Chernobyl (Ukraine)

Description: The Chernobyl accident in 1986 is the most serious radiological incident on record, a catastrophe that caused much human misery, vast economic disruption and had profound social and environmental consequences. It was also a disaster for the nuclear industry. However, as terrible a tragedy as it was, the accident has provided a unique opportunity for much to be learnt regarding the behaviour of radionuclides in the biosphere and the near surface environment. In particular, the widespread contamination of a large variety of different environments, natural, semi-natural and human-dominated, under different climatic conditions has provided opportunities to test many of the assumptions that have been employed in the construction and application of models designed to simulate the behaviour of radionuclides in the biosphere.

In some senses the Chernobyl accident may be regarded as an unplanned experiment carried out on a huge scale and affecting a wide diversity of environmental situations where the radionuclides deposited at a known date act as markers and tracers of the processes relevant to the forecasting of anthropogenic contaminant migration patterns. These opportunities for study have not been wasted and since the Chernobyl accident, many studies have been published on, for example, geochemistry, radioecology and health and environmental impacts. A number of the studies that have been carried out deal with landscape settings, soil and hydrogeochemical processes that are pertinent to the assessment of the potential future impacts of the geological disposal of radioactive wastes.

Observations of groundwater contamination

Shestopalov et al. (2003) state that sixteen years after the Chernobyl accident, the predominant part of the radionuclide activity is to be found in the upper 10 cm layer of natural soils or within the plough layer of agroecosystems. It has frequently been asserted that particle-reactive radionuclides deposited in fallout such as ^{137}Cs are rapidly and strongly ‘fixed’ within surface soils, that soil cores may be analysed to determine the local inventory of deposition and that the subsequent redistribution of activity occurs in association with soil or sediment particles (e.g. Walling and He, 1997a;b). These assertions may have general validity, but observations within the areas most affected by the Chernobyl accident reveal that ^{90}Sr and ^{137}Cs have penetrated to considerable depths below ground and have contaminated major aquifers over a wide area (Shestopalov et al. 2003). The appearance of ^{134}Cs, which has a half-life of 2 years, in groundwater soon after the accident was taken to confirm the Chernobyl origin of the contamination. According to Shestopalov et al. (2003) the annual vertical transfer from soil to the vadose zone exceeds by between 5-25 times the amount removed by the Prypyat River within the Chernobyl exclusion zone.

Figure 1 shows the distribution of ^{137}Cs in Quaternary and Eocene formations to a depth of 80 m south of Chernobyl. This distribution is qualitatively similar to that for ^{137}Cs deposition after the accident (De Cort et al. 1998). Figures 2 and 3 show the distribution of ^{90}Sr in Quaternary and Eocene aquifers respectively in 1996. The maximum contamination of groundwater in the Eocene aquifer is observed within the cone of depression created by the Kiev water-supply system which suggests a linkage which was established through a special investigation. However, Shestopalov et al. (2003) did not believe that ‘technogenic’ (i.e. man-made) pathways such as wells were responsible for the observed distributions. They also dismissed the likelihood of lateral migration over long distances due to watershed characteristics, although the exchange of aquifer and Dnieper River waters, both naturally and technogenically-induced is suggested by the pattern of contamination. They suggested instead that groundwater contamination results from the water percolating through the unsaturated zone from the land surface. In support of this, Shestopalov et al. (2003) reported measured values of ^{137}Cs in marl rock along a 1km transect of the Kiev metro system at 80m depth. The values ranged from 0.3 – 1.1 Bq/kg.

A complex pattern of radionuclide contamination resulted from the Chernobyl accident because the release was extended over 10 days and involved both gaseous and particulate forms. The initial large release was due to the mechanical fragmentation of fuel, whilst the second large release between day 7 and 10 was associated with the high temperatures reached in the core melt (NEA, 2002). During the period of release there were frequent changes in meteorological conditions and patterns of deposition were dependent on wind direction and dispersion parameters, particle size and the occurrence of rainfall. Subsequent to deposition, Shestopalov et al. (2003) found that the vertical distribution of radionuclides within the soil, and hence by implication, of groundwater contamination, was influenced by the form of deposition, landscape situation, land use, soil type and soil hydrological and chemical parameters. The transfer of radionuclides from fuel particles to soil solution was found to be directly preconditioned by the dissolution of the particle matrix. This depended on the oxidation state and chemical stability of the particles and on soil acidity. In acid soils the rate of dissolution is significantly greater than in neutral soils where the maximum soil solution concentration may not arise until 10-20 years after the accident.

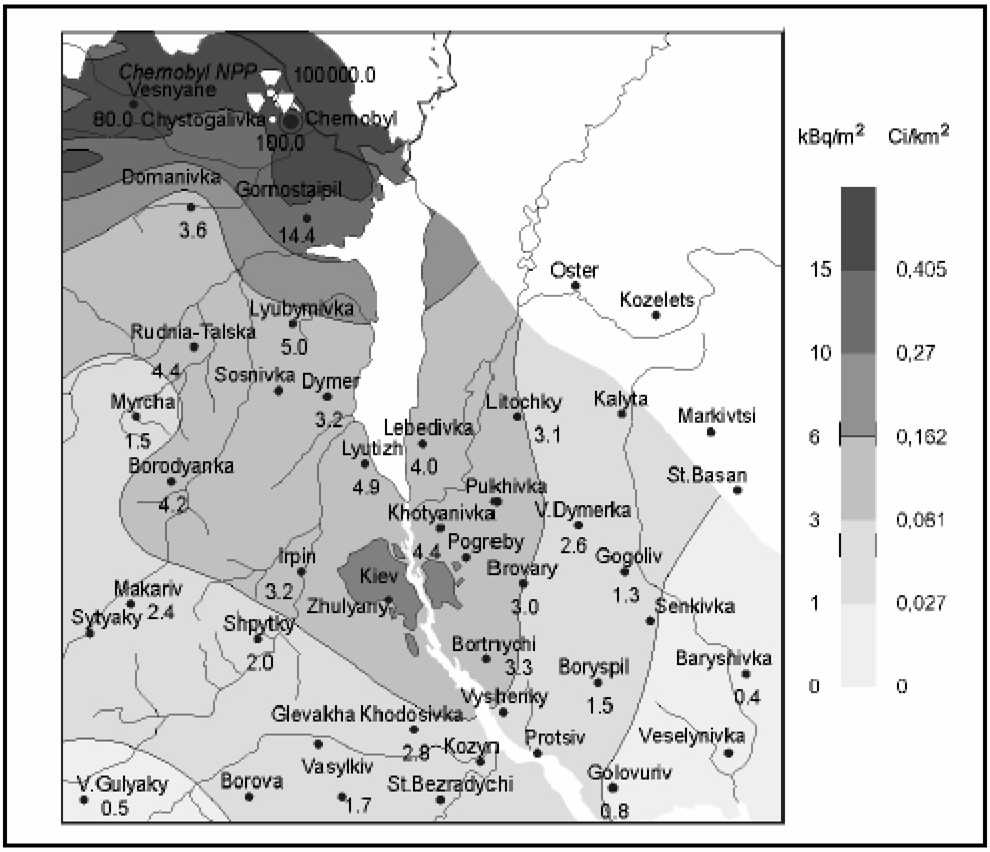

Figure 1. Total ^{137}Cs accumulation in Quaternary and Eocene formations (depth to 80m) within the Kiev region (Shestopalov et al. 2003).

The distribution of landforms, soils and vegetation is significant in terms of the vulnerability of groundwaters to contamination. The highest rate of ^{90}Sr vertical migration was found on immature sandy soils with a low humic content associated with river floodplains. High rates of migration were also observed on arable lands with low levels of organic matter in the Ap horizon. At the scale of a soil profile, Figure 4 shows the vertical distribution of ^{90}Sr within a sandy soil with an intrinsically low moisture retaining capability, a low organic matter content and no vegetation cover. The higher rates of vertical radionuclide migration were attributed to non-equilibrium soil conditions and convectional flow in zone of high local radioactivity (Shestopalov et al. 2003). Figure 5 illustrates the very heterogeneous distribution of contaminant that may arise within a soil profile under these conditions. At the time of sampling there was no correlation between radionuclide concentration and soil moisture content.

Rogachevskaya and Zektser (2003) presented the results of an investigation of shallow groundwater vulnerability within the Dnieper artesian basin, which was one of the areas most polluted by the Chernobyl accident. This study took into consideration the depth of the vadose zone, the hydraulic properties of the soils and the migration behaviour of contaminants, all of which determine the travel time to the water table. The vadose zone mainly comprises Quaternary sediments which are widely used as a decentralised water supply by small settlements. The sediments are very variable in composition but sandy types predominant. The soils and well developed surface water network are closely connected and favour the recharge and discharge of the aquifer. The predominant mode of recharge is local infiltration. Since the Chernobyl accident, Rogachevskaya and Zektser (2003) reported that ^{137}Cs and ^{90}Sr have migrated from the surface to both shallow unconfined and deep confined aquifers. Caesium migration velocities were reported to double below the top soil layer with effective retardation factors significantly lower than those reported in literature sources. Similar findings were reported by Holgye and Maly (2000) who studied the vertical movement of Chernobyl radionuclides in undisturbed grassland soils at three sites in Bohemia. They found that the convection transport rate of the plutonium and caesium increased with depth. Rogachevskaya and Zektser (2003) calculated a dispersion coefficient of 10 cm^2/yr from the vertical concentration distribution relative to the concentration mass-centre and concluded that the soil was not an absolute barrier to radionuclide migration through the vadose zone. The migration rate was observed to vary between types of regional landscape and soil compositional class. Areas subject to human intervention were considered to be particularly vulnerable as were sandy soils.

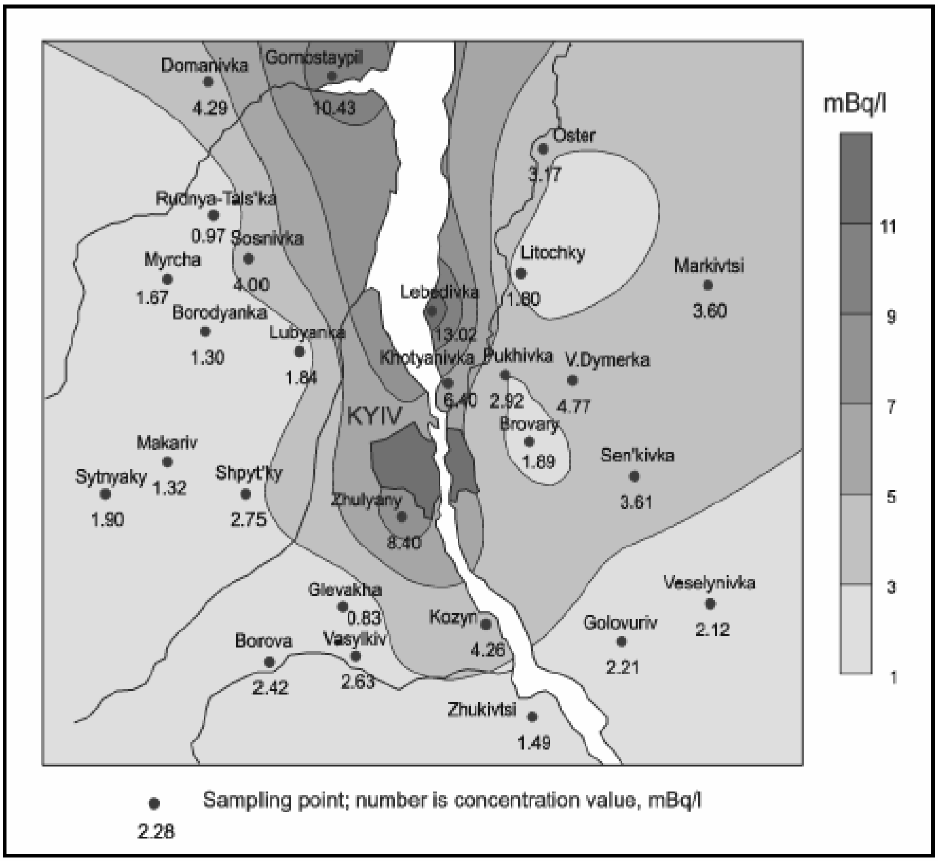

Figure 2. Distribution of ^{90}Sr in a Quaternary water-bearing complex (7-20 m) in 1996

In common with Shestopalov et al. (2003), Sobotovich et al. (2003) noted the penetration of radionuclides below the ground surface following the Chernobyl accident and suggested that as the potential for surface runoff was limited, the main process (other than radioactive decay) for natural attenuation of contamination was through vertical migration. They also noted the dependence of this process on landscape (relief and geographical feature, soil texture and vegetation cover), geochemical conditions (e.g. soil acidity) and on factors influencing the initial mobilization of radionuclides from fuel particles. Sobotovich et al. (2003) analysed observations of contaminant mobility in terms of kinetic-rate constants for radionuclide speciation with particular reference to rates of immobilization and remobilization.

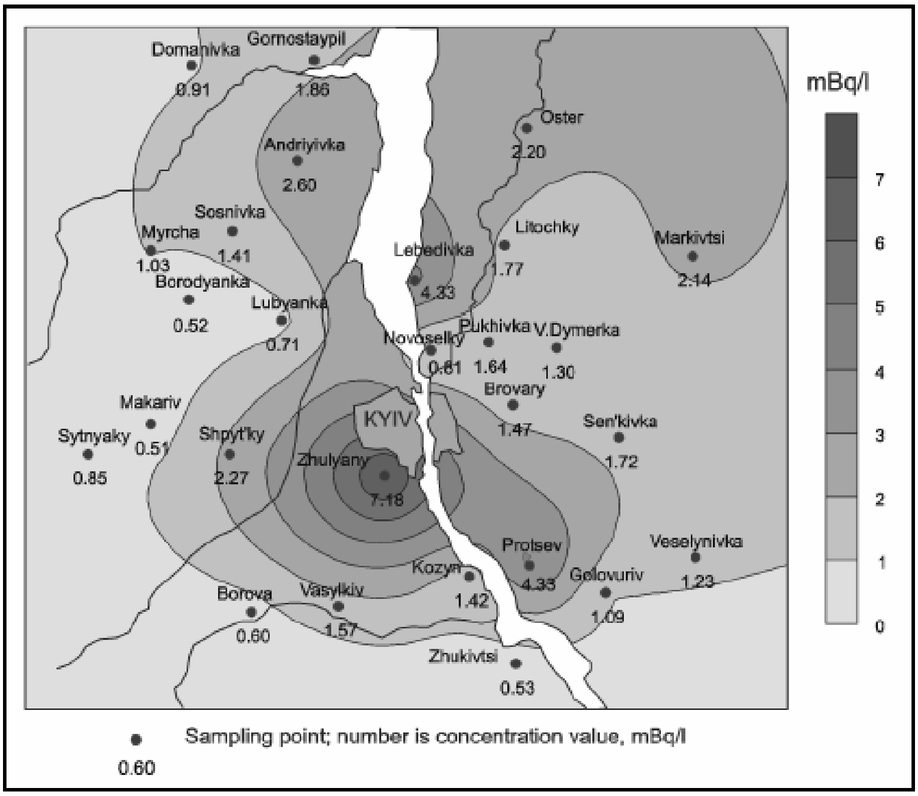

Figure 3. Distribution of ^{90}Sr in the Eocene water-bearing complex (30-60 m) in 1996

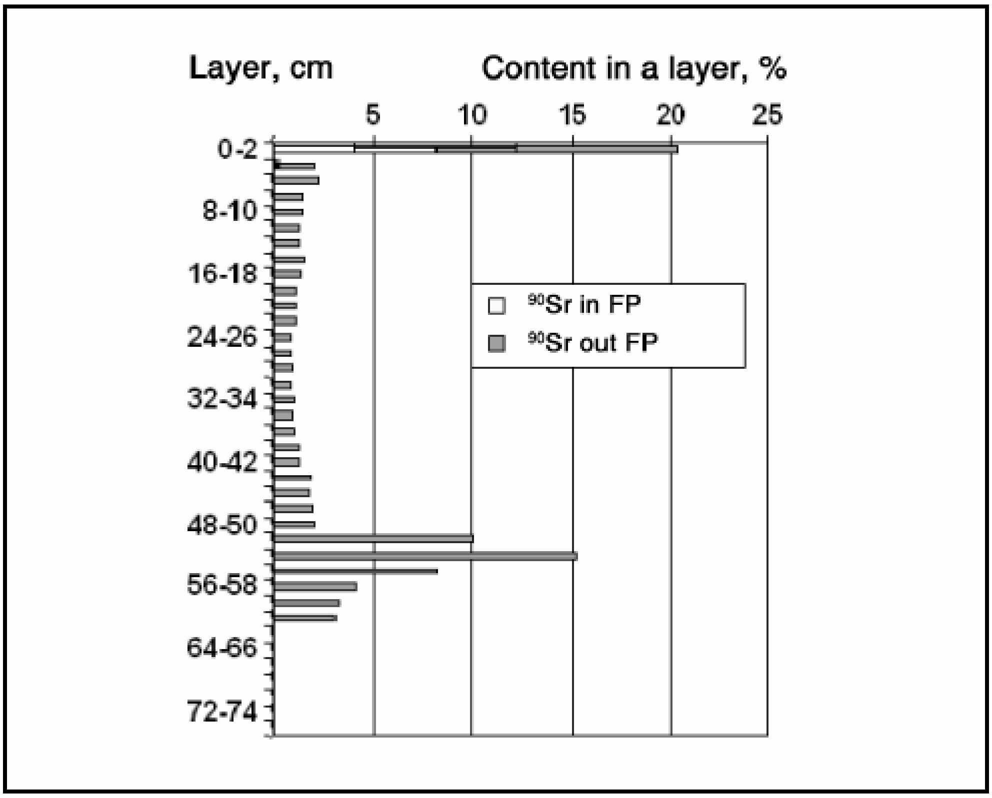

Figure 4 Vertical distribution of ^{90}Sr within a sandy soil in 1996.

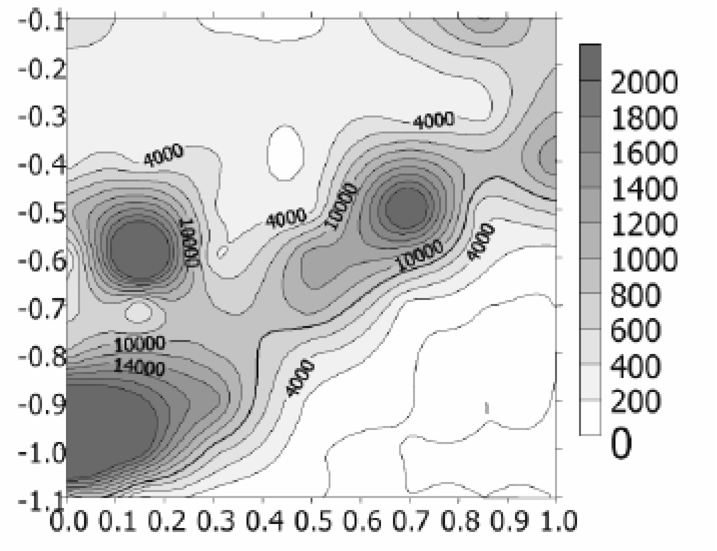

Figure 5. Two-dimensional distribution of ^{90}Sr (Bq/kg) in a pit face excavated into a sandy soil with a low organic matter content (Shestopalov et al. 2003).

Preferential flow is the rapid movement of water and solutes through soils that bypasses a large portion of the soil matrix. As a consequence, the residence time of solutes is shorter than expected based on the assumption of convective-dispersive flow. Field studies have shown that surface applied herbicides, fertilizers, heavy metals, radionuclides and microorganisms are transported faster and to greater depths than predicted, leading to the risk of groundwater contamination (Bevan and Germann, 1982). Hagedorn and Bundt (2002) used a dye tracer to identify preferential pathways in a fine-loamy Dystic Cambisol in a spruce-beech forest in Switzerland. Subsequent analysis showed that within preferential flow pathways, the activities of ^{137}Cs, ^{210}Pb and $^{239,240}Pu$ and concentrations of soil organic matter were substantially enriched relative to those of the soil matrix to depth 50cm, beyond which the differences were not statistically significant. The caesium originated mainly from the Chernobyl accident and most of the local inventory was deposited during a single rainstorm. In contrast, the lead derives from continuous natural atmospheric deposition and the plutonium from nuclear weapons fallout primarily during the 1950s and 1960s. This strongly suggests that the preferential flow paths in the soil studied are persistent for decades.

Speciation and mobility

The concept of the equilibrium distribution coefficient or K_d is almost universally employed in models of radionuclide transport in soils and rocks because it is convenient and because it can be shown to adequately describe the migration behaviour of contaminants under idealized equilibrium conditions. However, whilst a single value of retardation may be retrospectively obtained that adequately describes a known distribution for the bulk of a contaminant in geological media, K_d is often a poor predictor of radionuclide transport within natural or poorly characterized near-surface media. There are many reasons for this, but non-equilibrium conditions, especially within the soil and vadose zones, and speciation offer the most promising explanations. These factors may be particularly significant in accounting for the relative high mobility of small fractions of contaminant or heterogeneous patterns of transport. The potential significance of preferential pathways has been referred to above and clearly heterogeneity and anisotropy in soils physical properties, both intrinsic (e.g. hydraulic conductivity) and environmental (degree of saturation) may produce complex patterns of transport. However, the Chernobyl data suggest that paying more attention to speciation is likely to improve the success of predictive modeling.

Most long-lived radionuclides deposited on the soil from the atmosphere are sorbed rather efficiently by the various soil components (organic matter, clay minerals, sesquioxides). As a consequence, the concentrations of these radionuclides in the solution phase are usually quite low. Nevertheless, this quantity is of fundamental interest for a given radionuclide and for a given soil, because it is this fraction which is most readily available for ecological processes, such as plant uptake or for vertical migration in the soil. The soil solution, however, contains not only dissolved inorganic ions but also (depending on a variety of factors) various amounts of dissolved organic matter (e.g. fulvic or humic acids). The low capacity of many of the soil types characteristic of semi-natural ecosystems to immobilize radiocaesium whilst retaining it in the upper horizons of soil has proved to be the main reason for the continuing high activity concentrations in plants and animals in the UK. The retention within such soils is due to the high, but reversible, sorbing capacity of organic matter and to the low content of clay minerals (Howard, 1998). Desorbing caesium is available for plant uptake and for infiltration within the soil. Caesium associated with organic matter is generally likely to be less available for fixation and hence more available for transport. Burrough et al. (1999) reviewed evidence from research in the Pripyat catchment and found that radiocaesium is highly mobile in both river water and in poorly drained organic soils due to the ecological conditions prevalent in the area.

Speciation determines the potential mobility of radionuclides, but transport may occur in both dissolved and particulate forms and hence is influenced by physical and environmental factors. Radionuclides in dissolved and colloidal form are available for transport to rivers and via groundwaters. Radionuclides that are particle-associated and that reside at the ground surface are susceptible to erosion and overland transport. The coexistence of multiple species of radionuclides in soil solution has attracted considerable interest in explaining the different behaviours of radionuclides in natural systems. Chernobyl has provided valuable data in this regard because it highlighted several aspects of radionuclide behaviour. In particular, the rapid penetration of radionuclides within soil profiles at early times especially under conditions of wet deposition, and the difficulty of modelling the behaviour in soils of radionuclides associated with discrete particles of low initial solubility (SCOPE, 1993).

Amano et al. (1999) studied the availability for transport of Chernobyl radionuclides in undisturbed surface pine forest soils (sandy with surface organic layers) along the River Sahan within the exclusion zone. The speciation (by chemical extraction) of radionuclides (Cs, Sr, Pu and Am) in bulk surface soils was compared with the speciation (determine using ultrafiltration) in the water soluble fraction.

For bulk topsoil, about 90% of ^{90}Sr was found to be in the water soluble and the exchangeable fraction. Very little of the ^{90}Sr and ^{137}Cs were found in the free fulvic and humic acid fractions. Of these radionuclides, ^{90}Sr was mostly found in the water soluble and exchangeable fractions. The majority of ^{137}Cs, Pu isotopes and ^{241}Am in the surface soil were found to be in the humic plus insoluble fraction, but water soluble and exchangeable fractions of ^{137}Cs, Pu isotopes and ^{241}Am were also found. Pu isotopes and Am-241 were mostly present in comparable amounts in the free humic and free fulvic acid fractions, which can dissolve in water.

Within the soil solution phase, most of the Pu (79%) and Am (68%) exist in the molecular weight fractions beyond 10,000 Da (Daltons), in spite of the fact that most of the dissolved organic carbon fraction (69%) exist in the molecular weight below 10,000 Da. This means that transuranic elements, such as Pu and Am, are associated with high molecular weight materials containing carbon such as humic substances in water leachates. On the contrary, most of Cs (85%) and Sr (63%) exist in the molecular weight fraction of below 10,000 Da, reflecting the fact that they mostly exist in lower molecular weight forms or ionic forms associated with low molecular materials. However, some of Cs and Sr exist in the molecular weight band beyond 10,000 Da suggesting that some parts of Cs and Sr is associated with higher molecular materials.

Agapkina et al. (1995) investigated by means of gel filtration the association of radionuclides with soil organic matter in soil solution from forest topsoils from a location 6 km from Chernobyl. The litter, humus and mineral layers within the top 5 cm of the soil were distinguished and differences in speciation noted for the different layers. Plutonium and americium are generally associated with the poorly soluble humic fraction of topsoils which explains their generally low mobility, but they are also found to be mostly associated in solution with the high molecular weight fraction. Whilst americium was found to some extent in lower molecular weight fractions this was not the case for plutonium. Strontium was found to be mostly associated (86%) with low molecular weight organics within the litter layer but was essentially present in inorganic form in the other layers. Caesium in the litter layer was associated mostly (90%) with medium weight organics but in the deepest (mineral) layer was almost uniformly distributed between the 5 fractions distinguished in the analysis. It was concluded that caesium is most sensitive to the nature of the organic matter and plutonium was the least sensitive. Agapkina et al. (1995) felt that as K_d was probably an insufficient measure of mobility and that more attention could be paid to speciation within the solution phase. A further difficulty in employing the concept K_d simplistically is the tendency of radionuclides to become progressively fixed within the mineral phase and thus rendered less mobile over time (SCOPE, 1993).

Organic matter clearly plays a major role in radionuclide behaviour. For example, Epik and Yaprak (2003) suggested that fungal mycelium may play a role in retaining caesium within the upper humic-rich layers of forest soils and may be responsible for a significant fraction of the overall inventory. Kudelsky et al. (1996) looked at the behaviour of Chernobyl-derived caesium in a hydrologically isolated bog system in the catchment of the Pripyat River, Belarus. They found that the runoff coefficients for radiocaesium from peat bogs 8 years after the accident were about one order of magnitude greater than those from unsaturated soils of higher mineral content.

Sobotovich et al. (2003) provide kinetic-rate constants for 5 landscape categories and 4 soil types based on 15 years of observations of radionuclide speciation within the Chernobyl exclusion zone. They noted time-dependence in the rates and inferred changes in the descending and ascending radionuclide fluxes. Because of the nature of the Chernobyl accident, Sobotovich et al. (2003) and other workers devoted much study to the mobilization of radionuclides into soil solution from deposited fuel particles relative to the behaviour of condensed evaporation products. The ‘species’ involved were those identified through chemical extraction so that distinctions were made between water-soluble forms, exchangeable cations, organic-bound forms and ‘fixed’ forms. In this respect it is worth noting that these distinctions do not determine speciation as such, merely extractability and some workers such as Davydov et al. (2002) do not consider this to be scientifically valid or to provide quantification of ‘species’. Thus the kinetic-rate constants provided by Sobotovich et al. (2003) may also be ‘apparent’ rates in much the same way as K_d describes an apparent, net, retardation. Davydov et al. (2002) used an ultrafiltration method and concluded that caesium exists in soil in ionic (molecular) species.

Gri et al. (2000) also consider that the explanation to ^{137}Cs mobility lies in speciation and in the reactivity state of exchange ‘pools’. They were unable to correlate caesium behaviour with soil parameters such as cation exchange capacity and noted mechanisms by which sorption onto clays may be inhibited by organic matter complexes. They concluded that caesium may remain available for cation exchange and hence continue to be available for downward migration in soils. In this respect it is pertinent to note that Hilton et al. (1992) found that, compared with global fallout, the ^{137}Cs from Chernobyl (in far distant locations) was much more mobile in the first four years after the accident. They found that the water-soluble fractions of Chernobyl and global fallout were about 70% and 8% respectively. Later on, ageing processes were said to lead to similar mobility values for caesium from both sources.

Amano and Onuma (2003) employed speciation as determined by sequential extraction to analyse depth profiles of Chernobyl radionuclides in a range of undisturbed soils some 10 years after the accident. They discussed the relative mobility of caesium, strontium, americium and plutonium isotopes under varying soil conditions and found that penetrating fractions of caesium and the actinide elements were generally present. The authors’ identified acidic conditions and organic complexing as factors promoting mobility, but it is not clear whether they were also invoking colloidal phase transport.

The initial pattern of radionuclide deposition was determined by proximity to the Chernobyl site, wind conditions during and immediately after the accident and the incidence of local rainstorms. While these initial conditions determined the regional patterns of concentrations, subsequent work has demonstrated the local mobility of radiocaesium through erosion and deposition within polluted areas, within river systems and also within the food chain. Once adsorbed onto clay minerals,

^{137}Cs is strongly held and further transport in the landscape with mineral matter depends on the processes of erosion and sedimentation.

There have been a number of studies using global (weapons) fallout radionuclides (Cs and Pu) and Chernobyl deposition to determine areas of net erosion and deposition within the landscape (e.g. Panin et al. 2001). The methods used employ reference inventories, depth profiles of tracers and calibration models and generally seem to be more successful at indicating areas of net deposition than areas of net erosion. There will be situations where, for example, particle size fractionation and varying associations with organic matter cause caesium and plutonium to behaviour differently or to be lost from the system under study (e.g. Schimmack et al. 2001).

Radiocaesium deposited on organic soils may remain mobile (Howard, 1998) and may be lost through leaching to the river system. From there it may be redistributed in solution and become associated with suspended sediment (e.g. Walling et al. 1998). Seasonal flooding has been used historically to fertilise riparian areas of the landscape and this process may lead to the accumulation of contaminants (e.g. Walling and He, 1997a; b).

Burrough et al. (1999) investigated the hypothesis that there is a relationship between flood events and the distribution of radiocaesium within the Pripyat catchment of which organic peat soils occupy about 47% of the land area. They also set out to investigate the reasons why some areas within the catchment have seen marked increases in ^{137}Cs levels in milk since the initial deposition. For this purpose they prepared maps of the floodplain and of soil and land cover using a GIS system. They found that sites nearest to rivers generally had larger amounts of radiocaesium than sites further away and that in some areas concentrations in 1993 and 1994 exceed those measured in 1988. This was attributed to a major summer flood in 1993 which submerged a large area for 2-3 weeks. The land classed as rough pasture which occurs almost exclusively within the annually flooded zone seems particularly susceptible to the flood enhancement of ^{137}Cs. Burrough et al. (1999) were not able to establish the role of soil erosion and leaching via groundwater in supplying ^{137}Cs to the river during flood events, but did point to the large areas of organic soils as the source of caesium that was subsequently locally redeposited within the catchment.

Between the major flood in 1993 and 1994 there was a huge apparent loss of ^{137}Cs from the topsoils in the annually flooded zones. This was attributed to leaching from organic soils. However, the decline in ^{137}Cs concentrations in milk was greater than could be explained by leaching alone and progressive fixation by illite was implicated with the clay being derived by overbank sedimentation following flooding. The summer flood caused a brief, but substantial, increase in the ^{137}Cs content of milk. This phenomenon was not observed for spring floods that occur prior to the growing / grazing season. Burrough et al. (1999) suggest that during the 1993 summer flood, submerged plants took up the dissolved ^{137}Cs directly from the river water by root uptake. Another possibility is that cows directly ingested the ^{137}Cs by either drinking the floodwater or by eating contaminated silt which covered the vegetation immediately after the flood.

In temperate climatic regions, erosion by wind and running water is limited by vegetation cover. On arable lands, cultivation may increase the susceptibility to erosion, but it also mixes surface and subsoils and to some extent buries surface deposited contamination. In other areas, earthworms and trampling by cattle may have much the same effect. In forest ecosystems, the Chernobyl experience is of efficient interception of air-borne contaminant and the strong retention and recycling of radionuclides with little opportunity for redistribution by erosion (SCOPE, 1993). In arid areas where the vegetation cover is sparse, much soil can be moved by erosion and surface-derived contamination or contamination due to discharges to ephemeral streams will be retained within soils and sediments. These areas are frequently associated with high-intensity rainfall resulting in flash-flooding which is an efficient means of transporting sediment-associated contaminants. In contrast in upland ecosystems much surface-derived contamination may be retained in organic matter from which the losses (other than by radioactive decay) may be primarily by leaching. Under cold climates, surface erosion during snowmelt may be effective in transporting contaminants (SCOPE, 1993).

Testing of Models using Chernobyl Data

Chernobyl derived data have been used in three modelling studies that are considered here: the Chernobyl Pilot Site Project; the BIOMOVS II 'Wash-off' scenario and the BIOMASS Iput Catchment study.

Chernobyl Pilot Site Project

The Project involved a shallow disposal site located 2.5 km from Chernobyl (Bugai et al., 2002). The trench is about 70 m long, 8-10 m wide and 2-2.5 m deep. It is unlined. The 'waste' comprises heavily contaminated topsoil and pine tree trunks from the "red forest". Groundwater flow below the trench was approximately normal to the long axis of the trench. The Chernobyl Pilot Site Project had two main objectives. One objective was to study the dissolution mechanisms within the near-field of fall-out fuel particles and geochemical interactions within the trench between the soil and dissolved radionuclides. The second objective was to study the hydrodynamics of water and radionuclide transport within the unsaturated zone and in the aquifer underlying the burial trench. The geological situation is comparatively simple, comprising horizontally bedded aeolian (permeable) and alluvial (less permeable) sands of Quaternary age that form an unconfined sandy aquifer overlying, at 30 m, a low permeability Eocene marl layer. For 15 years radionuclides have been leached from the trench by rainwater (550-650 mm/y) and have penetrated through the unsaturated soil (2-3 m vertical distance) into the aquifer within which horizontal transport occurs. Annual recharge amounts to about 40% of precipitation. The inflow into the aquifer is event-based, with most of the recharge occurring during winter thaws and spring snowmelt and occasionally by large rainstorms during the summer-autumn period. The Project focused on the behaviour of ^{90}Sr.

Figure 6 shows the development of the ^{90}Sr plume as at June 2001. At the beginning of the project, it was believed that the site was suitable for a model validation exercise because it was considered to be 'simple' both hydrogeologically and in terms of waste characteristics. However, in both regards, the situation was revealed over time to be more complex than initially envisaged. For example, the aquifer flow was found to be transient and 3-dimensional. As the project progressed and more was learnt about the factors influencing release and transport in the particular situation of the trench and aquifer, the modelling became more successful. The authors were of the view that the experience would be useful in dealing with future accidents. It was clear that a model based on initial assumptions about the wastes and the hydrogeology and using standard literature data values would not have provided a satisfactory projection. Significant factors influencing the outcome were transient effects on sorption interactions within the unsaturated zone and the geochemical evolution of the organic-rich source term.

BIOMOVS II: 'Wash-off' scenario

Konoplev et al. (1996) reported on the results of a validation exercise using data from two experimental plots close to Chernobyl. The experiments were developed within the framework of the BIOMOVS II international study (BIOMOVS II, 1996, Konoplev et al., 1996). One plot was subjected to simulated heavy rain, whilst the other plot used snowmelt. Participants in the exercise were requested to estimate the vertical distribution of total ^{137}Cs and ^{90}Sr and their specific forms in the soil prior to the experiments, concentrations of each radionuclide in surface runoff (separately for particulate and dissolved forms) and the total amount of each radionuclide lost from one of the plots during the experiment.

The experiments provided an opportunity for (1) evaluation of the movement of contaminants from soil to water; (2) calculation of the alteration and migration of contaminants in soil over different time scales; (3) increased understanding of contaminant transport at a process level; and (4) development and use of methods for the estimation of key parameters. A specific objective was to take into account chemical speciation and its effect on the transfer of contamination from soil to water, as well as the geochemical and geophysical processes that affect such transfers.

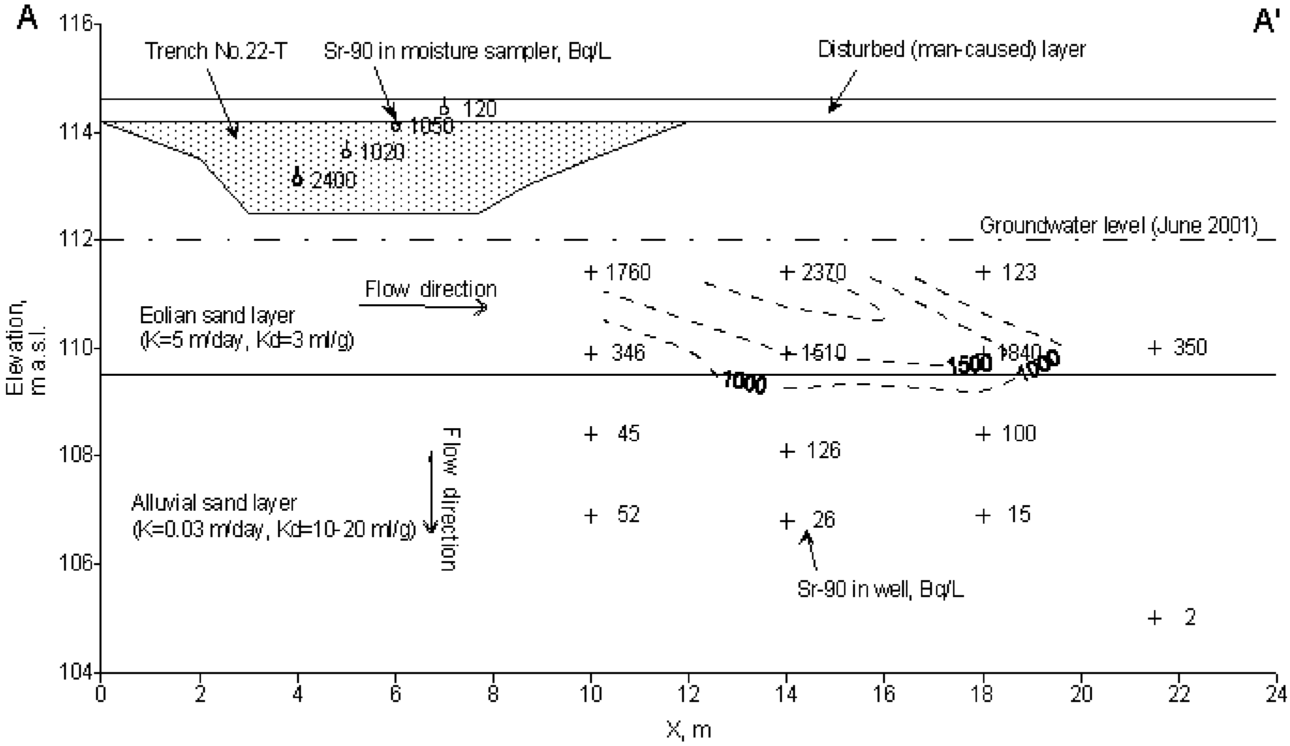

Figure 6. ^{90}Sr distribution in cross-section through the disposal trench and aquifer in June 2001. http://www.eurosafe-forum.org/down2001/semb3_10.doc

The results showed considerable variation between the models and discrepancies with field observations (Figure 7). In general the predictions about speciation chemistry were unrealistic and it was found to be necessary to determine rates constants for the fixation and remobilisation transfers and not to assume the extent to which a radionuclide was present in an exchangeable form. The model that had most success (MOVE) used a method for estimating site-specific the values of the vertical migration parameters and also included convective transport of the dissolved radionuclide fraction. It was concluded that the use of expert judgement based on literature data in the absence of test data reduced the accuracy of predictions and brought additional uncertainty. It was recommended that risk assessment models should include submodels for estimating key model parameters, taking account of local environmental characteristics and those of the radionuclides involved.

BIOMASS: Iput Catchment

The main aim of the Iput river catchment modeling exercise was the reconstruction of the radioecology and assessment of doses within an area that was highly contaminated as a result of the Chernobyl accident (IAEA, 2003). The exercise was focused on concentrations in foods and doses to humans rather than on radionuclide behaviour in soils. However, an adequate description of the behaviour of radionuclides in soils is essential for correctly estimating concentrations in food stuffs.

The models employed for the representation of radionuclide migration in soils varied from the simple empirical to the detailed dynamic. The models differed from each other in the number of physico-chemical processes considered, as well as in the number of model parameters used in the calculations. In the simple models, one or two empirical parameters were used; numerical values of these parameters were taken from the literature or estimated from available data on the vertical distribution of radionuclides in soil.

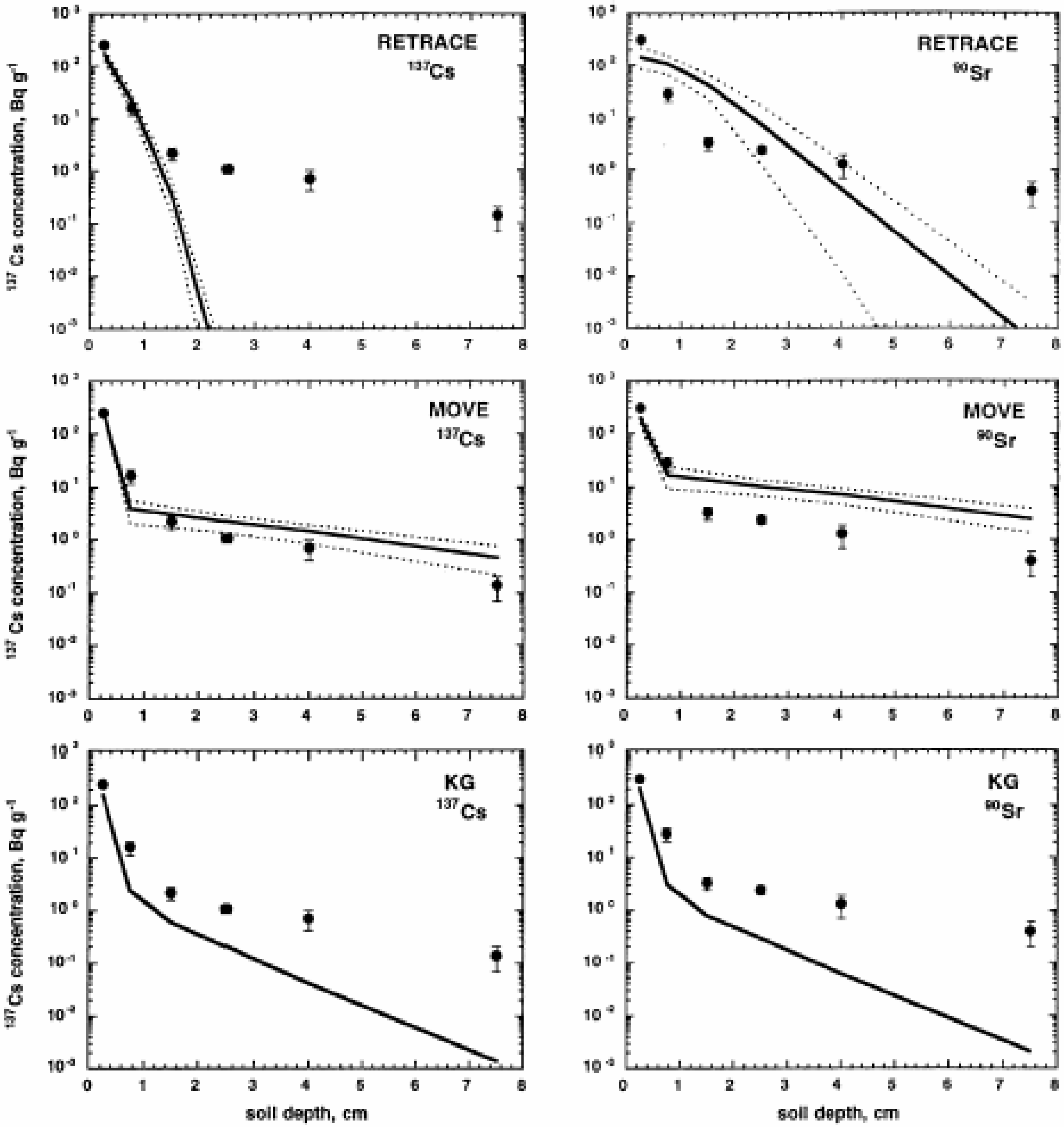

Figure 7. Comparison of predictions using the models RETRACE, MOVE and KG (solid lines) with observations (solid circles) for the vertical distribution of ^{137}Cs and ^{90}Sr in soil. Dashed lines represent 95% subjective confidence intervals on the predictions, vertical bars represent 95% confidence intervals on the observations.

The predicted dynamics of ^{137}Cs concentrations in soil, presented in Figure 8, clearly demonstrate the great variability of predictions obtained with different assumptions about the values of key parameters. The lack of data on site specific values of key parameters representing ^{137}Cs behaviour in soils may be considered as the main source of the discrepancies in the calculation of ^{137}Cs activity in the root zone of plants, which, in turn, results in differences in the prediction of concentrations in agricultural plants. In common with the findings from the BIOMOVS study reported above (Konoplev et al. (1996), radionuclide speciation and the kinetics of transfers between species was indicated to be the most significant consideration in the modelling of radionuclide mobility and plant availability.

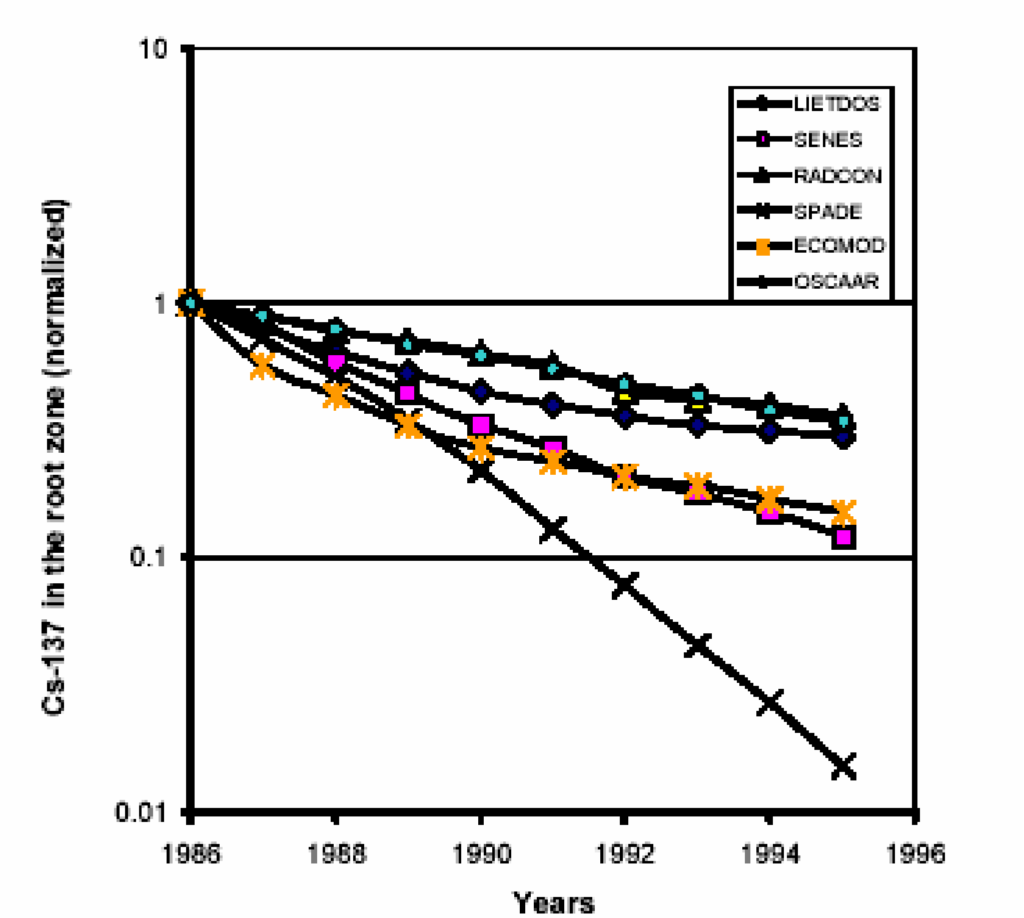

Figure 8. Predicted dynamics of a decrease of bioavailable ^{137}Cs in the Iput test area (arable soils, root zone). Data have been normalized to the predicted values of the initial contamination.

Relevance: The Chernobyl accident has provided many opportunities to study the behaviour of radionuclides under a very wide range of conditions. Because of the intensity of contamination and also because of the amount of research interest generated by the accident, much has been learn about radionuclide behaviour that is potentially relevant to the management of radioactive waste. Information on transport under non-equilibrium situations is particularly helpful as is information on transport within natural (i.e. heterogeneous) media. Although much of the information relates to ^{90}Sr and ^{137}Cs in the surface environment, it is capable of being generalized and has value in relation to other contaminants under different situations.

Research carried out since the Chernobyl accident has reinforced the commonly held view that river floodplains are the landscape setting that should be the focus of attention in near-surface and biosphere models developed for the purpose of assessing the groundwater pathway derived risks that might arise from the disposal of radioactive wastes underground. River floodplains are the areas in which groundwaters are likely to emerge and interact strongly with surface waters. They are the areas most likely to act as sinks for radionuclides. They are readily accessible to humans and are commonly used for pasture for grazing. They are areas where fine grained sediments tend to accumulate by overbank and downslope deposition and hence are favoured locations for the accumulation of strongly sorbed contaminants. Floodplains may be associated with organic soils and hence may retain or accumulate contaminants that tend to readily form organic complexes. The Chernobyl experience has emphasized the need to account for speciation in radionuclide transport models and to be able to predict speciation under different landscape, soil and environmental conditions. The ability to predict speciation under widely varying surface and near-surface conditions should lead to an improved ability to predict the behaviour of contaminants in groundwaters.

Chernobyl has provided new information on radionuclide behaviour in upland, forest and natural ecosystems and as a result, a large dataset exists. It is also the case that radioecological models are available for a wider range of environments than was previously available and they can be employed with greater confidence. Although ^{90}Sr and ^{137}Cs are of limited interest in themselves, they have acted as tracers for transport mechanisms of potential interest to such deep geological disposal concepts. The Chernobyl experience is mainly relevant to an understanding of radionuclide behaviour under unsaturated conditions and within shallow unconfined aquifers. This hydrogeological setting would be suitable for evaluating the conduct of contaminant migration models.

Limitations: Much of the data relating to the Chernobyl accident concerns the behaviour of ^{90}Sr and ^{137}Cs. These radionuclides are of limited interest in terms of the risks associated with geological disposal of solid radioactive waste, but they have acted as tracers for transport mechanisms of potential interest to such disposals. The Chernobyl accident involved deposition of radionuclides to surface soils in the form of particles. This mode of contamination has limited relevance to assessments of the potential risks that may arise in the biosphere as a result of the geological disposal of radioactive wastes, but it provides some insight into near-field processes in shallow disposal facilities. Subsequent dissolution of components within the contaminant particles and their migration through soils has more relevance. The environmental research following the Chernobyl accident was unfocused in terms of the situations relevant to deep disposal, although it does include floodplain and aquatic environments and stream-aquifer interactions.

Although the Chernobyl dataset is extensive and is available for evaluating the predictive power of models, they can generally be made to replicate field data through an iterative process of trial and error during which 'effective' values of parameters are determined and retrospectively justified. This does not necessarily increase the preparedness of models and modellers to deal with future situations.

Quantitative information: Data are generally available on radionuclide concentrations and depth profiles in a variety of soils, as a function of both depth and time. These data are available for testing hypotheses about radionuclide behaviour. The data on radionuclide sorption, speciation and kinetics are probably the most valuable in this regard. The data on soil characteristics are variable as are the data on contaminant speciation with different quantities being reported using different methods.

The model testing and intercomparison exercises provide sources of data that may be helpful in the development of models designed to predict radionuclide speciation under a variety of conditions.

Uncertainties: The Chernobyl accident was, in effect, an uncontrolled experiment with regard to radioecological research. Natural systems are complex and their interpretation is associated with uncertainty. In the case of Chernobyl this uncertainty is increased by lack of information on the form, composition and spatial variation in contaminant deposition. Natural systems are anyway complex and their interpretation tends to be ambiguous. The data on 'speciation' acquired by means of chemical extraction procedures does not provide unambiguous information of chemical species.

Time-scale: The time-scale is that since the Chernobyl accident in 1986 and has provided valuable insights into short and medium term radionuclide mobility.

PA/safety case applications: Experience from the Chernobyl accident has improved knowledge of the ways in which soils influence rates of transfer of contaminants between solid and solution phases and influence the mobility and bioavailability of contaminants. The Chernobyl accident has also shed light on the processes that cause radionuclides to be redistributed within the biosphere. Experience from the Chernobyl accident continues to improve the models and data available for safety assessments.

References:

Agapkina G, Tikhomirov F and Shcheglov A. 1995. Association of Chernobyl-derived $^{239+240}$Pu, ^{241}Am, ^{90}Sr and ^{137}Cs with organic matter in the soil solution. J. Environ. Radioactivity, 29, 3, 257-269.

Amano H, Matsunaga T, Nagao S, Hanzawa Y, Watanabe M, Ueno T and Onuma Y. 1999. The transfer capability of long-lived Chernobyl radionuclides from surface soil to river water in dissolved forms. Organic Geochemistry, 30, 437-442.

Amano H and Onuma Y. 2003. Depth profiles of long lived radionuclides in Chernobyl soils sampled around 10 years after the accident. J. Radioanalytical and Nuclear Chemistry, 255, 1, 217-222.

Bevan K and Germann P. 1982. Macropores and water flow in soils. Water Resour. Res. 18, 1311-1325.

BIOMOVS II. 1996. Wash-off of ^{90}Sr and ^{137}Cs from two experimental plots: Model testing using Chernobyl data. BIOMOVS II Technical Report No. 9, Swedish Radiation Protection Institute, Stockholm, Sweden.

Bugai D, Dewiere L, Kashparov V, Skalskyy A, Levchuk S and Barthes V. 2002. Chernobyl case study increases confidence level in radionuclide transport assessment in the geosphere. http://www.eurosafe-forum.org/ipsn/pdf/euro2_3_9_chernobyl_cases_geosphere.pdf

Burrough P, van der Perk M, Howard B, Prister B, Sansone U and Voitsekhovitch O. 1999. Environmental mobility of radiocaesium in the Pripyat catchment, Ukraine/Belarus. Water, Air and Soil Pollution, 110, 35-55.

Davydov Y, Voronik N, Shatilo N and Davydov D. 2002. Radionuclide speciation in soils contaminated by the Chernobyl accident. Radiochemistry, 44, 3, 307-310.

De Cort et al. 1998. Atlas of caesium deposition on Europe after the Chernobyl accident. EUR report nr. 16733, EC, Office for official publications of the European Communities, Luxembourg [reproduced in part in: NEA 2002].

Epik O and Yaprak G. 2003. Distribution of radiocaesium in coniferous forest soils around Izmir. Fifth General Conference of the Balkan Physical Union (Poster presentation).

Gri N, Stammose D, Guillou Ph. and Genet M. 2000. Mobility of ^{137}Cs related to speciation studies in contaminated soils of the Chernobyl area. J. Radioanalytical and Nuclear Chemistry, 246, 2, 403-409.

Hagedorn F and Bundt M. 2002. The age of preferential flow paths. Geoderma, 108, 119-132.

Hilton J, Cambray R and Green N. 1992. Chemical fractionation of radioactive caesium in airborne particles comtaining bomb fallout, Chernobyl fallout and atmospheric material from the Sellafield site. J. Environ. Radioact. 2: 103-111.

Holgye Z and Maly M. 2000. Sources, vertical distribution and migration rates of $^{239, 240}$Pu, ^{238}Pu and ^{137}Cs in grassland soil in three localities in central Bohemia. J. Environ. Radioactivity, 47, 2, 135-147.

Howard B. 1998. ITE scientific report 1997-1998.

IAEA, 2003. Testing of environmental transfer models using Chernobyl fallout data from the Iput river catchement area, Bryansk region, Russian Federation. Report of the dose reconstruction working group of BIOMASS Theme 2. IAEA-BIOMASS-4, IAEA, Vienna.

Konoplev A, Bulgakov V, Popov AA, Popov VE, Scherbak OF, Shveikin AV, V Yu. and Hoffman FO. 1996. Model testing using Chernobyl data: I. Wash-off of ^{90}Sr and ^{137}Cs from two experimental plots established in the vicinity of the Chernobyl reactor. Health Physics, 70, 8–12.

Kudelsky A, Smith J, Ovsiannikova S and Hilton J. 1996. Mobility of Chernobyl-derived ^{137}Cs in a peatbot system within the catchment of the Pripyat River, Belarus. Sci. of the Total Environ., 188, 2-3, 101-133.

NEA. 2002. Chernobyl – Assessment of radiological and health impacts. 2002 update of Chernobyl: Ten years on. NEA / OECD.

Panin A, Walling D and Golosov V. 2001. The role of soil erosion and fluvial processes in the post-fallout redistribution of Chernobyl-derived ^{137}Cs: a case study of the Lapki catchment, Central Russia. Geomorphology, 40, 3-4, 185-204.

Rogachevskaya L and Zektser I. 2003. Assessment of groundwater vulnerability to radionuclide contamination after the Chernobyl accident with the Dnieper artesian basin. Environ Sci & Pollut Res Special Issue 57-62.

Schimmack W, Auerswald K and Bunzl K. 2001. Can $^{239+240}$Pu replace ^{137}Cs as an erosion tracer in agricultural landscapes contaminated with Chernobyl fallout. J. Environ. Radioactivity, 53, 1, 41-57.

SCOPE. 1993. Radioecology after Chernobyl. Scientific Committee on Problems of the Environment. IUGG.

Shestopalov V, Kashparov V and Ivanov Y. 2003. Radionuclide migration into the geological environment and biota after the Chernobyl accident. Environ Sci & Pollut Res Special Issue 39-47.

Sobotovich E, Bondarenko G and Dolin V. 2003. Biogenic and abiogenic migration of ^{90}Sr and ^{137}Cs of Chernobyl origin in terretrial and aqueous ecosystems. Environ Sci & Pollut Res Special Issue 31-38.

Walling D and He Q. 1997a. Investigating spatial pattens of overbank sedimentation on river floodplains. Water, Air and Soil Pollution, 99, 9-20.

Walling D and He Q. 1997b. Use of fallout ^{137}Cs in investigations of overbank sediment deposition on river floodplains. Catena, 29, 263-282.

Walling D, Owens P and Leeks G. 1998. The role of channel and floodplain storage in the suspended sediment budget of the River Ouse, Yorkshire, UK. Geomorphology, 22, 225-242.

Added value comments: Each case study of radionuclide behaviour under different circumstances adds to the ability to respond to or anticipate radionuclide behaviour under future situations. The Chernobyl data clearly demonstrate that pathways exist for the rapid and widespread vertical transfer of radionuclides from soils to groundwaters via the vadose zone. This includes those radionuclides commonly considered to be strongly particle-reactive and hence with a strong tendency to be immobilised in geological media, The transport mechanisms of interest include convective flow under non-equilibrium conditions, the presence of preferential pathways and the existence of mobile species or of the conditions necessary to maintain mobility.

Potential follow-up work: Further consideration of the potential for developing assessment models that can more readily accommodate the complexity of natural systems in respect of both physical and chemical transport processes. The determination of specific radionuclide species in geological media to replace apportionment based on selective chemical extraction. Replacement of K_d-based models by models that employ kinetic speciation data and can take account of soil compositional and environmental information. There is a need to explore methods of determining radionuclide speciation in as close to an in-situ condition as is possible. The most commonly reported methods employ extractants that change the system being investigated. The early phase, high mobility of Chernobyl radionuclides has not been adequately explained.

The model tests that have been conducted using Chernobyl data indicate the need to improve the capability to predict radionuclide mobility under non-equilbrium conditions. There is a requirement to have available models for the prediction of radionuclide speciation under different and varying environmental conditions.

Keywords: Chernobyl, preferential flow paths, speciation, groundwater, soil, redistribution, model testing

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