

Characterisation of radioactive particles in the environment†

Brit Salbu, Trygve Krekling and Deborah H. Oughton

Laboratory for Analytical Chemistry, Agricultural University of Norway, N-1432 Aas, Norway

When radioactive particles are released into the environment, information on the size distribution pattern, radionuclide and matrix composition, morphology and structure is essential for assessing weathering and the subsequent mobilisation and biological uptake of associated radionuclides. The particle characteristics will depend on the source in question in addition to the release scenario, dispersion processes and deposition conditions. Following high temperature accident scenarios associated with nuclear installations (*e.g.*, Chernobyl accident) a range of different uranium fuel particles and condensed particles were observed, varying in composition, morphology and structure (*e.g.*, crystalline and amorphous phases). Following low temperature releases (*e.g.*, prefire Winscale releases), flake-like uranium fuel particles significantly different from those collected at Chernobyl were identified. Furthermore, a major fraction of radionuclides in effluents from reactors and reprocessing plant during normal operations are associated with particles and colloids. Hence the presence of radioactive particles or colloids in releases from nuclear sources occurs more frequently than usually expected. After deposition, weathering of particles occurs and associated radionuclides are mobilised with time. Hence the transfer of mobilised radionuclides within the ecosystem will be delayed until weathering takes place, and the assessment of short term consequences of releases may be overestimated if particles are present. The weathering rate will depend on the particle composition (*e.g.*, UO_2 fuel), structural changes occurring during the event (*e.g.*, transformation from UO_2 to U_3O_8) and chemical conditions after deposition (*e.g.*, pH, redox). Unless the impact of particle weathering is taken into account, assessment of mobilisation, transfer and long-term consequences of radionuclide releases may be underestimated. In order to improve the predicting power of transport models, ecosystem transfer models and dose assessment models, experimental information is required with respect to source term radionuclide speciation and in particular the association of radionuclides with particles and colloids, influencing mobility and biological uptake. This work was therefore focused on analytical techniques applicable to the fractionation, identification and characterisation of radioactive particles and colloids (*e.g.*, hollow fibre fractionation, electron microscopic techniques and reactivity studies) released from a source and deposited in the environment. Results from research carried out during several years at the authors' laboratory are presented to illustrate the usefulness of the techniques.

Keywords: Radioactive particles; radionuclides; source term; weathering; mobility

In order to assess short- and long-term consequences of discharges, atmospheric fallout or leakage of radionuclides from a source, information on the source term is essential. Usually source term characteristics are restricted to inventory estimates, activity levels or activity concentrations of released or deposited radionuclides, while information on the physico-chemical forms (*e.g.*, ions, colloids, particles) is scarce. However, the information on speciation is essential for assessing the mobility and bioavailability of radionuclides in various ecosystems and potential biological effects.

Released radionuclides may be present in different physico-chemical forms¹ varying in size (molecular mass distribution), structure and charge properties. These factors will influence mobility and biological uptake. Radionuclides associated with colloids and particles are relatively inert whereas low molecular mass (LMM) species are believed to be mobile and bioavailable. Particles containing actinides and fission and/or activation products are most often related to accident scenarios, *e.g.*, the Winscale pile fire in 1957,² the B-52 aircraft crash at Thule in 1968,³ the satellite Cosmos 954 crash in Canada in 1978⁴ and the Chernobyl accident in 1986.^{5–7} However, particle releases during normal operations at nuclear installations are also reported, *e.g.*, in atmospheric releases at Winscale⁸ during the early 1950s and in effluents from reprocessing plants^{9,10} (*e.g.*, Sellafield, UK, or Mayak Production Association, Russia) and nuclear reactors.¹¹ The presence of refractory radionuclides (*e.g.*, ^{95}Zr , ^{144}Ce) may indicate release of fuel particles, and the activity ratios may reflect fuel burn-up. Fuel particles or clusters may, however, be depleted to a variable extent in volatile radionuclides (*e.g.*, ^{137}Cs and ^{90}Sr), depending on release temperature. Owing to condensation of volatiles on available particle surfaces during release and on aerosols during transport, condensed particles may also form. Hence, the particular source and release scenario, course of events, distance from the source, dispersion and deposition conditions will influence the size distribution pattern, composition and structure of released 'hot' particles observed in the environment.

After deposition, weathering of particles occurs and associated radionuclides are mobilised with time. The particle composition (*e.g.*, UO_2 fuel), structural changes (*e.g.*, transformation of UO_2 to U_3O_8 during release) and the chemical conditions (pH, redox, microbial activity) at the deposition site will influence the weathering rate. Mobilised radionuclides may also interact with soil or sediment components (*e.g.*, reversible and irreversible sorption). Hence the relationship between the weathering rate of particles and rate of irreversible sorption (fixation) on soils is an essential parameter for estimating the potential transfer of released radionuclides within the ecosystem over time. However, most models assessing the transfer and consequences of radionuclide contamination include processes relevant to LMM ionic species only. If inert particles are present, the transfer of mobile radionuclide species will be delayed until weathering occurs, and the assessments of short-term consequences will be overestimated. Furthermore, the assessments of long-term consequences will be underestimated if time-dependent particle weathering and radionuclide mobilisation are not taken into account. Unless the impact from

† Presented at The Third International Symposium on Speciation of Elements in Toxicology and in Environmental and Biological Sciences, Port Douglas, Australia, September 15–19, 1997.

particles is included, model predictions may suffer from large uncertainties.

Source term information, including particle characteristics, serves as input to dispersion or transport models. Thus, experimental information on the size distribution pattern, composition and structures of radioactive particles and colloids released from different sources is needed for modelling purposes. In this paper we discuss available analytical techniques which can be utilised for the fractionation, identification and characterisation of radioactive particles and colloids. Results from research carried out during several years in our laboratory are presented to illustrate the techniques discussed and to highlight the impact of radioactive particles on mobility and biological uptake.

Size distribution pattern and structure analysis

Radionuclides associated with colloidal or particle material in waters can be collected by using *in situ* or on-site fractionation techniques such as tangential flow systems (*e.g.*, hollow fibre ultrafiltration). For soils and sediments, an inhomogeneous distribution of radionuclides indicates the presence of radioactive particles. This can most often be confirmed using autoradiography, followed by electron microscopy (EM) and X-ray microanalysis to obtain information on size, structure and surface elemental distributions.

Size fractionation techniques for aquatic samples

In aquatic systems, several techniques are available for the fractionation of radionuclides associated with colloidal or particulate material. Among the techniques, hollow fibre ultrafiltration or tangential flow systems are advantageous as sampling and fractionation can take place on-site.^{11,12} By means of a peristaltic pump, the water is sampled and transferred directly into the molecular mass discriminator with pore size cut-off values in the range 1–10⁵ Da and ultrafiltrates can be collected. For volumes in the range 1–100 l, hollow fibre ultrafiltration can be utilised, while tangential cross-flow cassettes can be applied for larger volumes. The pore size distribution of hollow fibre membranes has been proved to be relatively narrow, clogging is negligible owing to the high internal fibre flow and sorption is of no significance when a sample aliquot is used for conditioning purposes.¹³ When hollow fibre ultrafiltration is interfaced (on-line) with ion-exchange chromatography (cation, anion, active carbon resins), radionuclides associated with charged and neutral colloidal species can be differentiated simultaneously. The cassette membranes are, however, less well defined and memory effects reflecting sorption processes may influence the results. Following fractionation, further analysis (*e.g.*, EM with X-ray microanalysis and α -, β - or γ -spectrometry) provides information on particle structure, size distribution, elemental composition and radionuclide species.

The systems described have been utilised for determination of radionuclides associated with colloidal material in river and lake waters,¹⁴ soil waters¹⁵ and waters from the Irish Sea and Kara Sea.¹⁶ Hollow fibre ultrafiltration has also been used for the fractionation of radionuclides in effluents from Swedish reactors¹¹ and from the Sellafield nuclear installation in the UK.⁹ The results demonstrated that a major fraction of γ -emitting radionuclides and transuranics were associated with colloidal or particulate material. Similar results have recently been obtained for radionuclides in effluent from the nuclear installation at La Hague, France. As these sources represent the major input of radionuclides to waters adjacent to the North Sea, it is essential that models assessing transport, biological uptake and consequences for the marine ecosystem includes the impact of colloidal and particulate fractions.

Autoradiography

Following the Chernobyl accident, 'hot spots' have been identified in the field close to Chernobyl (Ukraine) and further away (*e.g.*, Norway) using portable GM tubes or NaI detectors. Autoradiography has been used to obtain radioactive particles in the soil samples prior to EM analysis. Dried samples (soil, sediment, vegetation) collected from hot spots are thinly spread on paper or plastic foils and the X-ray film (*e.g.*, Kodak X-OMAT AR-5) is placed in close contact for varying exposure times. For activity levels in the range 1–10 Bq the exposure time is several months. Alternatively, film sensitive to α -radiation is applied if Pu particles are expected. Hot spots on the film reflect particles and further separations of subsamples collected from the foils are made with the aid of a GM tube or α -detector and light microscope prior to the use of EM techniques.

Structure analysis by scanning electron microscopy (SEM) and X-ray microanalysis (XRMA)

Subsamples containing particles are mounted on stubs, usually carbon coated to avoid artefacts due to charging, and analysed at different magnifications by scanning electron microscopy (SEM) with X-ray microanalysis (XRMA) [Fig. 1(a)]. Using the backscattered electron imaging (BEI) mode, bright areas reflect the presence of high atomic number elements on particle surfaces [Fig. 1(b)] and the distribution of elements in the upper particle surface layer is attained using X-ray mapping [Fig. 1(c)]. XRMA also provides information on the elemental composition [Fig. 1(d)]. These techniques have been utilised to characterise different types of radioactive particles, *e.g.*, flake-like uranium fuel particles released under low temperature conditions from Winscale, UK, and large aggregates of fuel granulates and crystalline and amorphous single fuel particles released at high temperature from the Chernobyl reactor, in addition to small, spherical uranium particles collected in soil samples from Norway about 2000 km from Chernobyl.^{17,18}

Although all these particles are uranium fuel particles, their behaviour in the environment is expected to be different; the transport of spherical particles is different from that of flakes, and the weathering rates of amorphous structures are significantly higher than those of crystalline phases. Unless these processes are taken into account, model predictions on soil-plant transfer may suffer from large uncertainties. Radioactive particles, different from those associated with accident scenarios, are also identified in contaminated sediments, *e.g.*, in the Irish Sea due to effluents from the Sellafield nuclear installation and in the Abrosimov Fjord at Novaya Zemlya¹⁶ due to dumped radioactive waste (Fig. 2). To assess whether the contaminated sediments will act as a diffuse source of mobilised radionuclides in the future, information on particle characteristics and weathering rates is needed.

Structure analysis by transmission electron microscopy (TEM)

For colloidal radioactive material, transmission electron microscopy (TEM) with XRMA can be utilised for structure and elemental analysis. For unfiltered or fractionated water samples, droplets are transferred to Formvar-coated grids and carefully dried under a UV lamp prior to analysis. In TEM, electron dense structures can be recognised, when compared with a blank (distilled water). As demonstrated in Fig. 3, colloidal material with particle sizes close to 20 nm and pseudocolloids in the region of 100 nm were present in the Sellafield effluents.⁹ The colloidal material was obtained by hollow fibre fractionation. The radionuclide measurements demonstrated that a major fraction of ^{239,240}Pu, ⁹⁵Nb/⁹⁵Zr and ¹⁰⁶Ru/¹⁰⁶Rh was associated with particles which may settle outside the pipeline; a minor fraction of ^{239,240}Pu and ⁹⁰Sr was present as colloids which are

transported in the water volume and are of relevance to uptake by filtering organisms; and ^{99}Tc , ^{137}Cs and ^{135}Sb were predominantly present as LMM forms and are of relevance to uptake by fish, for instance.

Particle influence on reactivity

When weathering of radioactive particles occurs in heterogeneous systems, *e.g.*, soil–water or sediment–water systems, mobilised radionuclides may distribute between LMM and HMM forms in soil water, or be reversibly or irreversibly bound to solid phases (Fig. 4). Thus, the distribution of radionuclide species will change with time owing to climatic conditions (weathering due to acidification, freezing), microbial activity and interactions of mobilised radionuclides with naturally occurring components (*e.g.*, humic substances, clay material). The chemical behaviour and reactivity of mobilised radionuclide species will be different from those of inert radioactive particles, and the rate of weathering relative to the rate of soil association will determine the transfer of radionuclides within the ecosystem. Within the 30 km zone of Chernobyl, the rate of ^{137}Cs fixation to soil components is more rapid than the weathering rate of fuel particles, whereas for ^{90}Sr the rate of fixation is slower than the rate of release.¹⁹ Hence the mobility and biological uptake of ^{90}Sr have been expected to increase with time.²⁰

Information on charge properties of radionuclides, interactions and degree of binding to hot particles or to natural components in soils can be obtained from reactivity studies, *e.g.*, investigations of reversible and irreversible sorption processes (physical sorption, electrochemical sorption and chemisorption) by extraction procedures, sorption isotherms, distribution coefficients and mobility factors. When sequential extraction of soils or sediments containing radioactive particles is performed, the distribution of radioactive and stable isotopes of, *e.g.*, Cs and Sr, in the extracted fractions will be significantly different.

Sequential extraction of soil and sediments

Sequential extractions^{17,20–22} are useful techniques for studying reversible or irreversible interactions of radionuclides with solid phases. The reagents should be chosen in order to differentiate between binding mechanisms: (a) reversible physical sorption by using inert electrolytes, *e.g.*, water, followed by 1 M NH_4OAc at soil pH; (b) reversible electrochemical sorption by using the pH effect, *e.g.*, 1 M NH_4OAc at pH lower than the soil pH; or (c) irreversible chemisorption by using redox systems, *e.g.*, hydroxylamine, H_2O_2 in 1 M HNO_3 and 7 M HNO_3 , sequentially. The fraction of radionuclides which is easily displaced (*i.e.*, H_2O , NH_4OAc extractable) can readily be distinguished from that being released after dissolution (break-

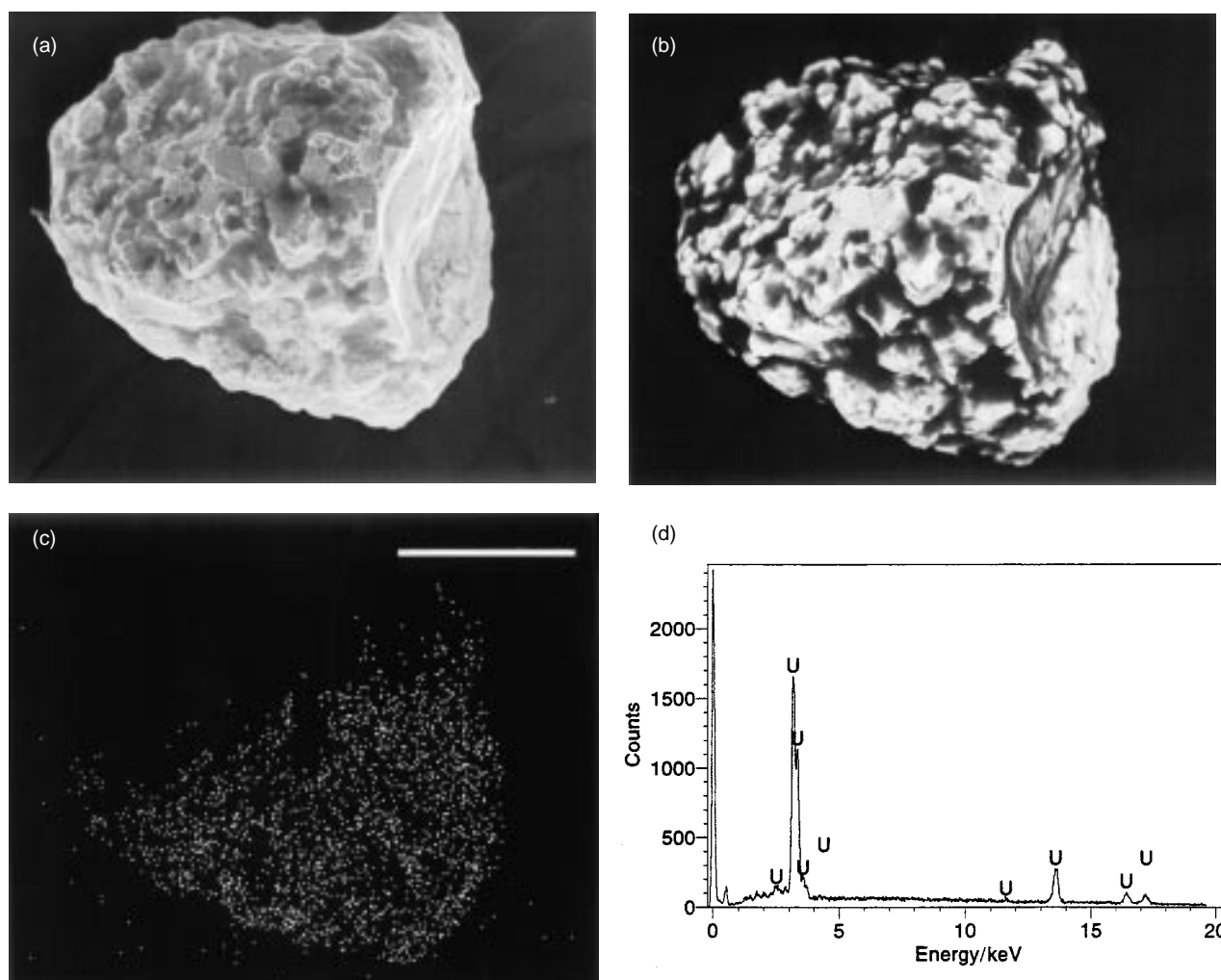


Fig. 1 Fuel particles collected within the 30 km zone of Chernobyl, 1987: (a) secondary electron imaging (SEI) mode, (b) backscattered electron imaging (BEI) mode, bright areas reflect high atomic number elements; (c) X-ray mapping of uranium; (d) elemental analysis by XRMA. Bar = 100 μm .

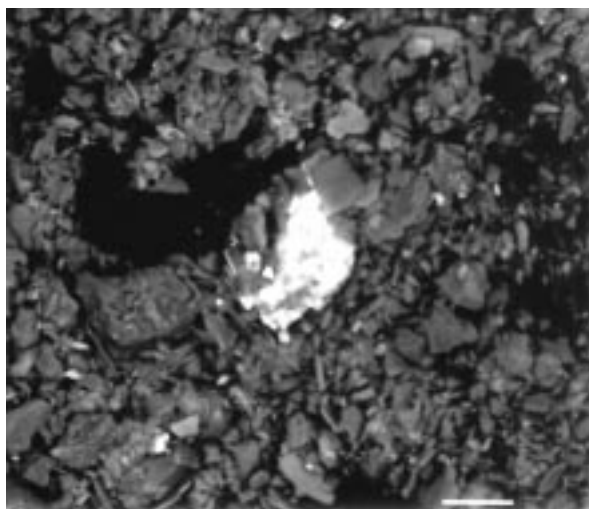


Fig. 2 Radioactive particles containing ⁶⁰Co in sediments from the Stepovogo Fjord, Novaya Zemlya (bright area, BEI mode). X-ray microanalysis showed that the corroded particle (about 10 × 20 μm) contained stable Co, Fe, Mn and Cr (*i.e.*, crude).

age of bonds due to redox agents). As reversibility is a matter of kinetics, the time needed for reversible processes (*e.g.*, displacement reactions, physical or electrochemical sorption) to reach equilibrium may be considerable. Hence the chosen contact time should at least allow rapid surface processes to occur. However, attention should be paid to the slow diffusion processes when long-term assessments are made. Another crucial point concerns the separation of solids and solution, whether colloidal material is included in the aqueous or solid phase. Using centrifugation at 10 000g or more, most of the colloidal and particulate material in soils is retained in the solid phase. Based on representative sample aliquots, the precision of the sequential extraction method has proved to be within 10%.²⁰⁻²² To account for differences in the soil chemistry between individual samples, it is beneficial to include measurements of stable isotopes or analogues in addition to the radionuclides in the fractions obtained. Comparison of the extraction yields gives information on particle association and the degree of isotopic exchange.

For reversibly (physical or electrochemical sorption) associated radionuclides, the effect of dilution can be demonstrated by repeating the H₂O or the inert electrolyte extraction. By extracting contaminated soils, swamp and sediments collected at Mayak, Urals, 10 times with the same reagent, a maximum of

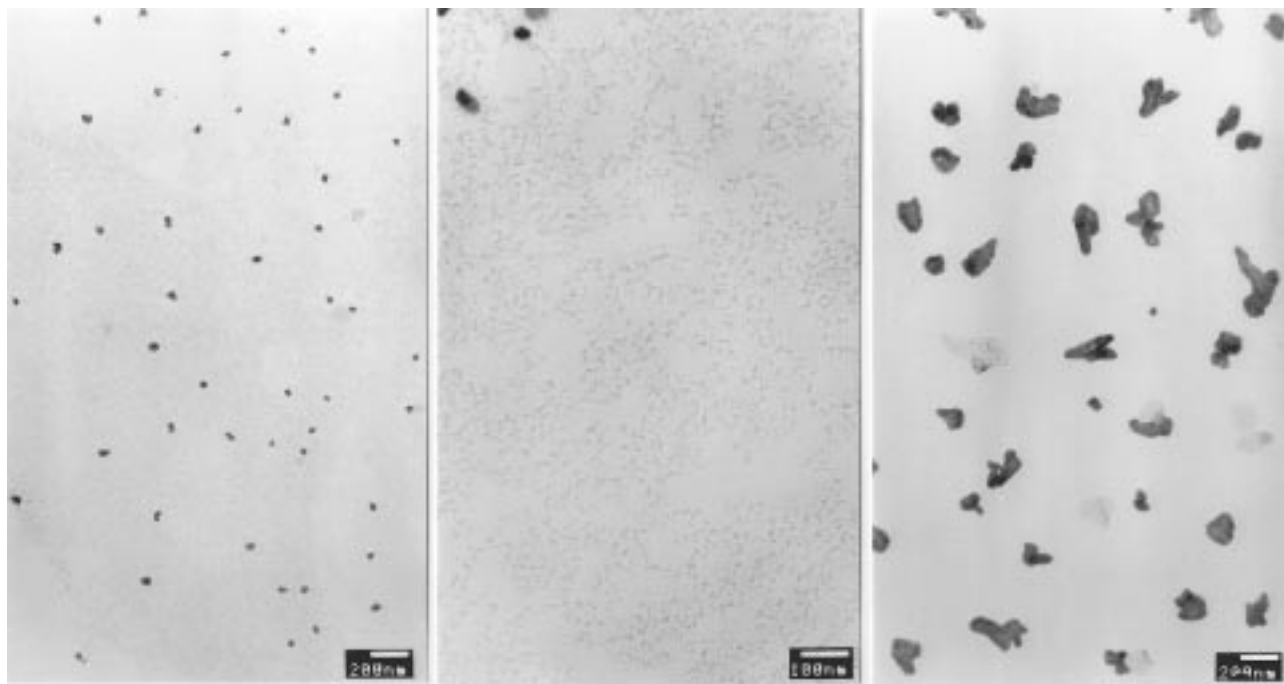


Fig. 3 TEM of colloidal material in effluents from Sellafield reprocessing plant, UK.⁹ Left, effluent from SIXEP unit; middle, distilled water; right, effluent from Seatank Unit.

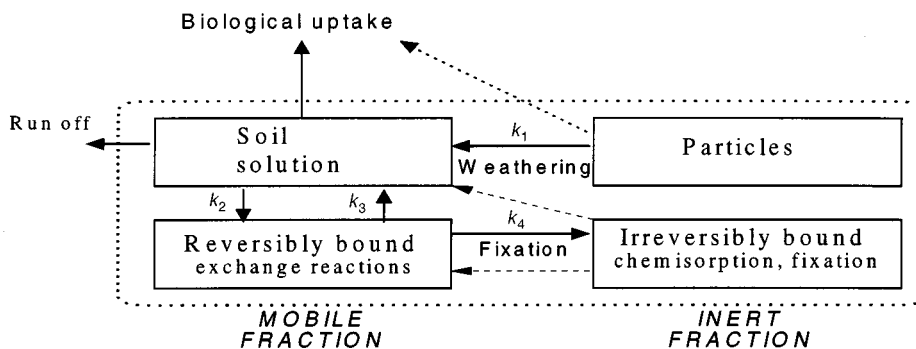


Fig. 4 Distribution of radionuclide species in soil–water (sediment–water) systems.

1–3% of the ^{137}Cs and 90–100% of the ^{90}Sr could be considered reversibly sorbed and exchangeable, *i.e.*, as mobile species.¹⁰ For Chernobyl soils collected in 1989, the fraction of extractable ^{90}Sr was, however, surprisingly low. Detailed EM studies and sequential leaching of individual Chernobyl particles and soils containing particles confirmed a very low extractability.¹⁷ By following the increase in the NH_4OAc -extractable ^{90}Sr in Chernobyl soils with time after deposition, the environmental weathering rates of uranium oxide fuel particles could be determined. Based on a series of extraction studies on Chernobyl soils collected over a 10 year period,²³ the particle weathering constant ranged from 0.04 to 0.42 yr^{-1} . The weathering rates were primarily dependent on particle composition (*e.g.*, crystalline, amorphous structure and the degree of oxidation), but were also influenced by soil pH.

For irreversibly sorbed species (chemisorption) or species incorporated in the mineral lattice, rupture of chemical bonds is needed for the species to be released. Within the sequential extraction scheme, pH is lowered and the redox power is gradually increased. Usually, the most effective extracting agent for ^{137}Cs and stable Cs in soil or sediment is 7 M HNO_3 , reflecting a strong interaction (fixation) to clays, whereas Sr is usually reversibly associated in soils. For ^{137}Cs and ^{90}Sr associated with uranium fuel particles, however, the particles are oxidised by H_2O_2 in HNO_3 and the nuclides are mobilised. In these cases, the distributions of extractable radioactive and stable isotopes of Cs and Sr are significantly different. Extraction of samples contaminated with fuel particles collected close to the Chernobyl reactor (Fig. 5) revealed a significant enriched H_2O_2 - HNO_3 fraction for ^{137}Cs and ^{90}Sr , whereas stable Sr was weakly (NH_4OAc) associated and stable Cs was strongly (7 M HNO_3) bound to the soils.^{17,20,22,24} Hence the weathering rates of fuel particles are a key factor for uptake of ^{90}Sr in vegetation and a key factor for fixation of ^{137}Cs in clays.

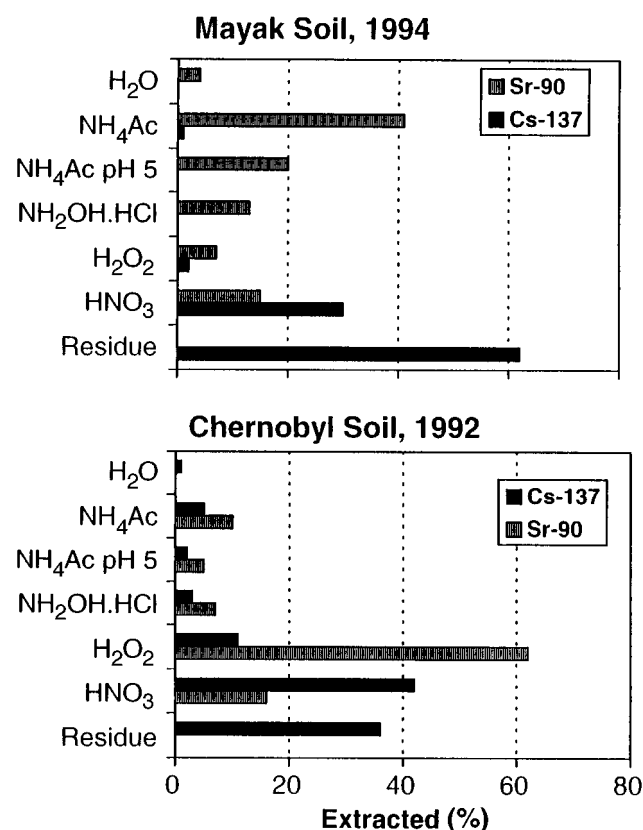


Fig. 5 Sequential extraction of ^{137}Cs and ^{90}Sr in soils from Chernobyl.

Particle influence on mobility

When weathering of radioactive particles occurs, inert and mobile radionuclide species are released. The term 'inert species' includes colloidal and particulate material in addition to radionuclides irreversibly bound to or incorporated in mineral lattices. The term 'mobile species' refers to LMM species, *e.g.*, ions or complexes soluble in water or species reversibly associated with solid surfaces. Hence a fraction of the mobile species may be available for active biological uptake.

The distribution coefficient, K_d (Bq kg^{-1} soil or sediment per Bq l^{-1} water at equilibrium) is an essential parameter for modelling transfer in the ecosystem. Usually K_d is based on activity measurements of contaminated soils or sediments and waters. If radioactive particles are present in soils or sediments, K_d may be very high, indicating that soils or sediment will act as an efficient sink. However, as weathering occurs and radionuclides are mobilised over time, the distribution of nuclides between solid and solution, and hence K_d , will change. Hence K_d values should be monitored by adding chemically well defined radioactive tracers to the heterogeneous systems (*e.g.*, soil–water or sediment–water systems). By following the interaction between specific radionuclide species and solids over time, K_d can be calculated and compared with those obtained from measurements of contaminated samples. For particle-contaminated soils and sediments, K_d could be orders of magnitude higher than that obtained from tracer experiments. So far, soils from the Ukraine and Nordic countries and sediments from the Irish Sea, Kara Sea, Abrosimov Fjord and Ob and Yenisey estuaries have been included in dynamic model experiments.^{22,26}

Alternatively, analytical results obtained from sequential extractions can be utilised to calculate mobility factors (MF):

$$MF = \frac{\text{mobile species (Bq m}^{-2}\text{)}}{\text{total deposition (Bq m}^{-2}\text{)}} \times 100 (\%)$$

where the mobile species include the mobile reversible (H_2O and NH_4OAc extractable) fraction of contaminated soils and that taken up by vegetation from the same site (Bq m^{-2}).²² By including the mobile fraction taken up by vegetation, MF , is less sensitive to seasonal variations due to vegetation growth. For particle-contaminated soils, the extraction yield, and hence MF , are small. For samples collected in Ukraine, Belarus and Norway, the calculated MF demonstrated a low mobility of ^{90}Sr close to the reactor whereas the mobility increased with increasing distance from the reactor.^{20,24}

Particle influence on bioavailability

When inhaled, radioactive particles in fallout or resuspension pose a well documented risk to animals and humans. Close to accident sites (*e.g.*, Chernobyl and Mayak in Russia), heavy surface contamination of radioactive particles has also caused severe biological effects on vegetation. However, colloids and particles are relatively inert and active biological uptake in plants and animals should be low, hence the concentration factor (CF) reflecting the transfer to biological systems (*e.g.*, vegetation, animals) is low. Therefore, in areas contaminated with radioactive particles, K_d is high, whereas MF and CF are low. Experiments performed within the Chernobyl 30 km zone have demonstrated that the 'relative biological availability' of ^{137}Cs from soil to grass was low compared with that of an added ionic Cs tracer.²⁷ Furthermore, studies have confirmed that particle weathering and the subsequent increases in ^{90}Sr mobility are accompanied by increases in soil-to-plant transfer factors in the 30 km zone.²³ In contaminated soils and sediments, bioerosion of radioactive particles by certain fungi in soils and bioerosion of radioactive particles by certain fungi in soils and bioerosion of radioactive particles may also represent an important pathway.

In most models the digestion by animals of fodder contaminated with radioactive particles is not taken into account, *i.e.*, *CF* is expected to be low. However, animal experiments in which individual fuel particles from Chernobyl were administered orally to six lactating goats demonstrated that fuel particles could be retained as a point source in the gastrointestinal (GI) tract much longer than previously expected (more than 3 months). Furthermore, ^{137}Cs (and ^{134}Cs) were mobilised, absorbed and excreted through the milk and urine. Using SEM, one can see that the mineral surfaces were clearly influenced by the transport through the GI tract (Fig. 6). About 20% of ^{137}Cs and ^{106}Ru and 10% of ^{144}Ce were released. Hence the low *CF* predicted was underestimated.²⁸

Conclusions

Key factors contributing to major uncertainties in the assessment of the consequences of radionuclide releases are source term characteristics including speciation and the presence of radioactive particles and colloids, time dependent distribution coefficients (K_d) for transfer between water and solids (soil or sediments) and concentration factors (*CF*) for transfer to biological systems. If inert radioactive particles represent a major fraction of the release, the observed K_d values will be higher and *CF*s will be lower than expected. When weathering occurs and mobile species are released, K_d will decrease whereas *CF* will increase with time.

The identification and characterisation of radioactive particles and colloids are still a challenge in analytical chemistry.

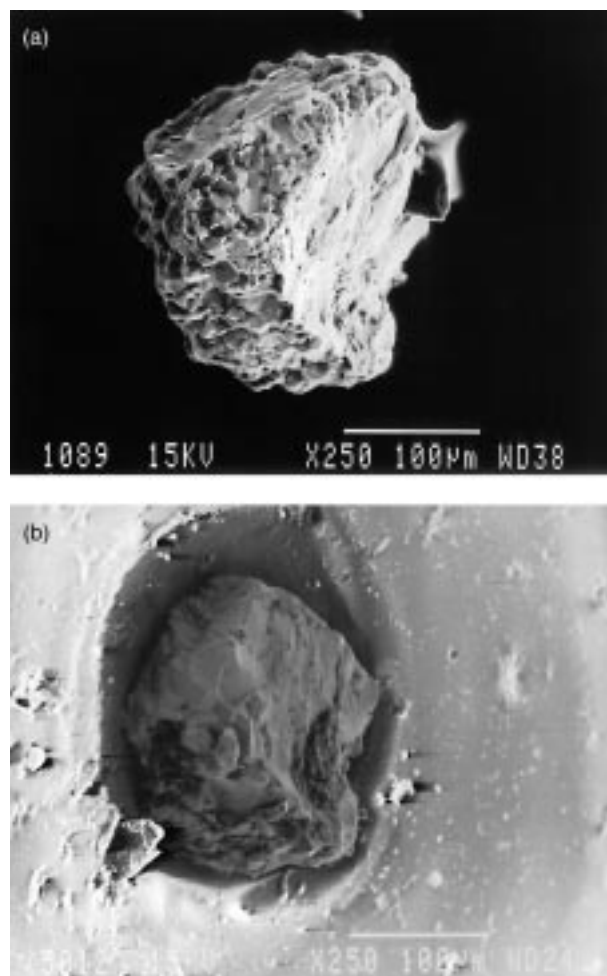


Fig. 6 Radioactive particles from Chernobyl administered to a goat (SEI mode), (a) prior to oral intake and (b) after transport in the GI tract.²⁸

Most emphasis has so far been put on the characterisation of radioactive particles which are easily identified from the emission of high energy γ - or β -radiation. When particles are obtained, the use of SEM and TEM combined with XRMA has proved very valuable for the characterisation of particles with respect to size and volume estimates, surface composition and structure. However, other surface analytical techniques should be utilised to obtain information about the oxidation states of the matrix elements. Prior to EM, samples must also be fractionated to obtain the relevant particles. In water, fractionation of radioactive particulate or colloidal materials using hollow fibre ultrafiltration is most appropriate. In soils and sediments, sequential extraction techniques provide useful information about radionuclides reversibly or irreversibly associated with particles.

By following the time dependent mobilisation of radionuclides from particles, essential information on weathering rates can be included in prognostic models. However, for small-sized particles with very low activities, especially of α -emitters only (*e.g.*, Pu isotopes), more sophisticated fractionation techniques are still needed.

Close collaboration with P. Strand, Norwegian Radiation Protection Agency, J. P. Day, University of Manchester, and scientists from Russia, Ukraine and Belarus has been very valuable, especially with respect to the collection of relevant sample materials.

References

- Salbu, B., and Oughton, D. H., in *Trace Elements in Natural Waters*, ed. Salbu, B., and Steinnes, E., CRC Press, Boca Raton, FL, 1994, pp. 41–69.
- Arnold, L., *The Windscale Fire 1957. Anatomy of a Nuclear Accident*, Macmillan, London, 1992.
- Smith, J. N., Ellis, K. M., Aarkrog, A., Dahlgard, H., and Holm, E., *J. Environ. Radioact.*, 1994, **25**, 134.
- UNSCEAR, *Sources and Effects of Ionizing Radiation*, United Nations Scientific Committee on the Effects of Atomic Radiation, New York, 1993.
- Loschilov, N. A., Kasparov, V. A., Yudin, Ye. B., Protsak, V. P., Zurba, M. A., and Parshakov, A. E., in *The Radiobiological Impact of Hot Beta-Particles from the Chernobyl Fallout. Risk Assessment*, IAEA, Vienna, 1992, Part I, pp. 34–39.
- Devell, L., Tovedal, M., Bergström, U., Applegren, A., Chussler, J., and Andersson, L., *Nature (London)*, 1986, **321**, 817.
- Raunemaa, R., Saari, H., Loukkanen, S., and Lehtinen, S., in *Hot Particles from the Chernobyl Fallout*, ed. von Philipsborn, H., and Steinhäuser, F., Bergbau und Industrimuseum, Theuern, 1988, p. 77.
- Jakeman, D., *Notes of the Level of Radioactive Contamination in the Sellafield Area Arising from Discharges in the Early 1950s*, UKAEA, AEEW Report 2104, Atomic Energy Establishment, Winfrith, 1986.
- Salbu, B., Bjørnstad, H. E., Sværen, I., Prosser, S. L., Bulman, R. A., Harvey, B. R., and Lovett, M. B., *Sci. Total Environ.*, 1993, **130/131**, 51.
- Joint Norwegian–Russian Expert Group for Investigation of Radioactive Contamination in the Northern Areas, *Sources Contributing to Radioactive Contamination of the Techa River and Areas Surrounding the 'Mayak' Production Association, Urals, Russia*, Østerås, Norway, 1997.
- Salbu, B., and Bjørnstad, H. E., *J. Radioanal. Nucl. Chem. Articles*, 1990, **138**, 337.
- Deguelde, C., Longworth, G., Moulin, V., Vilks, P., Ross, C., Bidoglio, G., Cremers, A., Kim, J., Pieri, J., Ramsay, J., Salbu, B., and Vuorinen, U., *PSI-Ber.*, 1989, No. 39.
- Lydersen, H. E., Bjørnstad, Salbu, B., and Pappas, A. C., in *Speciation of Metals in Water, Sediments and Soil Systems*, ed. Landner, L., Lecture Notes in Earth Sciences, vol. 11, Springer, Berlin, 1987, pp. 85–97.
- Salbu, B., Bjørnstad, H. E., and Brittain, J. E., *J. Radioanal. Nucl. Chem.*, 1989, **156**, 7.

- 15 Nisbet, A. F., Salbu, B., and Shaw, S., *J. Environ. Radioact.*, 1993, **18**, 71.
- 16 Salbu, B., Nikitin, A. I., Strand, P., Christensen, G. C., Chumichev, V. B., Lind, B., Fjellidal, H., Bergan, T. D. S., Rudjord, A. L., Sickel, M., Valetova, N. K., and Føyn, L., *Sci. Total Environ.*, 1997, **202**, 185.
- 17 Oughton, D. H., Salbu, B., Brand, T. L., Day, J. P., and Aarkrog, A., *Analyst*, 1993, **118**, 1101.
- 18 Salbu, B., Krekling, T., Oughton, D. H., Østby, G., Kashparov, V. A., Brand, T. L., and Day, J. P., *Analyst*, 1994, **119**, 125.
- 19 Bobovnikova, T. T., Virchenko, Y. P., Konoplev, A. V., Siverina, A., and Shkuratova, I., *Sov. Soil. Sci.*, 1991, **23**, 5.
- 20 Salbu, B., Oughton, D. H., Ratnikov, A. V., Zhigarev, T. L., Kruglov, S. V., Petrov, K. V., Grebenshakiyeva, N. V., Firsakova, S. K., Astasheva, N. P., Loschilov, N. A., Hove, K., and Strand, P., *Health Phys.*, 1994, **67**, 518.
- 21 Riise, G., Bjørnstad, H. E., Lien, H. N., Oughton, D. H., and Salbu, B., *J. Radioanal. Nucl. Chem.*, 1990, **142**, 531.
- 22 Oughton, D. H., and Salbu, B., in *Nordic Radioecology: the Transfer of Radionuclides Through Nordic Ecosystems to Man*, ed. Dahlgaard, H., Elsevier, Amsterdam, 1994, pp. 165–184.
- 23 Kashparov, V. A., Oughton, D. H., Zvarich, S. I., Protsak, V. P., and Levchuk, S. E., *Health Phys.*, in the press.
- 24 Oughton, D. H., Salbu, B., Riise, G., Lien, H., Østby, G., and Nøren, A., *Analyst*, 1992, **117**, 481.
- 25 Oughton, D. H., Børretzen, P., Salbu, B., and Tronstad, E., *Sci. Tot. Environ.*, 1997, **202**, 155.
- 26 Børretzen, P., Fjellidal, H., Lien, H., Oughton, D. H., and Salbu, B., in *Environmental Radioactivity in Arctic*, ed. Strand, P., and Cooke, A., Norwegian Radiation Protection Authority, Østerås, 1995, pp. 168–172.
- 27 Bondar, P. F., Ivanov, Yu, A., and Ozornov, A. G., in *The Radiobiological Impact of Hot Beta-Particles from the Chernobyl Fallout. Risk Assessment*, IAEA, Vienna, Part I, 1997, pp. 58–67.
- 28 Salbu, B., Krekling, T., Hove, K., Oughton, D. H., Kashparov, V. A., and Astasheva, N., *Proceedings of International Symposium on Environmental Impact of Radioactive Releases*, IAEA, Vienna, 1995, pp. 312–313.

Paper 8/003141

Received January 12, 1998

Accepted March 27, 1998