

*Appeared as:* A.V. Kudelsky, J.T. Smith, S.V. Ovsianikova, J. Hilton (1996) The mobility of Chernobyl-derived Cs-137 in a peatbog system within the catchment of the Pripyat River, Belarus. *Sci. Tot. Environ.* 188, 101-113.

**The mobility of Chernobyl-derived  $^{137}\text{Cs}$  in a peatbog system within the catchment of the Pripyat River, Belarus.**

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## Abstract

The behaviour of Chernobyl-derived  $^{137}\text{Cs}$  in a hydrologically isolated bog system in the catchment of the Pripyat river in Belarus was investigated. Measurements were made of  $^{137}\text{Cs}$  activities in the solids and pore waters of the bog soils, as well as the variability in activity in water draining from the bog. It was found that the radiocaesium activity of the pore water, and hence the measured distribution coefficient,  $K_d$ , was dependent upon the pressure at which the water was removed from the soil. Measured values of  $K_d$  were of order  $10^2 \text{ l.Kg}^{-1}$  at an extraction pressure of 0.8MPa, approximately one order of magnitude lower than those measured in a similar system, Devoke Water, UK [1]. Results of comparative measurements suggested that this was a result of the different pore water extraction techniques used. The vertical migration of radiocaesium was modelled using a solution of the advection-diffusion equation. Using a mass balance approach, it was estimated that  $^{137}\text{Cs}$  was removed from the system at a rate of 0.3% of the catchment inventory per year, approximately 8 years after the Chernobyl accident. It was shown that both vertical migration and removal of  $^{137}\text{Cs}$  is best modelled using a  $K_d$  based on a measurement of pore water held at low pressure in the soil, around  $10^3 \text{ l.Kg}^{-1}$ .  $^{137}\text{Cs}$  activities in soil pore waters and in drainage waters were very strongly related to the aqueous potassium concentration, and both showed concentration minima in drainage water during the spring. It was shown that runoff coefficients of radiocaesium from peat bogs 8 years after the Chernobyl accident were approximately one order of magnitude greater than those from unsaturated soils of higher mineral content.

## 1. Introduction

Large areas of land in Belarus, Ukraine and Russia which were contaminated by fallout radiocaesium from the Chernobyl accident are covered by peat bogs and highly organic soils. Peat bogs are high in organic matter content, resulting in a high availability of  $^{137}\text{Cs}$ , and they are saturated with water, allowing high mobility of  $^{137}\text{Cs}$  in solution. During the years following the Chernobyl accident it has been found that highly organic soils (particularly saturated peats) release much more  $^{137}\text{Cs}$  to water bodies than the majority of mineral soils [2]. A large proportion of the runoff to the Dnieper reservoir system is associated with the bog covered lands of the radionuclide-contaminated territories of Belarus. It is believed (O.V. Voitsekhovitch, pers. comm.) that remobilisation from peat bogs in Belarus, Russia and the Ukraine was an important source of secondary contamination to the Kiev Reservoir during the years following the Chernobyl accident. Because they are permanently saturated, bog areas are of particular importance during periods of low river flow.

In studies of  $^{137}\text{Cs}$  mobility in a peatbog in the catchment of Devoke Water, Cumbria, UK [1], it was shown that both the vertical migration of  $^{137}\text{Cs}$  and its release to streams could be related in a simple manner to the solids-aqueous distribution coefficient ( $K_d$ ) of  $^{137}\text{Cs}$  in these soils. Model-derived  $K_d$  values were found to be of order  $10^3 \text{ l.Kg}^{-1}$ , in good agreement with *in situ* measurements. We have carried out a similar study in a peat bog area (the "Opromokh" site) within the catchment of the Pripjat River, Belarus, in order to determine the chemical behaviour of  $^{137}\text{Cs}$  in this system and to allow us to make predictions of its removal to surface waters. In order to make comparisons with the

previous, more comprehensive studies at Devoke Water [1-3], samples from this site were also analysed as part of the study.

The binding of Cs in mineral soils has been shown to be a result of specific sorption to frayed edge sites (FES) on illitic clay minerals [4]. Cs which is sorbed to these sites is unavailable for exchange with many of the major cations which occur in natural waters (e.g. Ca, Mg) because of their large hydrated radius, although it will exchange with  $\text{NH}_4^+$  and  $\text{K}^+$  [5]. In very highly organic soils, however, it has been shown [6] that there is often too little illite to bind all of the  $^{137}\text{Cs}$  on specific sites, so that a large fraction may be resident on regular cation exchange sites (RES). It has also been established that  $^{137}\text{Cs}$  can migrate into non-exchangeable sites in illitic clays (so-called “fixed” sites). In very low mineral content soils, fixation of Cs is also expected to be low. In apparent contradiction to this, studies on the highly organic soils in Devoke Water [3] showed that around 80% of the  $^{137}\text{Cs}$  is in non-exchangeable forms. We will present further measurements of  $^{137}\text{Cs}$  exchangeability in the Opromokh soils, and attempt to relate its binding and mobility to that of other cations in solution.

## 2. Site description

The Opromokh peat bog is situated in the Lelchitsy region of the Gomel district of Belarus, approximately 150 km from the Chernobyl NPP (Fig. 1).  $^{137}\text{Cs}$  deposition due to the Chernobyl accident in this area is within the range 37-185  $\text{kBq}\cdot\text{m}^{-2}$ . The peat bog is associated with a sink in the relief (absolute elevations are 133-134 m) incised in the Pripyat second fluvial terrace above the flood plain. Crescentic dunes of alluvial-eolian

origin border the Opromokh bog from the north-northwest and northeast, with gentle slopes at the other sides. The area of the bogland outlined by an isohypse of 135 m is 6 km<sup>2</sup>. 5 km<sup>2</sup> are covered by small forest and shrubs and the remaining 1 km<sup>2</sup> is an open water surface bog (Figure 2). The catchment area is 9.1 km<sup>2</sup>.

Pine forest with sphagnum is found within the bogland. The pinewood surface is dominated by oligotrophic plants and hydrophytes (*Sphagnum*, *Eriophorum vaginatum* L., *Oxycossus quadripetalus* Gilib., *Ledum palustre* L., *Drosera rotundifolia* L.). Peat and roots of the modern vegetation are covered by standing surface water. In peripheral parts of the bog, ground water occurs in alluvial, eolian and aqueoglacial sands at a depth of 0.1 to 0.2 m below the surface within the lowlands and at a depth of 8-10 m deep within crescentic dunes. The soils surrounding the bog are peat podsols overlying a sand substrate (humus content of substrate 2%, clay fraction 5-9% and cation exchange capacity 5.3 meq/100g). Peat deposits are up to 4 m thick, consisting of roots and decomposing vegetation to a depth of 20cm below the surface overlying decomposed humic material and finely-dispersed mineral particles. Peat contains mobile phosphorus (from 3.5 to 10.1 mg/100g of soil) and exchangeable potassium (from 5.5 to 22.2 mg/100 g of soil). pH<sub>KCl</sub> ranges from 2.82 to 2.86 and the hydrolytic acidity is up to 207.0 meq/100 g soil. Water is input by both atmospheric precipitation and additional groundwater inflow from the adjacent dry valleys. The inflow rate is 0.73 l.s<sup>-1</sup>.km<sup>-2</sup>, the evaporation rate is 0.58 l.s<sup>-1</sup>.km<sup>-2</sup>, and the water supply due to net precipitation is 0.15 l.s<sup>-1</sup>.km<sup>-2</sup>. The bogland water is discharged through an old drainage canal. The water discharge shows seasonal variations.

The climate of the region is temperate continental. Mean air temperature of the warmest month (July) is +18.6°C and of the coldest month (January) is -5.8°C. The winter in the region begins in the middle of November and lasts for 105-130 days. The snow cover persists for 80-100 days. Maximum ice thickness in rivers is as high as 40-65 cm (in February), and from 20 to 30 cm in bogs with an open water surface. Average annual precipitation is 545 mm; once per 10 years the precipitation exceeds 660 mm, and in drought years decreases to 450 mm. The evapotranspiration from the water surface is 537 mm.y<sup>-1</sup> and from grassland 386 mm.y<sup>-1</sup>. The evaporation from the bog surface is of the same order as the mean precipitation.

### **3. Materials and Methods**

Peat cores were taken using 22 cm diameter stainless steel peat corers. The cores were sliced in the field at 5-10 cm intervals to a depth of 30-60 cm, the samples being returned to the laboratory in sealed plastic bags. Samples of surface soils (5-10cm) were also taken from other parts of the catchment area outside the peat bog. Samples of surface standing water, and from the outflow of the bog, were taken at various dates between 1993 and 1995.

Pore waters from peat cores were removed by squeezing the samples using plastic press moulds at a pressure of 0.8 and 9.8 MPa. The remaining solids were removed by filtration through 3.0 µm then 0.2 µm cellulose acetate filters. Drymass per unit wet volume was approximately 0.1 g.cm<sup>-3</sup>, and approximately 50-60% of the total water was removed by the press, an additional 10% being removed at the higher extraction

pressure. A sub-sample of the total solids was taken and analysed for  $^{137}\text{Cs}$  on a low-background Ge-Li co-axial  $\gamma$ -detector. Solids within the range  $0.2\ \mu\text{m} - 3.0\ \mu\text{m}$  were counted separately. A sub-sample of the pore water was removed for measurement of the major cations ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ,  $\text{Na}^+$ ) and ammonium. The pore water from each core was also analysed for anions ( $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$ ,  $\text{HCO}_3^-$ ), pH, DOC and alkalinity. The pore water was then preconcentrated by evaporation and analysed for  $^{137}\text{Cs}$ . For the purpose of intercomparison of results, the above experimental analyses were also carried out on a core from the Devoke Water site. In order to estimate water discharge from the Opromokh system, a rectangular spillway was constructed on the bog outflow (Rybnikov canal, Fig.2, site #7). Surface standing water and drainage waters were analysed in the same manner as the extracted soil pore water.

Three of the samples of bog soil from Opromokh site #6 were subjected to three sequential 1M ammonium acetate ( $\text{NH}_4\text{Ac}$ ) extractions at a solid-liquid ratio of approximately 1:10. Equilibration time for each extraction was 24hrs, the 3 extractant solutions being bulked prior to  $\gamma$ -counting.

## 4. Results

*Soil, peatbog soil and peat interstitial water measurements.*

$^{137}\text{Cs}$  inventories in the catchment soils and peatbogs are shown in Table 1. Surface samples (0-5cm) only were taken in the unsaturated sandy soils at sites 1-5, deeper cores (20-48 cm) being taken elsewhere. The mean inventory for the catchment is  $88.5\ \text{kBq.m}^{-2}$  within the range  $37\text{-}185\ \text{kBq.m}^{-2}$  expected for this area. The greatest part of the  $^{137}\text{Cs}$

inventory is within a depth of 20cm, the distribution of  $^{137}\text{Cs}$  along vertical peat soil profiles showing maximum values at depths of 7.5 cm (core #8 and #9) and 15 cm (core #6). Vertical migration of  $^{137}\text{Cs}$  appears to be dependent upon soil density: in the areas with more compact peats (sites 8, 9, 10), most of the inventory is retained within the 0-20 cm surface layer, whereas in the less dense peat soil (site 6) high concentrations are observed down to a depth of 40-45 cm, with a maximum value in the 10-20 cm layer. This vertical migration suggests that the inventories at sites 1-5 may have been underestimated, since only the surface 5 cm was measured, although the greater soil density at these sites would inhibit migration.

It is interesting to compare the distribution of  $^{137}\text{Cs}$  along the bog soil profile in summer and winter periods (frozen soil). The content of  $^{137}\text{Cs}$  in the solids and interstitial waters of two cores taken at site 6 (12.10.1993 and 2.06.1994) are shown in Fig. 3. As can be seen from Fig. 3, the profiles of solid-sorbed  $^{137}\text{Cs}$  from the two cores are in excellent agreement. An average of 3.7 % of the solid-sorbed  $^{137}\text{Cs}$  was retained on small particulates ( $0.2 < d < 3.0 \mu\text{m}$ ). The aqueous phase (i.e. passing through  $0.2 \mu\text{m}$  filter) activities in the winter core are much higher than those in the summer core. We believe that this may be due to variations in pore water chemistry and degree of dilution of the pore waters during the different periods of the year. A release of  $^{137}\text{Cs}$  and  $\text{K}^+$  could be caused by break up of the soil structure as a result of freezing [7].

$^{137}\text{Cs}$  concentrations in pore water (Table 2) vary from 1.2 to 48.0 Bq/l, the highest nuclide concentrations being found in the soil water from the upper parts of the profiles. *In situ*  $K_d$  values for the five cores taken at sites 6, 8, 9, 10 and the replicate core taken from Devoke Water, are given in Table 3. A number of the samples were pressed at



higher pressure (9.8 MPa), removing more water from the soil. The  $K_d$  values given in Table 3 represent the distribution of  $^{137}\text{Cs}$  between solid and solution extracted at low pressure (0.8 MPa) and, for selected samples, the distribution between solid and the additional water extracted at the higher pressure (9.8 MPa). The additional water extracted at high pressure had activities approximately 3 times higher than that extracted at low pressure, leading to a  $K_d$  which is around a factor of 3 lower.

Ammonium acetate extractions were carried out on three of the samples from core 6 (02/06/94). Exchangeable fractions,  $f_{\text{ex}}$  were relatively constant at 13.3% at the peak (13-18cm), 13.5% half way down the profile (23-28cm) and 17.2% at the bottom (43-48cm). The majority of the solid-sorbed  $^{137}\text{Cs}$  is therefore (at least on the extraction timescale of days) in non-exchangeable forms.

Interesting data were obtained by studying the pore water chemical composition (see Table 2). Their dissolved solids concentration varies from 19 to 200 mg/l, the sums of cations ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{NH}_4^+$ , etc.) and anions ( $\text{Cl}^-$ ,  $\text{HCO}_3^-$ ,  $\text{SO}_4^{2-}$ , etc.) being greatly out of balance (a mean of 30% greater cations than anions) in the upper parts of the hydrochemical profiles. The difference of sums of cations and anions is the greatest near the open surface of the soil and bog substratum and reduces as the depth increases. A deficiency of anions is compensated for by organic acids, the content of which can be judged from the value of bichromate oxidiseability ( $\text{mgC.l}^{-1}$ ). This latter varies from 24 to 274  $\text{mgC.l}^{-1}$ , the content of  $[\text{K}^+]$  from 0.6 to 68  $\text{mg.l}^{-1}$ ,  $[\text{NH}_4^+]$  from traces to 14  $\text{mg.l}^{-1}$  and more (46  $\text{mg.l}^{-1}$ , core #10, depth 0-5 cm). It is remarkable that the greatest values of

dissolved solids,  $[\text{NH}_4^+]$ ,  $[\text{K}^+]$ ,  $^{137}\text{Cs}$  and organic carbon ( $\text{C}_{\text{org}}$ ) concentrations are related to the upper (0-5 cm) parts of flooded bog soil.

*Surface standing water & drainage water.*

The  $^{137}\text{Cs}$  aqueous activity in the peat bog surface standing water is significantly lower (mean  $1.6 \text{ Bq.l}^{-1}$ , range  $0.74\text{-}2.52 \text{ Bq.l}^{-1}$ ) than was observed in the soil interstitial water (mean  $10.6 \text{ Bq.l}^{-1}$ , range  $1.6\text{-}48.6 \text{ Bq.l}^{-1}$ ). Small particulates ( $0.2 < d < 3.0 \mu\text{m}$ ) contribute a mean of  $0.05 \text{ Bq}$  of activity per litre of standing water solution (3.1%). Compared with the bog soil porewater, the drainage water (Table 4) is also characterised by rather low  $^{137}\text{Cs}$  concentrations ( $0.24 - 2.66 \text{ Bq.l}^{-1}$ ), similar to standing water activities.

Fig. 4 shows the variation of  $^{137}\text{Cs}$  activity and  $[\text{K}^+]$  in the drainage water over time. It is evident that the variation in concentrations of these elements is strongly related and follows a seasonal pattern with a minimum during the spring and early summer period. The rate of total loss from the bog (activity or concentration per litre multiplied by discharge), however, shows no minimum in spring.

## 5. Modelling

In a similar manner to the approach taken in previous studies on the Devoke Water site [1], we will use a model based on the solids - aqueous distribution coefficient ( $K_d$ ) to describe the vertical migration of  $^{137}\text{Cs}$  in this system.

The transport of a tracer with concentration,  $C$ , within a soil column can be described by the advection-diffusion equation:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} - v \frac{\partial C}{\partial x} - \lambda C \quad (1)$$

Where  $v$  is an effective velocity, dependent on the growth rate of the soil and the advection of pore water, and  $\lambda$  is the decay constant of the radionuclide.

Assuming that all of the sorbed  $^{137}\text{Cs}$  is exchangeable and that the exchange process is in equilibrium,  $D$  is an effective diffusion coefficient, given by:

$$D \approx \frac{\psi D_0}{1 + sK_d / \phi} \quad (2)$$

[8] where  $D_0$  is the molecular diffusion coefficient ( $1.45 \times 10^{-5} \text{ cm}^2/\text{s}$  at  $10^\circ\text{C}$ , [9]) of Cs in water,  $\phi$  is the porosity,  $s$  is the drymass per unit wet volume and  $\psi$  is a tortuosity factor, approximated by  $\psi = \phi^2$  [10]. For an impermeable upper boundary (ie diffusion can only take place downwards from the soil surface), the solution to equation (1) for a decay-corrected tracer can be approximated by a gaussian profile reflected at the upper boundary [11]:

$$C = \frac{M}{\sqrt{4\pi D_{eff} t}} \left\{ \exp\left(-\frac{(x-vt)^2}{4D_{eff} t}\right) + \exp\left(-\frac{(x+vt)^2}{4D_{eff} t}\right) \right\} \quad (3)$$

where  $t$  is the time since the fallout event. The exact solution to equation (1) for the impermeable boundary condition has been given by [1], however, for the low pore water velocities found in peat bogs, and given the numerous other simplifications of the model, the simpler equation given above is adequate.

Equation (3) was curve-fitted to the activity-depth profiles resulting from the Chernobyl fallout, the input being a spike at time corresponding to April 1986 using  $D$  and  $v$  as fitting parameters to obtain the best least-squares fit. An example of the curve fit for one of the cores from the Opromokh site is given in Fig. 5, showing the similarity between equation (3) and the exact solution given in [1]. The  $K_d$  values were obtained from the best fit  $D$  values using equation (2) (see Table 5). The parameter values obtained from the single core taken from Devoke Water are also shown in Table 5. The presence of a  $^{137}\text{Cs}$  Weapons Test peak in the profile can interfere with the curve fitting of the soil profiles. For low deposition areas such as Devoke Water it is sometimes possible to distinguish between Weapons Test and Chernobyl fallout using measurements of  $^{134}\text{Cs}$  [1]. In this case, however, because of the difficulties in counting the low levels of  $^{134}\text{Cs}$  this was not possible, so it was assumed that the whole profile was due to Chernobyl fallout. Since, in the Devoke Water area, Chernobyl inventories were around 3-5 times higher than those from bomb fallout, the Weapons Test profile has only a small effect on the shape of the profile. At the Opromokh site the Weapons Test fallout is negligible. We have not attempted to interpret the velocity values (Table 5), since this parameter is dependent in a complex manner upon both peat growth and (retarded) movement in the

pore waters. It should further be noted that the above equations strictly only apply to saturated conditions.

## 6. Discussion

### *Cs-137 in solution and vertical migration.*

The  $^{137}\text{Cs}$  activity measured in the aqueous phase is dependent upon the pressure at which the water was extracted from the soil. At the Opromokh site,  $C_{\text{aq}}$  values were found to be much lower (mean  $1.6 \text{ Bq.l}^{-1}$ ) in standing water (assumed to be held at zero pressure) than at  $0.8 \text{ MPa}$  (mean  $10.6 \text{ Bq.l}^{-1}$ ), which is itself a factor of 2 lower than that extracted at  $9.8 \text{ MPa}$  (mean  $21.3 \text{ Bq.l}^{-1}$ ). The "standing water" measurement is the closest to concentrations of activity removed in drainage water, which have mean  $1.14 \text{ Bq.l}^{-1}$ . The water removed at higher pressure from the soil originates in layers closely associated with solid soil surfaces and bog vegetation, showing higher isotope concentrations than that which is relatively distant from solid surfaces. Water more closely associated with soil and plant material is therefore a source of  $^{137}\text{Cs}$ , which can be desorbed and lost from the bog via drainage water.

Similar results are observed at the Devoke Water site. Activities in pore solution extracted at  $0.8 \text{ MPa}$  pressure had mean  $1.4 \text{ Bq.l}^{-1}$ , whilst that fraction held at pressure

between 0.8 and 9.8 MPa had activities almost an order of magnitude higher (mean 12.5 Bq.l<sup>-1</sup>). <sup>137</sup>Cs removed by hand squeezing (at a pressure lower than 0.8 MPa) had mean of 0.3 Bq.l<sup>-1</sup> [1]. As observed at the Opromokh site, streamwater flowing from the Devoke site has significantly lower activity (~ 0.1 Bq.l<sup>-1</sup>, assuming dilution of approximately 50% of the measured stream concentration of 0.05 Bq.l<sup>-1</sup> by water with very low concentrations of <sup>137</sup>Cs activity flowing from mineral soils, [2]) than that which is extracted at 0.8 or 9.8 MPa, but is comparable to that removed by hand squeezing.

Model-derived K<sub>d</sub> values for the Opromokh site are of order 10<sup>3</sup> l.Kg<sup>-1</sup>, whereas the *in situ* measurements were significantly lower, usually of order 10<sup>2</sup> l.Kg<sup>-1</sup> (Table 5). A similar effect is seen at Devoke Water. The *in situ* K<sub>d</sub> values determined at the Devoke site using a pressure of 0.8 MPa (10<sup>2</sup>- 10<sup>3</sup> l.Kg<sup>-1</sup>) are much lower than those observed by Smith *et al.* (1995) (10<sup>3</sup>- 10<sup>4</sup> l.Kg<sup>-1</sup>), the higher values being in better agreement with modelling [1]. It therefore appears that this higher pressure may under-estimate the *in situ* K<sub>d</sub> relevant for vertical migration. The <sup>137</sup>Cs which is transported both down the profile and in drainage water appears to be that fraction which is held at very low pressure.

#### *The influence of water chemistry on Cs-137 sorption.*

As discussed in the Introduction, on illitic clay minerals the Cs<sup>+</sup> ion competes for specific exchange sites (so called "Frayed-edge sites", FES) with similarly-sized NH<sub>4</sub><sup>+</sup> and K<sup>+</sup> ions. In highly organic peat bogs (Organic Matter content > 80%), however, there may not be enough illitic clay minerals to specifically sorb all of the Cs, so that a large fraction would reside on regular ion-exchange sites available to all cations [6]. In

this case, the Cs concentration in solution would be controlled by the ionic composition and strength of the solution.

We do not have measurements of illite content in the peat bog soils: it is expected that the content would be significantly less than 5%, making measurement extremely difficult. We can, however, test the hypothesis that the exchangeable FES on illites control caesium sorption in this system. If this were the case, then the  $^{137}\text{Cs}$   $K_d$  should be controlled by competition with  $\text{NH}_4^+$  and  $\text{K}^+$  in solution. Assuming that the majority of the Cs resides on exchangeable FES then the  $^{137}\text{Cs}$   $K_d$  is given by [3]:

$$K_d = \frac{K_c(Cs / K)[FES]}{[K^+] + K_c(NH_4 / K)[NH_4^+]} \quad (4)$$

where  $K_c(A/B)$  is the ion A to ion B selectivity coefficient of the FES and  $[FES]$  is the FES capacity, usually expressed in  $\text{eq.Kg}^{-1}$ . Assuming that  $[FES]$  can be considered to be constant for the peat profile (measurements of  $[FES]$  in the Devoke Water peat [12] showed no clear changes with depth), and that the  $\text{NH}_4^+$  -  $\text{K}^+$  selectivity coefficient is around 5.0 [13], then the  $K_d$  should be inversely proportional to  $[K^+] + 5.[NH_4^+]$ . A linear regression was carried out of  $\log_{10}(K_d)$  versus  $\log_{10}([K^+] + 5.[NH_4^+])$  for all of the  $K_d$  measurements made at the Opromokh site (0.8 MPa), however, no significant correlation was found ( $R^2 = 0.01$ ). In addition, no correlation was found between  $K_d$  and other ions in solution.

It is clear that the  $^{137}\text{Cs}$   $K_d$  is unrelated to competitor ion concentrations ( $[K^+]$  and  $[NH_4^+]$ ). This suggests that the binding of  $^{137}\text{Cs}$  is not controlled by fast ion-exchange to

specific sorption sites (FES) in the bog soil, as expected in these highly organic systems [6], and as implied by the low ammonium exchangeabilities (< 20%) we have observed. In addition, if simple ion-exchange were the dominant sorption mechanism, then a negative relationship between  $K_d$  and other major cations would be expected. The lack of any such relationship implies that the measured  $K_d$  values do not represent a true ion-exchange equilibrium between sorbed and aqueous phases.

It may be that the fact that the majority of  $^{137}\text{Cs}$  is unavailable for exchange with ions in solution (measured “exchangeable” fractions were less than 20%), is the cause of the lack of correlation between  $K_d$  and competing ions. We do, however, find such a correlation in mineral systems with much lower exchangeable  $^{137}\text{Cs}$  in which specific sorption takes place. This may suggest non-specific sorption in our highly organic system.

We can gain some insight into the behaviour of  $^{137}\text{Cs}$  in this system by studying the relationships between  $^{137}\text{Cs}$  in solution and concentrations of other ions in solution. As shown in Fig. 6, the relationship between (aqueous  $^{137}\text{Cs}$ ,  $\text{Bq.l}^{-1}$ ) and  $[\text{K}^+$ ,  $\mu\text{M}]$  in pore and drainage waters is strong, and can be represented by a power law relation:

$$[^{137}\text{Cs}] = 0.12[\text{K}^+]^{0.81} \quad (5)$$

with correlation coefficient  $R^2 = 0.84$ . The power of  $[\text{K}^+]$ , 0.81, is relatively close to 1, implying near linearity: indeed, a linear regression between  $[^{137}\text{Cs}]$  and  $[\text{K}^+]$  gives a good correlation ( $R^2 = 0.8$ ). The linear regression is, however, not sensitive over such a wide range of values of the variables (more than two orders of magnitude), causing the



*relative* deviation of the regression from the data to be high at the lower end of the range. Some relationship was also observed between  $\log_{10}$ (aqueous  $^{137}\text{Cs}$ ) and  $\log_{10}([\text{K}^+] + 5[\text{NH}_4^+])$ , implying a power law relationship, however the correlation coefficient was much lower ( $R^2 = 0.54$ ). Similarly, correlation between the logarithm of the aqueous activity of  $^{137}\text{Cs}$  and  $\log_{10}[\text{NH}_4^+]$  had coefficient  $R^2 = 0.42$ , with  $\log_{10}[\text{Na}^+]$ ,  $R^2 = 0.62$ , with  $\log_{10}[\text{Mg}^{2+}]$ ,  $R^2 = 0.54$ , with  $\log_{10}[\text{Cl}^-]$ ,  $R^2 = 0.48$ , and with the logarithm of the sum of cations in solution the correlation coefficient was  $R^2 = 0.63$ . No correlation was observed between aqueous  $^{137}\text{Cs}$  and  $[\text{Ca}^{2+}]$  or pH.

Despite the lack of any correlation between  $K_d$  values and ions in solution, the activity of  $^{137}\text{Cs}$  in solution is strongly related to  $[\text{K}^+]$  and, to a lesser extent, other ions in solution. The difference in  $^{137}\text{Cs}$  activities (and the concentrations of other ions) between the soil and drainage waters is around one order of magnitude. We believe that this is primarily a dilution effect: rainwater  $^{137}\text{Cs}$  activities are zero, and  $[\text{K}^+]$  concentrations are much lower than those found in the soil water. The large variations in  $^{137}\text{Cs}$  activities and ion concentrations in a given soil water profile may be due to differences in soil structure between the vegetation dominated surface and the decomposed soil at greater depths. Water extracted from surface samples (0-10cm), even at the lower pressure of 0.8 MPa, may be originating from within the surface vegetation, thus having high concentrations of, in particular,  $^{137}\text{Cs}$  and  $\text{K}^+$ .

#### *Availability of Cs-137 in bog soils*

Chemical extractions with  $\text{NH}_4\text{Ac}$  showed that the availability of  $^{137}\text{Cs}$  is similar to that observed at the Devoke Water site [3], being within the range 13.3-17.2%. These

exchangeabilities are significantly higher than those found in mineral soils (typically a few percent), but are surprisingly low given the extremely low mineral content (< 5%), and therefore fixation potential, of these soils. This result, combined with our suggestion above that the  $^{137}\text{Cs}$  in solution is not in equilibrium with (at least the majority of) the solid sorbed activity may imply that  $^{137}\text{Cs}$  is partly “fixed” to organics, either by diffusion into particles, or by uptake by biota. In this case, the  $^{137}\text{Cs}$  would not be irreversibly fixed, but would slowly cycle within the soil-plant-water system.

#### *Cs-137 removal from the Opromokh system*

Fig. 4 shows that the  $^{137}\text{Cs}$  activity and the  $\text{K}^+$  concentrations in drainage water are strongly related and both show a minimum during the spring and early summer. This appears to be unrelated to the minimum in stream discharge during the summer. There is, however, some relationship between concentration of  $^{137}\text{Cs}$  activity in the stream and discharge, as shown in Fig. 7. Although the correlation between  $^{137}\text{Cs}$  activity and discharge is very weak ( $R^2 = 0.15$ ), it is clear from Fig. 7 that there is a tendency to lower activities at high discharges, suggesting some dilution effect.

We can use a simple "mass balance" calculation to estimate the current rate of removal of  $^{137}\text{Cs}$  from the Opromokh catchment during the period 1993-95, approximately 8 years after the Chernobyl accident. The mean inventory in the catchment, calculated from the mean of the inventories of the soil cores, is  $88.5 \text{ KBqm}^{-2}$  giving a total inventory of 0.8 TBq over the whole ( $9.1 \text{ Km}^2$ ) catchment. The mean flow rate of the outflow is  $65.0 \text{ l.s}^{-1}$ , having mean concentration  $1.14 \text{ Bq.l}^{-1}$ , resulting in a loss rate from

the catchment of  $2.3 \text{ GBq.y}^{-1}$ . This loss rate represents a loss of 0.3% of the catchment inventory per year.

We can compare the removal of activity from the Opromokh system with other catchments by calculating a radiocaesium runoff coefficient,  $R$ , which is defined as:

$$R(t) = \frac{C_a}{I} \quad (6)$$

where,  $C_a$  is the activity of the radionuclide in the aqueous phase of runoff water ( $\text{Bq.m}^{-3}$ ) and  $I$  is the mean activity deposited on the catchment ( $\text{Bq.m}^{-2}$ ). The value of  $R$  will vary over time following an accident as a result of reduction in chemical availability of the  $^{137}\text{Cs}$ , as well as transport down the soil profile. Using the measurements of  $^{137}\text{Cs}$  in runoff water from peat bog areas, we can estimate long term (approximately 8 years after the Chernobyl release) runoff coefficients, and compare these with estimates for mineral catchments.

The mean  $^{137}\text{Cs}$  activity in the outflow of the Opromokh bog was  $1.14 \text{ kBq.m}^{-3}$ , which, with a fallout value,  $I$ , of  $88 \text{ kBq.m}^{-2}$  gives  $R = 12.9 \times 10^{-3} \text{ m}^{-1}$ . We can compare these values with measurements carried out in 1990 of dissolved  $^{137}\text{Cs}$  activity in two streams draining sub-catchments of Devoke Water [2] which gave values of 65 and 85  $\text{Bq.m}^{-3}$ . We can extrapolate these values to 1994 (for comparison with the Opromokh measurements, which were mainly taken during 1994) using the measured decline in stream concentrations in the Devoke catchment:  $C_a(t) = C_a(0)\exp(-0.001t)$  where  $t$  is measured in days [2]. This gives estimated values of 15 and 20  $\text{Bq.m}^{-3}$  respectively. Assuming that the majority of the activity is being released from the areas of peat bog in each sub-catchment (46.8% and 38.1% of each sub-catchment respectively), taking

account of dilution,  $^{137}\text{Cs}$  concentration in runoff from each peat bog should be 32 and 52  $\text{Bq}\cdot\text{m}^{-3}$  respectively. With a surface deposition of Chernobyl  $^{137}\text{Cs}$  on this catchment of  $10 \text{ kBq}\cdot\text{m}^{-2}$  [2] this gives R values of 3.2 and  $5.2 \times 10^{-3} \text{ m}^{-1}$ , of the same order as the Opromokh results.

Activities were also measured in streams draining sub-catchments of Devoke Water which contain only small areas of saturated peat soils, being mainly covered by unsaturated podzolic soils [2]. In these streams, activities were around  $20 \text{ Bq}\cdot\text{m}^{-3}$  in 1990, leading to a runoff coefficient of  $0.5 \times 10^{-3} \text{ m}^{-1}$  in 1994, assuming the above rate of decline. Since these catchments contained small areas of saturated peat which may also have contributed to runoff, this must be considered as a maximum value of runoff coefficient for these soils. A further measurement of activity [14] in a stream draining a catchment consisting of shallow mineral soils (Brotherswater, Cumbria) gave an activity of  $0.86 \text{ Bq}\cdot\text{m}^{-3}$ , and with  $I = 1.5 \text{ kBq}\cdot\text{m}^{-2}$  for this catchment, a value of R of  $0.6 \times 10^{-3} \text{ m}^{-1}$ . Estimates in the unsaturated soils of greater mineral content, therefore, give values of the runoff coefficient around one order of magnitude lower than the saturated peats.

Given a knowledge of R values for different soils, long-term concentrations of  $^{137}\text{Cs}$  activity in runoff water could be estimated from the areal deposition of activity and soil type, providing a useful tool for modelling in an emergency situation. This is, however, a very crude approach, and the variability of R in different soil types, and in a given soil type over time, should be investigated further.

## 7. Conclusions

*In situ* measurements of radiocaesium activities in the flooded peat bog soils of Opromokh give  $K_d$  values in the range  $10^2 - 10^3 \text{ l.Kg}^{-1}$ , although these values may be overestimates of the  $K_d$  relevant for radiocaesium migration. Mobility of  $^{137}\text{Cs}$  appears to occur in that fraction of water extracted at low pressure from the soil (i.e. in soil macropores). The measurements of the core taken from Devoke Water have shown that the pressure used in the pore water press (0.8 & 9.8 MPa) releases higher activities of  $^{137}\text{Cs}$  than the hand-squeezing technique used by [1], and thus gives lower values of  $K_d$ . Given the observed dependence of  $K_d$  on pore water extraction pressure, it is necessary to define a pressure at which to determine *in situ*  $K_d$  values. For the purpose of studying the  $K_d$  which is relevant to the vertical migration and removal of radiocaesium from these systems, a low removal pressure (of order 0.1 MPa; a few atmospheres) is appropriate. At both sites, the mobile fraction is that which is held at low pressure in the soil. The curve-fitted  $K_d$  values are around  $10^3 \text{ l.Kg}^{-1}$  in the Opromokh system, of the same order as those estimated at Devoke Water (this study, and [1]).

The fraction of exchangeable  $^{137}\text{Cs}$  in the peat bog soils of the Opromokh catchment is less than 20 %, significantly higher than in mineral soils (typically a few percent). The exchangeabilities are, however, lower than one might expect given the extremely low mineral contents of these soils (< 5%). In line with the conclusions of [1] and [3], it appears that the Cs must be fixed in some way to organic matter. In the Opromokh system, the relation between aqueous  $^{137}\text{Cs}$  and  $[\text{K}^+]$  is very strong, and applies both to soil pore water and discharge water. This, coupled with the similar seasonal variation in

the concentration of these ions in discharge water, shows that their mobility is very strongly linked. We believe that the large differences in  $^{137}\text{Cs}$  and  $[\text{K}^+]$  between soil water and drainage waters is primarily a dilution effect.

The study of runoff coefficients for the Devoke Water and Opromokh catchments has shown good agreement between these two peat bog systems. We have confirmed that the long term removal of radiocaesium from saturated organic soils is much higher (around one order of magnitude) than from unsaturated mineral soils.

### **Acknowledgements**

We would like to thank Rob Comans for constructive criticism of earlier versions of this manuscript. This work was funded by the European Commission International Collaboration on the Consequences of the Chernobyl Accident contract COSU-CT94-0079, and by the UK Natural Environmental Research Council. We would also like to thank The Royal Society for a Travel Award.

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Site	Date	Description	Depth cm	<sup>137</sup> Cs KBq/m <sup>2</sup>
1	02/06/93	Organic root layer, plant debris	0-5	149 ± 14.6
2	"	Soil with vegetation and roots	"	46.1 ± 4.7
3	"	Sphagnum moss	"	64.0 ± 6.6
4	"	Soil with moss & roots	"	112 ± 11.2
5	"	Soil with shreber moss	"	92.9 ± 9.3
6	"	Soil with sphagnum, cranberry	0-30	83.8 ± 8.4
6	10/10/93	"	"	75.1 ± 7.5
6	02/06/94	"	0-48	64.3 ± 6.4
8	19/03/94	Soil with shreber moss	0-20	90.1 ± 9.0
10	29/05/94	Soil with vegetation	0-25	107 ± 10.7

Depth	NH <sub>4</sub> <sup>+</sup>	K <sup>+</sup>	Na <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	Cation sum	Anion sum	Organ. carbon	pH	Cs-137
cm	μM					μE/l	μE/l	mg/l		Bq/l
Opromokh, Belarus. Core #6. 12.10.1993										
0-10	400.0	1320	522.6	86.3	90.3	2393	-	102.7	5.1	55.80
10-20	83.3	244.9	187.0	774.6	-	517.0	1362	69.0	6.7	24.30
20-30	344.4	161.8	56.5	-	-	566.7	-	55.2	4.9	16.30
Opromokh, Belarus. Core #6. 2.06.1994										
0-8	166.7	345.9	232.2	329.2	130.3	1203	-	124.0	4.4	10.50
8-13	27.8	72.3	87.0	329.2	335.8	850.4	-	54.6	6.1	5.30
13-18	111.1	63.8	61.3	197.5	150.3	595.0	614.9	58.8	6.4	4.70
18-23	55.6	80.8	119.6	131.7	130.3	555.6	383.8	45.2	6.4	3.00
23-28	88.9	50.0	87.0	91.3	75.3	391.9	197.7	42.3	5.3	6.90
28-33	27.8	68.2	172.6	197.5	220.5	685.3	-	75.9	5.9	3.10
33-38	55.6	58.5	200.0	131.7	195.5	640.1	-	74.7	5.8	1.60
38-43	0.6	58.5	183.9	167.1	155.3	568.8	-	105.0	5.7	2.80
43-48	83.3	66.9	159.6	157.1	185.3	651.7	-	63.0	6.0	3.40
Opromokh, Belarus. Core #8. 19.03.1994										
0-5	777.8	1590	409.6	232.9	461.0	3433	-	273.8	4.6	43.70
5-10	430.6	638.5	161.3	222.9	110.3	1550	1028	135.4	5.9	14.40
10-15	83.3	159.7	100.0	70.8	145.3	556.2	348.3	37.4	6.4	3.70
15-20	69.4	53.3	97.0	70.8	155.3	469.6	224.1	32.7	5.9	2.90
Opromokh, Belarus. Core #9. 29.05.1994										
0-5	222.2	1745	500.0	212.5	310.5	2948	2182	129.9	6.2	46.49
5-10	194.4	388.5	229.1	172.1	180.3	1155	-	96.0	5.4	22.35
10-15	125.0	119.2	166.1	182.1	320.8	911.4	421.8	63.6	6.1	11.95
15-20	163.3	42.6	216.1	212.5	280.5	914.5	-	26.4	4.0	9.11
20-25	166.7	53.3	253.0	152.1	230.5	858.7	456.3	60.0	5.6	3.00
25-30	111.1	42.6	253.0	222.9	320.8	950.2	723.0	60.0	7.0	2.45
30-35	111.1	15.9	151.7	80.8	180.3	630.6	-	36.1	5.1	2.82
35-40	138.9	15.9	143.9	131.7	400.8	833.1	525.5	36.0	6.6	1.52
40-45	72.2	16.9	132.2	70.8	150.3	442.1	196.2	24.0	5.8	1.79
Opromokh, Belarus. Core #10. 29.05.1994										
0-5	2556	1649	643.5	263.3	370.8	5449	3468	144.0	6.3	25.90
5-10	208.3	164.9	430.9	242.9	205.5	1249	478.2	78.0	6.2	15.83
10-15	111.1	91.5	174.3	227.9	285.5	889.9	-	76.9	4.9	5.57
15-20	94.4	26.7	104.8	131.7	180.3	537.6	235.7	39.1	5.7	1.23
20-25	72.2	27.7	135.7	106.3	220.5	562.3	365.6	27.0	6.5	1.85

Depth cm	Kd ( <sup>137</sup> Cs) core #6 12.10.1993		Kd ( <sup>137</sup> Cs) core #8 19.03.1994		Kd ( <sup>137</sup> Cs) core #9 29.05.1994		Kd ( <sup>137</sup> Cs) core #10 29.05.1994		Kd ( <sup>137</sup> Cs) core #6 2.06.1994		Kd ( <sup>137</sup> Cs) core #7 Peat bog Devoke 12.09.1993	
	Pressure, MPa		Pressure, MPa		Pressure, MPa		Pressure, MPa		Pressure, MPa		Pressure, MPa	
	0.8	9.8	0.8	9.8	0.8	9.8	0.8	9.8	0.8	9.8	0.8	9.8
0-5	130	-	127	67	150	-	433	-	483	-	585	-
5-10			468	243	389	-	676	-	985	368	372	80
10-15	465	-	781	188	311	-	725	-	2149	935	1037	38
15-20			231	64	236	-	260	-	1586	-	578	51
20-25	177	-			194	-	60	-	393	-		
25-30					129	-			325	-		
30-35					73	-			218	-		
35-40					75	-			46	-		
40-45					49	-			45	-		

Date	NH <sub>4</sub> <sup>+</sup>	K <sup>+</sup>	Na <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	Cation sum	Anion sum	pH	Org. carbon	Cs-137
	μM					μE/l	μE/l		mg/l	Bq/l
19.02.93	15.0	40.0	70.0	183.8	t.	365.5	-	4.70	26.40	-
25.04.93	t.	t.	40.0	133.8	6.0	256.0	-	4.45	20.40	0.30
2.06.93	10.0	t.	20.0	466.7	18.3	651.1	500.0	6.55	30.00	0.24
10.11.93	115.6	14.9	63.0	208.8	39.5	606.5	-	4.51	-	-
12.12.93	41.7	32.1	64.3	100.0	24.3	418.3	-	4.10	33.60	1.15
12.01.94	76.7	14.9	61.3	858.3	45.5	1335	844.0	7.40	25.20	0.83
15.02.94	106.1	30.0	60.0	167.1	6.0	496.0	-	4.10	27.60	0.99
15.03.94	70.0	40.0	50.0	675.0	15.3	1021	579.9	6.90	10.25	0.66
10.04.94	-	-	-	-	-	-	-	-	-	0.25
12.05.94	103.9	4.9	20.0	133.8	15.3	371.7	-	4.50	21.05	0.33
13.06.94	10.0	10.0	26.1	483.3	6.0	646.2	499.7	6.92	20.65	0.51
11.07.94	40.0	20.0	150.0	300.4	48.5	733.5	400.7	5.86	24.40	0.82
12.08.94	40.0	20.0	50.0	233.8	36.5	523.4	-	4.95	35.15	1.58
15.09.94	50.0	30.0	60.0	150.4	30.5	440.4	-	4.74	36.75	1.62
15.10.94	50.0	50.0	70.0	183.8	30.5	555.3	-	4.20	27.10	2.63
14.11.94	12.2	60.0	92.2	183.8	51.8	606.4	-	4.31	22.5	4.51
13.12.94	17.8	60.0	80.0	133.8	39.5	492.6	-	4.37	18.20	2.20
14.01.95	20.0	50.0	80.0	133.8	39.5	502.4	-	4.22	25.50	2.66
15.02.95	20.0	40.0	60.0	150.4	18.3	404.0	-	4.37	21.00	1.78
15.03.95	30.0	20.0	50.0	100.0	36.5	389.5	-	4.32	21.2	1.98
11.04.95	40.0	30.0	60.0	100.0	36.5	430.6	-	4.23	22.8	1.09
15.05.95	40.0	4.1	30.0	100.0	36.5	363.6	-	4.32	25.7	0.52

Core	D cm <sup>2</sup> /y	v cm/y	Model Kd l/Kg	<i>In Situ</i> Kd
Devoke	2.7	1.0	2200	643
6a	3.3	1.8	1460	300
6b	3.9	1.8	2460	1300
8	1.3	0.8	2210	400
9	1.4	0.7	3680	270
10	1.9	0.9	1510	520

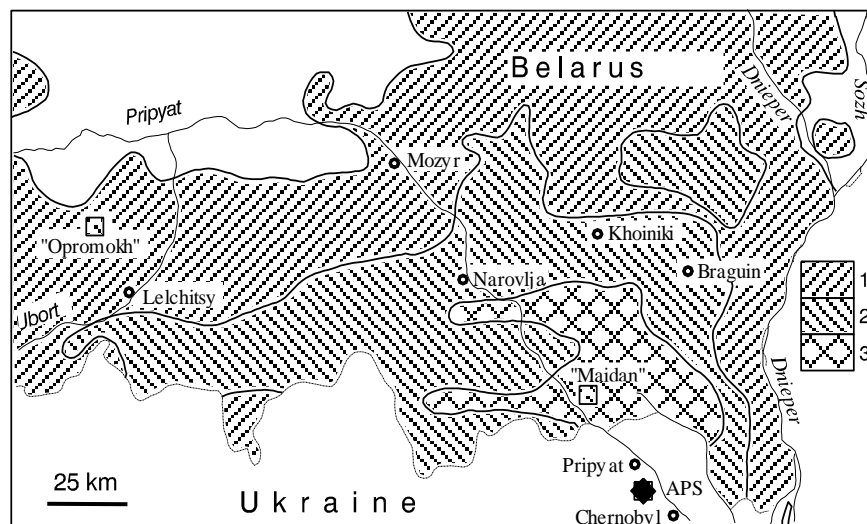
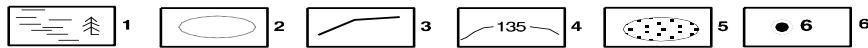
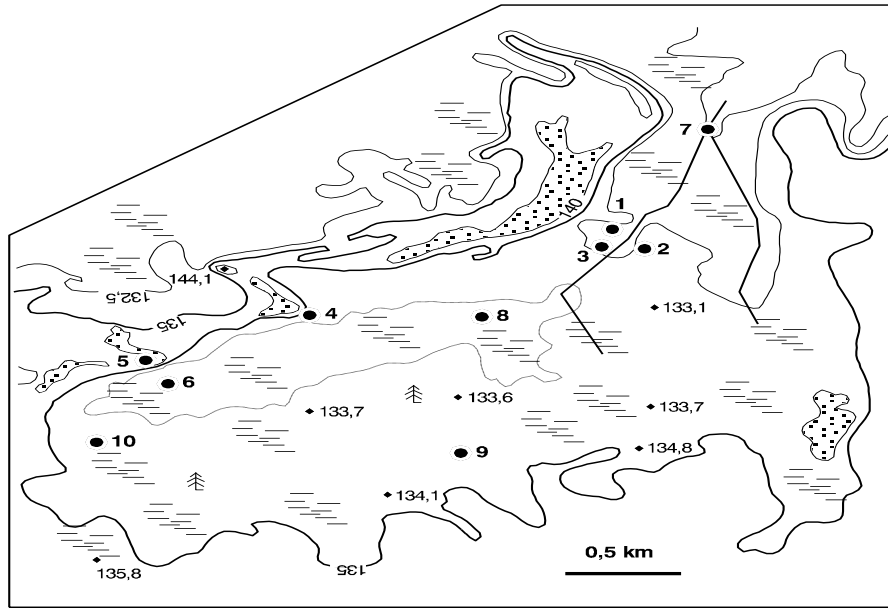


Figure 1.





1 = Bugged and Woded Territories  
 3 = Drainage Canal  
 5 = Sandy Hills

2 = Boundaries of Open Surface Bog  
 4 = Hydrologic limit of the Peat Bog  
 6 = Sampling sites and their numbers

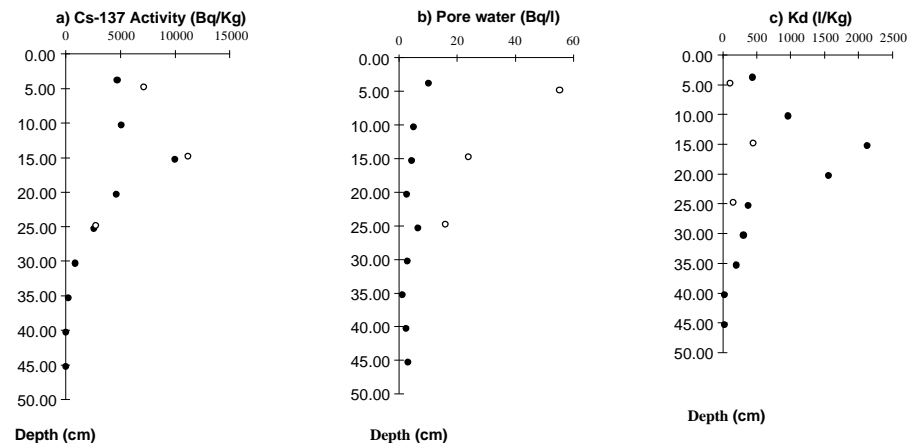


Fig. 3 a) total Cs-137 in solid phase; b) Cs-137 in pore water; c) distribution coefficient. Cores from site 6 Opromokh, taken during (o) winter, 12/10/93 and (o) summer, 02/06/94.

Fig. 4  
 (a) Seasonal change in Cs-137 and Potassium in drainage water.

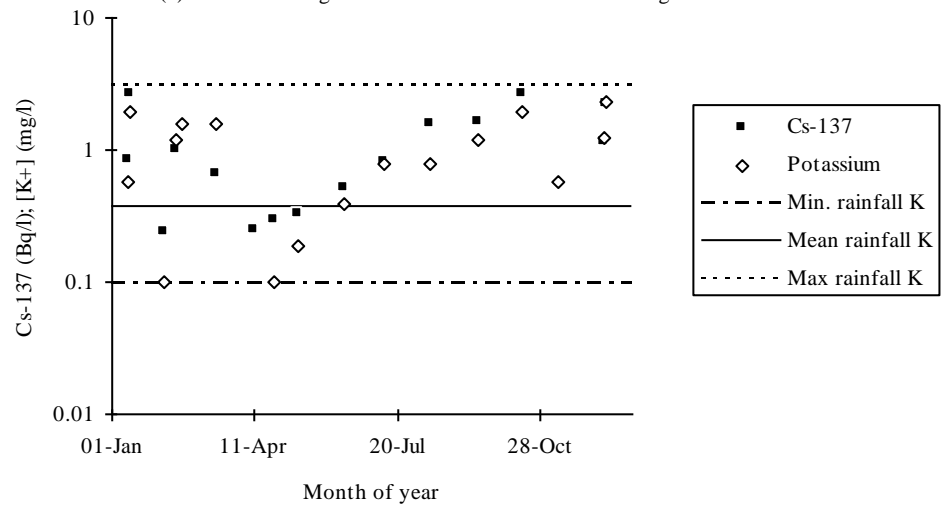


Fig. 5 Log(Aqueous Cs-137) vs Log(Potassium)  
In Pore & Drainage Waters. Opromokh.

