

Similarities in the behaviour of Chernobyl derived Ru-103, Ru-106 and Cs-137 in two freshwater lakes

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ABSTRACT

Measurements of the concentrations of Ru-103 and Ru-106 in the water column and sediments of two productive temperate lakes were made during an eight month period following the Chernobyl accident. Ru-103 accumulation in the sediments began simultaneously with that of Cs-134 and a constant ratio (approx. 2.0 on a decay corrected basis) of Ru-103/Cs-137 was observed in surface waters and sediments throughout the measurement period. The ratio is not statistically different from that reported for atmospheric input and for epilimnetic water. The results suggest that the processes of removal of radioactive ruthenium and caesium from the mixed water column to the sediments are the same.

Enhancement of Cs, compared with Ru, in the isolated bottom waters of one of the lakes in the summer months provided evidence for Cs release from the sediments during a period of hypolimnetic anoxia.

Introduction

The explosion at the Chernobyl reactor ejected into the atmosphere a cocktail of radionuclides which were subsequently deposited onto terrestrial and aquatic environments. For public health reasons the majority of post-Chernobyl monitoring centred on I-131, Cs-134 and Cs-137. However, significant quantities of Ru-103 and Ru-106 were also deposited. As ruthenium can be present as Ru(III), Ru(IV), or as various oxy-anions, its redox chemistry is more diverse than that of iron which is immediately above it in the Periodic Table. In its stable form it is only present naturally in the environment in low concentrations and all radioactive isotopes are man-made. The Chernobyl accident thus provided an opportunity to investigate the

pathways of Ru in the environment. We obtained a limited data set which is, nevertheless, valuable in view of the scant environmental literature on Ru (Coughtrey and Thorne, 1983; Mangini et al., 1990). Over the same time period considerable data were collected concerning changes in Chernobyl-derived Cs concentrations in the waters and sediments of the same lakes (Davison et al. 1993). The present paper reports data on Ru concentrations in lake waters and sediments after Chernobyl and compares the nuclide's behaviour with that of caesium isotopes over the same time period.

Methods

Using a Jenkin corer, samples were taken from the sediments at the deepest points of the North Basin of Windermere (mean depth 25.1 m, maximum depth 64 m, mesotrophic) and Esthwaite Water (mean depth 6.4 m, maximum depth 15.5 m, eutrophic). Collection began on 8th May 1986, almost immediately after the major deposition of Chernobyl material in Cumbria, England which occurred on 3rd May, 1986. Samples were initially collected at two weekly intervals but the sampling interval was increased as time from the deposition increased (Table 1). Because the samples were collected from a boat tied to permanently located buoys, all samples from a given lake originated from within an area of about 300 m².

On return to the laboratory the cores were sliced down to ten centimetres at one centimetre intervals, dried at 60 °C and analysed on a gamma ray spectrometer. More detailed descriptions of the lakes and the methods of sample collection and analysis are reported elsewhere (Davison et al., 1993).

Originally the data were analysed using Canberra Spectrum-F software. However, considerable inaccuracies were observed, both in the program's ability to identify multi-peak isotopes and in its ability to quantify correctly. As a result all data reported here were calculated by hand from the stored, original spectra. Because of the heavy pressure on counting facilities at this time, data for Ru-103, which has a 40 day half-life, were only obtained for the period from May to early July. Data for Ru-106 were only obtained for later samples (generally from mid July to December 1986) when counting times were longer as quantification was carried out on a low (9.8%) abundance line at 622 keV.

Ru-103 measurements were made on one water sample from the epilimnion and one water sample from the anoxic hypolimnion of Esthwaite Water, which was collected on 27th August, 1986. Cs-134 and Cs-137 measurements were made on all samples. Activities of radionuclides in rainfall collected in Cumbria during the deposition event were obtained from the Department of Agriculture for Northern Ireland et al. (1986). Ru and Cs activities in all samples were corrected to the 3rd May, 1986.

Results and discussion

Activity levels and associated counting errors for Ru-106, Ru-103, Cs-134 and Cs-137 in Windermere and Esthwaite sediments are given in tables 1a and b. Cs-137

Table 1. Cs-137, Cs-134, Ru-103 and Ru-106 activity levels and associated standard deviations, measured in the sediments of (a) Windermere and (b) Estwaite Water. The date of sampling and the depth of each slice are indicated. Slices with Cs-137/Cs-134 greater than 3.5 (*) probably contain significant amounts of weapons Cs-137 and were not included in further analysis

Date	Days from 06-May-86	Slice cm	Ru103 Bq/kg	SD	Ru106	SD	Cs-137	SD	Cs-134	SD	Cs-137 Cs-134
(a) WINDERMERE											
08-May-86	2	0-1	<22				81.4	12	<11		
13-May-86	7	0-1	<22				152	14	<11		
20-May-86	14	0-1	<22				311	16			
03-Jun-86	28	0-1	4144	249	880	210	1162	45	492	42	2.36
03-Jun-86	28	1-2	278	21			266	12	39	6	6.82*
17-Jun-86	42	0-1	2630	113	980	141	1728	33	696	26	2.48
17-Jun-86	42	1-2	280	16			359	12	115	9	3.12
01-Jul-86	56	0-1	2900	157	810	190	966	22	339	17	2.85
01-Jul-86	56	1-2	492	38	110	26	281	11	48	9	5.85*
15-Jul-86	70	0-1					1560	100	675	40	2.31
29-Jul-86	84	0-1			2550	550	3690	140	1620	60	2.28
29-Jul-86	84	1-2			557	145	521	24	217	12	2.4
14-Aug-86	100	0-1					1020	85	403	27	2.53
03-Sep-86	120	0-1			605	161	922	36	401	16	2.3
03-Sep-86	120	3-4			481	114	209	12	32.9	5.3	6.35*
23-Sep-86	140	0-1					725	66	303	22	2.39
21-Oct-86	168	0-1			992	298	2080	80	935	33	2.22
21-Oct-86	168	1-2			793	203	1480	60	638	22	2.32
18-Nov-86	196	0-1			1090	250	1870	70	854	29	2.19
18-Nov-86	196	1-2			1000	220	1736	64	797	26	2.18
18-Nov-86	196	2-3			515	149	959	38	386	15	2.48
09-Dec-86	217	1-2			729	18	1070	40	450	17	2.38
02-Apr-87	331	0-1			739	246	1540	57	676	23	2.28
02-Apr-87	331	1-2			626	173	856	33	344	14	2.49
03-Jun-87	393	0-1			966	281	1711	64	722	26	2.37

Table 1 (continued)

Date	Days from 06-May-86	Slice cm	Ru-103 Bq/kg	SD	Ru-106	SD	Cs-137	SD	Cs-134	SD	Cs-137 Cs-134
(b) ESTHWAITE											
08-May-86	2	0-1	<22				159		<11		5.24*
13-May-86	7	0-1	677	47			407	13	77.7	10.8	2.28
21-May-86	15	0-1	4995	245	1343	171	2479	34	1088	29	2.47
04-Jun-86	29	0-1	2146	103	477	116	1225	186	496	15	2.83
16-Jun-86	41	0-1	1400	59			684	14	242	10	2.47
02-Jul-86	57	0-1	3885	167	984	112	2357	27	955	16	2.21
14-Jul-86	69	0-1			2180	400	3730	140	1690	50	2.2
14-Jul-86	69	1-2			1240	190	2810	100	1280	30	2.15
30-Jul-86	85	0-1			1550	370	3480	110	1620	60	3.02
30-Jul-86	85	1-2			688	90	3080	100	1020	24	2.27
11-Aug-86	97	0-1			1330	290	2360	80	1040	30	2.25
03-Sep-86	120	0-1			483	107	1180	43	524	15	2.74
22-Sep-86	139	0-1					520	18	190	8	2.27
20-Oct-86	167	0-1			1160	210	1550	60	683	24	2.19
03-Nov-86	181	0-1			737	167	1060	40	483	19	2.26
03-Nov-86	181	1-2			749	112	1460	50	647	17	2.52
19-Nov-86	197	0-1			392	101	751	30	298	11	2.38
15-Dec-86	223	0-1			371	30	751	30	315	12	2.61
15-Dec-86	223	1-2			218	64	477	19	183	7	

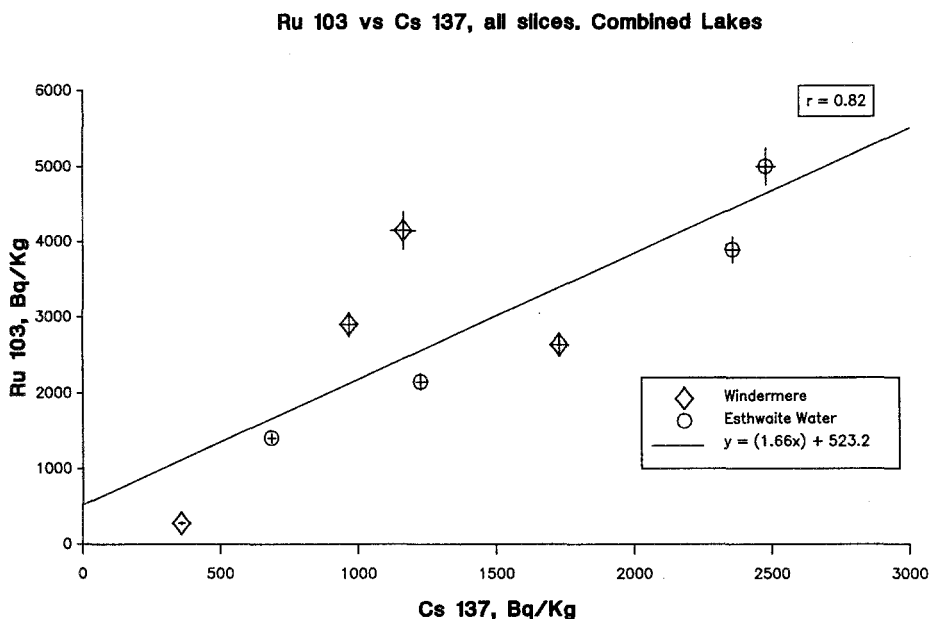


Figure 1. Plot of Ru-103 against Cs-137 (uncorrected for any residual weapons testing radio-caesium) for Windermere (diamonds) and Esthwaite (open circles). The regression line is given by $Ru-103 = 1.66 Cs-137 + 523$ ($r = 0.82$)

originated from a combination of weapons fallout or from Chernobyl deposition. Ru-103, Ru-106 and Cs-134 all resulted from Chernobyl deposition alone. There is no evidence of input of any radionuclides to these sediments from the nearby Sellafield site. Two of the Chernobyl specific nuclides, Ru-103 and Cs-134, were first observed, simultaneously, in the sediments of Esthwaite Water on 13th May, 1986, and on 3rd June, 1986, in Windermere, i.e. 10 days and 31 days respectively after atmospheric deposition. Prior to these dates neither nuclide had been observed separately in the sediments, suggesting some similarity in the mode of transport. Ru-106 activity levels were too low to be detected in these samples.

In Fig. 1, specific activities of Ru-103 in the sediments of Windermere and Esthwaite Water are plotted against those of Cs-137 measured in the same samples. Similarly, specific activities of Ru-106 in samples from the sediments of both Windermere and Esthwaite Water are plotted against those of Cs-137 in Fig. 2. In both cases the combined data sets are well represented by single straight lines ($r (Ru-103/Cs-137) = 0.82$ and $r (Ru-106/Cs-137) = 0.86$), with slopes and intercepts which do not differ significantly from those obtained for data from the lakes taken individually (Fig. 1 and 2). The straight lines demonstrate the relative constancy of the ratios over a wide range of concentrations and with time.

Levels of activity in different media, e.g. water, sediment, etc. cannot easily be compared. Unit-less ratios are more appropriate for this task. However, previous studies (Hilton et al., unpublished) showed that, in surface sediments, Cs-137 activity levels resulting from weapons fallout were equivalent to approximately 190

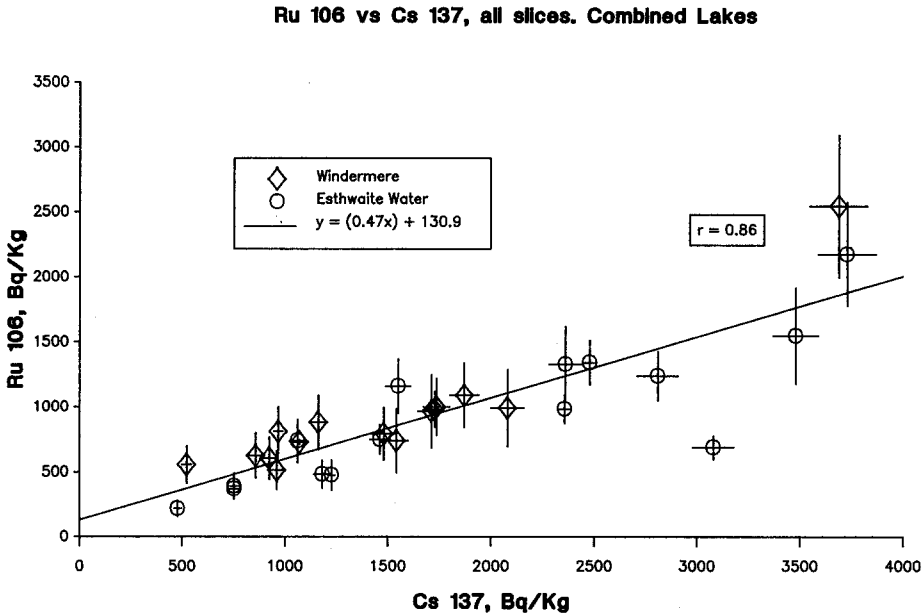


Figure 2. Plot of Ru-106 against Cs-137 (uncorrected for any residual weapons testing radio-caesium). Samples from Windermere are diamonds, those from Esthwaite are open circles. The regression line is given by $\text{Ru-106} = 0.47 \text{ Cs-137} + 131$ ($r = 0.86$)

and 160 Bq/kg in Windermere and Esthwaite respectively. Hence, in slices with a low Chernobyl contribution, the Ru-/Cs-137 ratio could be biased by the presence of a relatively large weapons component. There is good evidence that, in the UK the Cs-137/Cs-134 ratio in the Chernobyl deposition was approximately 2 (Cambrey et al., 1987). If a significant amount of weapons Cs-137 were present then the ratio would be higher than the expected ratio of about 2. On this basis four slices were found with Cs-137/Cs-134 ratios significantly greater than 3.12, the upper range of the cluster of values around a mean of 2.40 (SD = 0.24; $n = 30$). Data from these outlying slices were not considered in further analysis.

Because of the way in which the analyses were carried out in this work, Ru-103 data give information on the pattern of events during the two months immediately following deposition, while Ru-106 data generally give information on the longer time scale, i.e. from July to December, 1986. The timescale of the separate Ru-103 and Ru-106 data sets can be extended to cover the whole period from the initial deposition to 8 months afterwards by combining the two. Apparent Ru-103/Cs-137 ratios were derived from Ru-106/Cs-137 ratios using the mean Ru-103/Ru-106 ratio (3.86 ± 0.7 ; $n = 6$) observed in those samples from Windermere and Esthwaite Water where both isotopes were measurable in the same sample (Table 1). No systematic changes could be observed when the observed and calculated ratios of Ru-103/Cs-137 in the sediments of Esthwaite Water were plotted against time (Fig. 3b). The effect is less clear for Windermere (Fig. 3a) but is probably still the case.

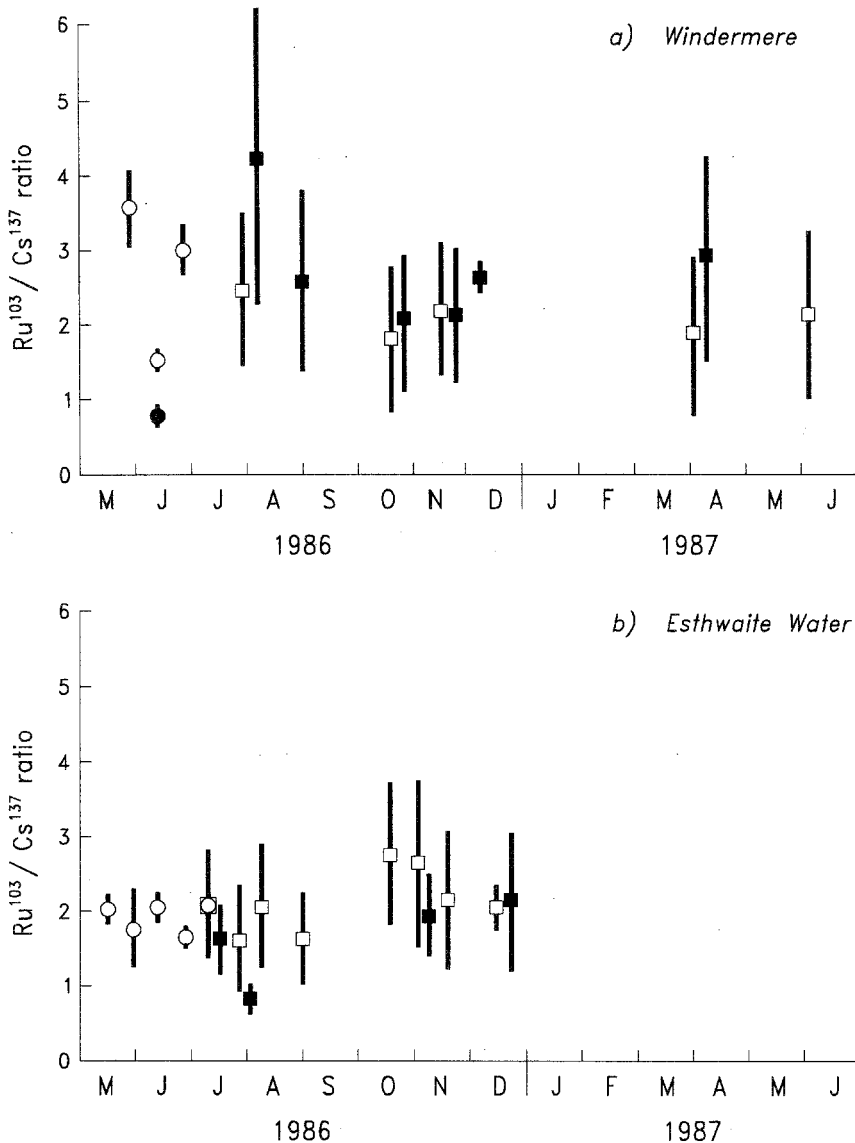


Figure 3. Ratios of Ru-103/Cs-137 measured in the sediments of (a) Windermere and (b) Esthwaite Water plotted against time. Square symbols are for data calculated from measured activities of Ru-106. Measurements made on the surface 1 cm of sediment are shown by open symbols and those from 1–2 cm depth by filled symbols. 95% confidence intervals for counting errors are shown

Table 2. Ru/Cs ratios in different parts of the ecosystem after Chernobyl. All data are corrected to 3rd May, 1986

		Ru-103	Ru-106	Ru-103
		Cs-137	Cs-137	Cs-137
Rainfall ¹	Mean	1.64		
	S.D.	0.57		
	n	4		
Water (Est) Epilimnion	mean	1.70		
	S.D.	0.22		
	n	1		
Water (Est) Hypolimnion	mean	0.74		
	S.D.	0.11		
	n	1		
Esthwaite Sediments	mean	1.87	0.50	1.94*
	S.D.	0.17	0.12	0.45
	n	4	15	15
Windermere Sediments	mean	2.22	0.65	2.38*
	S.D.	1.12	0.15	0.70
	n	4	13	13
Miscellaneous ² Atmospheric Particles	mean	1.23	0.33	
	S.D.	0.98	0.33	
	n	6	6	
	range	0.14–2.90	0.08–1.00	
Atmospheric particulates Chilton, UK. ²	single	1.33	0.35	
	single	1.73	0.39	
Soil (deposition) Switzerland ³	mean	1.26	0.29	
	S.D.	0.11		

* Equivalent long term ratio estimated from Ru-103/Ru-106 mean = 3.86; sigma = 0.7; n = 6 (Combined from Windermere and Esthwaite); range = 2.68–4.71.

¹ Dept. of Agriculture for Northern Ireland et al. (1986).

² Cambray et al. (1987)

³ Mangini et al. (1990)

Ru/Cs ratios observed in our data for waters and sediments of Esthwaite Water and Windermere are given in Table 2. They range from 0.78–3.57 for Ru-103/Cs-137 and from 0.22–0.84 for Ru-106/Cs-137. These values lie within the range of values reported for the Chernobyl plume and local rainfall during the deposition event (Table 2). However, Ru/Cs ratios in the atmosphere varied considerably with both the distance from the Chernobyl reactor and with time, hence the wide range of ratios given by Cambray et al. (1987) from miscellaneous sources such as dust from a ship in the Baltic on an unspecified date; material collected on the tail of an aircraft in Sweden and for atmospheric particles at Chilton, UK on the 2nd and 3rd

of May 1986. Our mean values were slightly higher than the ratios 1.26 ± 0.11 (Ru-103) and 0.29 (Ru-106) reported by Mangini et al. (1990) from soil profiles around Lake Constance, Switzerland. The slightly lower levels measured in Switzerland probably result from radionuclide fractionation in the atmospheric aerosol during the different times of travel of the radioactive cloud from Chernobyl. Although for some sub-sets standard deviations are high, overall the data show similar Ru-103/Cs-137 ratios in Cumbrian rainfall; epilimnetic lake water and sediments, whether they were derived from Ru-103 or Ru-106 measurements.

It is generally accepted that, except in shallow lakes where resuspension dominates, the removal of radionuclides from the water column proceeds via either: hydraulic flushing of particulate and dissolved radionuclides; sorption to particles followed by settlement to the sediment and/or direct sorption onto the sediment across a static hydraulic boundary layer (Hesslein et al., 1980; Davison et al., 1993). Hydraulic flushing will act equally on both radionuclides. Assuming no difference in the particle composition with time, the relative rates of transport of radionuclides to the sediment will be directly proportional to their adsorption coefficient, K_d . K_d s for Cs in these waters are typically of the order, $10^4 - 10^5$ (Davison et al., 1993; J. Smith, personal communication). K_d of $10^3 - 10^6$ are not unusual for Ru (Robbins et al., 1992; Aston and Duursma, 1973). If K_d^{Ru} were as high as 10^6 then it would disappear much faster from the water column than Cs, decreasing the Ru/Cs ratio in the water and increasing it in the sediment. Conversely, the rate of uptake of radionuclide by the sediment across the boundary layer is controlled by the diffusion coefficient. Since Ru ions are likely to carry a higher charge than monovalent Cs ions they are likely to have a higher diffusion constant. However, diffusion constants for small ions seldom vary by more than a factor of 3 from the fastest to the slowest (Li and Gregory, 1974). Hence, in this case, it would be expected that the Ru/Cs ratio would increase in the water and decrease in the sediment relative to the ratio in the initial deposition, although the rate of change may be slow. The constancy of the Ru/Cs ratio in Esthwaite (in both sediment, deposition and water) and Windermere (deposition and sediment) and their appearance in the sediments at the same time suggests that the transport processes and, in particular the K_d and, possibly, the diffusion coefficient are similar for the two nuclides.

Hesslein et al. (1980) found in a Canadian lake that two different elements, Co-60 and Cs-134, were removed from the water column at the same rate, i.e. a constant ratio was observed in the water column. There, however, it was considered that different mechanisms were involved for each element, but operating at similar rates; the faster rate of direct sorption onto the epilimnetic sediments by Cs-134 being countered by an increased rate of sedimentation loss to the deep sediments of Co-60. As a result, ratios in both the epilimnetic and hypolimnetic sediments differed from those in the water column. In our study, a constant ratio was observed in both water and sediments. Since the ratio of Ru/Cs in various environmental compartments at different times after the main deposition event will depend on the transport mechanisms of both Ru and Cs, a constant ratio throughout the lacustrine system (deposition, sediment and water) suggests that a similar mechanism for transport to the sediment exists for these two elements, despite their different chemistries. The ratios could be changed by the addition of radionuclides after the

deposition by transport from the catchment but there was no evidence for such inputs from any to the nuclides.

Elsewhere, in Swiss lakes, a variable ratio has been found. Mangini et al. (1990) observed mean values for Ru-103/Cs-137 of 1.26 ± 0.1 in the soils around Lake Constance (assumed to be the same as the original atmospheric deposition), 2.1 ± 0.3 in sediment traps over the period from the end of April until the end of September and 2 for deep water sediments, six months after Chernobyl. They used the difference between the mean ratios observed in the different environmental compartments and the systematic changes with respect to time in the Ru-103/Cs-137 ratio in the trap material to infer that fractionation of the two radionuclides had occurred.

Apparently the relative post-Chernobyl behaviour of ruthenium and caesium in the Swiss lakes was different from that observed in the English lakes. In the Swiss lakes, Ru was assumed to take part in coprecipitation reactions with autochthonous calcium carbonate, which constituted the majority of the settling particles in Constance, whereas Cs was sorbed by illite minerals, whose flux remained constant throughout the observation period. Although no Ru/Cs ratios were given by Sant-schi et al. (1990), qualitative comments suggest that a similar effect was also observed for sediment trap material in Lake Zurich, i.e. a decrease in Ru/Cs ratio with time. Robbins et al. (1992) analysed the trap data of Mangini et al. (1990) further by mathematical modelling. Their results suggested that Ru sorbed preferentially to organic matter with a K_d 4–10 times that of radiocaesium which sorbed to clays, particularly illite. Hence, changes in the Ru/Cs ratio would reflect changes in organic/illite ratios. Variable calcium carbonate precipitation throughout the period of trap exposure diluted the sorbing particles, reducing the apparent K_d of both nuclides, but the ratio would be unaffected by this effect. In Lake Constance, illite constitutes about 3.5% of the sediment dry matter, whereas in Esthwaite it constitutes approximately an order of magnitude more (Spezzano et al., 1993). The higher concentration of illite in settling particles in Esthwaite could be expected to increase the overall sediment K_d of radiocaesium compared to that observed in Lake Constance, while the K_d of Ru is most probably related to the organic content (Robbins et al., 1992) and is likely to be similar in both lakes.

Hence, it is quite possible, that while fractionation occurred in lake Constance, it did not occur to any appreciable extent in Esthwaite Water or Windermere. It is also possible that some of the variation in Ru/Cs ratios in Constance was the result of fuel particle settlement. Robbins et al. (1992) reported that the trap catches of Ce-144 were erratic. The authors attributed this behaviour to the presence of Ce-144 as hot particles (small particles of solid core material, rather than the more usual deposition products caused by condensation of gaseous radioactive discharges from the reactor). It is well known that fuel particles contained significantly greater amounts of Ru isotopes compared to radiocaesium isotopes (e.g. Schubert and Behrend, 1987; Misaelides et al., 1987).

Another feature of our data is the difference in the epilimnion and hypolimnion ratios for Esthwaite. The Ru-103/Cs-137 ratio in the water sample taken on August 1986 from the hypolimnion of Esthwaite Water was half the observed ratio in the epilimnion at that time. Ru concentrations were virtually the same in the epilimnion and the hypolimnion (10.7 ± 1.25 and 8.7 ± 1.3 mBq/l respectively), whereas Cs concentrations in the hypolimnion were twice those in the epilimnion (39.5 ± 4.4 and

72.0±15.0 mBq/l respectively). Cs concentrations in the bottom waters of both Windermere and Esthwaite Water remained fairly constant after thermal isolation during the summer of 1986 (Davison et al., 1993). Hence, unless there was a hypolimnetic removal mechanism that was unique to Ru, there was either a release of Cs from the sediment into the water column or conditions were such that Cs adsorption to particles or the sediment was inhibited in the anoxic hypolimnion. It is well known that, in anaerobic sediments, radiocaesium is displaced from radio-caesium specific sorption sites by ammonium ions (Evans et al., 1983), whereas no equivalent mechanism is known for ruthenium. The hypolimnion of Esthwaite Water regularly becomes deoxygenated soon after stratification. Hence, these data are compatible with other data which support a mechanism of remobilisation of Cs from the sediment in the hypolimnion of Esthwaite Water during summer stratification (see Davison et al., 1993).

In conclusion, the observations suggest that, in the two lake systems of our study, Ru and Cs behave similarly with respect to the recruitment rate to the sediment deposits. Modelling of elemental ratios in lacustrine systems (Hilton et al., in preparation) indicates that a constant ratio is possible if removal of radionuclides results from either a combination of hydraulic removal and particle sedimentation or a combination of hydraulic removal, particle sedimentation and direct diffusion into the sediment.

REFERENCES

- Aston, S.R. and E.K. Duursma, 1973. Concentration effects on Cs-137, Zn-65, Co-60 and Ru-106 sorption by marine sediments, with geochemical implications. *Netherlands J. Sea Res.* v. 6: 225–240.
- Cambray, R.S., P.A. Cawse, J.A. Garland, J.A.B. Gibson, P. Johnson, G.N.J. Lewis, D. Newton, L. Salmon and B.O. Wade, 1987. Observations of radioactivity from the Chernobyl accident. *Nuclear Energy*, v. 26: 77–101.
- Coughtrey, P.J. and M.C. Thorne, 1983. Radionuclide distribution and transport in terrestrial and aquatic ecosystems: a critical review of the data. volume 1. 304–320. A.A. Balkema (pub.), Rotterdam.
- Davison, W., J. Hilton, J. Hamilton-Taylor, F. Livens and M. Kelly, 1993. Measurement, interpretation and modelling of Cs-137 transport through two freshwater lakes after Chernobyl. *J. Environ. Radioactivity*, v. 19: 125–153.
- Department of Agriculture for Northern Ireland et al, 1986. Levels of radioactivity in the U.K. from the accident at Chernobyl USSR, on 26th April, 1986. HMSO.
- Evans, D.W., J.J. Alberts and R.A. Clark, 1983. Reversible ion-exchange fixation of cesium-137 leading to mobilisation from reservoir sediments. *Geochim. Cosmochim. Acta* v. 47: 1041–1049.
- Hesslein, R. W., W.S. Broecker and D. W. Schindler, 1980. Fates of metal radio tracers added to a whole lake: Sediment – water interactions. *Can. J. Fish. Aquat. Sci.* v. 37: 378–386.
- Hilton, J., E. Rigg, W. Davison, J. Hamilton-Taylor, M. Kelly, F. R. Livens, D. L. Singleton, (in preparation). Modelling and interpreting element ratios in water and sediments: a sensitivity analysis of post-Chernobyl Ru, Cs ratios.
- Li, Y-H and S. Gregory, 1974. Diffusion of ions in sea water and deep sea sediments. *Geochim. Cosmochim. Acta*, v. 38: 703–714.
- Mangini, A., U. Christian, M. Barth, W. Schmitz and H. H. Stabel, 1990. Pathways and residence times of radio tracers in Lake Constance. In „Large Lakes.“ Ed. M. M. Tilzer and C. Serruya. Springer Verlag. 265–287.

- Misaelides, P., C. Sikalidis, R. Tsitouridou and C. Alexiades, 1987. Distribution of fission products in dust samples from the region of Thessaloniki, Greece, after the Chernobyl nuclear accident. *Environ. Pollut.* v. 47: pp. 1–8.
- Robbins, J. A., G. Lindner, W. Pfeiffer, J. Kleiner, H. H. Stabel and P. Frenzel, 1992. Epilimnetic scavenging of Chernobyl radionuclides in Lake Constance. *Geochim. Cosmochim. Acta.* v. 56: pp. 2339–2361.
- Santschi, P.H., S. Bollhalder, S. Zingg, A. Lueck and K. Farrenkothen, 1990. The self-cleaning capacity of surface waters after radioactive fallout. Evidence from European waters after Chernobyl, 1986–1988. *Environ. Sci. Technol.*, v. 24: 519–527.
- Santschi, P.H., S. Bollhalder, K. Farrenkothen, A. Lueck, S. Zingg and M. Sturm, 1988. Chernobyl Radionuclides in the Environment: Tracers for the Tight coupling of Atmospheric, Terrestrial, and Aquatic Geochemical Processes. *Environ. Sci. Technol.* v. 22: 510–516.
- Schubert, P. and U. Berhend, 1987. Investigation of radioactive particles from the Chernobyl fallout. *Radiochem. Acta* v. 41, pp. 149–155.
- Spezzano, P., J. Hilton, J. P. Lishman and T. R. Carrick, 1993. The variability of Chernobyl Cs retention in the water column of lakes in the English Lake District, two years and four years after deposition. *J. Environ. Radioactivity* v. 19, pp. 213–232.

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