Bull. Environ. Contam. Toxicol. (1990) 44:535–541

Bulletin of Environmental Contamination and Toxicology

Distribution and Transport Kinetics of Radionuclides ^{99}Mo and ^{131}I in a Simulated Aquatic Ecosystem

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Radioactive liquid wastes from nuclear power stations increase the activity not only of water in the recipient but also of other components such as the sediment and aquatic and shore plants and animals. On average, the majority of the total radioactivity brought to the aquatic system is absorbed by the sediment, the remaining fraction is distributed between water and biomass. The total activity distribution among the components of the hydrosphere depends not only on the radionuclide concerned and its physico-chemical state but also on the physico-chemical properties of water and sediment, kind and amount of biomass, stagnancy or motion of water, season of the year, etc.

For us to be able to assess the influence of the nuclear power station at Temelin in South Bohemia (which is under construction) on the nearby hydrosphere, we concentrated first on the experimental investigation of the distribution and transport kinetics of some radionuclides in a simulated aquatic system.

MATERIALS AND METHODS

The experiment was conducted for approximately one month (October) in two water reservoirs accomodated in a thermostated greenhouse. Each reservoir has the surface area 2.4 square meters and contained approximately 850 liters of water. Its bottom was covered with about 100 kg of sediment. Both the water and sediment were taken from the Vltava river at the locality near the Temelin nuclear power station.
Tufts of the macrophytic water plant Ranunculus (Batrachium) aquatilis were suspended in the water; because of macrophyte absence at the locality Temelin

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or in its vicinity, this water plant was taken at the locality near České Budějovice. And finally, fish (tenches) were added : 22 specimens to the first reservoir, to which radioactive solution of ^{99}Mo and ^{131}I radionuclides was later added, and 6 specimens to the other, control reservoir, free from radioactivity. The water was aerated by means of several air bubblers. Water temperature in the two reservoirs was held at $17\pm1^{o}C$ for the whole time of the experiment, pH in the active reservoir was 7.1 ± 0.3.

Water used in the experiment was sampled periodically for chemical, bacteriological and biological assays. The most marked was the difference in the nitrate and nitrite concentrations at the beginning and at the end of the experiment (see Table 1 for data of the active reservoir).No appreciable changes were found in the bacteriological assays. The microorganism life in the water of the Vltava river at Temelin is poor, and although it increased slightly during the experiment owing to the presence of fish and to favourable light and heat conditions in the reservoir, bioaccumulation by living microorganisms was negligible.

The granulometric analysis of sediment is: 9% of gravel >2mm, 10% of coarse sand >200μm, 27% of fine sand 50-200μm, 49% of clay 2-50μm, 3% of clay <2μm and 0.5% of organic (combustible) matter. pH of the leach of the sediment in destilled water and in 0.1M KCl is 5.94 and 4.90,respectively.

Radioactive solutions of ^{99}Mo and ^{131}I were added to reservoir in the form of potassium iodide and ammonium molybdate, respectively.

Water, sediment, plants and fish were sampled from the active reservoir periodically, first in shorter and later in longer time intervals. Fractions of water were filtered successively through Filtrak No.389, S5 membrane and S8 membrane filters, which had been boiled in distilled water, dried to constant weight and weighed. Pore diameter of S5 and S8 is 0.6 and 0.2 μm respectively. After filtration of a defined amount of active water (100 or 200 mL), the filters were again dried to constant weight and weighed, and their radioactivity was measured.

Plants were cleaned by a 30 min treatment in an ultrasonic cleaner, spread over a filter paper, allowed to dry for 30 min and weighed. After activity measurement, the plant samples were dried to constant weight at $105^{o}C$ and weighed.

Table 1. Analysis of water from the reservoir

Analysis	Water		
	river at Temelin	experimental reservoir	
	September	October	November
pH	6.77	7.02	6.82
Alkalinity (mval/L)	0.86	1.48	0.38
Acidity (mval/L)	0.27	0.16	0.15
Conductivity (μS/cm)	255	343	308
Hardness (mval/L)			
total	1.75	2.14	1.75
(mg/L):			
Undissolved matter	11	19	184
Dissolved matter	219	311	332
Sulphate	59.5	72.5	80.0
Chloride	15.0	20.6	20.9
Phosphate	0.16	0.79	0.78
Nitrite	0.007	0.083	0.074
Nitrate	2.9	25	45
Silicic acid	12.3	14.7	14.6
Calcium	25	25	16
Magnesium	6.0	11.0	11.5
Sodium	11.6	16.8	16.0
Potassium	5.7	52.0	40.0
Iron	1.07	4.58	7.00
Manganese	0	0.02	0.05
Ammonia	0	0.50	0
Bioch.oxygen demand	-	10	18
in 1 mL:			
Cyanophyceae		2	0
Chlorophyceae		4	100
Diatomae		1	100
Ciliatae		2	300
Flagellatae		8	500

Fish were so handled that the fins, heads, skin, flesh, bones and viscera were separated and each of the organs was weighed and mineralized. One fish weighed about 100 g in average.

All samples were subjected to gamma spectrometric analysis in 250 mL polyethylene bottles; filters were also measured, using a different geometry. Samples of the above kind were also taken twice from the control reservoir. No measurable activity was found in them, so that no background correction of the activity in the radioactive reservoir had to be made.

RESULTS AND DISCUSSION

Experimental values of activity concentration of unfiltered and filtered water ($Bq.L^{-1}$) in dependence on time were fitted by the function

$$A_w(t) = A_1*\exp(-A_2*t) + A_3*\exp(-A_4*t)$$

using the least squares method. For both ^{99}Mo and ^{131}I, the first halflife (i.e., the $\ln(2)/A_2$ value) was found about a quarter lower than and the second (i.e., the $\ln(2)/A_4$ value), approximately the same as the physical decay constant of the radionuclide. The regression curves for unfiltered water are shown in Fig. 1. For filtered water the kinetics is virtually the same (i.e. the A_2 and A_4 values are comparable to those for unfiltered water) within experimental error; the plot, however, starts in a different point (i.e. the A_1+A_3 sum is different for the filtered and unfilterd water). The deviation of the water activity decrease from the course accounted for by the radionuclide decay solely is mainly due to the sorption of the radionuclide on the sediment.

For the two radionuclides, Fig. 1. shows the time course of the distribution coefficients of the sediment, D, i.e. the values of the sediment-to-filtered water specific activity ratio for the given radionuclide at a given time. The distribution coefficients exhibited a constantly ascending trend and the equilibrium state never established during the experiment. The highest D values observed were about 1.6 for ^{99}Mo and about 0.6 for ^{131}I.

Fig.1. shows also the time course of the Ranunculus aquatilis-to-filtered water specific activity ratio, F. For obtaining specific activity data of the plants with respect to the dry plant matter, the values are to be multiplied by the fresh-to-dry plant weight ratio, which was 10.0 in average in our experiment. Since the equilibrium in the water-plant-radionuclide system was never reached, the bioaccumulation factor, i.e. the biomass-to-water specific activity ratio at equilibrium, is difficult to estimate accurately enough. A coarse estimate of this factor is on the order of hundreds, and it is higher for ^{99}Mo than for ^{131}I.

Whereas the time course of activities is similar for the two radionuclides if measured in water, sediment or plants, this is not true of the activity in fish. Radioactivity of ^{99}Mo in the various fish organs was virtually nil, only in the viscera a slight activity could be traced till approximately the 13th day that

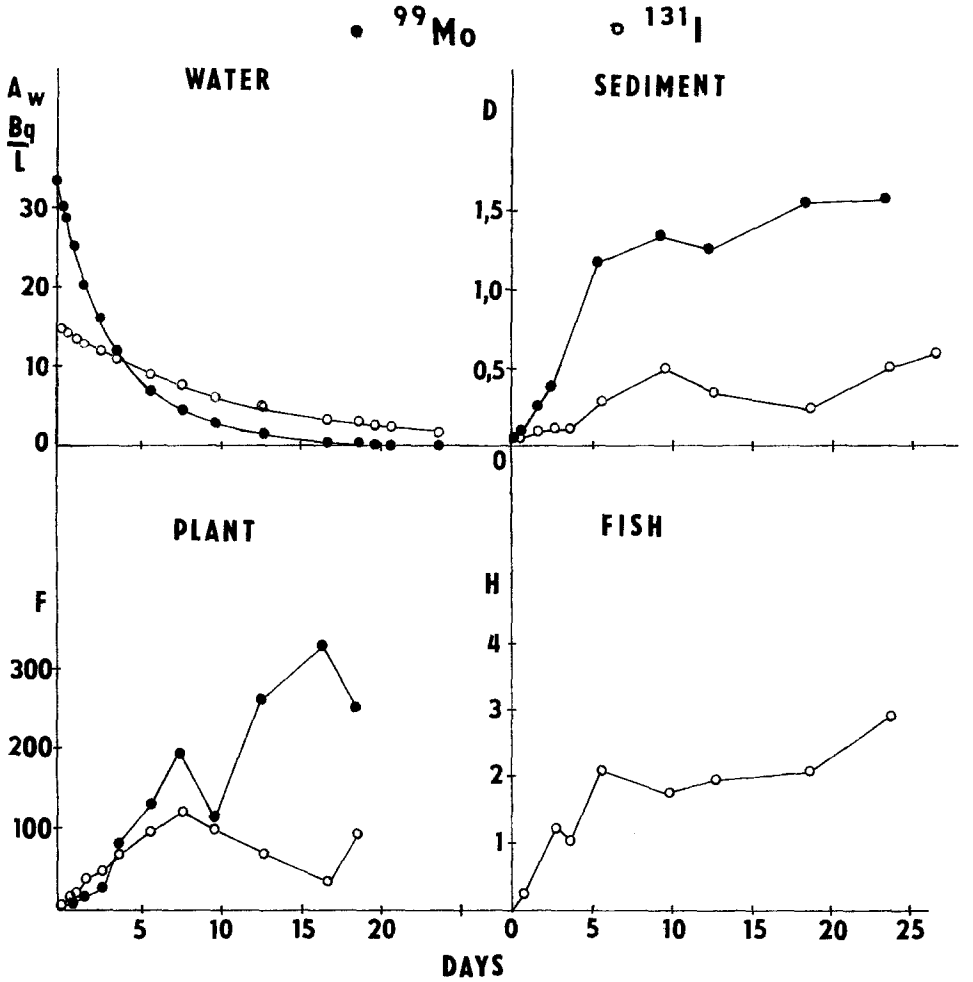

Figure 1. Time courses of activity concentration in the water, A_w, of distribution coefficient of the sediment, D, and of bioaccumulation factors for water plant Ranunculus aquatilis, F, and for fish - tench, H. Data are for radionuclides ^{99}Mo and ^{131}I. Means of 2-3 samples are represented; SE<30%.

the water activity decreased to below 1 kBq/L. For ^{131}I, on the other hand, the activity was measurable in all fish organs over the entire time of the experiment. The time course for ^{131}I of the whole fish or fish organ-to-unfiltered water specific activity ratios, H

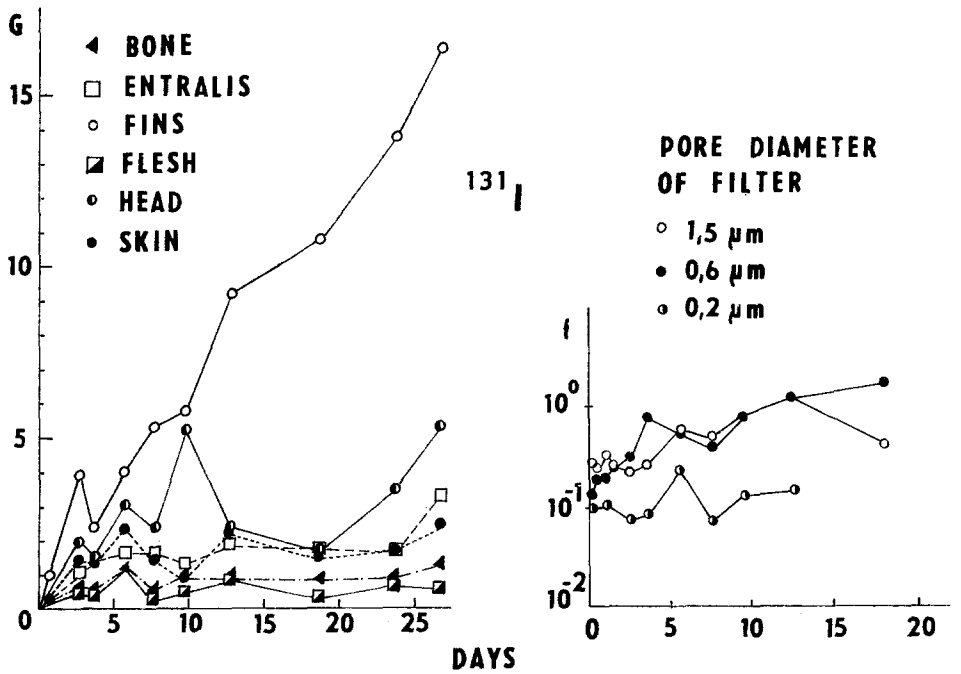

Figure 2. Time courses of bioaccumulation factor of fish organs,G, and of the filter/water total activity ratio,f. Data are shown for ^{131}I. Means of 2-3 samples are represented; SE<30%.

resp. G, are shown in Fig. 1. resp. Fig. 2. In this case the equilibrium state also never established, and so it can only be estimated that for ^{131}I the bioaccumulation factor is on the order of units for the whole fish and for the fish parts except the fins for which it is on the order of tens.

The bioaccumulation factors of ^{131}I for aquatic macrophytes and fish are reported in the literature (Polikarpov 1966; Vanderploeg 1975; NCRP 1984) and our estimates can be compared with them; this, however, does not apply to ^{99}Mo. The mean ^{131}I bioaccumulation factor value for aquatic macrophytes given in ref (Vanderploeg 1975) is 120. Our experimental result is close to that value. For fish, the bioaccumulation factor values given in refs (Vanderploeg 1975; NCRP 1984) are 40 and 8 - 110, respectively. Our data approach better the lower values.

For characterizing the time course of activity on the filters, two quantities were measured: one was the ratio of the total activity (Bq) of the filter after filtration of a defined amount of water to the total

activity of this amount of unfiltered water in the same time, denoted f, the other was the ratio of the specific activity ($Bq.kg^{-1}$) on the filter to the specific activity of the unfiltered water, g. The specific activity on the filter is the total activity of the filter divided by the increment of the filter weight after filtration of water (the filter being weighed as outlined above). The f and g values are corrected for self-absorption of the radionuclide on the filter material. The time course of the two quantities is similar for the two radionuclides, qualitatively as well as quantitatively. The f values for ^{131}I are plotted in dependence on time in Fig.2 for the various filters.

The activities of the filters showed that bioaccumulation by living microorganisms is negligible. This is also borne out by microbiological analysis of the water. If the water from the given locality contained more living microorganisms, the activities of the filters would respond accordingly because the bioaccumulation factors for phytoplancton and algae, as reported in refs (Vanderploeg 1975; NCRP 1984), lie in the region of 130 to 800.

Acknowledgement. We thank the laboratory staff of the Institute of Hygiene and Epidemiology, Č.Budějovice, for measure of samples radioactivity and Mrs.V.Hofmannová for technical assistance. We are also grateful to Dr.J.Justýn (Institute of Water Management, Prague) and Dr.J.Singer (Nuclear Power Station, Temelin) for their helpful comments.

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Received June 26, 1989; accepted October 15, 1989.