculatum* as an example, also reported that in higher plants, the greatest concentration occurs in the root systems, the coefficient in the roots being 1.5 times higher than the one in the aboveground parts. Paired t-test of common plant samples collected from rivers showed that Cs-137 concentrations are significantly higher in the river Axios than in the rivers Strymon and Aliakmon (Table 2).

The high cesium concentrations in the sediment of the Axios River (Cs-137 152.7 Bq/kg, Cs-134 7.2 Bq/kg), which originates from the former Yugoslavia, as well as those of Prespa Lake (Cs-137 96.2, Cs-134 5.29) and the Pinios River (Cs-137 75.0, Cs-134 6.5), could obviously be attributed to the Chernobyl reactor accident in April 1986. In the Greek part of the catchment area of the Axios River, the cesium-137 concentration in the air was 1.95 Bq m⁻³, whereas in the catchment area of Pinios, the concentration in the air was 1.80 Bq m⁻³ (Kritidis *et al.* 1986; Papastefanou *et al.* 1988a). Thermaikos gulf, into which both the Axios and Pinios rivers flow, showed the highest Cs-137 deposition (20–40 kBq m⁻²) than all other areas of the Aegean Sea. Deposition of Cs-137 in Strymonikos gulf, where the Strymon River flows, was 10–20 kBq m⁻² (Kritidis and Florou 1990).

Ra

The major reservoirs of radium in many specific areas of the earth's surface are geological deposits, especially natural phosphates of sedimentary origin (Altschuler *et al.* 1958; Nathan

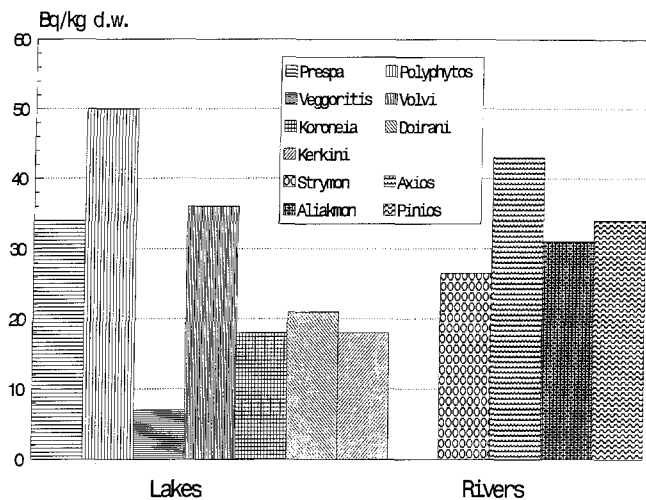


Fig. 5. Ra-226 concentrations in lake and river sediments. MSE \pm 2.0 Bq/kg

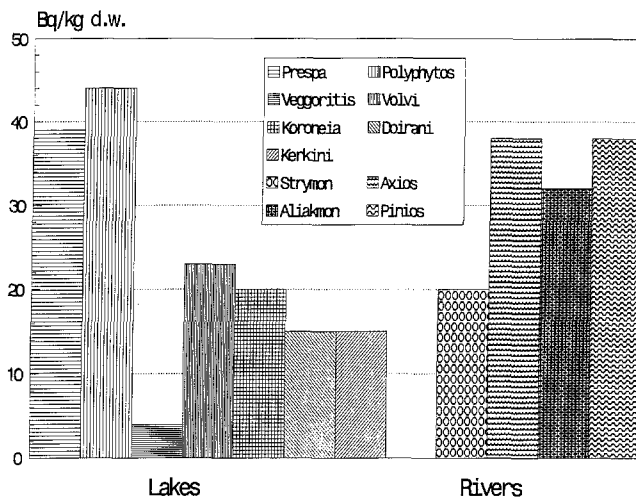


Fig. 7. Th-228 concentrations in lake and river sediments. MSE \pm 2.0 Bq/kg

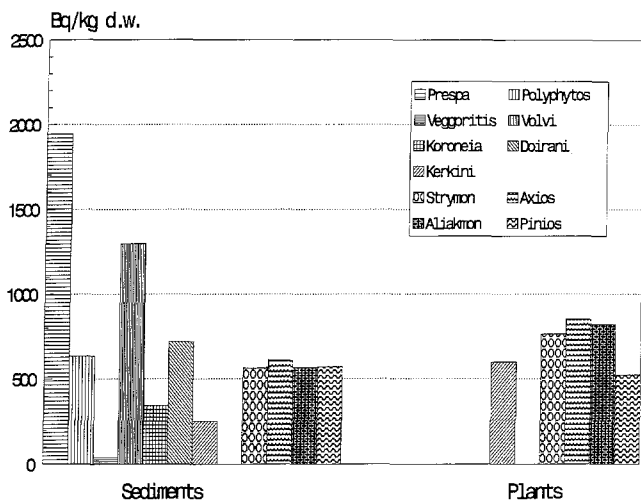


Fig. 6. K-40 concentrations in lake and river sediments and plants. MSE \pm 17.0 Bq/kg and \pm 51.4 Bq/kg, respectively

and Shiloni 1976; Slanski 1978; Altschuler 1980). Radium (Ra-226), a major uranium decay product, is found in natural phosphates which are processed for chemical fertilizers (Bunus 1994). Thus, radium is widely spread on agricultural lands and eventually ends up in lake or river sediments after leaching by rain water.

The highest Ra-228 and Ra-226 concentrations in the sediment were observed in Polyphytos artificial lake, the waters of which are used in the procedure of thermal electricity generation. High concentrations were also shown in Prespa Lake. Both lakes are situated in the vicinity of coal-fired plants where the combustion of lignites results in the release of natural radioactivity to the environment (Kirchner *et al.* 1974; Barber and Giorgio 1977; Papastefanou and Charalambous 1979; Papastefanou *et al.* 1988b). It is notable that when concentrations of Ra-226, Ra-228, and Th-228 in the sediment of the two lakes are elevated, K-40 values remain low, resulting in an inverse relationship. The same relationship among the above radionu-

clides (except Ra-228) was referred in the lignite concentrations from this area (Manolopoulou and Papastefanou 1992).

K

The greatest amount of natural radioactivity is due to K-40. There is no isotopic difference in chemical behavior between K-40 and stable K, and the isotopic abundance is preserved in biotic and abiotic materials. A slight superiority in plant material (collected from the rivers and Lake Kerkini) could be stressed compared to sediments. Pearson's correlation coefficient between sediments and 11 common plant samples collected from the rivers showed a correlation coefficient not significantly different than 0 in all cases except *Cyperus longus* (root) and *Paspalum pasalodes* (stem). The correlation coefficients between sediment and *Cyperus longus* (root) or *Paspalum pasalodes* (stem) are significantly different than 0 at the 95% confidence level.

Significant differences in metal concentrations were found among different plant organs from the same plant. Leaves, especially young ones, show higher K-40 activities than roots or stems. Heinrich *et al.* (1989) reported that the amount of K-40 decreases as leaves become mature and usually drawn off in autumn.

Paired T-test showed that K-40 in plant samples of the Axios River differed significantly at the 95% confidence level than in the rivers Aliakmon and Pinios, which indicated that K-40 was significantly higher in the Axios River than in the rivers Aliakmon and Pinios. The igneous rocks, over which the Axios River runs, contain high concentrations of potassium and its isotopes (Hughes 1982), and obviously this is the main reason of high K-40 activities both in the sediment (607.4 Bq/kg) and in aquatic plants (767.2 Bq/kg). The highest K-40 values are recorded in the sediment of Prespa (1942.6 Bq/kg) and Volvi (1298.6 Bq/kg) lakes, which also contain igneous rocks in their basement.

Th

Thorium, similar to the other actinides, is transported in ecosystems mainly by physical and sometimes chemical processes.

They tend to accumulate in soils and sediments which ultimately serve as storage reservoirs. Subsequent movement is largely associated with geological processes such as erosion and leaching (Whicker and Schultz 1982). An analogous distribution of Th-228 to Ra-226 or Ra-228 in the sediments is obvious in our results. Polyphytos Lake shows the highest concentrations followed by Prespa. Literature data have shown that thorium is a prevalent element in igneous apatites (Altschuler *et al.* 1958; Habashi 1962). These geological formations are present in both lakes mentioned above.

Apart from the geology of the area where both lakes are situated, the operation of the coal power plants is again an additional reason for the release and redistribution of natural radionuclides in the surrounding area (Georgakopoulos *et al.* 1992; Kassoli-Fourmaraki *et al.* 1993; Georgakopoulos *et al.* 1994).

The influence of the power stations on Lake Polyphytos is consistent with the wind-transported ash particles. On the other hand, Veggorititis Lake, situated on the opposite side of the power plants, shows much lower concentrations in the above radionuclides (Figure 1).

Among the rivers, Axios shows again the highest concentrations of Th-228 as in all other natural radionuclides, followed by Pinios and Aliakmon. It is inferred that geological deposits serve as the major reservoirs of natural radionuclides, and its geographical distribution is ubiquitous yet heterogenous with well-known "hot-spots" in many specific areas. Thus, the lake sediments are more representative for the radiological status of a specific area than the river sediments which are composed of various particles (small grain sizes) transported away by the erosive water flow.

Acknowledgments. The author thanks the E.R.P. of the N.C.S.R. "Democritus" for the measurements of the samples and especially to Dr. H. Florou for her valuable assistance. This work is a part of the INTERREG program (Code Nr. C91 1549/3/25-7-91) supported by the European Community.

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