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Radioactivity in aquatic systems

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4.1 INTRODUCTION

The Chernobyl Nuclear Power Plant (NPP) is situated next to the Pripyat River which is an important component of the Dnieper River–Reservoir system, one of the largest surface water systems in Europe (Figure 4.1). After the accident, radioactive fallout on the Pripyat catchment threatened to wash downriver into the Kiev Reservoir, a major source of drinking water for the city of Kiev. The radioactive contamination of aquatic systems therefore became a major issue in the immediate aftermath of Chernobyl.

Initial radioactivity concentrations in river water were relatively high as a result of direct fallout onto the river surfaces and washoff of contamination from the surrounding catchment area. During the first few weeks after the accident, however, activity concentrations in river waters rapidly declined because of the physical decay of short-lived isotopes and as radionuclide deposits became absorbed to catchment soils. In the longer term, relatively long-lived radiocaesium and radiostrontium formed the major component of river water contamination. Though long-term levels of these isotopes in rivers were low, temporary increases in activity concentrations during flooding of the Pripyat River caused serious concern in Kiev over the safety of the drinking water supply.

Lakes and reservoirs around Europe were contaminated by fallout to lake surfaces and transfers of radionuclides from their surrounding catchments. Radioactivity concentrations in water declined relatively rapidly in reservoirs and in those lakes with significant inflows and outflows of water, as radionuclides were 'flushed' out of the system. In the long term, most lakes and reservoirs showed similar radiocaesium and radiostrontium activity concentrations to those of their inflowing rivers and streams. In the areas around Chernobyl, however, there are many lakes with no inflowing and outflowing streams ('closed' lake systems). Cycling of radiocaesium in these closed systems led to much higher activity

Figure 4.1. Pripyat–Dnieper River–Reservoir system showing Chernobyl and Kiev with the Kiev Reservoir in between.

concentrations in water and aquatic biota than were seen in open lakes and rivers.

Bioaccumulation of radionuclides (particularly radiocaesium) in fish resulted in activity concentrations (both in western Europe and in the former Soviet Union, fSU) which were in many cases significantly above maximum permissible levels for consumption. In some lakes, particularly in the Ukraine, Belarus and Russia, these problems have continued to the present day and evidence suggests that they will continue for the foreseeable future. Freshwater fish provide an important food source for many of the inhabitants of the contaminated regions of the Ukraine, Russia and Belarus. Prior to the Chernobyl accident, 17% of the population of the Bryansk region of Russia consumed fish from local rivers and lakes (Balonov and Travnikova, 1990).

4.1.1 Distribution of radionuclides between dissolved and particulate phases

The fraction of a radionuclide which is absorbed to suspended particles in surface waters strongly influences both its transport and bioaccumulation. This fraction is expressed as the distribution coefficient (K_d), the radionuclide activity per kg of solid matter divided by the activity per litre of water. Table 4.1 shows a selection of estimated K_d values for some radiologically important radionuclides (^{131}I, ^{90}Sr, ^{134,137}Cs and Pu isotopes). These emphasise measurements made *in situ*, usually with a long contact time between radionuclide and sediment. For short contact times, K_d values may be lower by an order of magnitude or more. It is clear that ranges in reported values are very large, sometimes covering several orders of magnitude.

It is useful also to consider the fraction f_p of radioactivity which is sorbed to the solid phase (e.g., Håkanson, 1997). Defining C_{aq} (Bq l^{-1}) as the aqueous phase activity of a given radionuclide and C_s (Bq kg^{-1}) as the solid phase activity we can write:

$$K_d = \frac{C_s}{C_{aq}} \quad \text{and} \quad f_p = \frac{sK_d}{(1 + sK_d)} \tag{4.1}$$

where s is the suspended solids concentration (kg l^{-1}), K_d is the solids–aqueous distribution coefficient (l kg^{-1}) and f_p is the (dimensionless) fraction of the total activity which is in the solid phase. The relationship between K_d, s and f_p is illustrated in Figure 4.2.

In freshwaters, values of suspended solids concentrations typically range between 0.5–50 mg l^{-1}. Assuming the 'best estimate' K_d values given in Table 4.1, for example, suspended solids concentrations within this range lead to particulate sorbed fractions (expressed as a percentage) of 0.05–5% for ^{90}Sr, 4–80% for ^{134,137}Cs and 5–83% for Pu. We have not included a 'best estimate' K_d for ^{131}I since values reported are even more variable than for the other radionuclides. Field measurements in an experimental enclosure (Milton *et al.*, 1992) and in water bodies after Chernobyl (Kryshev, 1995), suggest values of f_p in the range 2–16% and 5–36% respectively.

In marine systems, generally lower particle sorption capacities and higher concentrations of competing ions tend to make radionuclide particle sorbed fractions significantly lower than in freshwaters. In the Baltic Sea after Chernobyl, less than 10% of ^{137}Cs was bound to particles (Carlson and Holm, 1992) and estimates put the average particulate sorbed fraction at approximately 1% (Carlson and Holm, 1992; Knapinska-Skiba *et al.*, 2001).

Table 4.1. K_d values for radiostrontium, radioiodine, radiocaesium and plutonium in freshwaters. 'Best estimate' values are given where appropriate. Note that different review estimates may contain some of the same data.

RN	Reported (K_d l kg^{-1})	Source	Best estimate	Comments
^{89,90}Sr	8–4×10^3	IAEA (1994)	10^3	Review.
	10^3	Coughtrey and Thorne (1983) Coughtrey *et al.* (1985)		Review.
	10^2–10^3	Mundschenk (1996)		Measurements at various suspended solids concentrations.
	250–500	Chittenden (1983)		'*In situ*' K_d from suspended sediment and river water.
	380–730	Joshi and McCrea (1992)		Two K_ds in the Ottawa River
	750–1,800	Konoplev *et al.* (1992a)		L. Lelev, 5 km from Chern. NPP
	1,600	Zeevaert *et al.* (1986)		'*In situ*' K_d from suspended sediment and river water.
Sorption is proportional to the cation exchange capacity (CEC) of the sorbent and inversely proportional to the strength of competing cations (usually Ca, Mg) in solution (Yasuda and Uchida, 1993). Relatively good agreement between different studies.				
^{131}I	0–80	IAEA (1994)		Review.
	3×10^2	Coughtrey *et al.* (1983, 1985)		Review. Based on limited data.
	10.5×10^4 and 8.2×10^5	Milton *et al.* (1992)		Two experimental enclosures, Perch Lake, Canada.
	2.2×10^3 and1.3×10^5	Estimated from data in Kryshev (1995) and assuming s values given in Sansone and Voitsekhovitch (1996)		Pripyat River, Kiev Reservoir, 1 May, 1986. May be influenced by hot particles, but most ^{131}I was in aerosol form.
Limited data and high variation between reported values.				
^{134,137}Cs	5×10^1 to 8×10^4	IAEA (1994)		Review.
	2×10^4	Coughtrey and Thorne (1983), Coughtrey *et al.* (1985)		Review. K_d presented as an estimated value only.
	$(3.7$–$9.4) \times 10^3$	Konoplev *et al.* (1992a)		L. Lelev, 5 km from Chern. NPP
	4.6×10^4 to 2.7×10^6	Konoplev *et al.* (2002)		L. Constance, Alpine Rhine, Rhine below L. Constance.
	8×10^3 to 4.2×10^5	J.T. Smith, unpubl. res. (see Figure 4.2)	8×10^4	Review of long term *in situ* measurements in 18 rivers and lakes.
Preferential sorption to illitic clay minerals. K_d believed to be inversely related to potassium and ammonium concentrations in water and to increase significantly over time. Best estimate is for long times after fallout.				
^{238}Pu	10^2–10^7	IAEA (1994)	10^5	Review.
^{239,240}Pu	10^4–10^6	Coughtrey *et al.* (1984); Coughtrey *et al.* (1985)		Review.
	6×10^4	Allard *et al.* (1984)		Hudson river
	1.4×10^5	Zeevaert *et al.* (1986)		'*In situ*' K_d from suspended sediment and river water.
	2×10^2 to 6×10^5	Murdock *et al.* (1995)		'*In situ*' distribution from contaminated stream suspended sediment and water.
Most (~80%) Pu in Lake Michigan lake water was found to be in the dissolved phase (Sholkovitz, 1983) Possible remobilisation of Pu from anoxic sediments.				

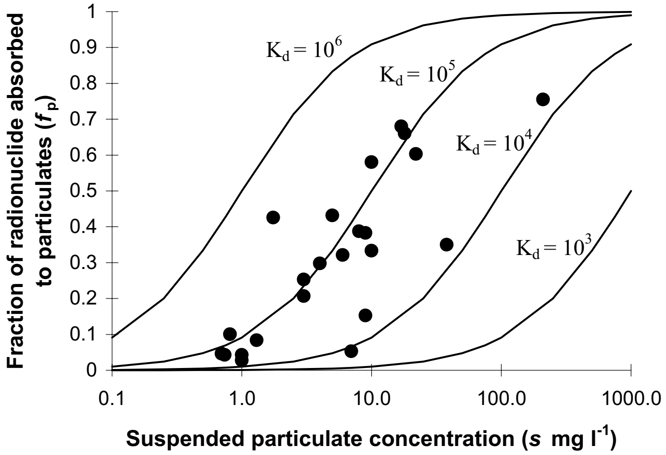


Figure 4.2. Fraction of a radionuclide absorbed to particulates as a function of suspended solids concentration in water for different values of K_d. The relationship is illustrated using measurements of the f_p for ^{137}Cs in 18 European rivers and lakes. In more than 75% of cases, most of the ^{137}Cs was found in the dissolved phase ($f_p < 0.5$).
From J.T. Smith, unpubl. res.

4.2 RADIONUCLIDES IN RIVERS AND STREAMS

The processes which control the radioactive contamination of rivers and streams have been identified by studies into the fate of ^{137}Cs and ^{90}Sr deposited as a result of atmospheric nuclear weapons testing (e.g., Carlsson, 1978; Helton *et al.*, 1985). Following a radioactive fallout, deposition of radionuclides onto the water surface is combined with runoff of radioactivity from the catchment. These transfers of radioactivity from the catchment are due to washoff from plant surfaces and from the surface soils. After some weeks/months, infiltration of radionuclides to deeper soil layers, and binding to soil particles, considerably decreases the rate of radionuclide runoff.

In streams and small rivers, maximum radioactivity concentrations in water were observed during, and shortly after, the Chernobyl accident, with levels declining rapidly over the first few weeks. However, radionuclide deposition in large river catchments was often non-uniform, so in some rivers 'polluted' water took some time to travel downriver from contaminated to less contaminated areas. For example, radioactivity deposited on the upper Elbe River catchment on 29 April, 1986 took approximately 8 days to reach a sampling station near the river mouth at Hamburg (Schoer, 1988).

Over longer time periods after fallout, radionuclides held in catchment soils are slowly transferred to river water by erosion of soil particles and (in the dissolved phase) by desorption from soils. The rates of transfer are influenced by the extent of soil erosion, the strength of radionuclide binding to catchment soils and migration down the soil profile. The time changes in radionuclide activity concentrations in rivers can be modelled by a series of exponential functions, as shown in Box 4.1.

Box 4.1. Modelling time changes in radionuclide contamination of rivers

The time changes in radionuclide activity concentrations in rivers may be modelled by a series of exponential functions (Monte, 1997; Smith *et al.*, 2004), as illustrated in Figure 4.3 for the Pripyat River.

For radiocaesium (Figure 4.3(a)), the radionuclide concentration in runoff or river water C_R (Bq m^{-3}) is given by:

$$C_R(t) = D_c(\alpha e^{-(\lambda+k_1)t} + \beta e^{-(\lambda+k_2)t} + \gamma e^{-(\lambda+k_3)t})$$

where λ (y^{-1}) is the decay constant of the radionuclide and D_c is the radionuclide deposition to the catchment (Bq m^{-2}). α, β, γ (m^{-1}) and k_1, k_2, k_3 (y^{-1}) are empirically determined (radionuclide-specific) constants. The k values may be expressed as effective ecological half-lives T_{eff} where $T_{eff} \approx \ln 2/(k + \lambda)$. The three exponential terms represent, respectively: the fast ‘flush’ of activity as a result of rapid washoff processes; a slow decline as a result of soil fixation and redistribution processes; and the very long-term ‘equilibrium’ situation.

For radiostrontium, the activity concentration in runoff water is also given by the above equation, though the parameter values are different to those for radiocaesium (Figure 4.3(b)).

For ^{131}I (and other short-lived isotopes), the half-life is so short that there is no long-term component to the decline, so the model is a simple exponential decay:

$$C_R(t) = D_c \alpha e^{-(\lambda+k_1)t}$$

4.2.1 Early phase

To our knowledge, there are few data of radionuclide concentrations in small streams in the Chernobyl area during the early phase of the accident. Most available data is for large rivers. Table 4.2 shows a summary of available measurements of radionuclide activity concentrations in a large river (the Pripyat) at Chernobyl at various times after the accident. Temporarily allowable levels of radionuclides in drinking water in the Ukraine at different times after Chernobyl are shown in Table 4.3.

Maximum radioactivity concentrations in rivers close to Chernobyl (the Pripyat, Teterev, Irpen and Dnieper) were approximately proportional to the amount of radioactivity released from the reactor (Figure 4.4). This relationship also approximately held for radionuclides in rivers in western Europe (the Glatt, Danube and Po) though it was less strong than in the the Ukrainian rivers.

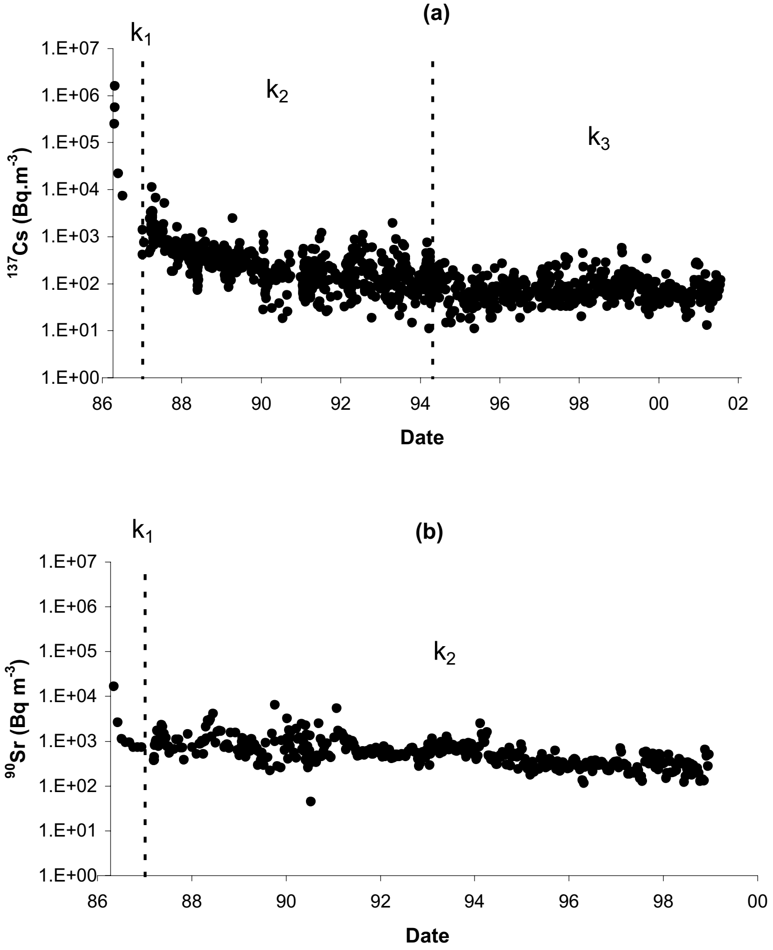


Figure 4.3. The change in activity concentration of ^{137}Cs and ^{90}Sr in the Pripyat River over time after the accident. The different phases in the exponential decline in activity concentrations (Box 4.1) are illustrated by the dotted lines and the '*k*' values. ^{137}Cs shows a three-component exponential decline whilst, over this period, ^{90}Sr shows a two-component decline. Data from Ukrainian Hydrometeorological Institute, Kiev.

In the UK, maximum activity concentrations of ^{131}I in surface waters in most regions were of the order 10 Bq l^{-1}, though one measurement of ^{131}I in surface water in the Strathclyde region of Scotland gave 1,315 Bq l^{-1} (c.f. 9,400 Bq l^{-1} observed in rainwater during 3–5 May in this region) (Jones and Castle, 1987).

As with initial maximum concentrations (Figure 4.4), a similar initial behaviour of different radionuclides was observed in the rate of their decline in concentrations in river water during the early phase after the accident. The rate of decline of radionuclide activity concentration in water is commonly measured as an effective

Table 4.2. Radionuclide levels (dissolved phase) in the Pripyat River at Chernobyl.[†] For some (radiologically important) radionuclides, doses are calculated assuming consumption at these concentrations over a one year period after the accident using ingestion rates and dose coefficients given in (NRPB, 1996).*

[†] From Vakulovsky *et al.* (1990), Voitsekhovitch *et al.* (1991), Vakulovsky *et al.* (1994), Kryshev (1995).

		Radionuclide concentration in water ($Bq\,l^{-1}$)							
RN	Half-life	01/05/86	02/05/86	06/05/86	03/06/86	16/07/86	09/08/86	1987 (mean)	Committed effective dose during 1st year (mSv)
^{137}Cs	30.2 y	250	555	1591	22.2	7.4		1.8	0.57 (a)
^{134}Cs	2.1 y	130	289[1]	827[1]	11.5[1]	3.8[1]		0.94[1]	0.43 (a)
^{131}I	8.1 d	2100	4440	814	33.3	<0.82[2]			4.2 (i)
^{90}Sr	28 y	30			1.9			1.5	0.049 (c)
^{140}Ba	12.8 d	1400							
^{99}Mo	3 d	670							
^{103}Ru	40 d	550	814	170	26	15			0.053 (i)
^{106}Ru	365 d	183[3]	271[3]	57[3]	8.7[3]	5[3]			0.29 (i)
^{144}Ce	284 d	380				37			
^{141}Ce	33 d	400		89		14.8			
^{95}Zr	65 d	400	1554	167	11	37			
^{95}Nb	35 d	420							
^{241}Pu	13 y	33[4]					0.6[4]		0.072 (a)
$^{239+240}$Pu	2.4×10^4 y 6.6×10^3 y	0.4					0.0074		0.0046 (a)

* Doses from each radionuclide were calculated for infants (i), children (c) and adults (a), the result for the age group showing the highest dose is shown for each radionuclide. Note that radionuclide concentrations in water at the point of consumption, and consequently doses, are likely to be much lower than these measurements in rivers owing to dilution and water treatment.

[1] From ^{137}Cs measurement and a ^{134}Cs : ^{137}Cs ratio ~0.52; [2] Assuming a decline from the 3 June, 1986 value by radioactive decay only; [3] From a ^{103}Ru measurement and assuming a ^{103}Ru : ^{106}Ru ratio (~3) for Chernobyl fallout; [4] From a ^{239,240}Pu measurement and a ^{241}Pu : ^{239,240}Pu ratio (~82) for Chernobyl fallout.

Table 4.3. Temporary allowable levels of radionuclides in drinking water in the Ukraine at different times after Chernobyl. The dates refer to the time at which each new regulation was implemented.

From Los *et al.* (1998).

Radionuclide ($Bq\,l^{-1}$)	6 May, 1986	30 May, 1986	15 Dec, 1987	22 Jan, 1991
^{131}I	3,700			
Total beta activity		370		
$^{137}Cs + {}^{134}Cs$			18.5	18.5
^{90}Sr				3.7

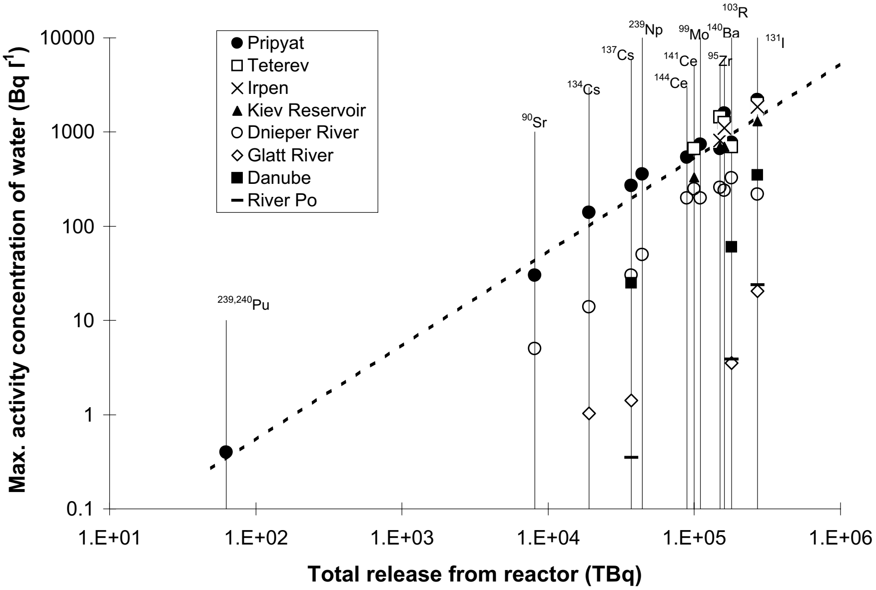


Figure 4.4. The initial activity concentrations of radionuclides in various rivers vs. the total amount released from the reactor. The measurements from the Pripyat, Irpen, Teterev and Dnieper Rivers show an approximately constant ratio between amount released and initial river water activity concentration.

From data in Voitsekhovitch *et al.* (1991), Kryshev (1995), Waber *et al.* (1987), Foulquier and Baudin-Jaulent (1990).

ecological half-life (Box 4.1), the time taken for the radioactivity concentration in water to decline by one-half. Measurements of the change in ^{137}Cs activity concentrations as a function of time after fallout for six European rivers were obtained from the literature. These gave values (Table 4.4) of effective ecological half-lives during the initial period after the accident of approximately 1–3 weeks. An additional study on measurements from the Rhine in Germany (Monte, 1995) gave a T_{eff} of 12.3 days for radiocaesium.

Table 4.4. Estimates of the initial rate of decline of radionuclides in river water (dissolved phase, except where indicated) after Chernobyl. Declines include a radioactive decay component: ecological half-lives (i.e., excluding radioactive decay) are given in brackets.

	Effective ecological half-life of decline in activity concentration (days)**					
RN	Pripyat, Ukraine[1]	Dnieper, Ukraine[1]	Po, Italy[2]	Glatt, Switzerland[3]	Danube, Hungary[4]	Elbe, Germany[5]
^{137}Cs	11.2 (11.2)	9.0 (9.0)	34.7* (34.7)	19.2 (19.2)	–	17.8† (17.8)
^{90}Sr	10.4 (10.4)	15.4 (15.4)	–	–	–	–
^{131}I	5.2 (14.3)	–	7.3* (72.3)	5.7 (19.5)	3.2 (5.2)	5.9† (21.4)
^{144}Ce	22.4 (24.3)	–	–	–	–	–
^{141}Ce	19.3 (46.0)	–	–	–	–	–
^{95}Zr	16.5 (22.2)	–	–	–	–	–
^{132}Te	–	–	–	2.3 (7.9)	1.9 (4.7)	–
^{103}Ru	13.8 (21.3)	–	17.1* (29.9)	28.7 (101)	5.3 (6.2)	7.5† (12.4)
^{241}Pu	17.3 (17.3)	–	–	–	–	–

* Measurements not begun until 16–20 May (Monte, 1995). † Dissolved and particulate phases. ** From data in [1] Voitsekhovitch *et al.* (1991); [2] Monte (1995); [3] Waber *et al.* (1987); [4] German (1986) quoted in Foulquier and Baudin Jaulent (1990); [5] Schoer (1988).

Though there is significant variation, there is little evidence of systematic differences in rates of decline between the different radionuclides. Generally faster rates of decline in ^{132}Te and ^{131}I are largely due to their rapid physical decay ($T_{1/2} = 3.2$ and 8.05 d respectively) rather than to any obvious differences in their washoff and deposition behaviour. The observation of similar half-times of ^{137}Cs and ^{131}I, for example, is supported by measurements of the changes in their activity concentrations in grass in the UK (Cambray *et al.*, 1987) which gave a removal half-time by washoff (excluding physical decay) of approximately 11 days for both elements (see also Chapter 3).

All of the radionuclides studied had effective ecological half-lives of less than one month (Table 4.4) in the initial period after the accident. The only estimate greater than one month is ^{137}Cs in the River Po ($T_{eff} = 34.7$ d), though this value is likely to be an overestimate of the initial effective ecological half-life since in this case measurements were not begun until around 3 weeks after the accident.

The consistent initial concentrations of different radionuclides in river water (per TBq released), and their similar rates of decline, would not be expected if different chemical interactions of the various radionuclides with the soil strongly controlled transport in the early period. Although there is significant variation, the measurements imply that in the early stages, physical transport processes such as rainfall onto the river surface, washoff from plants and from easily available fractions in the soil were more important than differing individual behaviours of the various radionuclides.

4.2.2 Intermediate phase

Following the initial rapid decline in radioactivity in rivers, longer term contamination was primarily due to ^{137}Cs and ^{90}Sr. Both radionuclides have relatively long half-lives (30.1 and 28.8 years, respectively) and were released from the reactor in significant quantities (Table 1.2). The temporal change in activity concentration of ^{137}Cs and ^{90}Sr in rivers is illustrated with measurements from the Pripyat River (Figure 4.3). Both radionuclides show a large decline in activity concentrations over time after fallout.

In the first few years after Chernobyl, the rate of decline in radiocaesium concentrations in river water (dissolved phase) was observed to be remarkably similar in the Dnieper, Pripyat, Rhine, Teterev and Uzh Rivers (Monte, 1995). This observation was confirmed by studies in a number of other surface waters in Europe (Smith *et al.*, 1999a). Estimates of the rates of decline of radiocaesium activity concentrations during a five-year period after the accident are presented in Table 4.5. Almost all (95%) of the measurements show effective ecological half-lives within the range of 1–4 years in the intermediate period after the accident.

The decline in ^{90}Sr activity concentrations in rivers was much slower than for ^{137}Cs (Figure 4.3 and Table 4.5) with most estimates of T_{eff} being in the range 5.6–11.7 years. This rate of decline of ^{90}Sr did not change significantly over a 15 year period after the accident (1987–2001). Because the vast majority of ^{90}Sr fallout was close to the reactor and in the form of fuel particles, these measurements could have been affected by the change in the chemical availability of ^{90}Sr as the fuel particles broke down (Konoplev *et al.*, 1992b; Kashparov *et al.*, 1999). Surprisingly, however, measurements of T_{eff} for (chemically available) ^{90}Sr from nuclear weapons test (NWT) fallout showed similar rates of decline to the post-Chernobyl studies. In five catchments in Finland, mean T_{eff} was 7.7 y (Cross *et al.*, 2002, from data in Salo *et al.*, 1984) and in 11 Italian rivers the rate of decline showed relatively little variation, having mean value $T_{eff} = 5.3$ y (Monte, 1997). The low ^{90}Sr fallout at long distances from Chernobyl meant that measurements in areas unaffected by fuel particles are scarce, though data from two rivers in Finland give $T_{eff} = 4.9$ and 9.9 y (Cross *et al.*, 2002) in general agreement with the pre-Chernobyl (NWT) studies.

4.2.3 Long-term ^{137}Cs contamination of water

The rate of decline in ^{137}Cs activity concentrations in the Pripyat river water (in contrast to ^{90}Sr) has slowed in recent years (Figure 4.3). The effective ecological half life of 1.2 years (dissolved phase) and 1.7 y (particulate phase) in the period 87-91, increased to 4.3 y (dissolved phase) and 11.2 y (particulate phase) between 1995 and 1998. This increase in T_{eff} has also been observed in rivers in Belarus (Kudelsky *et al.*, 1998), Ukraine (Voitsekhovitch, 1998) and Finland (R. Saxén, pers. commun.) as illustrated in Table 4.5.

Table 4.5. Rates of change in ^{137}Cs and ^{90}Sr activity concentrations in different rivers in the medium to long term (1987–2001) after Chernobyl.

Study	Time period	Rate of decline k (y^{-1})	Effective ecological half-life (T_{eff}) (years)
^{137}Cs in dissolved phase (intermediate time period)			
9 Ukrainian rivers[1]	1987–1991	0.3–0.65	1.1–2.3
5 Finnish rivers[2]	1987–1991	0.14–0.39	1.8–5.0
5 Belarussian rivers[3]	1987–1991	0.49–0.65	1.1–1.4
Forest catchment in Sweden[4]	1987–1993	0.18	3.8
Inlet to Lake Sälgsjön, Sweden[5]	1987–1990	0.23	3.0
Upland catchment in the UK[6]	1987–1989	0.39	1.8
Dora Baltea River, Italy[7]	1987–1991	0.34	2.0
Rhine River, Germany[8]	1987–1991	0.52	1.3
^{137}Cs on particulates (intermediate time period)			
Rhine River, Germany[8]	1987–1991	0.35	2.0
Pripyat River, the Ukraine[9]	1987–1991	0.41	1.7
^{137}Cs in dissolved phase (long-term change)			
Pripyat River, the Ukraine[9]	1995–1998	0.16	4.3
Dnieper River, the Ukraine[9]	1995–1998	0.17	4.1
Desna River, the Ukraine[9]	1995–1998	0.047	14.7
5 Rivers in Finland[10]	1995–2002	0.07–0.11	6.3–9.9
5 Rivers in Belarus[3]	1994–1998	0.13–0.3	2.3–5.3
^{137}Cs on particulates (long-term change)			
Pripyat River, the Ukraine[9]	1995–1998	0.062	11.2
Dnieper River, the Ukraine[9]	1995–1998	0.07	9.9
Desna River, the Ukraine[9]	1995–1998	0.24	2.9
^{90}Sr in dissolved phase			
10 Rivers in the Ukraine[9]	1987–2001	0.025–0.124	5.6–28.8
Kymijoki (Finland)[11]	1987–1995		9.9
Kokemäenjoki (Finland)[11]	1987–1995		4.9

[1] From data in Vakulovsky *et al.* (1994); [2] Smith *et al.* (2000a); [3] from data in Kudelsky *et al.* (1998); [4] Nylén (1996); [5] Sundblad *et al.* (1991) quoted in Nylén (1996); [6] Hilton *et al.* (1993); [7] data from L. Monte, ENEA, Italy (pers. commun.); [8] Monte (1997); [9] Kanivets and Voitsekhovitch (2001) in Smith *et al.* (2001); [10] from data in Saxén and Ilus (2001) and Smith *et al.* (2004); [11] Cross *et al.* (2002).

4.2.4 Processes controlling declines in ^{90}Sr and ^{137}Cs in surface waters

There are three main mechanisms which may contribute to the decline in radionuclide transfers (in dissolved form) to runoff water. These are: (1) loss of radioactivity from the catchment; (2) vertical migration to deeper layers of soil; (3) slow chemical 'fixation' in the soil. The first studies of radionuclide washoff after Chernobyl were carried out by Borzilov *et al.* (1988), Bulgakov *et al.* (1990) and Konoplev *et al.* (1992b). These studies showed significant removal of radioactivity in both dissolved and particulate phases. Long-term estimates of rates of ^{137}Cs removal from catchments (e.g., Smith *et al.*, 1999a; Helton *et al.*, 1985; Kudelsky *et al.*, 1998) show that losses of ^{137}Cs from catchments are very slow – being at most around 0.5–2% of the total amount in the catchment per year. Removal rates of <2% (typically, 0.1–1%) of the radiocaesium in the catchment per year are not sufficient to cause a decline in activity concentration in river water by one-half every 1.5–2 years (as observed in Table 4.5). Thus the observed rates of change in activity concentrations in surface waters between 1987 and 1991 cannot be due simply to loss of the store of radiocaesium in the catchment.

Another possible mechanism for reduction in transfers of radionuclides to surface waters is transport into deeper layers of the soil, thus reducing the concentration in more erodible surface layers. If vertical migration were the controlling mechanism, however, we would expect to see more rapid declines in radiostrontium activity concentrations than radiocaesium since radiostrontium migrates in the soil more rapidly than radiocaesium (Chapter 2). From the measurements presented in Table 4.5, this is not the case, at least in the first five years after fallout. The observed slower declines in radiostrontium in river water compared to radiocaesium suggest that the change in radiocaesium concentration is primarily controlled by fixation to soil particles rather than vertical migration during the intermediate period after the accident (Smith *et al.*, 1999a).

The gradual slowing of the rate of decline (increase in T_{eff}) of radiocaesium in rivers (Table 4.5) is attributed to a long-term equilibration of sorption and desorption processes in the soil (Chapter 2). In other words, the rate of decrease in radiocaesium availability in the soil has slowed. In the coming decades, it is expected that ^{137}Cs activity concentrations in rivers will decline at a slow rate determined by physical decay and slow physical redistribution processes in the catchment (Smith *et al.*, 2000b).

Time changes in particulate phase radiocaesium appear to be similar to those observed for the dissolved phase (Table 4.5). We would expect radiocaesium absorbed to particles to change only by physical redistribution in the catchment, since the amount absorbed to particles (in the long term) is not significantly affected by sorption and desorption processes. The similar rates of decline of particulate and dissolved phase radiocaesium may reflect an equilibration of the two phases in river water (so that, on average, particulate phase concentrations are a constant multiple of dissolved, i.e., the two phases are 'coupled'). On the other hand, radiocaesium absorbed to particles may be controlled by entirely different mechanisms of erosional transport in the catchment, and the similarity in rates of decline may be coincidental. It is plausible that radiocaesium attached to the more 'erodible' soils in the catchment is lost in the early years after fallout, leading to declining radiocaesium erosion rates over time.

As discussed above (see Section 4.1), radiostrontium is found almost entirely in the dissolved phase in river water. The steady decline in radiostrontium concentrations in river waters is not expected to be due to long-term changes in the strength of its sorption to soils since it is generally assumed that strontium is rapidly and reversibly sorbed to soils. The relatively slow declines in ^{90}Sr concentrations in rivers ($T_{eff} \sim 5$–10 years) may be partly explained by loss of the inventory of ^{90}Sr in the catchment. Studies of nuclear weapons test (NWT) and Chernobyl ^{90}Sr in rivers in Finland (Saxén and Ilus, 2001; Cross *et al.*, 2002) suggest loss rates of the order of 1–2% per year, leading to a half-time of decline of approximately 35 years (including decay, $T_{eff} > 16$ years). This is higher than typical observed T_{eff} values (Table 4.5), implying that vertical migration in soils and/or slow 'fixation' of ^{90}Sr may also play a role in its declining concentration in surface waters.

4.2.5 Influence of catchment characteristics on radionuclide runoff

The concentration of a radionuclide in surface water is usefully expressed as a concentration per unit of deposition ('normalised concentration', cf. aggregated transfer coefficient, Chapter 3):

$$R_c(t) = \frac{\text{Concentration of radionuclide in water (Bq m}^{-3})}{\text{Radionuclide deposition to catchment (Bq m}^{-2})} \text{ m}^{-1} \qquad (4.2)$$

Since the radionuclide concentration in water declines significantly over time, the normalised concentration also declines at the same rate. For simplicity, the fallout to the catchment is usually the value estimated for the time of the accident and is therefore constant.

Measurements of the normalised ^{137}Cs activity concentration in many different rivers (Figure 4.5(a)) show a range of approximately a factor of 30, even when temporal changes in concentrations have been accounted for. A number of studies (Hansen and Aarkrog, 1990; Hilton *et al.*, 1993; Nylén, 1996; Kudelsky *et al.*, 1996) have attributed this variation to the types of soil in the catchment, in particular, the proportion of highly organic peat bog soils. In mineral soils, ^{137}Cs is sorbed to highly selective 'Frayed Edge Sites' (FES) on the illitic clay fraction (Cremers *et al.*, 1988) and becomes 'fixed' in the mineral lattice (Comans and Hockley, 1992). Work has shown, however, that in highly organic soils FES concentrations are low, leading to a reduced binding of ^{137}Cs to the solid phase (Valcke and Cremers, 1994). In field studies on small catchments (Hilton *et al.*, 1993; Nylén, 1996; Kudelsky *et al.*, 1996) it was found that highly organic soils (particularly saturated peats) released up to an order of magnitude more radiocaesium to surface waters than some mineral soils. The relationship between radiocaesium in surface water and the percentage of organic soils in the catchment is illustrated in Figure 4.5(b).

As discussed above, the assessment of concentrations of radiostrontium in surface waters is complicated by a lack of data for systems outside the fSU, and the large component of fallout in the form of fuel particles. In river catchments in Finland, it was found (Saxén and Ilus, 2001) that (per unit of deposition) the runoff of both NWT and Chernobyl radiostrontium was approximately one order of magnitude greater than for radiocaesium. In studies of NWT ^{90}Sr, runoff was highest in catchments with organic soils (Linsley *et al.*, 1982; Hansen and Aarkrog, 1990) and those with a high proportion of surface waters (rivers, bogs and lakes) in the catchment (Salo *et al.*, 1984; Smith *et al.*, 2004).

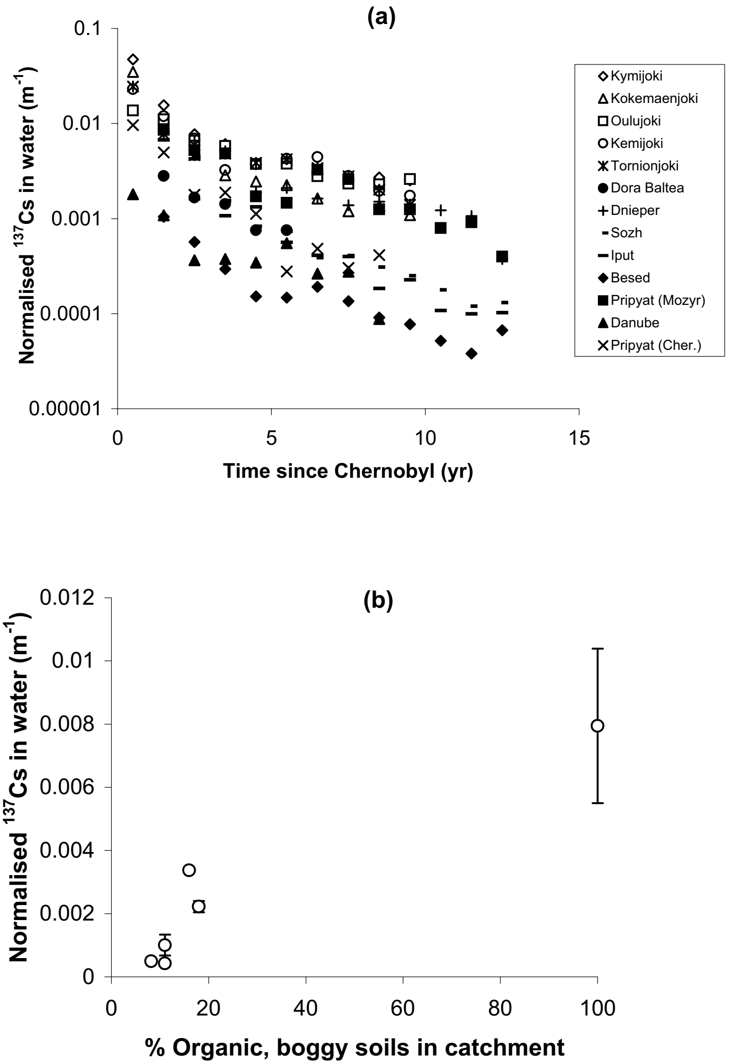


Figure 4.5. (a) Normalised activity concentration of ^{137}Cs in the dissolved phase of different rivers after Chernobyl. (b) Correlation between the normalised ^{137}Cs activity concentration and the percentage catchment coverage of organic, boggy soils in six different catchments.
(a) From data reviewed by Smith *et al.* (2004). (b) From data in Hilton *et al.* (1993); Kudelsky *et al.* (1996); Nylén (1996); Kudelsky *et al.* (1998).

Radionuclide activity concentrations in rivers can vary significantly throughout the year as a result of changing river and catchment conditions. For example, flooding of the Pripyat River, caused by blockages of the river by ice in late winter, led to temporary increases in ^{90}Sr activity concentrations in this system, but did not significantly affect ^{137}Cs concentrations (Vakulovsky *et al.*, 1994). The flooding caused increased washoff of ^{90}Sr from a highly contaminated flood plain area within the 30-km zone. For example, during winter 1991, concentrations of ^{90}Sr in the river water increased from around 1 Bq l^{-1} to approximately 8 Bq l^{-1} for a 5–10 day period (Vakulovsky *et al.*, 1994).

4.3 RADIOACTIVITY IN LAKES AND RESERVOIRS

Maximum activity concentrations of radionuclides in lakes and reservoirs occured during and shortly after the accident as a result of direct deposition of activity to the water surface and (initially, to a much lesser extent) transport of radioactivity from the catchment (Figure 4.6). In most lakes, radionuclides were well mixed throughout the lake water during the first days/weeks after fallout. In deep lakes such as Lake Zurich (mean depth of 143 m), however, it took several months for full vertical mixing to take place (Santschi *et al.*, 1990). In some areas of northern Europe, lakes were covered in ice at the time of the accident, so maximum concentrations in lake waters were only observed after the ice melted.

The initial concentration of radionuclides in lake and reservoir waters can be assessed by estimating the dilution of the surface deposited radioactivity in the body of water. The initial average activity concentration in the lake water C_T (Bq m^{-3}) is therefore estimated by:

$$C_T(t=0) = \frac{DA_L}{V_L} = \frac{D}{d} \tag{4.3}$$

where D is the fallout (Bq m^{-2}), V_L is the lake volume, A_L the lake surface area and d $(= V_L/A_L)$ the lake mean depth. It should be noted that C_T represents the total activity concentration in solid and aqueous phases and should not be confused with C_{aq} (the dissolved phase activity concentration).

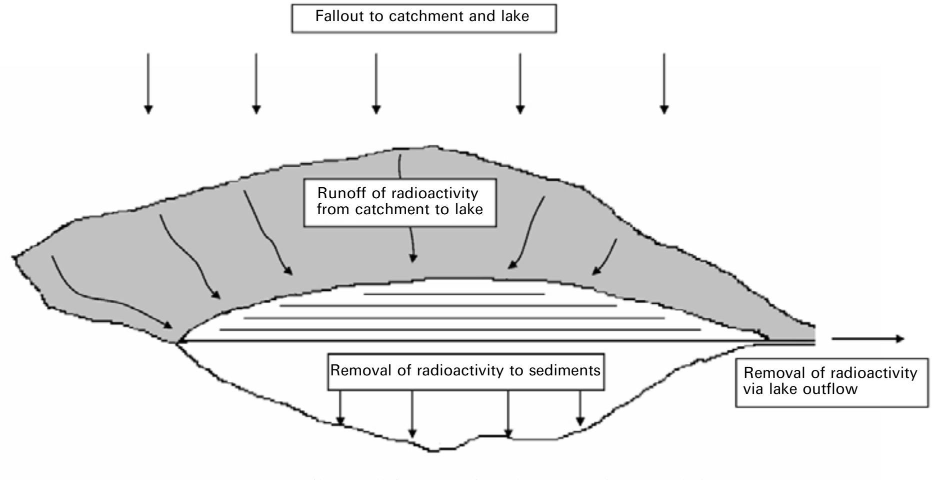


Figure 4.6. Radionuclide transfers in a catchment–lake system.

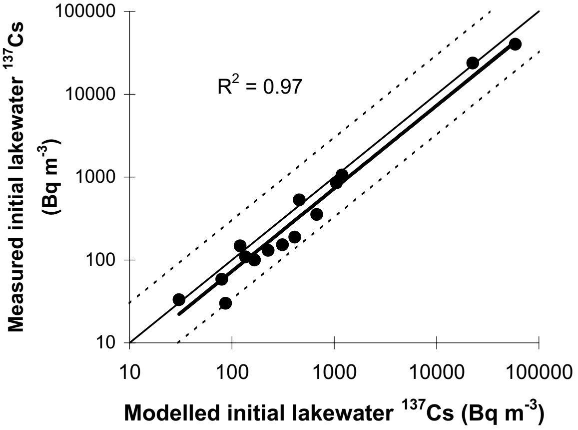


Figure 4.7. Comparison of initial ^{137}Cs activity concentration in 15 lakes determined from measurements with that estimated from a simple dilution model (Equation 4.3). Thick solid line shows best-fit regression line, thin solid line shows 1 : 1 relationship. Dotted lines show factor of 3 deviation from 1 : 1 relationship.

Estimates of initial ^{137}Cs in the water of a number of European lakes show that this simple dilution model (Equation 4.3) gives good estimates ($R^2 = 0.97$, $n = 15$, $p < 0.001$) of the initial average activity concentration, as shown in Figure 4.7 (Smith *et al.*, 1999b). In general, however, estimates made on the basis of Equation 4.3 are slightly higher than the measurements (most points are below the $x = y$ line). The mean ratio of predicted C_T (at time = 0) to values extrapolated from measurements is 1.35. This difference is partly due to the fact that most of the measurements presented in Figure 4.7 are of radiocaesium in the dissolved phase only, whereas the model predicts the concentration in both phases. There may also be underestimation of initial measured values since most measurements were begun a few days after fallout and estimates by extrapolation to time zero may be slightly low. In spite of these differences, it is clear that initial radiocaesium activity concentrations in these lakes were determined primarily by dilution of radioactivity directly deposited on the lake surface.

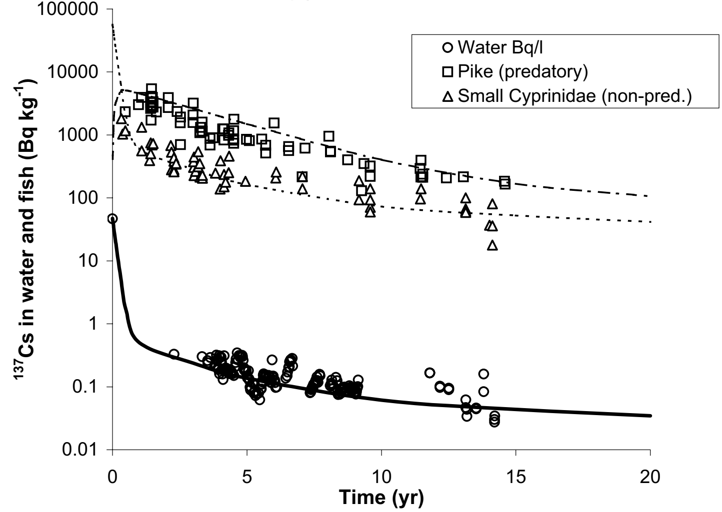
It should be noted that Equation 4.3 predicts average activity concentrations in the whole lake. Activity concentrations in the surface waters of lakes are likely to be higher than the average concentration in the lake during the first few weeks after fallout. In addition, if the lake is stratified, radionuclides may initially be rapidly mixed in the upper layer of water, taking some time to mix throughout the whole lake (Santschi *et al.*, 1990; Davison *et al.*, 1993).

4.3.1 Initial removal of radionuclides from the lake water

Following deposition of radionuclides onto the lake surface, the concentration in lake water declines approximately exponentially (Figure 4.8):

$$C = C(0)\exp(-Kt) \tag{4.4}$$

(a) Lake Vorsee



(b) Lake Constance

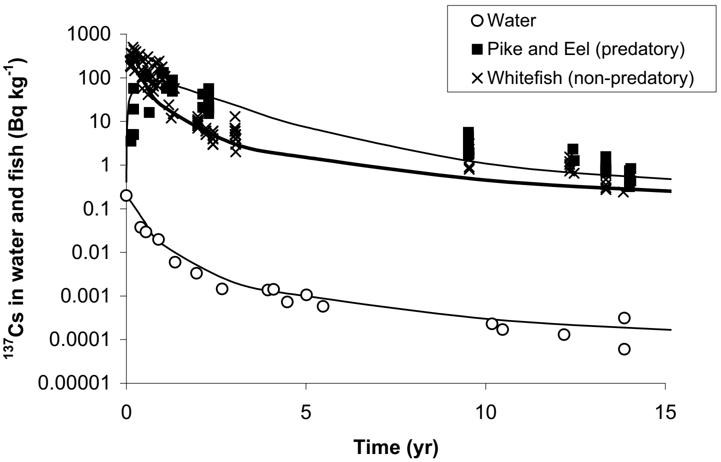


Figure 4.8. Change in the ^{137}Cs activity concentration in water and fish of: (a) a small shallow lake in Germany, Lake Vorsee and; (b) the large, deep Lake Constance (Bodensee).
Adapted from Klemt *et al.* (1998) and Zibold *et al.* (2002) using data kindly supplied by Gregor Zibold, Fachhochschule Weingarten.

where inputs of radionuclides to the lake from the catchment have been ignored since they are rarely significant in this early period. The rate of decline K (d^{-1}) in radionuclide concentration in water, termed the 'self-cleaning' capacity of a lake (Santschi *et al.*, 1990) is determined by losses through the lake outflow, transfers of radioactivity to the bed sediments and physical decay with rate constant λ (d^{-1}). The removal rate K is given by (see, e.g., Smith *et al.*, 1999b):

$$K = \frac{1}{T_w} + \frac{1}{T_s} + \lambda \tag{4.5}$$

where T_w (d), the water residence or 'turnover' time of the lake, is defined as the ratio of the lake volume to the rate of water discharge through the outflow:

$$T_w = \frac{V_L}{q_o} \tag{4.6}$$

and T_s is the transfer time of ^{137}Cs to bottom sediments. Rates of removal of radioactivity from the lake water typically vary in the range 4–25 $\times$ $10^{-3}\,\mathrm{d}^{-1}$ (i.e., T_{eff} ($\ln 2/K$) = 28–170 days (Santschi *et al.*, 1990; Smith *et al.*, 1999b)).

The process which predominantly determines pollutant transfers to sediments is still open to question. Most models assume that removal to bed sediments occurs primarily by absorption of the radionuclide to suspended particulate matter which subsequently falls to the lake bed. But some workers (Santschi *et al.*, 1986; Hesslein, 1987) have shown that direct diffusion across the sediment–water interface may also be important for some radionuclides. Direct diffusion to bottom sediments is, however, ignored here since correct modelling of this process is complex (Smith and Comans, 1996). In addition, a study (Smith *et al.*, 1999b) suggests that it is of minor importance for radiocaesium transfers to sediments.

The rate of transfer of radioactivity to bottom sediments by settling of suspended particles is given by:

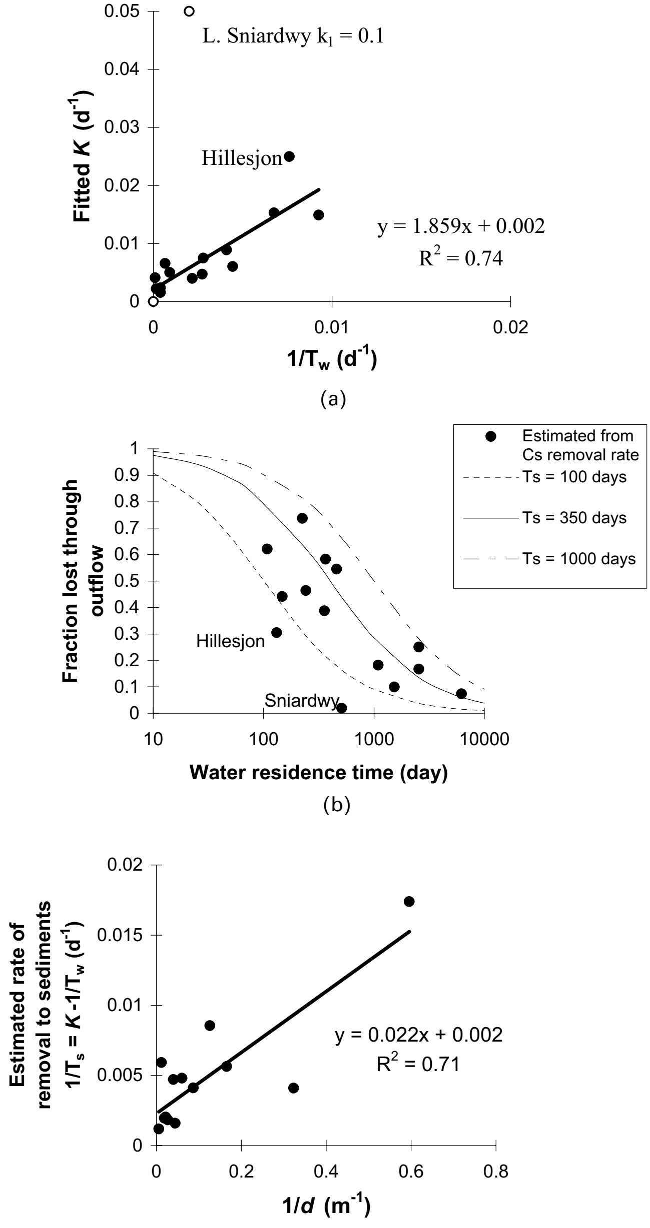
$$\frac{1}{T_s} = \frac{f_p v_p}{d} \tag{4.7}$$

where v_p ($\mathrm{m\,d^{-1}}$) is the mean settling velocity of suspended particles, d is the lake mean depth, and f_p is the fraction of activity absorbed to suspended particles, defined in Equation 4.1.

The importance of different environmental processes to the transport of radionuclides in lakes was illustrated by a study of radiocaesium removal rates in 14 different European lakes following Chernobyl (Smith *et al.*, 1999b). For each of these lakes, the rate of removal of radiocaesium from the lake water (K, units: d^{-1}) was estimated using time series measurements of ^{137}Cs in the lake water. This removal rate was then correlated with different lake characteristics to determine their influence on radiocaesium removal.

4.3.2 The influence of lake water residence time

The influence of water outflow on radiocaesium removal from the lake is demonstrated in a plot of the radiocaesium removal rate K (derived from field measurements) against the inverse of the lake water residence time ($1/T_w$), as shown in Figure 4.9(a) ($R^2 = 0.74$, $n = 14$, $p < 0.005$). The graph illustrates that, although there is a strong inverse correlation between removal rates and water residence times, the rate of ^{137}Cs removal is significantly higher than the rate of loss of



(a)

(b)

(c)

Figure 4.9. (a) The relationship between ^{137}Cs removal rate from 14 lakes and the removal rate of water through the outflow (the open circle shows an outlier, Lake Sniardwy, see text). (b) The relationship between the fraction of the total ^{137}Cs transferred to the outflow and the lake water residence time. (c) The relationship between ^{137}Cs removal rate (from lake water to sediments) and the lake mean depth.

Data is from a literature review carried out by Smith *et al.* (1999b).

water through the outflow (given by the inverse of the water residence time). Thus, as expected, lake water turnover ('flushing') alone is not sufficient to explain observed rates of ^{137}Cs removal, so transfers of activity to bottom sediments must be important.

It is useful to estimate the fraction f_o of the total activity deposited on the lake surface which was eventually lost via the outflow (and, therefore, the fraction $f_s = 1 - f_o$ of activity transported to the bottom sediments). This fraction can be estimated from the measured rate of removal to sediments ($1/T_s = K(\text{observed}) - 1/T_w$):

$$f_o = \frac{T_s}{T_s + T_w} \tag{4.8}$$

The fraction of activity lost through the outflow $f_o(= 1 - f_s)$ is shown as a function of T_w in Figure 4.9(b). The graph shows, unsurprisingly, that more radioactivity is lost through the outflow in lakes with shorter water residence time. Lakes which show unusually low losses through the outflow and hence unusually high transfers to sediments (Lakes Hillesjøn: Brittain *et al.*, 1997; and Sniardwy: Robbins and Jasinski, 1995) are relatively shallow.

4.3.3 The influence of lake mean depth *d*

The influence of lake mean depth *d* on radiocaesium removal to sediments is demonstated by a plot of the rate of removal of activity to sediments ($1/T_s = K - 1/T_\mathrm{w} - \lambda$, Equation 4.5) vs. $1/d$ (Figure 4.9(c)), showing removal rates in inverse proportion to the lake mean depth ($R^2 = 0.71$, $n = 14$, $p < 0.001$). Removal rates of radiocaesium to sediments are therefore greatest in shallow lakes, as implied by Equation 4.7.

4.3.4 The influence of sediment–water distribution coefficient K_d

Using measured values of T_w, s and v_p, Smith *et al.* (1999b) estimated the K_d value required to produce the observed radiocaesium removal rate K for different lakes. In Table 4.6, we compare these K_d values estimated from the ^{137}Cs removal rates with measured K_d values in six lakes. As shown in Table 4.6, the measured K_d values tend to be higher (but are of the same order as) than those required to give the observed rates of radiocaesium removal to sediments. Radiocaesium distribution coefficient (K_d) measurements in freshwaters are known to vary widely as a result of experimental error, environmental variability (water chemistry, clay content of particulate matter) and 'fixation' of ^{137}Cs over time (Comans and Hockley, 1992). The use of the K_d to represent the many processes that govern the partitioning of ^{137}Cs inevitably leads to discrepancies between model parameters (i.e., the K_d used in the model) and field or laboratory measurements (the measured K_d). However, the comparison in Table 4.6 indicates that the sorption of radiocaesium to solids is sufficiently strong (i.e., K_d is sufficiently high) to explain the rates of radiocaesium removal to sediments observed in these lakes.

Table 4.6. Comparison of radiocaesium K_d determined from removal rate measurements (assuming the particulate settling model) with K_d measured in the field or laboratory.

Lake	K_d required for observed removal (1/kg)	Measured K_d (1/kg)	Source and notes
Constance	9.0×10^4	2.0×10^4	(1) Laboratory measurement
Sniardwy	0.6×10^4	1.5×10^4	(2) Laboratory measurement
Zurich	4.3×10^4	4.5×10^4	(3) *In situ* measurement
Devoke	4.3×10^4	6.0×10^4	(4) *In situ* measurement
Esthwaite	1.5×10^4	1.4×10^5	(4) *In situ* measurement
Windermere	3.3×10^4	1.3×10^5	(4) *In situ* measurement

[1] Robbins *et al.* (1992); [2] Robbins and Jasinski (1995); [3] Santschi *et al.* (1990); [4] Smith *et al.* (1997).

4.3.5 Transport of ^{90}Sr in lakes

The water–sediment distribution coefficient (K_d) of radiostrontium is relatively low so the fraction absorbed to particulates f_p is expected to be less than 5% (see Section 4.1), assuming a suspended matter concentration $s < 50\,\mathrm{mg\,l^{-1}}$. The low affinity of ^{90}Sr for particles compared to radiocaesium suggests that transfers to sediments will be relatively low in comparison with removal through the outflow in most lakes. This is illustrated by a comparison of removal of ^{137}Cs and ^{90}Sr in the Dnieper River–Reservoir system. The different affinities of these radionuclides for suspended matter influenced their transport through the 6 reservoirs of the system (Voitsekhovitch, 2001). Caesium-137 tends to become fixed onto clay sediments which are deposited in the deep sediments of the reservoirs, particularly in the Kiev Reservoir. Because of this process, very little ^{137}Cs flows through the cascade of reservoirs, the majority being trapped in the reservior sediments. On the other hand, although ^{90}Sr concentration decreases with distance from the source (mainly due to dilution by water inflowing from less contaminated areas), about 40–60% passes through the cascade and reaches the Black Sea. Figure 4.10 shows the trend in average annual ^{90}Sr and ^{137}Cs concentrations in the Dnieper Reservoirs since the accident. As shown in Figure 4.10, ^{137}Cs is trapped by sediments in the reservoir system, so activity concentrations in the lower part of the system are orders of magnitude lower than in the Kiev Reservoir. ^{90}Sr is not strongly bound by sediments, so concentrations in the lower part of the river–reservoir system are much closer to those measured in the Kiev Reservoir.

The peaks in ^{90}Sr activity concentration in the reservoirs of the Dnieper cascade (Figure 4.10) were caused by flooding of the most contaminated floodplains in the Chernobyl exclusion zone. For example, concentrations of ^{90}Sr in the river water increased from around $1\,\mathrm{Bq\,l^{-1}}$ to approximately $8\,\mathrm{Bq\,l^{-1}}$ for a 5–10-day period during the winter of 1991 (Vakulovsky *et al.*, 1994) as a result of flooding due to blockages of the river by ice. Caesium-137 activity concentrations were unaffected.

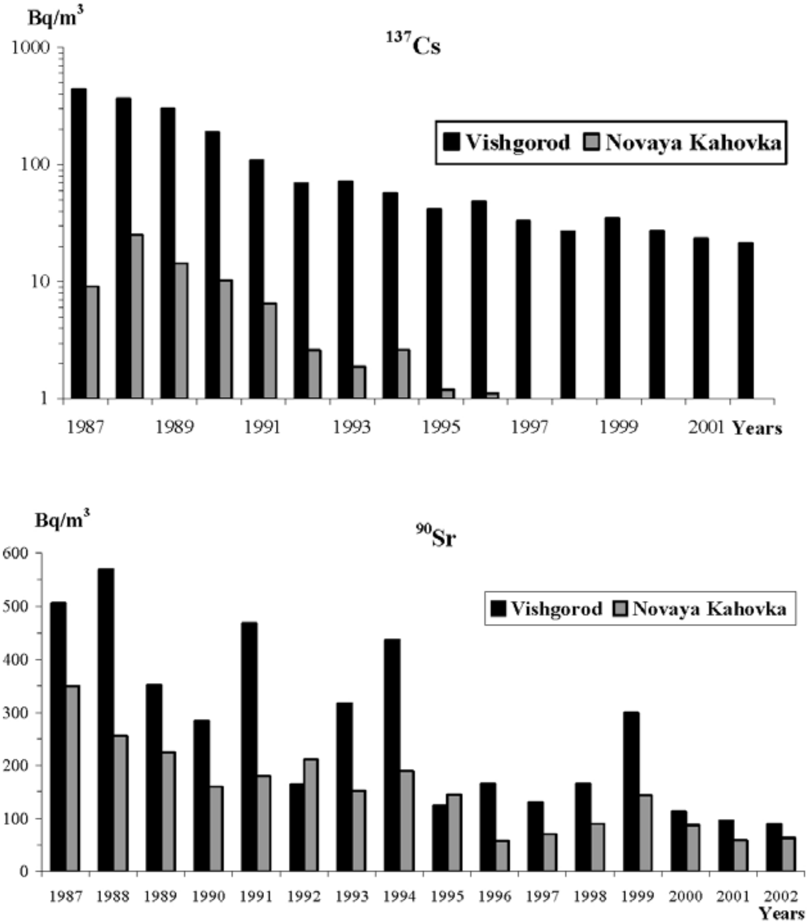


Figure 4.10. Changes in average annual content of ^{137}Cs and ^{90}Sr in the water of the first (Vishgorod, Kiev Reservoir) and last (Novaya Kahovka, Kahovka Reservoir) reservoirs of the Dnieper cascade.
From Voitsekhovitch (2001); Voitsekhovitch and Smith (2005).

Similar flood events took place during the winter flood of 1994, during summer rainfall in July 1993 and during the high spring flood in 1999.

4.3.6 Transport of ^{131}I in lakes

The rapid physical decay rate of ^{131}I (physical half-life, 8.02 days) controls its rate of removal from lake waters since, relative to physical decay, rates of transfers to sediments are low. In tracer studies in experimental lakes, it has been shown that, although ^{131}I can associate with large organic molecules in solution, the transfer to bed sediments was at a rate of only 0.018–0.033 d^{-1} (Milton *et al.*, 1992). This is significantly lower than the ^{131}I physical decay rate of 0.086 d^{-1}. The importance of physical decay in determining ^{131}I removal from lakes is confirmed by measurements after Chernobyl. In the Kiev Reservoir, ^{131}I, with an initial concentration of 500 Bq l^{-1} declined to 20 Bq l^{-1} an estimated 37 days after the accident giving a rate of decline (K) of 0.087 d^{-1} (calculated from data in Kryshev, 1995). This rate of decline is close to the physical decay rate, suggesting negligible removal of radioactivity to the sediments during this period. Studies of transfers of radionuclides to bed sediments in the Chernobyl Cooling Pond showed that only 11% of the total inventory of ^{131}I was found in bed sediments one month after the accident (Kryshev, 1995; see also Table 4.9, p. 165).

4.3.7 Transport of ruthenium in lakes

There are relatively few data concerning ^{103,106}Ru in lakes following Chernobyl. In two lakes in the English Lake District (Windermere and Esthwaite Water), it was found that the ratio of ruthenium isotopes to ^{137}Cs in sediments during 1986 was not significantly different to the Ru : ^{137}Cs ratios in fallout (Hilton *et al.*, 1994). These workers concluded that in these lakes the rate of Ru transfer to sediments was similar to that of ^{137}Cs. This conclusion was supported by similar reported distribution coefficients (K_ds) of the two radionuclides (Hilton *et al.*, 1994). In studies on Lake Constance (Bodensee), however, ruthenium was removed from the water column two times faster than radiocaesium (Mangini *et al.*, 1990). The K_d of ruthenium in this lake was of order 10^5 l kg^{-1}, significantly higher than the 2.0×10^4 l kg^{-1} measured for radiocaesium (Robbins *et al.*, 1992).

4.3.8 Radionuclide balance in water of open lakes

Once the radioactivity which had directly deposited to the lake surface had been removed to the bed sediments or the outflow, the catchment (and potentially sediments) were a long-term source of radionuclides to the lake. In lakes with a relatively rapid inflow and outflow of water, it is postulated that average long-term radioactivity concentrations were primarily controlled by inflows in runoff water from the catchment. Under this assumption, in the long term (months/years) after fallout, the bed sediments would not (on average) act as a major source or sink of radionuclides. Therefore, average activity concentrations in the lake waters and outflow would be similar to average activity concentrations in inflowing streams.

Table 4.7 summarises measurements of the ^{137}Cs and ^{90}Sr balance in a number of European lakes. There are clearly variations in the ratio of activity concentrations in inflows compared to lake or outflow waters. A ratio greater than 1 implies a net loss of radioactivity to sediments, a ratio of less than 1 implies a net remobilisation of radioactivity from sediments. In Lake Hillesjøn in Sweden, for example, there appears to have been a significant remobilisation of ^{137}Cs from the sediments, at least during the spring period studied. In contrast, the Kiev Reservoir sediments act

Table 4.7. Mean ^{137}Cs and ^{90}Sr activity concentration (in dissolved and particulate phases) in inflow streams compared with concentrations in the lake water/outlet of different lakes.

Lake	Mean inlet conc. (Bq m^{-3})		Mean lake or outlet conc. (Bq m^{-3})		Ratio inlet/(lake or outlet)		Sampling date
	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	
Brotherswater[1]	1.22	–	1.44	–	0.85	–	Spring/summer 1992
Devoke Water[1]	70.3	–	81.0	–	0.87	–	Spring/summer 1992
Loweswater[1]	12.0	–	12.2	–	0.98	–	Spring/summer 1992
Vorsee[2]	100*	–	151	–	0.66	–	Mean 1990–1994
Hillesjøn[3]	39.9	13.7	522	20.8	0.08	0.66	Spring 1991
Saarisjärvi[3]	372	4.4	215	3.8	1.73	1.15	Spring 1991
Øvre Heimdalsvatn[3]	251	13	113	9	2.22	1.44	Spring 1991
Örtrsket[4]	–	–	–	–	1.13	–	Total input 1987–1991
Kiev Reservoir[5]	369	476	225	384	1.64	1.24	Mean 1987–1993

* Approximate average of northern and southern inflows weighted by flow rate (G. Zibold, Fachhochschule Weingarten, pers. commun.).

[1] Smith *et al.* (1997); [2] G. Zibold, Fachhochschule Weingarten (pers. commun.); [3] Brittain *et al.* (1997); [4] Malmgren and Jansson (1995); [5] Sansone and Voitsekhovitch (1996).

as a sink for ^{137}Cs and, to a much lesser extent, ^{90}Sr (Voitsekhovitch, 2001), as discussed above.

Some of the variation in the ratios presented in Table 4.7 is probably due to sampling errors. For example, activity concentrations in inflow waters in particular may vary significantly, so averages of a few samples over a relatively short period of time (as is the case in some of the studies presented in Table 4.7) may be inaccurate. In spite of these problems, it can be concluded that there is little evidence of systematic differences in inlet compared to lake water/outflow activity concentrations across a number of lakes. This observation supports the hypothesis that, in the long term in most lakes, net transfers of radioactivity to and from the sediments do not have a major influence on activity concentrations in the lake water. Thus, activity concentrations in the lake and outlet are (on average) similar to those in inflowing water in the long term after fallout.

Because of the dominance of inputs of radioactivity from the catchment, long-term declines in radionuclide concentrations in open lakes are similar to those in rivers (Table 4.5). Thus, T_{eff} values for 9 open lakes were in the range 1–4 years during the period 1987–1992, as seen for rivers in Table 4.5 (Smith *et al.*, 1999a).

4.3.9 Closed lake systems

In some lakes, where there is no (or only minor) surface inflow and outflow of water, the bed sediments play a major role in controlling radionuclide activity concentration in the water. Such lakes have been termed 'closed' lakes (Vakulovsky *et al.*, 1994; Bulgakov *et al.*, 2002) and, as shown in Table 4.8, have relatively much higher

Table 4.8. Normalised water concentrations (R_c, m^{-1}) of ^{137}Cs and ^{90}Sr in various water bodies 4–10 years after fallout.

Water body	Type	Date	R_c (m^{-1})
^{137}Cs after Chernobyl			
Iso Valkjärvi, Finland[1]	Closed lake	1990	26×10^{-3}
Lake Svyatoe, Bryansk, Russia[2]	Closed lake	1993–1994	19×10^{-3}
Lake Kozhanovskoe, Bryansk, Russia[2]	Closed lake	1993–1994	14×10^{-3}
Devoke Water, UK[3]	Open lake	1990–1995	2.2×10^{-3}
Ennerdale Water, UK[3]	Open lake	1990–1995	0.5×10^{-3}
Lake Constance, Switzerland[4]	Open lake	1990–1992	0.08×10^{-3}
Stream draining Opromokh peat bog, Belarus[5]	River	1993–1995	7.9×10^{-3}
River Sozh, Belarus (mineral catchment)[6]	River	1990–1995	0.4×10^{-3}
Range in 13 European rivers[7]	River	1990–1995	$0.15–5.0 \times 10^{-3}$
Near-surface groundwater, Belarus (24 sites)[8]	Groundwater	1991–1997	$0.03–0.7 \times 10^{-3}$
^{90}Sr after various fallout incidents			
Lake Svyatoe, Bryansk, Russia after Chernobyl[2]	Closed lake	1993–1994	14×10^{-3}
Lake Kozhanovskoe, Bryansk, after Chernobyl[2]	Closed lake	1993–1994	70×10^{-3}
Lake Uruskul, Siberia, Kyshtym accident 1957[9]	Closed lake	1962–1964	55×10^{-3}
Haweswater Reservoir, UK after NWT[10]	Open lake	1967–1971	13×10^{-3}
Range in 19 European rivers after NWT[7]	River	1968	$2.1–31 \times 10^{-3}$
Two rivers in Finland after Chernobyl[11] *	River	1990–1994	$30–40 \times 10^{-3}$

[1] IAEA (2000); [2] Sansone and Voitsekhovitch (1996); [3] Smith *et al.* (1997); [4] Zibold *et al.* (2002); [5] Kudelsky *et al.* (1996); [6] Kudelsky *et al.* (1998); [7] Smith *et al.* (2004); [8] Kudelsky *et al.* (2004); [9] Monte *et al.* (2002); [10] Linsley *et al.* (1982); [11] Cross *et al.* (2002). * These two rivers were observed to have high ^{90}Sr runoff compared to other European rivers after NWT, so these normalised concentrations for Chernobyl fallout are likely to be higher than in the majority of rivers.

^{137}Cs and ^{90}Sr activity concentrations (in the long term) than most rivers and open lake systems. There are a number of such lakes in the areas of the fSU most affected by Chernobyl, and fish from these lakes played an important part in radiocaesium intakes by some rural populations living nearby.

The most highly contaminated water bodies in the Chernobyl affected areas are the closed lakes of the Pripyat flood plain within the 30-km exclusion zone. There are many such lakes in the areas surrounding Chernobyl. During 1991, ^{137}Cs levels in these lakes were up to $74\,\mathrm{Bq\,l^{-1}}$ in Glubokoye Lake and ^{90}Sr activity concentrations were between 100 and $370\,\mathrm{Bq\,l^{-1}}$ in 6 of 17 studied water bodies (Vakulovsky *et al.*, 1994).

As with other surface water systems, radionuclide activity concentrations in closed lakes tended to decline over time after fallout. In the early period, contamination declined only as a result of transfers to bed sediments since there are negligible losses through the lake outflow in these systems. In Lake Iso Valkjärvi in Finland, the rate of ^{137}Cs transfer to sediments K was 4.1×10^{-3} d^{-1} ($T_{eff} = 169$ days) after Chernobyl (Smith *et al.*, 1999b) which is of the same order as rates observed in tracer experiments in Canadian Experimental Lakes of $K = 1/T_s = 14.5 \times 10^{-3}$ (Lake 226NE) and 8.4×10^{-3} (Lake 226SW) d^{-1} ($T_{eff} = 48$ and 83 days respectively, calculated from data in Hesslein, 1987). In the same study, the ^{89}Sr removal rate due to transfers to sediments was $K = 1/Ts = 10 \times 10^{-3}$ (L226NE) and 2.0×10^{-3} (L226SW) d^{-1} ($T_{eff} = 70$ and 347 days respectively). These removal rates are slower than those typically observed for open lakes of similar size because, by definition, there is no significant outflow of radioactivity in closed lake systems.

Over longer time periods after fallout, the activity concentration in closed lakes continues to decline at a slow rate as more radioactivity becomes incorporated in sediments and due to (slow) losses of water from the system. In Lake Svyatoe, Russia, it was observed that between 1993 and 1999 ^{137}Cs activity concentrations declined with an effective ecological half-life of 6.9 years (Bulgakov *et al.*, 2002). In contrast, between 1993 and 1998, there was no significant decline in ^{137}Cs in the water of Lake Kozhanovskoe (A.V. Konoplev, A.A. Bulgakov, SPA Typhoon, unpubl. res.).

4.4 RADIONUCLIDES IN SEDIMENTS

Bed sediments are an important long term sink for radionuclides. In the Chernobyl Cooling Pond, approximately one month after the accident, most of the radioactivity was found in bed sediments (Table 4.9). In this area (i.e., within 10 km of the power plant), the majority of radionuclides were associated with hot particles (see Chapter 2), so the rapid transfer to bed sediments was largely due to sedimentation of these dense particles. This is illustrated by the contrasting behaviour of ^{90}Sr (89% in bed sediments) and ^{131}I (11% in bed sediments). Both isotopes have relatively low affinity for sediments when deposited in dissolved form. However, the majority of ^{90}Sr was deposited as fuel particles, whereas the volatile ^{131}I was primarily discharged as a vapour.

In the Cooling Pond, at present most radioactivity is found in the fine sediments in deeper areas (Table 4.10); sandy sediments along the shoreline have much lower radionuclide activity concentrations (Voitsekhovitch *et al.*, 2002).

In the long term, approximately 99% of the radiocaesium in a lake is typically found in the bed sediment. From measurements in Lake Svyatoe (Kostiukovichy, Belarus), during 1997, it was estimated that there was 3×10^9 Bq in water and approximately 2.5×10^{11} Bq in sediments (A.V. Kudelsky, unpubl. res.). In Lake Kozhanovskoe, Russia, approximately 90% of the radiostrontium was found in the bed sediments during 1993–1994 (estimated from measurements of ^{90}Sr in water and sediment presented in Sansone and Voitsekhovitch, 1996).

Table 4.9. Radionuclides in Chernobyl Cooling Pond bed sediments approximately one month after the accident, expressed as a percentage of the total amount in both sediments and water.

From data in Kryshev (1995).

Date	^{90}Sr	^{95}Zr	^{95}Nb	^{103}Ru	^{106}Ru	^{131}I	^{134}Cs	^{137}Cs	^{140}Ba	^{140}La	^{141}Ce	^{144}Ce
30/5/86	89	96	94	95	92	11	67	65	77	78	93	97

Table 4.10. Typical radionuclide activity concentrations in the most contaminated silty sediments of the Cooling Pond (from data in Voitsekhovitch *et al.*, 2002).

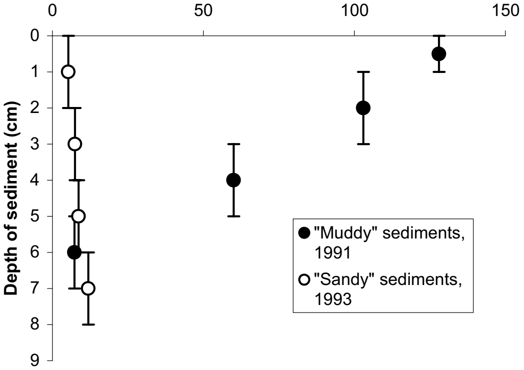
RN	Activity concentration in sediment (kBq kg^{-1})
Caesium-137	600
Strontium-90	110
Americium-241	5
Plutonium-239/240	2.4
Plutonium-238	1.2

Radiocaesium is relatively immobile in lake sediments, though some dispersion may occur by physical mixing of the sediments (by biota and water currents) or diffusion in the sediment pore waters. Studies into the sorption of Chernobyl radiocaesium to sediments have shown that the solid–aqueous distribution coefficient (K_d) is inversely proportional to the content of competing ions (specifically K^+ and NH_4^+) in the sediment pore waters (Comans *et al.*, 1989). In stratified lakes, NH_4^+ in anoxic sediments and bottom waters can lead to remobilisation of radiocaesium from contaminated sediments (Evans *et al.*, 1983), although it has been estimated that less than 3% of the sediment inventory could be remobilised per year by this mechanism (Smith and Comans, 1996).

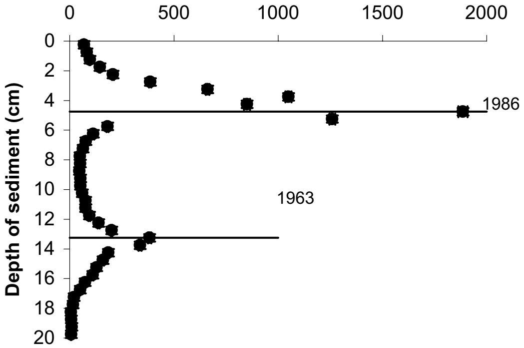
The sediment composition also plays an important role in determining radiocaesium mobility. Figure 4.11 shows examples of ^{137}Cs activity–depth profiles in different sediments of marine and freshwater systems. In the Baltic Sea (Figure 4.11(a)), ^{137}Cs deposition was associated mainly with fine sediments, and dispersion in the sediment is relatively low in the muddy (fine) sediment, as indicated by a sharp decline in activity concentration with depth from the sediment surface. In Lake Constance, where bed sediments are relatively rich in illite clay minerals, clear peaks resulting from NWT and Chernobyl depositions are observed in the ^{137}Cs activity–depth profiles. The immobility of radiocaesium in many lake sediments means that the depth of the peaks representing the maxima of Chernobyl (1986) and NWT (1963) depositions (illustrated in Figure 4.11(b)) can be used to estimate the rate of sediment accumulation in lakes (e.g., Appleby, 1997).

The sediment profile from Lake Svyatoe (Kostiukovichy, Belarus, Figure 4.11(c)), in contrast to that from Lake Constance, shows a maximum ^{137}Cs concentration near the sediment surface, indicating that there has been little or no net sediment accumulation in this lake during the 14 years between fallout and the sampling date (16 February, 2000). The Lake Svyatoe activity–depth profile is also much more disperse (i.e., the change in activity concentration with depth is much less steep) than that from Lake Constance. This may be attributed to physical mixing of the sediments of this shallow lake, and/or greater molecular diffusion of radiocaesium since the sediments are high in organic matter and sand content and hence do not strongly 'fix' radiocaesium (Ovsiannikova *et al.*, 2004). We would

(a) Chernobyl^{137}Cs in Baltic Sea sediments (Bq kg^{-1})



(b) Chernobyl and NWT ^{137}Cs in Lake Constance sediment, 1999 (Bq kg^{-1})



(c) Chernobyl ^{137}Cs in Lake Svyatoe (Belarus) sediment, year 2000 (kBq kg^{-1})

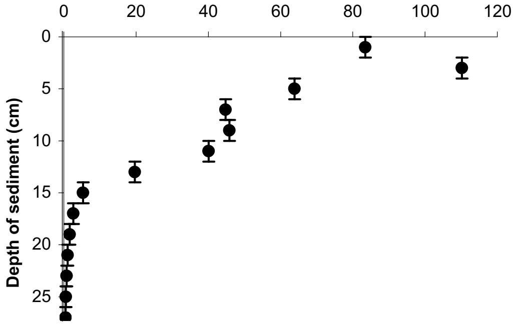


Figure 4.11. Graphs of ^{137}Cs activity–depth profiles in sediments in (a) Baltic Sea, muddy and sandy sediments; (b) Lake Constance; (c) Lake Svyatoe, Kostiukovichy, Belarus.

(a) Knapinska-Skiba *et al.* (2001). (b) G. Zibold and E. Klemt, Fachhochschule Weingarten (pers. commun.). (c) A.V Kudelsky, unpubl. res.

not expect to see any contribution from NWT in the Lake Svyatoe sediments as it is obscured by the much higher Chernobyl fallout.

Radionuclides which were deposited in the form of fuel particles are generally less mobile than those deposited in dissolved form since transport only occurs by physical mixing and accumulation of the sediment and not by molecular diffusion. In sediments of Glubokoye Lake (in the 30-km zone) in 1993, most fuel particles remained in the surface 5 cm of sediment (Sansone and Voitsekhovitch, 1996). It has also been observed (see Chapter 2) that fuel particle breakdown was at a much lower rate in lake sediments compared to soils.

4.5 UPTAKE OF RADIONUCLIDES TO AQUATIC BIOTA

4.5.1 ^{137}Cs in freshwater fish

There have been many studies of the levels of radiocaesium contamination of freshwater fish during the years after the Chernobyl accident. As a result of high radiocaesium bioaccumulation factors, fish have remained contaminated despite relatively low radiocaesium levels in water. In some cases, activity concentrations in fish have greatly exceeded the European Union intervention level for radiocaesium activity in fish of 1,250 Bq kg^{-1} wet weight (w.w.).

In the Chernobyl Cooling Pond, ^{137}Cs levels in carp (*Cyprinus carpio*), silver bream (*Blicca bjoerkna*), perch (*Perca fluviatilis*) and pike (*Esox lucius*) were of the order of 100 kBq kg^{-1} w.w. in 1986, declining to a few tens of kBq kg^{-1} in 1990 (Kryshev and Ryabov, 1990; Kryshev, 1995). In the Kiev Reservoir, activity concentrations in fish were in the range 0.6–1.6 kBq kg^{-1} w.w. (in 1987) and 0.2–0.8 kBq kg^{-1} w.w. (from 1990–1995) for adult non-predatory fish and 1–7 kBq kg^{-1} (in 1987) and 0.2–1.2 kBq kg^{-1} (from 1990–1995) for predatory fish species. In lakes in the Bryansk region of Russia, activity concentrations in a number of fish species varied within the range 0.215–18.9 kBq.kg^{-1} w.w. during the period 1990–1992 (Fleishman *et al.*, 1994; Sansone and Voitsekhovitch, 1996). It was estimated that about 14,000 lakes in Sweden had fish with ^{137}Cs concentrations above 1,500 Bq kg^{-1} (the Swedish guideline value) in 1987 (Håkanson *et al.*, 1992). In a small lake in Germany, levels in pike were up to 5 kBq kg^{-1} shortly after the Chernobyl accident (Klemt *et al.*, 1998). In Devoke Water in the English Lake District, perch and brown trout (*Salmo trutta*) contained around 1 kBq kg^{-1} in 1988 declining slowly to a few hundreds of Bequerels per kg in 1993 (Camplin *et al.*, 1989; Smith *et al.*, 2000b).

The contamination of fish following the Chernobyl accident was a cause for concern in the short term (months) for less contaminated areas (e.g., parts of the UK and Germany) and in the long term (years-decades) in the Chernobyl affected areas of the Ukraine, Belarus, Russia and parts of Scandinavia.

It is known that the bioaccumulation of radioactivity in fish is determined by numerous ecological and environmental factors such as the trophic level of the fish species, the length of the food chain, water temperature and the water chemistry.

Uptake may be via ingestion of contaminated food or direct transfers from the water via the gills. For many radionuclides, including radiocaesium, the food chain is the primary uptake pathway, so a food uptake model is usually used to estimate uptake rates. Since fish feeding rate is strongly influenced by temperature (Elliot, 1975), the uptake rate of radionuclides absorbed through food tends to be faster at higher water temperatures.

The level of radioactive contamination of aquatic biota is commonly defined in terms of a concentration factor (CF) where

$$\mathrm{CF} = \frac{\text{Activity concentration per kg of fish (wet weight)}}{\text{Activity concentration per litre of water}}\ \mathrm{l\,kg^{-1}} \tag{4.9}$$

Previous studies on the accumulation of radiocaesium in fish have focused on the prediction of CF (sometimes termed the bioaccumulation factor (BAF) or aggregated concentration factor (ACF)). The equilibrium CF modelling approach is appropriate for cases in which the radionuclide activity concentration in fish can be assumed to be in equilibrium with that of water, for example at long times (years) after radionuclide fallout, or for continuous releases of radionuclides to a river. For short-term releases of radionuclides to an aquatic system, however, dynamic models may be required.

The activity concentration of a radionuclide in fish C_f ($\mathrm{Bq\,kg^{-1}}$) is often modelled by a simple 'two-box' model describing uptake from the water C_w ($\mathrm{Bq\,l^{-1}}$) and release from the fish (Figure 4.12):

$$\frac{dC_f}{dt} = k_f C_w - (k_b + \lambda)C_f \tag{4.10}$$

where k_f ($\mathrm{l\,kg^{-1}\,y^{-1}}$) is the rate constant describing transfers of ^{137}Cs to fish through its food and k_b ($\mathrm{y^{-1}}$) is the backward rate constant describing excretion of radioactivity from the fish. The ratio of these rate constants gives the equilibrium CF ($\mathrm{l\,kg^{-1}}$) of the radionuclide in fish relative to water:

$$\frac{k_f}{k_b + \lambda} = \frac{C_f}{C_w}\,\text{(at equilibrium)} = \mathrm{CF} \tag{4.11}$$

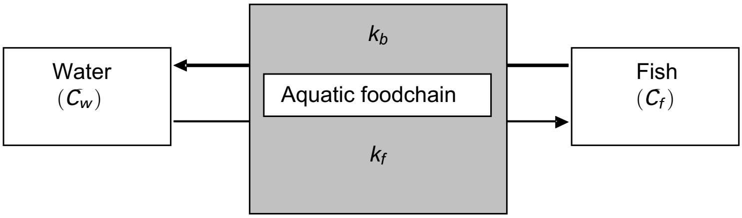


Figure 4.12. Illustration of a simple model for uptake in fish via the food chain.

4.5.2 Influence of trophic level on radiocaesium accumulation in fish

Maximum radiocaesium activity concentrations in non-predatory fish were observed within the first few months after the accident (Kryshev, 1995), indicating a rapid uptake. In predatory fish, however, there is a delay in the transfer of radiocaesium as it takes some time to accumulate up the food chain. In many systems, maximum ^{137}Cs activity concentrations in predatory fish were not observed until a period of between several months and one year after the accident (Brittain *et al.*, 1991; Elliot *et al.*, 1992; Kryshev, 1995). The dynamics of radiocaesium in water and fish are illustrated using data from Lake Vorsee and Lake Constance after Chernobyl (Figure 4.8; see Section 4.3). At long times (years) after fallout, the radionuclide uptake and excretion processes reached a steady state, so activity concentrations in fish changed at the same rate as those in water (Figure 4.8; Equation 4.11).

A comprehensive study of radiocaesium bioaccumulation in fish was carried out in Canadian aquatic systems contaminated by NWT fallout. In this study (Rowan *et al.*, 1998) it was shown that there was a clear 'biomagnification' effect in radiocaesium activity concentrations as trophic level increased. In the Ottawa River, different organisms were assigned different trophic positions according to their diet. Zooplankton and molluscs were assigned position 2.0, planktivorous fish (one level higher in the trophic chain) occupied position 3.0 and wholly piscivorous fish occupied position 4.0. Omnivorous species were assigned positions between 3.0 and 4.0 according to the composition of their diet. It was found (Rowan *et al.*, 1998) that there was an approximately four-fold difference in ^{137}Cs with each unit increase in trophic position (e.g., planktivorous fish had approximately four times as much ^{137}Cs (per kg) as zooplankton and molluscs). It was further observed in this study that, for a given trophic level, organisms with diets composed of benthic (bottom-dwelling) species had approximately 1.7 times greater ^{137}Cs activity concentration compared to organisms feeding from the open water food chain. This reflected the high accumulation of radiocaesium in bed sediments.

Similar observations have been made of radiocaesium accumulation in fish contaminated by the Chernobyl accident. In 1988, for example, radiocaesium concentration factors for predatory fish in the Chernobyl Cooling Pond and the Dnieper River were 4–5 times higher than for non-predatory fish (Kryshev and Sazykina, 1994).

4.5.3 Size and age effects on radiocaesium accumulation

The 'size effect' of radiocaesium accumulation in fish results in an increasing contamination (per unit weight of fish) with increasing fish size (Elliott *et al.*, 1992; Hadderingh *et al.*, 1997). In a study of ^{137}Cs in fish in the Pripyat River and the Kiev Reservoir during 1992, Hadderingh *et al.* (1997) found that CFs in non-predatory fish were generally independent of fish size. In contrast, predatory perch and pike showed an increase in ^{137}Cs concentration with increasing weight of fish (Figure 4.13). Increasing ^{137}Cs CFs with increasing fish size may be a result of environmental factors (e.g., different feeding habits, differing biological retention

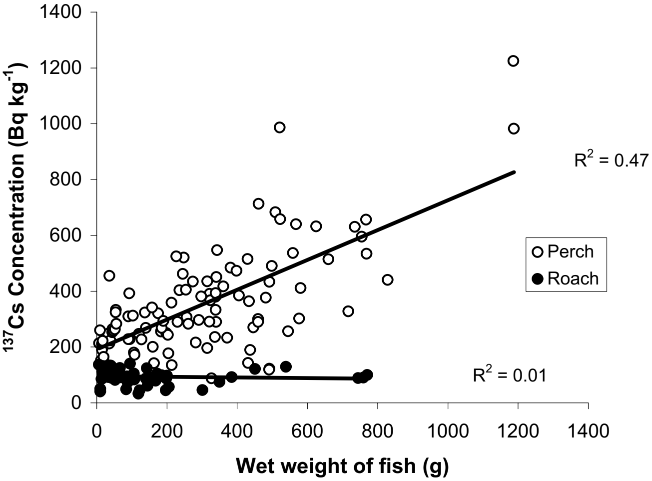


Figure 4.13. Radiocaesium in fish in the Kiev Reservoir after Chernobyl, illustrating the 'size effect' in predatory perch, but not in the non-predatory roach.
Adapted from Sansone and Voitsekhovitch (1996).

times in different sized fish), or the effect may be caused by non-equilibrium in the fish–water system. If radiocaesium concentrations in a water body are declining with time, then older, and hence larger, fish may retain relatively higher radiocaesium concentrations because they grew up in a scenario of higher water concentrations than younger fish.

The 'size effect' may also be caused by changes in fish diet as they grow. Ugedal *et al.* (1995) observed an increase in ^{137}Cs activity concentration in Arctic charr (*Salvelinus alpinus*) as the diet shifted from zooplankton in small fish to zoobenthos in larger fish. In perch, it was found that much of the 'size effect' could be explained by differences in feeding habits between small and large fish (Rowan *et al.*, 1998; Smith *et al.*, 2002). As perch mature, their diet changes from eating mainly invertebrates to being primarily piscivorous, though the size at which they do this varies depending on available food, competition and other environmental factors.

4.5.4 Influence of water chemistry on radiocaesium accumulation in fish

Because of its chemical similarity to potassium, radiocaesium can concentrate in organisms via the same accumulation mechanisms as potassium, an important nutrient. Potassium concentrations are regulated in organisms, so in waters with abundant potassium, bioaccumulation of potassium is weaker than in waters where it is scarce. Since caesium is bioaccumulated by organisms in the same way as potassium, caesium concentration factors tend also to be high in waters with low potassium. Studies on NWT derived ^{137}Cs found that the CF of radiocaesium in fish was inversely proportional to the potassium content of the surrounding water

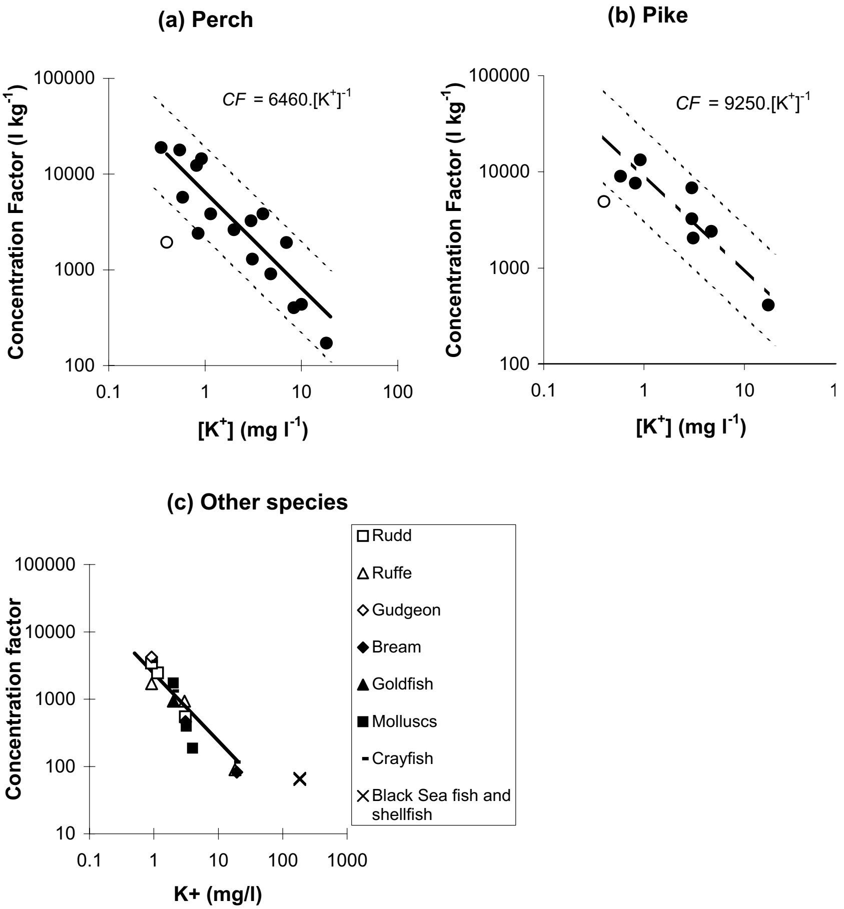


Figure 4.14. Relationship between ^{137}Cs concentration factor (w.w.) in fish and the potassium concentration in 17 lakes around Europe.* The open circles in (a) and (b) show data from the low pH Lake Iso Valkjärvi, see text. Notice that the measurement for the Black Sea (c)† does not follow the inverse trend observed for freshwaters (see Section 4.6.2).

* From Smith *et al.* (2000c, 2002). † From Baysal and Tunçer (1994; converted to w.w. basis).

(Kolehmainen, 1967; Fleishman, 1973; Blaylock, 1982; Rowan and Rasmussen, 1994). This relationship was also observed in fish contaminated after Chernobyl (Smith *et al.*, 2000c), as shown in Figure 4.14.

It is likely that other chemical conditions in water also affect radiocaesium uptake to fish, though the effect of potassium is dominant. In Lake Iso Valkjärvi in Finland, it was found that the radiocaesium CF was lower than expected for this low potassium concentration lake. This lake has low pH (high H$^+$) and low calcium concentration (IAEA, 1994). Interference in the potassium uptake mechanism (and therefore in caesium accumulation) has been hypothesised in conditions of low pH or low calcium concentration (Smith *et al.*, 2002).

4.5.5 ^{131}I in freshwater fish

Kryshev (1995) presented measurements of ^{131}I in fish in the Kiev Reservoir shortly after Chernobyl. Iodine-131 was rapidly absorbed, maximum concentrations in fish being observed at the beginning of May 1986. The fish–water CF was around $10\,\mathrm{l\,kg^{-1}}$, and activity concentrations in fish muscle declined from around $6{,}000\,\mathrm{Bq\,kg^{-1}}$ on 1 May, 1986 to $50\,\mathrm{Bq\,kg^{-1}}$ on 20 June, 1986. This represents a rate of decline similar to the rate of physical decay of ^{131}I ($T_{1/2} = 8.1$ days). As with other vertebrate species, there is a tendency for fish to concentrate iodine in the thyroid gland (Vinogradov, 1953).

4.5.6 ^{90}Sr in freshwater fish

Strontium behaves chemically and biologically in a similar way to calcium. Strontium is most strongly bioaccumulated in low calcium ('soft') waters (this is analogous to the relationship between caesium and potassium discussed above). There is less quantitative information available for uptake and retention rates of ^{90}Sr in fish than for ^{137}Cs, however it has been shown that bioaccumulation is inversely proportional to the calcium concentration of water (Vanderploeg *et al.*, 1975, quoted in Blaylock, 1982). Like calcium, strontium is primarily absorbed in the bony parts of the fish (skeleton, fins, scales). Using CFs estimated by Vanderploeg *et al.* (1975, quoted in Blaylock, 1982), it is estimated that approximately 95% of the strontium in a fish is found in the bony parts and only 5% in the soft tissues. It is known (Chowdhury and Blust, 2001) that strontium can be absorbed through the gills of fish, though it is believed that intake of food plays an important role in strontium uptake (Kryshev, 2003).

Estimates of ^{90}Sr CF values for fish in some lakes in the Ukraine, Russia and Belarus are given in Table 4.11. The model of Vanderploeg *et al.* (1975, quoted in

Table 4.11. ^{90}Sr concentration factors in freshwater fish after Chernobyl (whole fish, w.w.).

Water body	Sampling date	^{90}Sr CF ($\mathrm{l\,kg^{-1}}$)	Calcium conc. in water ($\mu\mathrm{M\,l^{-1}}$)
Cooling Pond	July–December 1986	100[1]	1,175[2]
Kiev Reservoir	1987–1988	99[1] (Pike-perch) 46[1] (Bream)	1,300[3]
Lake Kozhanovskoe	1993–1994	140[2] (Crucian carp) 173[2] (Goldfish) 365[2] (Perch)	1,010
Lake Perstok	2003	452 (Roach, Rudd) 239 (Perch)	660
Glubokoye Lake	2003	190 (Roach)[4]	738[2]

[1] Kryshev (1995); [2] I.N. Ryabov, Severtsov Institute, Moscow (pers. commun.); [3] Sansone and Voitsekhovitch (1996); [4] N.V. Belova, Severtsov Institute, Moscow (pers. commun).

Blaylock, 1982) using the observed Ca concentration in these lakes predicts CF values in the range 30–70 l kg^{-1} which are significantly lower than the measurements in Table 4.11, particularly for measurements made many years after the accident. This suggests that accumulation in bones may continue to increase ^{90}Sr activity concentrations in fish in the long term.

In general, whole fish–water CFs of ^{90}Sr (of order 10^{2} l kg^{-1}; Table 4.11) were significantly lower than for radiocaesium. In addition to its generally lower fallout, this meant that ^{90}Sr levels in fish were typically much lower than those of ^{137}Cs. In the Chernobyl Cooling Pond, ^{90}Sr activity concentrations were around 2 kBq kg^{-1} in fish during 1986, compared with around 100 kBq kg^{-1} for ^{137}Cs (Kryshev, 1995). Freshwater molluscs showed significantly stronger bioaccumulation of ^{90}Sr than fish. In the Dnieper River, molluscs had approximately 10 times more ^{90}Sr in their soft tissues than was found in fish muscle (Kryshev and Sazykina, 1994).

4.5.7 Radiocaesium and radiostrontium in aquatic plants

The accumulation of radiocaesium in aquatic plants in various freshwater and marine systems is summarised in Table 4.12. CFs in aquatic plants tend to be lower than in fish (a result of the ‘biomagnification’ effect with increasing trophic level), but, as with fish, a strong inverse relationship with potassium concentration is observed.

CFs of ^{137}Cs and ^{90}Sr in various aquatic plants in lakes in the 30-km zone were studied by Gudkov *et al.* (2001). Accumulation of both radionuclides varied between species. ^{137}Cs accumulation was higher in summer than in spring or autumn, though there was little obvious pattern in seasonal changes of ^{90}Sr. Mean CF values (w.w. basis) in different species of aquatic plants varied from around 130–580 l kg^{-1} for ^{137}Cs and from 8–55 l kg^{-1} for ^{90}Sr (Gudkov *et al.*, 2001).

4.5.8 Bioaccumulation of various other radionuclides

There are relatively few data available on the accumulation of radionuclides other than Cs, Sr and I in freshwater biota. For short-lived radionuclides, bioaccumulation factors must be interpreted with care: they are unlikely to represent an equilibrium condition since activity concentrations in water and biota are changing rapidly. It is also believed that accumulation of Cs, Sr and I is by biological uptake whereas other radionuclides are adsorbed to the surfaces of organisms (Kryshev and Sazykina, 1994).

Accumulation factors of different radionuclides in aquatic organisms of the Dnieper River illustrate the difficulties of predicting bioaccumulation of radionuclides other than the more intensively studied caesium, strontium and iodine (Table 4.13). Measurements in June 1986 of accumulation factors in the Dnieper (Kryshev and Sazykina, 1994) showed that isotopes of Ce, Zr, Nb, Ba, La and Ru had much higher accumulation factors in molluscs and aquatic plants than isotopes of Cs, Sr and I (Table 4.13). Kryshev and Sazykina (1994) noted that ‘owing to the processes of radioactive decay and deposition of radionuclides to the bottom with

Table 4.12. Mean CF of radiocaesium in aquatic plants (w.w. basis).

Site	Date	Species sampled	CF ($l\,kg^{-1}$)	$[K^+]$ ($\mu M\,l^{-1}$)	Reference
Cooling Pond	1986–1987	*Potamogeton pectinatus* *Potamogeton perfoliatus*	294*	103	(1)
Dnieper River	1986–1987	*Myriophyllum spicatum* *Ceratophyllum spicatum* *Cladophora glomerata Kuetz*	294*	82	(1)
Crummock water	1986–1987	Aquatic moss (unspecified) *Juncus Bulbous*	7,640	9.0	(2)
Devoke water	1986–1987	Aquatic moss (unspecified) *Fontinalis* *Juncus Bulbous* *Spirogyra* *Subularia Aquatica*	3,770	14	(2)
Loweswater	1986–1987	*Elodea canadensis* *Fontinalis* *Potamogeton gramineus* *Subularia aquatica*	4,750	21	(2)
Black Sea, Turkish coast	1992	*Zostera marina*	50*	4,750†	(3, 4)
Baltic Sea at Kämpinge, Sweden	1987–1988	*Fucus vesiculosus*	40*	2,370**	(5)

* Estimated from CF measured on a dry weight basis and converted to wet weight by assuming dry weight/wet weight = 0.07 for aquatic plants. † Estimated assuming a salinity of 17.0 in the Black Sea, Turkish coast. ** Estimated assuming a salinity of 8.5 in the Southern Baltic.

(1) Kryshev and Sazykina (1994); (2) Camplin *et al.* (1989); (3) Baysal and Tunçer (1994); (4) Vakulovsky *et al.* (1994); (5) Carlson and Holm (1992).

suspended matter, these estimates are highly arbitrary'. They also note, importantly, that '[the estimates] may differ … from the so called equilibrium accumulation coefficients which are measured in laboratory experimental conditions, but practically never occur in nature'. With these caveats, however, it appears that most of the measurements for fish are within or relatively close to the ranges of CF values for fish given by IAEA (1994).

4.6 RADIOACTIVITY IN MARINE SYSTEMS

Marine systems were not seriously affected by fallout from Chernobyl, the nearest seas to the reactor being the Black Sea (approximate distance 520 km) and the Baltic Sea (approximate distance 750 km). The primary pathway of contamination of these seas was atmospheric fallout, with smaller inputs from riverine transport occurring over the years following the accident. Surface deposition of ^{137}Cs was approximately

Table 4.13. Radionuclide CFs† in biota of the Dnieper River in June 1986. For fish, measurements are compared with estimated CF's from IAEA (1994).
From Kryshev and Sazykina (1994).

RN	Molluscs (w.w.)	Aquatic plants (w.w)*	Fish (w.w.)	Fish CF from IAEA (1994)
^{90}Sr	440	16.8	50	10^0–10^3
^{103,106}Ru	750, 1,000	770, 1190	120, 130	10^1–2×10^2
^{131}I	120	4.2	30	2×10^1–6×10^2
^{134,137}Cs	270–300	189, 210	300, 300	3×10^1–3×10^3
^{95}Zr	2,900	1,400	190	3–3×10^2
^{95}Nb	3,700	1,540	220	1×10^2–3×10^4
^{140}Ba	2,800	252	420	4–2×10^2
^{140}La	2,400	210	400	30
^{141,144}Ce	4,600	1,680	900	30–5×10^2

† Estimates are means of more than 20 samples. Standard deviations of measured values were typically in the range 20–50%. * Estimated from CF measured on a dry weight basis and converted to wet weight by assuming dry weight/wet weight = 0.07 for aquatic plants.

2.8 PBq over the Black Sea (Eremeev *et al.*, 1995) and 3.0 PBq over the Baltic Sea (Vakulovsky *et al.*, 1994). Using estimates of the seas' surface areas of 423,000 km^2 and 415,000 km^2, this gives average surface fallout of ^{137}Cs of 6,630 Bq m^{-2} and 7,130 Bq m^{-2} for the Black and Baltic Seas respectively. The total fallout of ^{137}Cs from Chernobyl to the world's seas and oceans was estimated to be 15–20 PBq (Aarkrog, 1998).

Fallout onto the seas' surface was not uniform: in the Black Sea surface water, concentrations of ^{137}Cs ranged from 14.8–503.2 mBq l^{-1} in June–July of 1986. By 1989, horizontal mixing of surface waters had resulted in relatively uniform concentrations in the range 40.7–77.7 mBq l^{-1} (Vakulovsky *et al.*, 1994).

The maximum radioactive fallout onto the Baltic Sea occurred in the northern Gulf of Bothnia and Gulf of Finland where ^{137}Cs activity concentrations of up to 930 mBq l^{-1} were observed (Vakulovsky *et al.*, 1994). By 1988–1989, ^{137}Cs activity concentrations were much more uniform being in the range 100–200 mBq l^{-1} (Vakulovsky *et al.*, 1994; Carlson and Holm, 1992), as shown in Figure 4.15. Vakulovsky *et al.* (1994) report only a minor increase of 20% in pre-accident levels of ^{90}Sr in this area. Holm (1995) observed that Chernobyl fallout was a less significant source of Pu in the Baltic Sea than NWT.

Vertical mixing of surface deposited radioactivity also reduced maximum radioactivity concentrations observed in water over the months to years after fallout. Removal of radioactivity to deeper waters steadily reduced ^{137}Cs activity concentrations in the surface (0–50 m) layer of the Black Sea (Figure 4.15). Profiles of ^{137}Cs with depth (Vakulovsky *et al.*, 1994) showed that by October of 1986 the fallout had mixed vertically within the surface 30–40-m layer of water and by June of 1987, significant amounts of radioactivity had penetrated below a depth of 50 m. The reduction in radiocaesium in the northern Baltic Sea, and the slight increase in

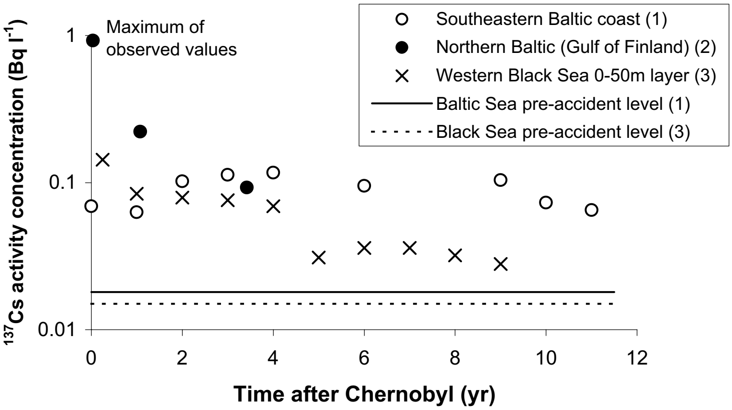


Figure 4.15. Radiocaesium in the Baltic and Black Seas.

From data in: (1) Styro *et al.* (2001); (2) Vakulovsky *et al.* (1994); (3) Kanivets *et al.* (1999).

concentrations in the southern Baltic (Figure 4.15) may be due to horizontal mixing of these differently contaminated waters.

The influence of transfers of Chernobyl radionuclides to seabed sediments has not to our knowledge been quantified. Low K_d's of ^{137}Cs and ^{90}Sr in saline waters suggest that the influence will be low for these isotopes. For example, it is estimated that <1% of radiocaesium in the Baltic is in the dissolved phase of seawater (Knapinska-Skiba *et al.*, 2001). These authors observed accumulation of radiocaesium in fine, muddy sediments but little accumulation in sandy sediments of the southern Baltic. In the Gulf of Finland, up to 80 kBq m^{-2} of ^{137}Cs was observed in bed sediments of coastal waters. In the Black Sea, anoxic bed sediments and bottom waters are likely to have resulted in low transfer rates of radiocaesium to bed sediments. Pu isotopes are more strongly bound to sediments, and studies in the Baltic Sea indicate a residence time of Pu in the water column of 8–10 years (Holm, 1995).

Outflow of water from the Baltic to the North Sea and from the Black Sea through the Bosphorus Strait had little effect on activity concentrations in the seas since water removal is slow. The residence time of water in the Baltic is 25–35 years and in the Black Sea it is of order 1,000 years (the latter estimated from data in Kanivets *et al.*, 1999).

4.6.1 Riverine inputs to marine systems

Riverine input of radioactivity (primarily from the Danube and Dnieper Rivers) to the Black Sea was much less significant than direct atmospheric fallout to the sea surface. Over the period 1986–1995, riverine input for ^{137}Cs was only 4% of the atmospheric deposition, though ^{90}Sr inputs were more significant, being approximately 25% of the total inputs from atmospheric deposition and rivers together (Kanivets *et al.*, 1999). For the Baltic Sea, riverine inputs were similar to those observed in the Black Sea, being approximately 4% and 35% of atmospheric fallout for ^{137}Cs and ^{90}Sr, respectively (Nielsen *et al.*, 1999). The greater relative riverine input of ^{90}Sr compared to ^{137}Cs is due to its weaker adsorption to catchment soils and to lake and river sediments. In addition, a greater proportion of the ^{90}Sr fallout was deposited near to the reactor compared with ^{137}Cs (Figure 2.1) so increasing the influence of transport from the catchment compared to atmospheric fallout to the (distant) Black and Baltic Seas.

4.6.2 Transfers of radionuclides to marine biota

Bioaccumulation of radiocaesium and radiostrontium in marine systems is generally lower than in freshwater because of the much higher concentration of competing ions in saline waters. This is illustrated by comparison of the CFs of ^{137}Cs in freshwater and marine systems for aquatic plants (Table 4.12) and fish (Figure 4.14). Notice, however, that the CF of marine plants and animals is only slightly lower than in freshwaters of high potassium concentration, even though the potassium concentration of marine systems is more than one order of magnitude higher than the maximum typically observed in freshwater. It appears that, relative to potassium, radiocaesium is more strongly accumulated in marine biota than in freshwaters. This confirms similar observations made prior to Chernobyl (Fleishman, 1973).

The relatively low fallout of ^{90}Sr on marine systems and the relatively low bioaccumulation of ^{90}Sr in high calcium content saline waters (Fleishman, 1973) imply that ^{90}Sr contamination of marine species was not significant after Chernobyl. Activity concentrations of other radionuclides in marine systems were also relatively low, though ^{110m}Ag, ^{95}Zr and ^{103,106}Ru were observed in marine macroalgae (*Fucus vesiculosus*) in the Baltic Sea during July 1986 (Carlson and Holm, 1992). Mean ratios of ^{134}Cs, ^{103,106}Ru and ^{95}Zr (corrected for decay) were similar to those observed in atmospheric fallout in this area (Table 4.14), though there was considerable variability in these ratios between different samples.

Table 4.14. Radionuclides in marine macroalgae and fallout compared to ^{137}Cs in July 1986 and August–September 1987.

Calculated from data in Carlson and Holm (1992).*

RN	Ratio RN: ^{137}Cs in macroalgae	Ratio RN: ^{137}Cs in fallout[†]
^{137}Cs	1.0	1.0
^{134}Cs (July 1986)	0.54	0.55
^{134}Cs (August–September 1987)	0.56	0.55
^{103}Ru (July 1986)	1.19	1.27
^{106}Ru (July 1986)	0.32	0.28
^{106}Ru (August–September 1987)	0.17	0.28
^{95}Zr (July 1986)	0.084	0.051

* We estimated ratios in marine macroalgae from averages of the ratio at 7 sites which showed the highest activity concentrations in macroalgae and were decay corrected to the date of fallout. [†] Calculated from relationships in Mück *et al.* (2002) for a distance 1,200 km from Chernobyl.

No evidence was found in the study of Carlson and Holm (1992) for increases in ^{99}Tc, ^{238}Pu, $^{239+240}$Pu and ^{241}Am in macroalgae (compared to background levels from other sources) following Chernobyl. It was, however, suggested that ^{241}Pu from Chernobyl could potentially be observable since the $^{241}\mathrm{Pu}:^{239+240}\mathrm{Pu}$ activity ratio was about 86 in Chernobyl fallout compared to 6 in the Baltic Sea before the accident (Carlson and Holm, 1992). Though ^{241}Pu was not directly measured, it was suggested that this may lead to an observable increase in ^{241}Am as a result of ingrowth from ^{241}Pu in the coming decades.

4.7 RADIONUCLIDES IN GROUNDWATER AND IRRIGATION WATER

4.7.1 Radionuclides in groundwater

Transfers of radionuclides to groundwaters has occurred from waste disposal sites in the exclusion zone. After the accident, radioactive debris as well as trees from the 'Red Forest' were buried in shallow unlined trenches. At these waste disposal sites, ^{90}Sr activity concentrations in groundwaters were in some cases of the order of 1,000 $\mathrm{Bq\,l^{-1}}$ (Voitsekhovitch *et al.*, 1996). Health risks from groundwaters to hypothetical residents of these areas, however, were shown to be low in comparison to external radiation and internal doses from foodstuffs (Bugai *et al.*, 1996). Although there is a potential for off-site (i.e., outside the 30-km zone) transfer of radionuclides from the disposal sites, these workers concluded that this will not be significant in comparison to washout of surface deposited radioactivity. Off-site transport of contaminated groundwater around the Sarcophagus is also expected to be insignificant since radioactivity in the Sarcophagus is separated from ground waters by an unsaturated zone of thickness 5–6 m, and groundwater velocities are low (Bugai *et al.*, 1996).

Radionuclides could potentially contaminate groundwater by migration of radioactivity deposited on the surface soils. It is known (see Chapter 2), however, that long-lived radionuclides such as ^{137}Cs and ^{90}Sr are relatively immobile in surface soils and transfers from surface fallout to deep groundwaters are expected to be very low in comparison to transfers from surface runoff to rivers and lakes. After fallout from NWT, it was observed that ^{90}Sr in Danish groundwater was approximately 10 times lower than in surface streams (Hansen and Aarkrog, 1990). These authors also observed that after Chernobyl, despite measureable quantities of ^{137}Cs in surface streams, activity concentrations were below detection limits in groundwater. Short-lived radionuclides are not expected to affect groundwater supplies since groundwater residence times are much longer than their physical decay time.

In Belarus, the ^{137}Cs activity concentration of water in the saturated zone of soils had no observable correlation with the water level (which was between 35 and 130 cm below the soil surface) but had a significant, but weak ($R^2 = 0.44$, $p < 0.05$) correlation with the ^{137}Cs contamination density of the soils (Smith *et al.*, 2001; Kudelsky *et al.*, 2004). At 10 different sites, the activity concentration in groundwater (per unit of radiocaesium deposition) was significantly lower than in most river and lake systems (Table 4.8).

4.7.2 Irrigation water

There is little information available on the use of contaminated water for irrigation purposes. If the water originates from contaminated areas and is used to irrigate similarly contaminated soils, it will add little to the radioactivity in crops since activity levels in the irrigation water will be comparable to those in the *in situ* soil water. In areas where water originating in a contaminated area is used to irrigate much less contaminated soils, there is a potential problem. This latter scenario is not unlikely, since upland reservoirs often supply irrigation water for lowland soils.

A large amount (1.8 million hectares) of agricultural land in the Dnieper basin is irrigated. Almost 72% of this area is irrigated with water from the Kakhovka Reservoir in the Dnieper River–Reservoir system. Accumulation of radionuclides in plants on irrigated fields can take place because of root uptake of radionuclides introduced with irrigation water and due to direct incorporation of radionuclides through leaves after sprinkling. However, recent studies (O.V. Voitsekhovitch, unpubl. res.) have shown that, in the case of irrigated lands of the southern Ukraine, radioactivity in irrigation water did not add significant radioactivity to crops in comparison with that which had been initially deposited in atmospheric fallout and subsequently taken up *in situ* from the soil.

4.8 RADIATION EXPOSURES VIA THE AQUATIC PATHWAY

Doses from ^{137}Cs and ^{90}Sr contamination of waterbodies in the countries of the fSU are difficult to quantify. Doses from the freshwater pathway (including fish and irrigation water) to the people of Kiev were relatively low, being around 5–10% of doses via terrestrial foodstuffs (Konoplev *et al.*, 1996). As radioactivity (mainly ^{90}Sr) was transferred to areas not significantly contaminated by fallout to terrestrial systems in the lower reaches of the Dnieper cascade of reservoirs, the relative contribution to dose via freshwaters increased to 10–20%.

Radionuclides in the Pripyat River could potentially have led to significant doses in the first months after the accident through consumption of drinking water (Table 4.2). The most significant potential dose calculated (assuming the drinking water activity concentration was equal to that in the river) was 4.2 mSv from ^{131}I (Table 4.2). It is likely, though, that there was significant reduction in activity concentrations within the water supply system, so these doses are likely to be overestimates. Studies in the UK after Chernobyl indicated that in one water supply area, 80% of the radioactivity in raw water was removed by the water treatment process (Jones and Castle, 1987). In an experiment carried out in a water treatment plant in Belgium, it was found that removal efficiencies by conventional water treatment were 73%, 61%, 17% and 56% for Ru, Co, I and Cs respectively (Goosens *et al.*, 1989).

Contamination of fish led to significant doses in some areas. The critical group amongst the 30 million users of Dnieper water were commercial fishermen in the Kiev Reservoir who, in 1986, received a dose of 5 mSv from ^{137}Cs contamination of fish (Berkovski *et al.*, 1996). These commercial fishermen were estimated to consume up to 360 kg of fish per year, fish being the main component of their diet. For the general population living around the Dnieper river-reservoir system, consumption (and hence ^{137}Cs ingestion from fish) was much lower, being around 5–7 kg per year. These consumption estimates can be compared with critical group freshwater fish consumption estimates in the UK of 20 kg per year (NRPB, 1996).

In rural parts of the Chernobyl contaminated areas of the fSU during 1994–1995, it was found that so-called 'wild foods' (mushrooms, berries, freshwater fish, game animals) had radiocaesium contents which were around one order of magnitude higher than agricultural products (e.g., milk or meat). Whole body monitoring of people living close to Lake Kozhanovskoe, Bryansk, showed (Travnikova *et al.*, 2004) that ^{137}Cs intake by the population was strongly correlated with levels of consumption of freshwater fish. In rare situations like this, where people consume fish from the (few) highly contaminated 'closed' lakes, the ingestion dose can be dominated by ^{137}Cs from fish.

In western Europe, consumption of freshwater fish does not form an important part of the diet, but sports and commercial fisheries may be of economic importance in some areas. Though not necessarily dependent on the consumption of fish, angling is one of Europe's most popular leisure activities. In the UK, fallout from Chernobyl had little effect on fisheries, though anecdotal evidence suggests that it was a cause of (unfounded) concern amongst anglers in the more heavily contaminated areas such as the western part of Cumbria. In Norway, where fallout levels were up to one order of magnitude higher, consumption of freshwater fish declined by up to 50% in the more contaminated areas, and the sale of freshwater fish to the general public was prohibited in these areas (Brittain *et al.*, 1991). These authors also reported that the sale of fishing licences in parts of Norway declined by 25% after Chernobyl.

Doses from various sources of radioactivity in the Baltic Sea have been estimated by Nielsen *et al.* (1999). The maximum dose from Chernobyl derived radionuclides to the critical group of seafood consumers was estimated to be 0.2 mSv in the Bothnian Sea and the Gulf of Finland during the first year after the accident. This was a factor of 3–4 times lower than estimated annual doses from naturally occuring ^{210}Po in the Baltic Sea (Nielsen *et al.*, 1999).

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