Review of Remediation Approaches Implemented in Radioactively Contaminated Areas

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1 Introduction

Closing of the nuclear facilities worldwide is a regulated process which includes their decommissioning or dismantling, followed by appropriate remedial actions to enable that remedial areas can be newly used for the different purposes with the safe levels of the residual radioactivity. Complexity of nuclear site remediation activities involves following steps: problem formulation, investigation before remediation, planning of remediation, actions of remediation, close out and post remedial control of the site. One of the main remediation goals at many nuclear sites is preventing uncontrolled dispersion and transport of radionuclides through the environment. Remediation projects in countries with significant legacy clean-up challenges usually considered an approach of remediation which is based on the risk and considered two main issues: sustainability and costs. International Atomic Energy Agency (IAEA) in International Basic Safety Standards set up a string of requirements to which attention must be paid in the time of the performance of remediation actions (IAEA 1996, 2011). The United States Environmental Protection Agency (US EPA) published technical guidance documents with the purpose to improve the effectiveness of decontamination technologies. In the document 'Technology Reference Guide for Radioactively Contaminated Media' different technologies suitable for treatment of radioactive contamination in different media, liquid (groundwater, surface and/or waste water) or solid (soil, sediment and/ or solid waste) are summarized (USEPA 2007). This Guide provides information on 21 technologies that could be evaluated and mutually compared for site-specific applications. Some additional

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information regarding the technologies which are not included in the EPA documents (ITRC 2008) could be found in the literature 'Decontamination and Decommissioning of Radiologically Contaminated Facilities'.

Radioactivity of naturally occurring radionuclides arises mostly from uranium and thorium decays. It is generally low-level and is not considered as contamination. In the area with industrial facilities, e.g. coal fired power plant, there is an increased probability for environmental media to become enriched with natural radionuclides (Dai et al. 2007; Charro et al. 2013; Lu et al. 2012; Ćujić et al. 2015). On the other side, anthropogenic levels of radioactivity in the environment originating from testing of nuclear weapons, nuclear disasters (e.g. Fukushima and Chernobyl) and routine authorised releases to the environment, are contamination, but it is either not under the control of the nuclear facilities operator or authorised by the national regulatory body. Major deposition on surface soils of anthropogenic radionuclide ^{137}Cs throughout Europe arises from nuclear accident in Chernobyl (Raffery et al. 2000; Petrović et al. 2013, 2016a, b; Dragović et al. 2015). Common radionuclides in radioactive wastes are: ^{137}Cs (physical half-life 30.5 year), ^{90}Sr (physical half-life 28.8 year),^{3}H (physical half-life 12.3 year), ^{14}C (physical half-life 5730 year), ^{60}Co (physical half-life 5.3 year), ^{99}Tc (physical half-life 2.1×10^5 year), ^{129}I (physical half-life 1.6×107 year), ^{226}Ra (physical half-life 1601 year), uranium nuclides (physical half-life 2.5×10^5 year for ^{234}U, 4.7×10^9 year for ^{238}U) and transuranic nuclides (longest physical half-life 2.4×10^4 year for ^{239}Pu). Some elements, such as C, Sr, Tc and I, show high mobility in the environment, and because of that they are of long-term environmental concern and their varying biogeochemical behaviour makes their co-treatment by traditional remediation technique challenging. Monitoring of remediated nuclear sites is an important issue as sometimes contamination from the sites is being spread beyond controlled areas and could potentially affect groundwater users. The selection of appropriate remediation technique is of special importance and it depends on several factors, e.g. existing problems, selection of the contaminants for removing, the soil type, the geology on the local level and goals which want to be reached. It sometimes happens that outside pressure and participation of stakeholders and regulators bring to the wrong finding. This was the case at the Hanford Site where a pump-and-treat system was installed and operated to remediate a ^{90}Sr plume adjacent into the Columbia River. In spite of scientists' warnings to regulators that the system would not be enough effective, it was operated almost 8 years and only less than 1% (67 GBq) of ^{90}Sr was removed. During the same period the activity of 11,840 GBq was removed by radioactive decay. At the end, the costly remedial system was replaced with the permeable reactive barrier to decrease flux to the river and the ^{90}Sr has been left to natural attenuation. Monitored natural attenuation could be an option of clean-up and IAEA recommended its applicability to media contaminated with different radionuclides (IAEA 2006a). The migration velocity of radionuclides which are not attenuated easily ought to significantly reduced and appropriate techniques for that purpose should be developed. *In situ* waste treatment can be an option as it avoids problems originating from 'dilute and disperse' or the 'redistribution' of exposures to settlers in the area surrounding the disposal site (Nisbet and Woodman 2000; Nisbet 2002;

Oughton 2013). The groundwater clean-up systems which are widely used are expensive and need to be applied in practice during long time period. One of the key roles in controlling the speciation and mobility of uranium and other redox sensitive radionuclides (e.g. Tc, Np, Pu) occurred via biogeochemical interactions through direct metabolic processes such as microbial respiration, or indirectly by changing ambient redox/pH conditions, producing ligands or new bio minerals, or altering mineral surfaces. Control of radionuclide mobility via natural attenuation can be stimulated by biogeochemical processes to accelerate clean-up of contaminated environments through bioremediation. Instead of using traditional methods in remediation of contaminated groundwater or surface water, could be used more efficient and cost-effective biological treatment methods. In the recent years, the remediation research is focused on development of new clean-up systems and improvement of the efficiency of the existing ones.

2 Remediation of Water: Case Studies

2.1 Pump and Treat System at Hanford Site (USA)

The US Department of Energy's Hanford Site is located in south-eastern Washington State along the Columbia River. At Hanford, the groundwater pump and treat system is projected to grab and treat contaminated groundwater plume emanating from a plutonium separations plant. It was estimated that approximately 180 billion kilolitres of contaminated water was discharged from the Site to the soil and that some of the contaminants have migrated to groundwater under the site. Contaminated groundwater with the radioactivity levels above groundwater protection standards spread over area of 207 km^2. The major constituents of concern (COC) for the Site are carbon tetrachloride and ^{99}Tc. The other COCs are total chromium, nitrate, trichloroethylene (TCE), ^{129}I, ^{3}H. With the desire to rich clean-up levels contaminants in 125 years, components are projected and applied in practice in combination with monitored natural attenuation. Concentrations of carbon tetrachloride in the groundwater over 100 mg L^{-1} roughly corresponds to 95% of the mass of carbon tetrachloride currently exists in the aquifer. To decrease the mass of carbon tetrachloride by 95%, the estimated pumping rate for this purpose should be 9500 L min^{-1} in the expected timeframe. After extraction, the COCs in groundwater is treated to reach the clean-up grade and returned to the aquifer out of injection wells. Natural attenuation processes are utilized and relied on to decrease concentrations to below the clean-up grade and parts of these processes include abiotic degradation, dispersion, sorption and, radioactive decay for tritium. Flow-path control is reached after the treated groundwater was syringed into the aquifer of the groundwater contamination.

The construction of the treatment facility included six process buildings: two extraction transfer buildings, two injection transfer buildings and the radiological and biological buildings. In the treatment system for the groundwater both

radioactive ion exchange system and a biological system in airless conditions to remove the non-radioactive contamination from the groundwater were implemented. Groundwater from extraction wells that feed extraction transfer building contained concentrations of ^{99}Tc higher than 33 Bq L^{-1} and in the radiological building using ion exchange resins the concentrations of radionuclides were reduced. Cartridge filters removed fine particulate matter from incoming groundwater. The ion exchange effluent flow through bag filters which serve as a resin trap, and for a next treatment it was directed to the biological building. When the ion exchange resin reached its loading limit, it was removed from the vessel and was treated for clean-up and returned to plant. After treatment the stripping water was pumped to the biological building for treatment. The resin was sluiced with finished water and placed in a container to allow drainage and the dewatered resin to be transported to the landfill for disposal (US DOE 2010; US DOE, US EPA, Ecology 2009).

2.2 In situ *Permeable Treatment Wall System at West Valley (USA)*

In West Valley (in western New York, USA) at a former commercial nuclear fuel recycling and vitrification test site *in situ* permeable treatment wall (PTW) was implemented with the aim to remove ^{90}Sr from groundwater. The potential of exploiting a permeable barrier was evaluated and found to be reproducible and a pump-and-treat system that was not satisfactory treating ^{90}Sr was replaced. Moor et al. (2000) provided a general overview of the pilot PTW project at the West Valley. Rabideau et al. (2005) reported the results of laboratory and numerical analysis carried out to characterize the potential performance of the West Valley Demonstration Project treatment wall. In the groundwater at the site a plume of ^{90}Sr with radioactivity in the range 37–3700 Bq kg^{-1} was detected. Groundwater flows to the north and east and appears at depths in the range from 3 m below ground level up to the near ground surface. The main remedial actions objectives were to reduce or eliminate presence of ^{90}Sr in groundwater leakage, leaving activity concentrations on the site at the minimum level as practically achievable; minimise the possibilities for expansion of the ^{90}Sr plume beyond its current limits in the future; ensure that a selected technology does not excludes any plans for addressing the plume during site decommissioning. Based on ion exchange reaction ^{90}Sr was removed *in situ* from progressing plume in groundwater. For that purpose thick zone of zeolite (granular formation made of approximately 85% of the mineral clinoptilolite) dimensions 260 m length by 1 m thick was constructed. In wastewater treatment systems zeolite materials have been used extensively as ion exchange media. Robinson et al. (1991) demonstrated that certain natural zeolites possessed a strong selectivity for the caesium radioisotopes and ^{90}Sr. Modelling approach in correlation with the treatment media assessment showed that a 1 m zeolite thickness could satisfactory the

desired design-life of 20 years. As the best installation solution for the 1 m thick PTW, single-pass trenching was applied. This technique includes a trencher moving along the alignment and bringing trench spoils to the surface using a chain-saw like cutting boom at the rear of the trencher. The trench was sustained and instantly backfilled behind the cutting boom using a delivery system. Some of the gains compared to other familiar methods are increased efficiency of installation, simply for construction and cost effectiveness.

The PTW performance monitoring system was installed on the PTW platform. Estimated hydraulic performance and ^{90}Sr removal from groundwater was in compliance with experimental results, according to obtained monitoring data. Activity concentrations of analysed ^{90}Sr within the PTW were mainly below 3.7 Bq L^{-1} and often below detectable level. It should be stated that activity concentrations of analysed ^{90}Sr immediately after installation of the PTW were higher than 370 Bq L^{-1}.

2.3 *Monitored Natural Attenuation at the Monticello Mill Tailing Site (USA)*

The Monticello Mill produced uranium and vanadium for military purposes in the period 1942–1960. Mill tailings placed of in piles along a small stream which flows through the site Montezuma Creek were the main source of contamination. Leaching of uranium from the tailings led to groundwater uranium plume within the underlying alluvial aquifer. Approximately 1.5 km down gradient of the former mill site reached the groundwater plume area where maximum contaminant levels were exceeded (US DOE 2009).

After the removal of mill tailings and an additional time of rating, an assessment of plume stability was carried out. The assessment covered the flow directions characterization of groundwater and analysis of contaminant concentrations in groundwater and aquifer solids (US DOE 2004). The removal of tailings piles has led to a significant decreasing of uranium concentration in groundwater of the investigated area. Applying model based on an equilibrium partitioning process between groundwater and aquifer materials, uranium attenuation was simulated and predicted activity concentrations of uranium were likened to observed trends at selected site locations to assess consistency. The solute transport model, with the lower K_d value of 2.8 kg L^{-1} foresee remediation by monitored natural attenuation and results pointed out uranium groundwater concentrations below maximum contaminant level values for approximately 42 years. Davis et al. (2004) reported that K_d values for uranium can vary several orders of magnitude over the pH range 6–9.

2.4 Enhanced Attenuation Technologies at Savannah River Site (USA)

Nuclear processing facilities operating in the period 1955–1988 produced seven billion litres of acidic aqueous waste storage in the F-Area Seepage Basins in Savannah River site. At the beginning it was assumed that under the basin head on connection between radionuclides and soil particles would be occurred. This assumption was correct for many disposed radionuclides, e.g. plutonium isotopes and ^{137}Cs, but for ^{90}Sr, ^{129}I, ^{99}Tc, ^{3}H, uranium isotopes migration into the groundwater in the direction of stream in its nearby has occurred. Heterogeneous geology exists at the site and the aquifer sediments underlying the basins were under the acidic solutions influence more than 40 years period, which contributed to modification in mineralogy of the sediments. The uppermost mechanism of attenuation for all radionuclide contaminants, excluding tritium, is adsorption, and attenuation at particular locations occurred due to dilution.

To measure radionuclide concentrations in the site, monitoring of wells was implemented. Radionuclide activity concentrations in groundwater from neighbouring wells and downgradient of the constrained basins indicated a major flux of radionuclides in the direction from the vadose zone below the basins to the saturated zone. Because of this, monitored natural attenuation was not a solution of the problem. One of the recommended solutions for contamination problem, the pump-and-treat system, was not acceptable because of relatively long period to meet regulatory goals for stream concentrations (several decades) and potentially radioactive solid waste in large quantities will be produced during that time. Because of that decision was made to apply *in situ* treatment technologies that would lead to the low pH and uranium, ^{90}Sr and ^{129}I plumes. Intensive research which included laboratory column studies followed by a field study, pointed out that relieved stabilisation will occurred after application of alkaline solutions with high buffering capacity. Implementation of the enhancement technology involves the assembly of a modified funnel-and-gate system. Monitoring downgradient of the treatment zone showed that adsorption of uranium and ^{90}Sr has occurred. This system showed satisfactory results regarding stabilization of both ^{90}Sr and uranium. The barrier wall was decreasing ^{3}H concentrations in the stream, while ^{129}I remained untreated. One of proposed solution for ^{129}I treatment in this site was interaction with silver chloride particles. Therefore, as ^{129}I in the groundwater interacts with these particles silver iodide is formed as precipitate, reacting with the silver chloride and releasing chloride ion. This access will lead to stabilization of the ^{129}I for a long-term period (PNNL 2009).

3 Microbial Bioreduction as the Potential Co-treatment of the Groundwater Radionuclide Contaminants

The biostimulation of anaerobic microbial communities was investigated as a method for removal of redox-sensitive radionuclides such as U(VI) from contaminated groundwater at nuclear sites. Stimulated uranium bioreduction has been

achieved by adding an electron donor to promote enzymatic reduction of aqueous U (VI) to insoluble U(IV). This has been demonstrated in laboratory experiments (Wilkins et al. 2007; Begg et al. 2011; Law et al. 2011) and *in situ* (Istok et al. 2004; Wu et al. 2007; Williams et al. 2011). The direct enzymatic reduction under ambient environmental conditions is the dominant mechanism which mediates the reduction of U(VI) (Williams et al. 2012; Bargar et al. 2013). Biosorption is the passive uptake of contaminants (e.g. radionuclides) to the surface of microbial cells. Suzuki and Banfield (1999) reported that microbial biosorption capacity of uranium uptake in bacteria ranged from 45 to 615 mg g^{-1} cell dry weight. It has been shown that prokaryotes could enzymatically reduce U(VI) (Williams et al. 2012). U (VI) reduction occurred in the sulfate-reducers *Desulfovibrio desulfuricans* and *Desulfovibrio vulgaris*. These species produced the U(IV) mineral uraninite using c-type cytochrome activity (Lovley and Phillips 1992a, b). Enzymatically induced bioreduction of U(VI) involved a wide spectrum of the sulfate-reducer species: *Anaeromyxobacter species* (Sanford et al. 2007), *C. ferrireducens* (Khijniak et al. 2005), *Clostridium* species (Francis et al. 1994; Suzuki et al. 2003; Madden et al. 2007), *Cellulomonas* species (Sivaswamy et al. 2011), *Desulfosporosinus* (Suzuki et al. 2003), *Paenibacillus* (Ahmed et al. 2012). The bioreduction mechanisms of U (VI) were not fully explained. Renshaw et al. (2005) used *Geobacter sulfurreducens* and concluded that U(VI) was decreased to the unstable intermediate U(V), which became disproportionate to the end product U(IV). Gram-positive bacteria may use cytochromes to reduce Fe(III) which could be correlated to U(VI) and radionuclide reduction (Carlson et al. 2012).

Thorpe et al. (2014) investigated potential of microbial enrichment cultures obtained from Sellafield UK representative sediments to reductively precipitate an Fe(II)-bearing biomineral assemblage at both neutral and alkaline pH. This approach was used to investigate effects of microbially-mediated Fe(III) cutting down on the mobility of ^{99}Tc and Sr (stabile Sr^{2+} and radioactive $^{90}Sr^{2+}$). In alkaline conditions occurs microbially-mediated Fe(III) reduction which leads to ^{99}Tc reduction and as a result appear less mobile Tc(IV) phases, and ^{90}Sr via increased sorption and/or precipitation formed at higher pH values. The mobility of redox active ^{99}Tc can be attenuated by reduction from soluble Tc(VII) to poorly soluble Tc(IV) (Lloyd et al. 2000; Burke et al. 2005). The mobility of the $^{90}Sr^{2+}$ ion is mostly controlled by sorption and mineral precipitation reactions (Langley et al. 2009; Thorpe et al. 2012; Wallace et al. 2012, 2013). It is known that species of the *Clostridiales* could reduce Fe(III) in freshwater and marine environments over a wide pH range either via dissimilatory Fe(III) reduction (Dobbin et al. 1999). It was demonstrated that during fermentation process reduction will appear if Fe(III) acts as a minor electron acceptor (Lovley and Phillips 1988; Lehours et al. 2010). It was demonstrated that TcO^{-}_{4} added at 1.6×10^{-6} M was eliminated (more than80%) from solution concurrent with Fe(III) reduction in enrichment culture experiments at pH 7 and pH 9. Reduction of Tc(VII) to Tc(IV) was confirmed by X-ray absorption spectroscopy of the reduced bio minerals. Microbial active systems demonstrated enhanced removal of ^{90}Sr at pH 7 in ultra-trace experiments, compared to the sterile control because of sorption of $^{90}Sr^{2+}$ to the Fe(II)-bearing bio minerals that generated *in situ*. Similar

removal of ^{90}Sr to the microbial active experiment shown at pH 9 in sterile control and was ascribed to Sr-sorption to mineral phases in the case of chemically precipitated in sterile controls, and also for the biologically precipitated in the microbial active systems. Throughout the different systems, the amount of the added ^{90}Sr was removed approximately 60–70% in bioreduction experiments. Different treatment approaches involving bioreduction and the promotion of Fe(III)-reducing conditions to remediate Tc(IV) are not coincide with treatment of groundwater ^{90}Sr contamination (Thorpe et al. 2014). The results obtained by Thorpe et al. (2014) pointed out the potential for removal of trace levels of ^{99}Tc and $^{90}Sr^{2+}$ from groundwater during stimulated bioreduction and pointed out that if there exists stable Sr^{2+}, than optimal removal for technetium and strontium is likely to occur under mildly alkaline, reducing conditions. Much greater quantities of Tc(VII) were removed by *Geobacter*, *Anaeromyxobacter* and *Shewanella* in the presence of ferrihydrite compared to experiments with just cells, indicate the importance of biogenic Fe(II) in Tc (VII) bioreduction (Plymale et al. 2011).

Experiments with enrichment cultures from the Oak Ridge site Nyman et al. (2007) found that the inhibition co-efficient for U(VI) was around 100 μM and at this concentration the effective yield and growth rate were reduced by 50%. Spain and Krumholz (2011) reported concentrations of U(VI) up to 11 μM in groundwater close to the S3 ponds at the Oak Ridge site, while Cho et al. (2012) reported value of 250 μM in well FW113–47.

The Old Rifle processing site was permitted to DOE for custody and long-term care and managed by the Office of Legacy Management. The site is under DOE jurisdiction from 1998 and requires routine inspection and maintenance, records-related activities and stakeholder support (USDOE 1999). Groundwater at this former uranium ore processing facility are contaminated with low levels of uranium, which leached from mill tailings into the unconfined aquifer, which is an alluvial deposit of the nearby Colorado River. Groundwater uranium concentrations inside of the test area ranged from 0.4 to 1.4 μM and were above the maximum contaminant limit of 0.18 μM (Anderson et al. 2003). It was also demonstrated that there is potential for *in situ* effective removing of uranium present in contaminated groundwater, by stimulating the activity of *Geobacter* species in the subsurface. When acetate was added to enhance the growth of *Geobacter* species, U(VI) was actively removed (Anderson et al. 2003). Zachara et al. (2013) pointed out that vertical migration of uranium-contaminated groundwater is limited by the Wasatch formation and that release of contaminant U(IV) from naturally reduced zones by oxidation, together with migration of up-gradient groundwater naturally high in U(VI), are responsible for the persistence of elevated concentrations of uranium in groundwater. Williams et al. (2011) carried out research at the Rifle site and concluded that concentrations of U decreased from 1–1.5 μM to 0.05–0.1 μM when acetate was supplied as an electron donor and U(VI) was in a complex uranyl-calcium-carbonate. These low concentrations were achieved by ensuring the concentration of acetate remained greater than the 10 mM present sulfate. Stable isotope probing and gene expression analysis confirmed *Geobacter* were active and oxidising acetate, even during sulfate reduction, and so are likely to be responsible for U(VI) reduction and

maintaining low concentrations of U(VI) in groundwater. Bopp et al. (2010) reported that uranium isotope ratios, $^{238}U/^{235}U$, in groundwater decreased significantly during *in situ* bioreduction.

Areas where research should continue include determination of dominant mechanisms in bacterial electron transport, in both natural environment and bio stimulation trials. Newsome et al. (2014) pointed out in their review paper that further work in this area should be directed towards: determination of the exact mechanism(s) of electron transfer to U(VI) in circum neutral aquifer sediments; the reduction of U (VI) at alkaline pH, especially the role played by Gram-positive bacteria and determining the longevity of bioreduced U(IV).

4 Remediation of Soil: Excavation and Disposal

Common clean-up method of radioactive contaminated soil involves excavation/removal, transportation and disposal of contaminated soil on site or off site (Environment Agency 2002; IAEA 1999, 2006b). During the radiological clean-up of Enewetak atoll of the Marshall Islands in the Pacific Ocean, former United States nuclear test site, conducted from 1972 to 1980, large amount of plutonium-contaminated soil from different islands was removed by excision and collected and transported to the Runit Island and deposited in the Cactus nuclear test crater (DNA 1981). Contaminated soil was mixed with cement to form a slurry and placed into the Cactus crater, formed in 1958 as a result of the nuclear explosion, and the rest of the contaminated material was mixed with concrete and put above ground over the crater in the shape of a dome, and finally the crater was sealed with concrete cap (DNA 1981; Paajanen and Lehto 1992; Lehto 1994; Noshkin and Robison 1997). After the nuclear accidents at Chernobyl NPP in Ukraine (1986) and Fukushima Daiichi NPP in Japan (2011) (Balonov 2007; Hardie and McKinley 2014) various environmental media (including soil) over large areas were contaminated with radionuclides and the different decontamination methods were used in order to reduce radiation exposure from contaminated soil. Vovk et al. (1993) gave an overview of mechanical and physical methods of decontamination of land after Chernobyl NPP accident. The clean-up of the Chernobyl NPP site included removal of highly radioactive soil surface layers (5–10 cm) and their transport to the solid waste storage vault of the fifth unit and also removal of the less radioactive soil and their transport to the disposal areas near to the site (IAEA 1989; Vovk et al. 1993; Rudy and Vovk 1996). A number of waste disposal pits and trenches were dug at different locations adjacent to the Chernobyl NPP (Vovk et al. 1993; Rudy and Vovk 1996). Removal of radioactive soil was commonly implemented as remediation measure in contaminated areas following the accident at the Fukushima Daiichi NPP (Wada et al. 2012; Hardie and McKinley 2014; EPA 2016). According to the results presented in the MOE (2015) report, the very high decontamination rate of surface contamination density (over 90%) was achieved after scraping topsoil by 5 cm from the playgrounds in a kindergarten facility and after scraping topsoil by

5 cm and ploughing of rice fields. Soil and waste generated from decontamination work can be stored in temporary storage site (no more than 3 years), interim storage facilities (less than 30 years) until its final disposal (disposal out of Fukushima) (MOE 2013, 2015; Hardie and McKinley 2014). Some temporary storage sites after the accident were established in Koori town, Yugawa village, Kawamata town, Ten-ei village, Miharu town, Tamakawa village etc. (MOE 2015). In 2012, during the decontamination of sports field, grassed and forest areas around the Yamakiya Elementary School, Kawamata town, located 40 km from the Fukushima Daiichi nuclear power plant, top soil from the sports field and grassed areas was removed and replaced with sand or fresh turf, and leaf litter from the forest areas was removed to a distance of 20 m from the forest edge (Cresswell et al. 2016). In 2014, test decontamination measure carried out at the lower part of the Kami-Oguni River, a tributary of the Abukuma River in Fukushima, located 55 km from the Fukushima Daiichi NPP, reduced the air dose rate by a factor of approximately two (Nishikiori and Suzuki 2017). During the decontamination test process all plants were removed from the flood channel and taken to a waste incineration plant, the upper 5 cm of soil was removed from the dike slopes using excavator and turf was put on the decontaminated slope, and sediments up to 15–35 cm in depth from the flood channel were removed. The removed soil and sediments were packed into flexible container bags, and transport to a temporary storage site for decontaminated soil (Nishikiori and Suzuki 2017). Before decontamination, the air dose rates at 1 and 100 cm above ground were 0.78 and 0.66 μSv h^{-1}, respectively, while after completion of decontamination air dose rates at 1 and 100 cm above ground were 0.34 μSv h^{-1} (Nishikiori and Suzuki 2017). Smaller nuclear accident in 1987 in Goiania, Brazil, compared to the accident at Chernobyl and Fukushima, resulted in the contamination of the environment and various remedial actions were undertaken (such as decontamination of property, collection of contaminated clothing, removal of contaminated soil etc.) in order to clean up the contamination (IAEA 1988). Topsoil was removed (about 60% of caesium total activity was retained in the top 1.5 cm of soil after the accident) and the surfaces were covered either with uncontaminated (clean) soil or concrete (Amaral et al. 1991). Radioactive waste generated during the decontamination was packed and over 200 lorry loads of waste were transported to the temporary disposal site (IAEA 1988). Contaminated waste arising from the clean-up of Salt Lake City, USA, processing site (from 1984 to 1989) in Port Hope, Canada (from 1976 to 1981) – has been transported over long distances to disposal sites (IAEA 1992; Paajanen and Lehto 1992; Lehto 1994). During the clean-up of – Salt Lake City processing site, large amount of contaminated waste (including tailings, contaminated soils and building debris) was transported by trains to the disposal site near Clive, Utah, 140 km away (IAEA 1992; Paajanen and Lehto 1992; Lehto 1994; DOE 2016). From the city of Port Hope more than 100,000 tons of radioactively contaminated soil was transported by trucks to a storage site at the Chalk River Nuclear Laboratories, 350 km away (IAEA 1989, 1992; Paajanen and Lehto 1992; Lehto 1994; Screening Report – Port Hope Project 2006).

5 Remediation of Soil: *In situ* Treatment Technologies

In situ remediation's technologies are used to treat contaminated soil on-site (IAEA 1999). Depending on whether *in situ* remediation technologies reduce mobility, toxicity and/or volume through the treatment, they can be categorized into following groups: (1) containment technologies; (2) stabilization technologies, and (3) treatment technologies (IAEA 1999; EPA 2007). *In situ* treatment technologies are source control technologies that apply various physical/chemical, biological or thermal processes in order to reduce the toxicity and/or volume of the contaminated soil by destroying or removing contaminants (IAEA 1999). According to EPA (2006, 2007) and IAEA (2006b) different *in situ* technologies (such as: electrokinetic remediation, *in situ* vitrification (ISV), phytoremediation etc.) are suitable for treatment of radioactively contaminated soil.

5.1 Electrokinetic Remediation

Separation and extraction of radionuclides from saturated or unsaturated soils by electrokinetic remediation process, requires the application of a direct electric current of low intensity (DC) across electrodes (anode – positive electrode and cathode – negative electrode) placed in the soil (EPA 1995, 2007; Cameselle et al. 2013a, b). Electromigration and electroosmosis are main mechanisms of contaminants' movement in an electric field involved in electrokinetic technology (Kim et al. 2010, 2016a; Cameselle et al. 2013a; Gill et al. 2014; Yang et al. 2014; Silva et al. 2017). Kim GN et al. (2008, 2010, 2011, 2012, 2015, 2016a); Kim SS et al. 2016, were studying electrokinetic-flushing, electrokinetic-electrodialytic and electrokinetic methods to decontaminate radioactive contaminated soil.

Laboratory-scale experiments of the removal of uranium from soil using electrokinetic method performed by Booher et al. (1997) demonstrated that this methods can be used for removal of uranium, in uranyl form (UO_2^{2+}), from unsaturated contaminated soil. Original and improved electrokinetic equipment was manufactured in order to remove uranium from soil (Kim et al. 2011). Original pilot-scale electrokinetic equipment of a 50 L size consisted of a reagent reservoir, an anode room (material for the anode electrode was titanium), an electrokinetic soil cell, a cathode room (material for the cathode electrode was DSA-Dimensional Stable Anode), an equipment support system, a power supply, pH controller. During the experiment with this equipment, lots of metal oxides were generated and covered the cathode plate. Therefore, original pilot-scale electrokinetic equipment was improved by manufacturing immersion-washing device, metal oxide separator and circulation system, and the remediation experiments were conducted again using improved pilot-scale electrokinetic remediation equipment (Kim et al. 2011). Remediation experiments show that when the initial uranium concentration of soil was 50, 75 and 100 Bq g^{-1} the electrokinetic remediation time required to remediate the uranium concentration below clearance concentration of 1.0 Bq g^{-1} was about

34, 42 and 49 days, respectively, and the created waste-solution was 3.8, 4.4 and 5.0 ml g^{-1}, respectively (Kim et al. 2011). The 50 L size electrokinetic remediation equipment (consisted of a electrokinetic soil cell, an anode/cathode rooms, a reagent reservoir, an equipment support system, a pH controller, a power supply) appropriate for the soil characteristics of a nuclear facility site in South Korea, was manufactured in order to remove ^{60}Co and ^{137}Cs from soil (Kim et al. 2010). Remediation experiments were carried out for soil contaminated with 0.01 M of Co^{2+} and Cs^{+} and radioactive soil from storage facility using manufactured electrokinetic remediation equipment. From contaminated soil the removal efficiencies of Co^{2+} was 98.4% and of Cs^{+} was 94.9%, after 15 days, and 3.4 ml g^{-1} of waste solution was generated (Kim et al. 2010). Experiment results for radioactive soil show that reagent type, soil radioactivity concentration, electric current in a soil cell and particle size of soil have influence on the removal efficiencies of cobalt-60 and cesium-137 from soil (Kim et al. 2010). Kim et al. (2010) found that electrokinetic remediation time required to reduce soil radioactivity concentration below clearance concentration level (100 Bq kg^{-1}) depends on initial radioactivity concentrations in soils. After an electrokinetic remediation experiment (conditions – remediation time: 55 days; electric current: 15 mA cm^{-2}) the total removal efficiency of cobalt-60 and cesium-137 from the radioactive soil of 2 kBq kg^{-1} was 95.8% (Kim et al. 2010). The study of the soil remediation using electrokinetic-flushing technology, show that electrokinetic-flushing remediation enhances the removal efficiencies of Co^{2+} and Cs^{+} from the contaminated soil when compared to the electrokinetic remediation (Kim et al. 2008). Electrokinetic method was used to remove cesium from the artificially contaminated Hanford sediment with nonradioactive ^{133}Cs (0.01 M $CsNO_3$) (Jang et al. 2015). After an electrokinetic remediation experiment for 68 days, the removal efficiency of Cs from the bulk sediment was 47% (312 mg kg^{-1}), and the higher removal efficiency of Cs was obtained from the silt-clay fraction (average 52%) than from the sand fraction (20–55%) (Jang et al. 2015).

5.2 In situ *Vitrification*

In situ vitrification (ISV) is a thermal treatment process that uses extremely high temperatures to melt and vitrify contaminated soil (Campbell and Koegler 1990; Spalding et al. 1992; IAEA 1999, 2006b; EPA 2007). Two methods can be used for producing heat for melting the contaminated soil, the older one uses electrodes and electrical resistance (traditional and planar *in situ* vitrification) while the second one uses plasma arc technology. *In situ* vitrification process is described in different studies (Campbell and Koegler 1990; Spalding et al. 1992; IAEA 2006b; EPA 2007).

In the 1980s, *in situ* vitrification process was developed by Pacific Northwest Laboratory (PNL) for the U.S. Department of Energy (DOE), primarily for treating soil contaminated with transuranic radionuclides (Dragun 1991). In 1987, joint ORNL (Oak Ridge National Laboratory) – PNL pilot-scale demonstration of *in situ* vitrification technology for the stabilization of radioactively contaminated soil

was completed after 110 h of operation and consumption of about 29 MWh of power, which led to the formation of 25 × 10^3 kg vitrified soil approximately 4.9 m long × 1.2 m thick × 2.1 m wide (Jacobs et al. 1988). A large-scale demonstration of the *in situ* vitrification process, using existing DOE large-scale ISV equipment, was performed on soils contaminated with mixed wastes on the116-B-6A crib at Hanford (Luey et al. 1992; Luey 1993). Test began 11 April 1990 at 2:30 p.m. and ended 23 April 1990 at 2:55 p.m., after 288 h of operation and consumption of about 550 MWh of electrical energy, which resulted in 850 ton block of vitrified soil, the retention of chromium, and lead and cesium-l37 in the vitrified block greater than 99.9% (Luey et al. 1992; Luey 1993). During the rehabilitation of the Maralinga, former British nuclear test sites, *in situ* vitrification had been identified by Technical Assessment Group – TAG (established by Australian Government to report on options, and associated costs, for the decontamination and rehabilitation of the former British nuclear test sites in Australia) as best options for stabilization a series of 21 burial pits containing soil and debris primarily contaminated with plutonium and uranium at Taranaki (MARTAC 2003). The GeoMelt-ISV project was developed through four phases – phase I testing and evaluation of Taranaki soils; phase II engineering and intermediate-scale ISV tests; phase III manufacture and testing of the full-scale ISV equipment; and phase IV operation of ISV on the Taranaki pits (MARTAC 2003). Prior to ISV operations the concrete pit caps were removed, the surface area and depth of pit was determined, and the refractory sand trenches and associated instrumentation as well as soil berm were installed. ISV equipment at Taranaki, 1999, is shown in Fig. 1 (MARTAC 2003). A total of 11 pits were treated

Fig. 1 ISV equipment at Taranaki, March 1999 (This figure has been obtained from the Commonwealth of Australia, Report titled "Rehabilitation of Former Nuclear Test Sites at Emu and Maralinga (Australia)", MARTAC 2003)

with the GeoMelt-ISV process until its suspension, the first melt was carried out on pit no. 19 from 21 May 1998 to 25 May 1998 and the last melt was carried out on pit no. 17 from 10 March 1999 to 21 March 1999 when the melt was terminated by the explosion (MARTAC 2003). The ISV materials were removed from Taranaki pits and disposed of either in ISV burial trench or debris burial trench (MARTAC 2003).

5.3 Phytoremediation

Phytoremediation is a bioremediation process that uses plants in order to extract, degrade, contain, or immobilize contaminants that are present in soil (IAEA 2004a, 2006b; Khan et al. 2004; EPA 2007). Among different mechanisms of phytoremediation, phytoextraction and phytostabilization can be used for remediation of radionuclides, since they cannot be biodegraded (EPA 2007). Phytoextraction or phytoaccumulation is a process that involves the uptake/absorption of radionuclides from soil through the plant root system and translocation/accumulation of radionuclides in the aboveground biomass, i.e. shoots and leaves (Dushenkov 2003; IAEA 2004a; EPA 2007). Phytoextraction removes radionuclides without destructive impact on soil structure and with a limited impact on fertility and it is especially suitable for treatment of large areas with a low level of radioactivity (Dushenkov 2003). Above-ground biomass of the plants loaded with radionuclides can be harvested and disposed of (IAEA 2004a; Saleh 2016; Sheoran et al. 2016). Identification of hyperaccumulator of radionuclides is a key step in successful application of phytoextraction. In 2009, the concentrations of U, Th, Ba, Ni, Sr and Pb in the tailings and dominant plant parts (stalk, shoot, root) from the uranium mill tailings repository in South China were determined (Li et al. 2011). Among studied plants, only *Cyperus iria* (U concentration in the shoot, root and tailings was 36.4, 2.43 and 6.03 μg g^{-1}, respectively) and *Parthenocissus quinquefolia* (Sr concentration in leaf, stalk and tailings was 190, 72.2 and 154 μg g^{-1}, respectively) satisfied the criteria for a hyperaccumulator of uranium and strontium (Li et al. 2011). Similar study was conducted in 2011, when the ^{226}Ra activity were determined in the tailings and dominant plant species (stalk, shoot, root) from the uranium mill tailings impoundment in South China (Hu et al. 2014). Among studied plants, only *Pteris multifida* (activities of ^{226}Ra and transfer factors (TFs) found in leaf, stalk and root were: ^{226}Ra – 150.6, 53.65 and 1.782 Bq g^{-1}, respectively, and TF – 9.131, 3.251 and 0.108, respectively), *Pteridium aquilinum* (activities of ^{226}Ra and transfer factors (TFs) found in leaf, stalk and root were: ^{226}Ra – 122.2, 42.36 and 13.14 Bq g^{-1}, respectively, and TF – 7.409, 2.568 and 0.797, respectively), and *Dryopteri sscottii* (activities of ^{226}Ra and transfer factors (TFs) found in leaf, stalk and root were: ^{226}Ra – 105.7, 29.685 and 5.699 Bq g^{-1}, respectively, and TF – 6.408, 1.800 and 0.346, respectively) satisfied the criteria for a hyperaccumulator for ^{226}Ra (Hu et al. 2014). According to Hu et al. (2014) and Li et al. (2011) these plants could be the candidates for phytoremediation of uranium, strontium and ^{226}Ra contaminated soils. Huang et al. (1998) found that citric acid was the most effective in enhancing

uranium accumulation in *Brassica juncea* and *Brassica chinensis*. Soudek et al. (2007) investigated the ^{226}Ra activities in the leaves, flowers and seeds of three woody species alder (*Alnus glutinosa*), birch (*Betula pendula*) and elder (*Sambucus nigra*) growing at a mill tailing dump K1 at a previous uranium mill in Mydlovary, South Bohemia. Plant samples were collected in period May–October during 3 years and results showed that^{226}Ra activities in the leaves generally increased during the vegetation periods and the highest ^{226}Ra accumulation occurred in the birch and lowest in alder. Soudek et al. (2007) suggested that these pioneer woody species can be used as remediation alternative to the use of herbs. By analysing plants grown in uranium mining-impacted soils, −, Chen et al. (2005) reported that the root uptake of ^{238}U, ^{226}Ra and ^{232}Th from the soil is plant-specific. The highest TF values for ^{238}U, ^{226}Ra and ^{232}Th were found for lupine (*Lupinus albus*) shoot, white clover (*Trifolium pratense*) shoot, and ryegrass (*Lolium perenne*) shoot, respectively (Chen et al. 2005). *Tridax procumbens L.*, *Azadirachta indica A. Juss.* and *Euphorbia hirta L.* were reported to be effective in phytoremediation of ^{90}Sr contaminated sites, based on the study conducted in the vicinity of Tummalapalle uranium mining site Kadapa district, A.P. India (Ahammad et al. 2015). Dushenkov et al. (1999) found that *Amaranthus retroflexus cvs. L. cv. aureus* and *PT-95* (Table 1) shows highest total removal of ^{137}Cs from soil in the vicinity of Chernobyl NPP. Sugiura et al. (2016)

Table 1 Bioaccumulation coefficient and total ^{137}Cs removal from soil[a]

Species and cultivars	Bioaccumulation coefficient	Total removal (Bq m^{-2})
Amaranthus retroflexus L. cv. PT-95	1.50	3225
Amaranthus retroflexus L. cv. aureus	1.90	2440
Amaranthus retroflexus L. cv. belozernii	1.41	1392
Amaranthus cruentus L.	1.32	1251
Helianthus tuberosum L. x Helianthus annuus L.	0.49	1221
Amaranthus caudatus L.	2.03	1144
Amaranthus cruentus L. cv. myronivka	1.07	1053
Helianthus tuberosus L.	0.30	846
Amaranthus hybridus L.	0.60	719
Amaranthus retroflexus L. cv. Antey	1.07	641
Amaranthus bicolor L.	0.59	417
Amaranthus cruentus L. cv. paniculatus	0.53	412
Zea mays L.	0.28	409
Helianthus annuus L.	0.24	319
Pisum sativum L.	0.48	44
Brassica juncea (L.) Czern.	0.47	194

Adopted from Dushenkov et al. (1999)

[a]Plants were grown at the experimental plot at the Northwest border of Chernobyl, Ukraine, approximately 10 km south of the ChNPP fourth reactor that was damaged in 1986. Bioaccumulation coefficient was calculated as a ratio of ^{137}Cs specific activity in the plant versus ^{137}Cs specific activity in the soil

reported that *Chengiopanax sciadophylloides* has an ability to accumulate radiocesium. The concentrations of ^{137}Cs in leaves of *Chengiopanax sciadophylloides* (28.1 kBq kg^{-1}) were higher comparing to those in leaves of *Quercus serrata* (4.13 Bq kg^{-1}), *Acer crataegifolium* (9.67 Bq kg^{-1}), *Fraxinus sieboldiana* (3.34 Bq kg^{-1}), and *Ilex macropoda* (2.55 Bq kg^{-1}) growing at same site near the town of Kawamata, Fukushima prefecture, located approximately 37 km northwest of F1 NPP.

6 Mathematical Modelling Approaches for Remediation of Contaminated Environments

In order to obtain the most efficient remediation strategy environmental decision aiding technologies are coupled with models for radionuclide transfer in different environmental compartments, models for radiation risk assessment and information on available regulatory and management strategies. A number of software based models (ERICA Tool, RESRAD Family of Codes, NORMALYSA, SYMBIOSE, CROM), internet based calculators (PRG, DCC) and excel based models (UK RCLEA, NJRaSoRs) have been developed so far for radiological risk and dose assessment (ANL 2001; US EPA 2002, 2004; Bureau of Environmental Radiation, NJ 2003; UK DEFRA 2003; Brown et al. 2008; CIEMAT 2011; IAEA 2015; IRSN n.d.). Following Chernobyl accident, the development of a wide range of programs has been started on the international level with the purpose to support the methods and models for assessing environmental impacts of radionuclides and other contaminants were designed, such as VAMP (Validation and Model Predictors), BIOMOVS (BIOsphericMOdel Validation Study) and BIOMASS (BIOsphere Modelling and ASSessments Methods). Within BIOMASS program the particular emphasis is directed towards the improvement of modelling techniques and accuracy of predicted results (IAEA 2004b). Among other important radioecological issues, the program addressed environmental assessment modelling in remediation of contaminated environment due to nuclear accidents, unlimited releases, even unsatisfactory management practices. To estimate the success of the remediation steps environmental transfer models were tested within two scenarios based on the results from the remediation program carried out on a radium extraction site in Olen, Belgium. Olen scenario Type A was aimed at testing the accuracy of predictions of different models used (CLRP, DOSDIM, OLENRAD-A, RISKOLEN and TAMDYN) based on data when remedial actions are performed. It was related to the influence of these actions on the ^{226}Ra concentrations in food chain. Within Olen scenario Type B the following remedial actions were evaluated: removal of the most contaminated soil layer (1 m depth) and capped with a uncontaminated soil layer (0.5 m depth), and their effectiveness assessed in terms of individual doses to an adult farmer arise from external irradiation, inhalation of resuspended particles and radon, consumption of soil particles, drinking water and food products. Six different

models (CLRP-RAD, DOSDIM, OLENRAD-B, RESRAD-OFFSITE, RESRAD-ONSITE and TAMDYN-UV) were tested within this type of Olen scenario. The analysis of the model prediction results pointed out that the different interpretations of the scenario contributed more to the differences among the model predictions than the differences in modelling approaches (IAEA 2004b).

As rural environment can remain polluted for long time of period after nuclear accident, the effective recovering strategies are highly needed to protect the population. Within STRATEGY project the model was used to help decision makers in recognising optimal agricultural countermeasure actions under user-defined requirements (Cox and Crout 2003). The application to the model to a hypothetical nuclear power plant accident scenario in Cumbria (UK) showed the significance of relevant information on local level useful for preparation of effective remediation (Cox et al. 2005). This model enabled decision makers to point out the optimal combination of ploughing, administration of ammonium-ferric-cyano-ferrate, clean feeding and food restrictions, taking into account local conditions and implementation costs.

After years of application of countermeasures in radioactively contaminated areas it was recognized that they are not quite effective for ecosystems and society in general. The effectiveness of remediation of radioactively contaminated areas varied with implemented management options (Table 2).A reliable evaluation of cost-benefit balance through predictive models was highly needed in optimization of remediation strategies. Under the European Commission project MOIRA (A Model based computerised system for management support to identify optimal remedial strategies for restoring radionuclide contaminated aquatic ecosystems and drainage areas) the software model was developed for the cost-benefit analysis of applied countermeasures in affected areas and identification of optimal remediation strategies in freshwater ecosystems and complex catchments (Appelgren et al. 1996). Because of site-specific models in some cases could be costly and time consuming there was the demand if predictive models have generic strictures, than can be used for different environmental conditions. The model for assessing radionuclide transport and countermeasure effects in complex catchments (MARTE) established on summarized collective parameters was developed to predict sediment removal, water

Table 2 Summary of reduction factors for selected management options to decrease contamination of agricultural products implemented after Kyshtym and Chernobyl accidents

	Reduction factor		
	Kyshtym accident (crops)	Chernobyl accident	
Management option	^{90}Sr	^{137}Cs	^{90}Sr
Normal ploughing	10–16	2.0–4.0	n.d.
Liming	upto 3	1.5–3.0	1.5–2.6
Application of mineral fertilizers	1.1–1.4	1.5–3.0	1.0–2.0
Clean feeding	3.0–4.0	2.0–5.0	2.0–5.0
Administration of Cs binders to animals	n.d.	3.0–5.0	n.d.
Administration of clay minerals to animals	n.d.	2.0–3.0	n.d.

Adopted from IAEA (2012a)
Note: *n.d.* no data

diversion from sub-catchments and cleaning up of sub-catchments (Monte 2001). The model included sub-models for: (1) radionuclide migration, (2) the assessment of the water balance and the water fluxes and (3) predicting the effects of countermeasures. It was used to a variety of European water bodies in Sweden, Italy and Ukraine contaminated by ^{137}Cs and ^{90}Sr and the obtained outcomes suggested the effectiveness of some remediation strategies, e.g. the control of water from polluted catchments, in complex catchments of radioactively contaminated areas (Monte 2001).

Taking into consideration temporal and spatial variations of radionuclide activities in areas of former Soviet Union affected by Chernobyl accident due to hydrochemical conditions, an environmental decision support system (EDSS) based on geographical information systems (GIS) which integrates all these factors was developed in the frame of the RESTORE project to enable local authorities to manage the remediation of contaminated land (Van der Perk et al. 1998). Data on surface contamination (maps of soil contamination produced by geostatistical interpolation), soil type, land use, food production rates, the density and structure of population were used as input for the EDSS. The EDSS integrated radioecological data (data on transfer of radionuclides to agricultural and wild food products, external doses etc.) with geographical database through dynamic modelling toolkit which included geostatistical analysis, conditional simulation, and topological modelling. The EDSS enabled an insight into radionuclide fluxes in the environment, particularly in the food chain, and also identification of areas vulnerable due to the presence of population groups which received an increased radiation doses.

After the decade of remediation in affected areas in Belarus, Russia and Ukraine it was noticed that due to a large variety of factors governing the radionuclide transfer in the environment the identical countermeasures are not appropriate for each contaminated area and that there is a need for integrated remediation strategies adapted to site-specific conditions. In a due course an approach was developed based on wide radioecological data collected from 70 representative contaminated colonies in affected areas taking into account the distribution of different soil types in these colonies (Jacob et al. 2001). For eight remedial actions for large-scale application (radical improvement, Prussian blue, supply clean milk, unpolluted feed for pigs, mineral fertilizer for potatoes, limit mushroom consumption, food monitors and removal of soil), were consider basic data on reduction factors, time periods of effectiveness and costs. Taking into account radioecological data for each settlement (^{137}Cs activity per unit area, transfer factors for foodstuffs) and data on fraction of the foodstuffs produced locally and on their annual consumption in affected areas, a dose model was established and representative remediation strategies were derived. These data were used to evaluate remediation strategies for different radioecological classes in which affected area was subdivided and results were then generalised to the total affected population (Jacob et al. 2001).

The experiences in implementation of decision aiding technologies in areas affected by Chernobyl accident pointed out that these technologies, to be effectively used, have to be supported by models for radionuclide transfer in the environment, radiation risk models and information on available management and decision

making options (IAEA 2012b). Two decades after Chernobyl accident there were a few hundreds of settlements in Belarus, Russia and Ukraine in which population suffered annual effective doses due to ionizing radiation caused by accident which exceeded 1 mSv. Therefore, the IAEA initiated the project Radiological support for the rehabilitation of the areas affected by the Chernobyl nuclear power plant accident. For the purpose of this project, a software tool ReSCA–Remediation Strategies after the Chernobyl Accident has been established (Ulanovsky et al. 2011) to facilitate optimization of remediation strategies in the rural settlements of the affected areas. The model used settlement-specific data on ground contamination and activity contents in foodstuffs (Jacob et al. 2001). Only exposures due to ^{137}Cs as only relevant radionuclide after decades from the accident were considered in the model. Based on radioecological data collected for rural settlements in which the annual effective dose exceeds 1 mSv annual effective doses were calculated with ReSCA and validated with data from whole body counter measurements (Jacob et al. 2009). The implementation of the model has shown that a substantial collective dose can be averted by quite cost-effective remediation strategies (Jacob et al. 2009). The FORESTLAND EDSS applied in Bryansk region of Russian Federation for justification of remediation strategies of forests affected by Chernobyl accident. The results indicated the ingestion of milk from cows grazing forest meadows as the main pathway of contamination and that forest management options are the most effective way to decrease the long term impact of radiocaesium contamination (IAEA 2012b).

With the aim to evaluate dose rates to marine biota after Fukushima accident Vives i Batlle et al. (2016) report an inter-comparison of eight models designed to predict the radiological exposure of radionuclides. In this work model predictions of radionuclide uptake and turnover using kinetic calculations based on biological half-life ($T_{b1/2}$) and/or more complex metabolic modelling approaches were used to predict activity concentrations and dose rates of ^{90}Sr, ^{131}I and ^{137}Cs to fish, crustaceans, macroalgae and molluscs under circumstances where the water concentrations are changing with time.

The modelling approaches were also successfully applied to resolve some important issues in optimization of bioremediation strategies. It has been widely recognized that dissimilatory metal reducing bacteria grown on acetate could be applied as a technique for remediation purpose to immobilize hexavalent uranium [U(VI)] as precipitated U(IV). However, Anderson et al. (2003) identified the inefficiency of U (VI) removal from groundwater when acetate-oxidizing sulfate reducers became dominant as an important consideration in the optimization of a bioremediation strategy. The multicomponent reactive transport model based on acetate-oxidizing iron- and sulfate reducing microbial populations was developed to picture the bioreduction of a continuous influx of soluble U(VI) in groundwater to insoluble and immobile U(IV). The model was applied for identification of the dominant transport and biological processes controlling uranium mobility throughout acetate biostimulation in the unconfined aquifer of the Old Rifle UMTRA field site in western Colorado (Yabusaki et al. 2007). It was further developed to involve abiotic reactions along with the biologically mediated reaction network in the multicomponent biogeochemical reactive transport simulator (Fang et al. 2009).

In planning, guiding and evaluating remediation, locally varying conditions such as temporal variation of radionuclide activity concentrations in different environmental compartments, need to be taken into account. The linking of radioecological models with GIS enables optimization of remediation strategies according to local variations of radionuclide activity concentrations. Following Chernobyl accident, a number of EDSSs such as FORCON (Fesenko et al. 1996), SAVE (Howard and Wright 1999), RESTORE (Van der Perk et al. 1998) and ReSCA (Ulanovsky et al. 2011; Fesenko et al. 2012), were developed to facilitate optimization of remediation strategies in contaminated areas.

The understanding of physical, chemical and biological processes governing behaviour of contaminants in aqueous environments is necessary for assessment of efficiency of terrestrial environmental remediation strategies. Therefore, a number of mechanistically-based reactive chemical transport models have been developed in the decades after Chernobyl accident (Yeh and Tripathi 1990; Pruess 1991; Zyvoloski et al. 1994; Fang et al. 2003; Zhang et al. 2007). Yeh et al. (2013) reviewed the development of mechanistically-based numerical models and demonstrated their flexibility and range of capabilities through the application to environmental remediation problems such as waste disposals and uranium mill tailings.

Mathematical modelling methods were successfully used for solving complex decision-making problems such as the selection of alternative remediation options. Jimenez-Martin et al. (2016) proposed an approach based on multi-criteria and fuzzy logic which takes into account environmental, radiological, social and economic impact as the main criteria for the appropriate management of the site (Perez-Sanchez et al. 2015). This approach was applied for evaluating different remediation options for the Zapadnoe uranium mill tailings in Ukraine (Jimenez-Martin et al. 2016).

One of the most challenging issues in remediation of closed uranium mining sites is the treatment of mine water. To ensure the sufficient removal of pollutants from these waters it is necessary to understand underlying reaction processes which could be interpreted by hydrogeochemical modelling. This approach was used at Wismut mine sites to investigate reaction processes during the treatment of mine water, i.e. for hydrogeochemical understanding of precipitation processes in a CO_2 stripping column with a focus on the influence of pH value and for characterization of uranium speciation of mine water (Lietsch et al. 2015). The modelling derived the prognosis of necessary pH values in order to prevent oversaturation inside the stripping column. The model outputs were tested at mine water treatment plant where lower pH values showed significant improvements. The application of hydrogeochemical modelling for identification of typical uranium species and complexes in mine water revealed the dominance of calcium uranyl carbonates in mine water at alkaline and neutral conditions and uranyl sulfates in acidic conditions (Lietsch et al. 2015). Authors emphasized the need for further investigation of thermodynamic parameters of uranyl species.

The widespread contamination in subsurface environments makes the remediation process economically challenging and in such cases there is a need for alternative approaches. In the case of uranium contamination such an approach implies

changes in physicochemical conditions of the environment to promote *in situ* immobilization of uranium, for example by addition of phosphate amendments (Wellman et al. 2008; Mehta et al. 2014). The understanding of processes governing U(VI) transport under the influence of phosphate amendments, can result in identification of conditions for reduction of its mobility. Mehta et al. (2015) used a reactive transport model to evaluate a transport of U(VI) through contaminated sediments. The application of the model in batch and column experiments with sediments from a Rifle site, Colorado, indicated that addition of phosphate could result in uranium retention in sediments to some extent.

Up to now, much effort has been devoted to explore both new technologies and new types of materials for remediation of contaminated wastewater. Modelling is often used in characterization of capacity and mechanisms of sorption of radionuclides on materials proposed to be used for environmental remediation. Zhang et al. (2015) investigated interaction between U(VI) and carbonaceous nanofibers by applying double layer sorption model. They found out that when co-existing with montmorillonite, carbonaceous nanofibers could extract the sorbed uranium onto their surface by a pseudo-second order kinetic process. The modelling results indicate carbonaceous nanofibers as a promising material for wastewater remediation. Different kinetic and equilibrium models are used to understand the mechanism of uranium adsorption from contaminated water. Naeem et al. (2017) investigated mechanism of uranium adsorption on green mung (*Vigna radiata*) by applying Freundlich isotherm and pseudo-second order kinetic model and proved its efficiency for uranium removal from aqueous solutions.

Within EMRAS (Environmental Modelling for Radiation Safety) Programme several mathematical models (EXPURT, METRO-K, CPHR, RESRAD-RDD) used for assessment related to the remediation of urban areas contaminated with radionuclides were tested (IAEA 2012a). The EXPURT model calculates surface activity densities and external gamma dose rates as a function of time in built environments and can represent the implementation of countermeasures such as decontamination and soil mixing. The EXPURT embedded within CONDO software tool can assist decision makers by providing estimations of normal living doses, doses from inhalation of resuspended radionuclides and waste activity concentrations (IAEA 2012a). The METRO-K model with a simple mathematical structure gives concentrations of radioactive materials and associate exposure doses as a function of time for different environments. Different countermeasures such as cutting and removal of grass, removal of trees, leaves, soil and washing of different surfaces could be considered within this model (IAEA 2012a). The compartment model CPHR is based on Ecolego tool[1] and it is used to assess time dependence of radioactive contamination and associated dose rates due to environmental processes (IAEA 2012a). The RESRAD-RDD compartment model is used to derive external exposure rates at receptor locations from different surfaces, radionuclide surface concentrations, annual and cumulative external doses for receptors

[1] http://www.facilia.se/products/ecolego.asp

(IAEA 2012a). It also considers different countermeasures such as cutting and removal of grass, removal of soil, washing and relocation. The models were tested on three hypothetical scenarios (point source, area source and area source + river). By comparing the model outputs a number of issues were derived where model capabilities could be improved and uncertainties reduced (information on initial distribution of contaminants and their transport in urban conditions, environmental data and data on radionuclides other than ^{137}Cs).

7 Conclusions

In the past few decades a much effort has been made to clean up the surrounding environment after decommissioning of nuclear installations. It is important to point out that there is no universal remediation plan how to remediate some media (water or soil). Because of plenty of factors that influence the outcome of the media rehabilitation process, it is important to made selection of optimal solution for each considered case study. International organisations (e.g. IAEA, EPA) made a lot of effort to provide guides and recommendation for different remediation approach based on research studies and the practical experience. More mobile radionuclides seek for more effective methods of remediation and that should be an area for coordinated international research in the future. It is of major importance in groundwater remediation strategy to develop new clean-up systems or to improve the efficiency of the existing ones with the goal to minimise the generation of secondary wastes. Monitored natural attenuation may be an alternative to clean-up radiologically contaminated media. Regarding soil remediation, developing remediation techniques for land underneath operating facilities is of major interest. Instead the most common conventional *ex situ* (soil removal, chemical extraction) remediation technique used for soil, *in situ* technologies (e.g. bioremediation, phytoremediation) is becoming increasingly used. Different approaches based on mathematical modelling methods have been proven to be successful in a case of complex decision-making problems, e.g. the selection of alternative remediation options.

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References

Ahammad JS, Sumithra S, Yamuna RT (2015) Screening for strontium accumulator plants in the vicinity of Tummalapalle uranium mining site Kadapa District, A.P. India. Int J Environ Sci 6:37–45

Ahmed B, Cao B, McLean JS, Ica T, Dohnalkova A, Istanbullu O, Paksoy A, Fredrickson JK, Beyenal H (2012) Fe(III) Reduction and U(VI) immobilization by *Paenibacillus sp.* strain

300A, isolated from Hanford 300A subsurface sediments. Appl Environ Microbiol 78:8001–8009

Amaral EC, Vianna ME, Godoy JM, Rochedo ER, Campos MJ, do Rio MA, Oliveira JP, Pereira JC, Reis WG (1991) Distribution of ^{137}Cs in soils due to the Goiânia accident and decisions for remedial action during the recovery phase. Health Phys 60:91–98

Anderson RT, Vrionis HA, Ortiz-Bernad I, Resch CT, Long PE, Dayvault R, Karp K, Marutzky S, Metzler DR, Peacock A, White DC, Lowe M, Lovley DR (2003) Stimulating the in situ activity of Geobacter species to remove uranium from the groundwater of a uranium-contaminated aquifer. Appl Environ Microbiol 69:5884–5891

ANL (2001) User's manual for RESRAD version 6. Environmental Assessment Division, Argonne National Laboratory, Argonne, https://web.evs.anl.gov/resrad/RESRAD_Family/

Appelgren A, Bergström U, Brittain J, Gallego Diaz E, Håkanson L, Heling R, Monte L (1996) An outline of a model-based expert system to identify optimal remedial strategies for restoring contaminated aquatic ecosystems: the project "MOIRA", ENEA, RT/AMB/96/17, Roma

Balonov MI (2007) The Chernobyl Forum: major findings and recommendations. J Environ Radioact 96:6–12

Bargar JR, Williams KH, Campbell KM, Long PE, Stubbs JE, Suvorova EI, Lezama-Pacheco JS, Alessi DS, Stylo M, Webb SM, Davis JA, Giammar DE, Blue LY, Bernier-Latmani R (2013) Uranium redox transition pathways in acetate-amended sediments. Proc Natl Acad Sci U S A 110:4506–4511

Begg JDC, Burke IT, Lloyd JR, Boothman C, Shaw S, Charnock JM, Morris K (2011) Bioreduction behavior of U(VI) sorbed to sediments. Geomicrobiol J 28:160–171

Booher WF, Lindgren ER, Brady PV (1997) Electrokinetic removal of uranium from contaminated, unsaturated soils. Prepared by Sandia National Laboratories for U.S. DOE. Sandia Report SAND97–0122

Bopp CJ, Lundstrom CC, Johnson TM, Sanford RA, Long PE, Williams KH (2010) Uranium 238U/235U isotope ratios as indicators of reduction: results from an in situ biostimulation experiment at Rifle, Colorado, USA. Environ Sci Technol 44:5927–5933

Brown JE, Alfonso B, Avila R, Beresford NA, Copplestone D, Pröhl G, Ulanovsky A (2008) The ERICA tool. J Environ Radioact 99:1371–1383

Bureau of Environmental Radiation, NJ (2003) Radioactive Soil Remediation Standards (RaSoRS). http://www.nj.gov/dep/rpp/rms/rad_cleanups.htm

Burke IT, Boothman C, Lloyd JR, Mortimer RJM, Livens FR, Morris K (2005) Effects of progressive anoxia on the solubility of technetium in sediments. Environ Sci Technol 39:4109–4116

Cameselle C, Chirakkara RA, Reddy KR (2013a) Electrokinetic-enhanced phytoremediation of soils: status and opportunities. Chemosphere 93:626–636

Cameselle C, Gouveia S, Akretche DE, Belhadj B (2013b) Advances in electrokinetic remediation for the removal of organic contaminants in soils. In: Rashed MN (ed) Organic pollutants – monitoring, risk and treatment. Intech Open Publisher, pp 209–229

Campbell BE, Koegler SS (1990) In situ vitrification of a mixed radioactive and hazardous waste site. Presented at the American Institure of Chemical Engineer 1990. Annual Meeting November 11–16, Chicago, Illinois

Carlson HK, Iavarone AT, Gorur A, Yeo BS, Tran R, Melnyk RA, Mathies RA, Aue M, Coates JD (2012) Surface multiheme c-type cytochromes from *Thermincola potens* and implications for respiratory metal reduction by Gram-positive bacteria. Proc Natl Acad Sci U S A 109:1702–1707

Charro E, Pardo R, Peña V (2013) Chemometric interpretation of vertical profiles of radionuclides in soil near a Spanish coal-fired power plant. Chemosphere 90:488–496

Chen SB, Zhu YG, Hu QH (2005) Soil to plant transfer of ^{238}U, ^{226}Ra and ^{232}Th on a uranium mining-impacted soil from southeastern China. J Environ Radioact 82:223–236

Cho K, Zholi A, Frabutt D, Flood M, Floyd D, Tiquia SM (2012) Linking bacterial diversity and geochemistry of uranium-contaminated groundwater. Environ Technol 33:1629–1640

CIEMAT (2011) Screening model for environmental assessment (CROM). ftp://ftp.ciemat.es/pub/CROM/CROM_8/

Cox GM, Crout NMJ (2003) A methodology for the selection and optimisation of countermeasure strategies, report for the STRATEGY project (CEC Contract No: FIKR-CT 2000-00018), University of Nottingham, UK

Cox G, Beresford NA, Alvarez-Farizo B, Oughton D, Kis Z, Eged K, Thørring H, Hunt J, Wright S, Barnett CL, Gil JM, Howard BJ, Crout NMJ (2005) Identifying optimal agricultural countermeasure strategies for a hypothetical contamination scenario using the strategy model. J Environ Radioact 83:383–397

Cresswell AJ, Kato H, Onda Y, Nanba K (2016) Evaluation of forest decontamination using radiometric measurements. J Environ Radioact 164:133–144

Ćujić M, Dragović S, Đorđević M, Dragović R, Gajić B, Miljanić Š (2015) Radionuclides in the soil around the largest coal-fired power plant in Serbia: radiological hazard, relationship with soil characteristics and spatial distribution. Environ Sci Pollut Res 22:10317–10330

Dai L, Wei H, Wang L (2007) Spatial distribution and risk assessment of radionuclides in soils around a coal-fired power plant: a case study from the city of Baoji, China. Environ Res 104:201–208

Davis JA, Meece DE, Kohler M, Curtis GP (2004) Approaches to surface complexation modelling of uranium (VI) adsorption on aquifer sediments. Geochim Cosmochim Acta 68:3621–3641

DNA (1981) The radiological cleanup of Enewetak Atoll. Defense Nuclear Agency, Washington, DC

Dobbin PS, Carter JP, San Juan CGS, von Hobe M, Powell AK, Richardson DJ (1999) Dissimilatory Fe(III) reduction by *Clostridium beijenrickii* isolated from freshwater sediment using Fe (III) maltol enrichment. FEMS Microbiol Lett 176:131–138

DOE (2016) Salt Lake City, Utah, processing and disposal sites –fact sheet, U.S. Department of Energy Office of Legacy Management

Dragović S, Petrović J, Dragović R, Đorđević M, Đokić M, Gajić B (2015) The influence of edaphic factors on spatial and vertical distribution of radionuclides in soil. In: Walther C, Gupta DK (eds) Radionuclides in the environment: influence of chemical speciation and plant uptake on radionuclide migration. Springer, Cham, pp 61–80

Dragun J (1991) Geochemistry and soil chemistry reactions occurring during in situ vitrification. J Hazard Mater 26:343–364

Dushenkov S (2003) Trends in phytoremediation of radionuclides. Plant Soil 249:167–175

Dushenkov S, Mikheev A, Prokhnevsky A, Ruchko M, Sorochinsky B (1999) Phytoremediation of radiocesium-contaminated soil in the vicinity of Chernobyl, Ukraine. Environ Sci Technol 33:469–475

Environment Agency (2002) Guidance on the characterisation and remediation of radioactively contaminated land

EPA (1995) In situ remediation technology: electrokinetics. EPA542-K-94-007. U.S. Environmental Protection Agency Office of Solid Waste and Emergency Response Technology Innovation Office Washington, D.C.

EPA (2006) In situ treatment technologies for contaminated soil. EPA 542/F-06/013. U.S. Environmental Protection Agency

EPA (2007) Technology reference guide for radioactively contaminated media. EPA 402-R-07-004. U.S. Environmental Protection Agency Office of Air and Radiation Office of Radiation and Indoor Air Radiation Protection Division Center for Radiation Site Cleanup

EPA (2016) Current and emerging post-Fukushima technologies, and techniques, and practices for wide area radiological survey, remediation, and waste management. EPA/600/R-16-140. U.S. Environmental Protection Agency

Fang Y, Yeh GT, Burgos WD (2003) A generic paradigm to model reaction-based biogeochemical processes in batch systems. Water Resour Res 33:1083–1118

Fang Y, Yabusaki SB, Morisson SJ, Amonette JP, Long PE (2009) Multicomponent reactive transport modeling of uranium bioremediation field experiments. Geochim Cosmochim Acta 73:6029–6051

Fesenko SV, Sanzharova NI, Wilkins BT, Nisbet AF (1996) FORCON: local decision support system for the provision of advice in agriculture–methodology and experience of practical implementation. Radiat Prot Dosim 64:157–164

Fesenko S, Jacob P, Ulanovsky A, Chupov A, Bogdevich I, Sanzharova N (2012) Justification of remediation strategies in the long term after the Chernobyl accident. J Environ Radioact 119:39–47

Francis AJ, Dodge CJ, Lu F, Halada GP, Clayton CR (1994) XPS and XANES studies of uranium reduction by Clostridium sp. Environ Sci Technol 28:636–639

Gill RT, Harbottle MJ, Smith JWN, Thornton SF (2014) Electrokinetic-enhanced bioremediation of organic contaminants: a review of processes and environmental applications. Chemosphere 107:31–42

Hardie SML, McKinley IG (2014) Fukushima remediation: status and overview of future plans. J Environ Radioact 133:75–85

Howard BJ, Wright SM (1999) Spatial analysis of vulnerable ecosystems in Europe: spatial and dynamic prediction of Radiocaesium fluxes into European foods (SAVE). Final Report, Commission of European Communities, Brussels

Hu N, Ding D, Li G, Zheng J, Li L, Zhao W, Wang Y (2014) Vegetation composition and ^{226}Ra uptake by native plant species at a uranium mill tailings impoundment in South China. J Environ Radioact 129:100–106

Huang JW, Blaylock MJ, Kapulnik Y, Ensley BD (1998) Phytoremediation of uranium-contaminated soils: role of organic acids in triggering uranium hyperaccumulation in plants. Environ Sci Technol 32:2004–2008

IAEA (1988) The radiological accident in Goiânia.International Atomic Energy Agency (IAEA). Vienna

IAEA (1989) Cleanup of large areas contaminated as a result of a nuclear accident. Technical Reports Series No. 300. International Atomic Energy Agency (IAEA), Vienna

IAEA (1992) Disposal of waste from the cleanup of large areas contaminated as a result of a nuclear accident. Technical Reports Series No. 330. International Atomic Energy Agency (IAEA), Vienna

IAEA (1996) International basic safety standards for protection against ionising radiation and for the safety of radiation sources. Safety Series No. 115. International Atomic Energy Agency (IAEA), Vienna

IAEA (1999) Technologies for remediation of radioactively contaminated sites, IAEA TECDOC-1086. International Atomic Energy Agency (IAEA), Vienna

IAEA (2004a) Remediation of sites with dispersed radioactive contamination. Technical Reports Series No. 424. International Atomic Energy Agency (IAEA),Vienna

IAEA (2004b) Testing of environmental transfer models using data from the remediation of a radium extraction site report of the remediation assessment working group of BIOMASS theme 2, IAEA-BIOMASS-7, Vienna

IAEA (2006a) Applicability of monitored natural attenuation at radioactively contaminated sites. Technical Reports Series No. 445. International Atomic Energy Agency (IAEA), Vienna

IAEA (2006b) Remediation of sites with mixed contamination of radioactive and other hazardous substances. Technical Reports Series No. 442. International Atomic Energy Agency (IAEA), Venna

IAEA (2011) Radiation protection and safety of radiation sources: international basic safety standards for protecting people and the environment INTERIM EDITION No. GSR Part 3 (Interim) General Safety Requirements. International Atomic Energy Agency (IAEA), Vienna

IAEA (2012a) Environmental modelling for radiation safety (EMRAS). A summary report of the results of the EMRAS Programme (2003–2007), Vienna

IAEA (2012b) Guidelines for remediation strategies to reduce the radiological consequences of environmental contamination. Technical Report Series No. 475. International Atomic Energy Agency (IAEA), Vienna

IAEA (2015) NORM and Legacy Site Assessment (NORMALYSA). http://project.facilia.se/normalysa/software.html

IRSN (n.d.) SYMBIOSE. https://gforge.irsn.fr/gf/project/symbiose/

Istok JD, Senko JM, Krumholz LR, Watson D, Bogle MA, Peacock A, Chang YJ, White DC (2004) In situ bioreduction of technetium and uranium in a nitrate contaminated aquifer. Environ Sci Technol 38:468–475

ITRC (2008) Decontamination and decommissioning of radiologically contaminated facilities. Interstate Technology and Regulatory Council (ITRC), Washington, DC

Jacob P, Fesenko S, Kirsakova SK, Likhtareva IA, Schotola C, Alexakhin RM, Zhuchenko YM, Kovgan L, Sanzharova NI, Ageyets V (2001) Remediation strategies for rural territories contaminated by the Chernobyl accident. J Environ Radioact 56:51–76

Jacob P, Fesenko S, Bogdevitch I, Kashparov V, Sanzharova N, Grebenshikova N, Isamov N, Lazarev N, Panov A, Ulanovsky A, Zhuchenko Y, Zhurba M (2009) Rural areas affected by the Chernobyl accident: radiation exposure and remediation strategies. Sci Total Environ 408:14–25

Jacobs GK, Spalding BP, Carter JG, Koegler SS (1988) In situ vitrification demonstration for the stabilization of buried wastes at the Oak Ridge National Laboratory. Nuc Chem Waste Manag 8:249–259

Jang HB, Yang JS, Um W (2015) Bench-scale electrokinetic remediation for cesium-contaminated sediment at the Hanford Site, USA. J Radioanal Nucl Chem 304:615–625

Jimenez-Martin A, Martin M, Perez-Sanchez D, Mateos A, Dvorzhak A (2016) Zapadnoe uranium mill tailings: a fuzzy approach. MVLSC 26:529–540

Khan FI, Husain T, Hejazi R (2004) An overview and analysis of site remediation technologies. J Environ Manag 71:95–122

Khijniak TV, Slobodkin AI, Coker V, Renshaw JC, Liven FR, Bonch-Osmolovskaya EA, Birkeland NK, Medvedeva-Lyalikova NN, Lloyd JR (2005) Reduction of uranium (VI) phosphate during growth of the thermophilic bacterium *Thermoterra bacterium ferri reducens*. Appl Environ Microbiol 71:6423–6426

Kim GN, Jung YH, Lee JJ, Moon JK, Jung CH (2008) Development of electrokinetic-flushing technology for the remediation of contaminated soil around nuclear facilities. J Indust Eng Chem 14:732–738

Kim GN, Lee SS, Shon DB, Lee KW, Chung US (2010) Development of pilot-scale electrokinetic remediation technology to remove ^{60}Co and ^{137}Cs from soil. J Indust Eng Chem 16:986–991

Kim GN, Shon DB, Park HM, Lee KW, Chung US (2011) Development of pilot-scale electrokinetic remediation technology for uranium removal. Sep Purif Technol 80:67–72

Kim GN, Kim SS, Park HM, Kim WS, Moon JK, Hyeon JH (2012) Development of complex electrokinetic decontamination method for soil contaminated with uranium. Electrochim Acta 86:49–56

Kim GN, Kim SS, Park UR, Moon JK (2015) Decontamination of soil contaminated with cesium using electrokinetic-electrodialytic method. Electrochim Acta 181:233–237

Kim GN, Kim I, Kim SS, Choi JW (2016) Removal of uranium from contaminated soil using indoor electrokinetic decontamination. J Radioanal Nucl Chem 309:1175–1181

Kim SS, Han GS, Kim GN, Koo DS, Kim IG, Choi JW (2016) Advanced remediation of uranium-contaminated soil. J Environ Radioact 164:239–244

Langley S, Gault AG, Ibrahim A, Takahashi Y, Renaud R, Fortin D, Clark ID, Ferris FG (2009) Strontium desorption from bacteriogenic iron oxides (BIOS) subjected to microbial Fe(III) reduction. Chem Geol 262:217–228

Law GTW, Geissler A, Burke IT, Livens FR, Lloyd JR, McBeth JM, Morris K (2011) Uranium redox cycling in sediment and biomineral systems. Geomicrobiol J 28:497–506

Lehours A, Rabiet M, Morel-Desrosiers N, Morel J, Jouve L, Arbeile B, Mailhot G, Fonty G (2010) Ferric iron reduction by fermentative strain BS2 isolated from an iron-rich anoxic environment (Lake Pavin, France). Geomicrobiol J 27:714–722

Lehto J (1994) Cleanup of large radioactive –contaminated areas and disposal of generated waste. Final report of the KAN2 project. Copenhagen Tema Nord 1994:567

Li GY, Hu N, Ding DX, Zheng JF, Liu YL, Wang YD, Nie XQ (2011) Screening of plant species for phytoremediation of uranium, thorium, barium, nickel, strontium and lead contaminated soils from a uranium mill tailings repository in South China. Bull Environ Contam Toxicol 86:646–652

Lietsch C, Hoth N, Kassahun A (2015) Investigation of phenomena in uranium mine waters using hydrogeochemical modeling – a case study, In: Merkel BJ, Arab A (eds) Uranium – Past and Future Challenges. Proceedings of the 7th international conference of uranium mining and hydrogeology, Springer

Lloyd JR, Sole VA, van Praagh CVG, Lovley DR (2000) Direct and Fe(II)-mediated reduction of technetium by Fe(II) reducing bacteria. Appl Environ Microbiol 66:3743–3749

Lovley DR, Phillips EJP (1988) Novel mode of microbial energy metabolism: organic carbon oxidation coupled to dissimilatory reduction of iron or manganese. Appl Environ Microbiol 54:1472–1480

Lovley DR, Phillips EJ (1992a) Reduction of uranium by *Desulfovibrio desulfuricans*. Appl Environ Microbiol 58:850–856

Lovley DR, Phillips EJP (1992b) Bioremediation of uranium contamination with enzymatic uranium reduction. Environ Sci Technol 26:2228–2234

Lu X, Li X, Yun P, Luo D, Wang L, Ren C, Chen C (2012) Measurement of natural radioactivity and assessment of associated hazards in soil around Baoji second coal-fired thermal power plant, China. Radiat Prot Dosim 148:219–226

Luey J (1993) In situ vitrification: demonstrated capabilities and potential applications. Presented at the Office of Technology Development 2nd semi-annual information meeting, January 25–27. Houston, Texas

Luey J, Koegler SS, Kuhn WL, Lowery PS, Winkelman RG (1992) In Situ Vitrification of a Mixed-Waste Contaminated Soil Site: The 116-B-6A Crib at Hanford. PNl-8281. Prepared for the U.S. Department of Energy under Contract DE-AC06-76RLO 1830

Madden AS, Smith AC, Balkwill DL, Fagan LA, Phelps TJ (2007) Microbial uranium immobilization independent of nitrate reduction. Environ Microbiol 9:2321–2330

MARTAC (2003) Rehabilitation of former nuclear test sites at Emu and Maralinga (Australlia). Report by the Maralinga Rehabilitation Technical Advisory Committee

Mehta VS, Maillot F, Wang Z, Catalano JG, Giammar DE (2014) Effect of co-solutes on the products and solubility of uranium(VI) precipitated with phosphate. Chem Geol 364:66–75

Mehta VS, Maillot F, Wang Z, Catalano JG, Giammar DE (2015) Transport of U(VI) through sediments amended with phosphate to induce *in situ* uranium immobilization. Water Res 69:307–317

MOE (2013) Decontamination guidelines. Ministry of the Environment. Retrieved from http://josen.env.go.jp/en/framework/pdf/decontamination_guidelines_2nd.pdf

MOE (2015) FY2014 decontamination report. Ministry of the Environment. Retrieved from http://josen.env.go.jp/en/cooperation/pdf/decontamination_report1503_01.pdf

Monte L (2001) A generic model for assessing the effects of countermeasures to reduce the radionuclide contamination levels in abiotic components of fresh water systems and complex catchments. Env Model Soft 16:669–690

Moore HR, Steiner RE, Fallon E, Repp CL, Hemann MR, Rabideau AJ (2000) Permeable treatment wall pilot project at the West Valley demonstration project. Proceedings of waste management 2000, 27 February–2 March, 2000. Tucson, AZ

Naeem H, Bhatti HN, Sadaf S, Iqbal M (2017) Uranium remediation using modified *Vigna radiata* waste biomass. Appl Radiat Isot 123:94–101

Newsome L, Morris K, Lloyd JR (2014) The biogeochemistry and bioremediation of uranium and other priority radionuclides. Chem Geol 363:164–184

Nisbet AF (2002) Management options for food production systems contaminated as a result of a nuclear accident. Radioprot Colloq 37:115–120

Nisbet AF, Woodman RFM (2000) Options for the management of Chernobyl restricted areas in England and Wales. J Environ Radioact 51:239–254

Nishikiori T, Suzuki S (2017) Radiocesium decontamination of a riverside in Fukushima, Japan. J Environ Radioact 177:58–64

Noshkin V, Robison W (1997) Assessment of a radioactive waste disposal site at Enewetak Atoll. Health Phys 73:234–247

Nyman JL, Wu HI, Gentile ME, Kitanidis PK, Criddle CS (2007) Inhibition of a U(VI)- and sulfate-reducing consortia by U(VI). Environ Sci Technol 41:6528–6533

Oughton DH (2013) Social and ethical issues in environmental remediation projects. J Environ Radioact 119:21–25

Paajanen A, Lehto J (1992) Disposal of Radioactive Wastes from the Cleanup of Large Areas Contaminated in Nuclear Accidents, A Literature Survay. Nordic Nuclear Safety Research Program 1990–93. Projekt KAN-2

Perez-Sanchez D, Jimenez A, Mateos A, Dvorzhak A (2015) Fuzzy MCDA for remediation of a uranium tailing. In: Merkel BJ, Arab A (eds) Uranium–past and future challenges. Proceedings of the 7th international conference of uranium mining and hydrogeology, Springer, Cham

Petrović J, Ćujić M, Đorđević M, Dragovic R, Gajic B, Miljanic S, Dragovic S (2013) Spatial distribution and vertical migration of ^{137}Cs in soils of Belgrade (Serbia) 25 years after the Chernobyl accident. Environ Sci Proces Imp 15:1279–1289

Petrović J, Dragović S, Dragović R, Đorđević M, Đokić M, Ćujić M (2016a) Spatial and vertical distribution of ^{137}Cs in soils in the erosive area of south eastern Serbia (Pčinja and South Morava River Basins). J Soil Sed 16:1168–1175

Petrović J, Dragović S, Dragović R, Đorđević M, Đokić M, Zlatković B, Walling D (2016b) Using ^{137}Cs measurements to estimate soil erosion rates in the Pčinja and South Morava River Basins, southeastern Serbia. J Environ Radioact 158–159:71–80

Plymale AE, Fredrickson JK, Zachara JM, Dohnalkova AC, Heald SM, Moore DA, Kennedy DW, Marshall MJ, Wang CM, Resch CT, Nachimuthu P (2011) Competitive reduction of pertechnetate ((TcO_4^-)-Tc-99) by dissimilatory metal reducing bacteria and biogenic Fe(II). Environ Sci Technol 45:951–957

PNNL (2009) Scientific opportunities to reduce risk in groundwater and soil remediation. PNNL 18516. Pacific Northwest National Laboratory (PNNL)

Pruess K (1991) TOUGH2: a general numerical simulator for multiple phase fluid and heat flow, report LBL-29400. Lawrence Berkeley Laboratory, Berkeley

Rabideau AJ, van Benschoten J, Patel A, Bandilla K (2005) Performance assessment of a zeolite treatment wall for removing Sr-90 from groundwater. J Contam Hydro 79:1–24

Rafferty B, Brennan M, Dawson D, Dowding D (2000) Mechanisms of ^{137}Cs migration in coniferous forest soils. J Environ Radioact 48:131–143

Renshaw JC, Butchins LJC, Livens FR, May I, Charnock JM, Lloyd JR (2005) Bioreduction of uranium: environmental implications of a pentavalent intermediate. Environ Sci Technol 39:5657–5660

Robinson SM, Arnold WD, Byers CH (1991) Multicomponent ion exchange equilibria in zeolite. In: Tedder DW, Pohland FG (eds) Emerging Technologies in Hazardous Waste Treatment. ACS Symposium Series No. 486. Kluwer Academic Publishers, New York

Rudy CG, Vovk IF (1996) Radioactive waste management issues related to the conversion of the Chernobyl Sarcophagus into an ecologically safe system. One decade after Chernobyl: summing up the consequences of the accident. IAEA-TECDOC-964. Vienna, Austria

Saleh HM (2016) Biological remediation of hazardous pollutants using water hyacinth – a review. J Biotech Res 2:80–91

Sanford RA, Wu Q, Sung Y, Thomas SH, Amos BK, Prince EK, Löffler FE (2007) Hexavalent uranium supports growth of *Anaeromyxobacter dehalogenans* and *Geobacter spp.* with lower than predicted biomass yields. Environ Microbiol 9:2885–2893

Screening Report –Port Hope Project (2006) The port hope long-term low-level radioactive waste management project. Natural Resources Canada, the Canadian Nuclear Safety Commission and Fisheries and Oceans Canada, Ottawa

Sheoran V, Sheoran AS, Poonia P (2016) Factors affecting phytoextraction: a review. Pedosphere 26:148–166

Silva EBS, Fernandes NS, Costa ECTA, Silva DR, de Araújo DM, Martínez-Huitle CA (2017) Scale-up on electrokinetic treatment of polluted soil with petroleum: effect of operating conditions. Int J Electrochem Sci 12:4001–4015

Sivaswamy V, Boyanov MI, Peyton BM, Viamajala S, Gerlach R, Apel WA, Sani RK, Dohnalkova A, Kemner KM, Borch T (2011) Multiple mechanisms of uranium immobilization by *Cellulomonas sp*. strain ES6. Biotechnol Bioeng 108:264–276

Soudek P, Petrova S, Benesova D, Tykva R, Vankova R, Vanek T (2007) Comparison of ^{226}Ra nuclide from soil by three woody species *Betula pendula*, *Sambucus nigra* and *Alnus glutinosa* during the vegetation period. J Environ Radioact 97:76–82

Spain AM, Krumholz LR (2011) Nitrate-reducing bacteria at the nitrate and radionuclide contaminated Oak ridge integrated field research challenge site: a review. Geomicrobiol J 28:418–429

Spalding BP, Jacobs GK, Dunbar NW, Naney MT, Tixier JS, Powell TD (1992) Tracer-level radioactive pilot-scale test of in situ vitrification for the stabilization of contaminated soil sites at ORNL. Prepared by the Oak Ridge National Laboratory, Oak Ridge

Sugiura Y, Kanasashi T, Ogata Y, Ozawa H, Takenaka C (2016) Radiocesium accumulation properties of Chengiopanax sciadophylloides. J Environ Radio 151:250–257

Suzuki Y, Banfield JF (1999) Geomicrobiology of uranium. Rev Mineral Geochem 38:393–432

Suzuki Y, Kelly SD, Kemner KA, Banfield JF (2003) Microbial populations stimulated for hexavalent uranium reduction in uranium mine sediment. Appl Environ Microbiol 69:1337–1346

Thorpe CL, Lloyd JR, Law GTW, Burke IT, Shaw S, Bryan ND, Morris K (2012) Strontium sorption and precipitation behaviour during bioreduction in nitrate impacted sediments. Chem Geol 306:114–122

Thorpe CL, Boothman C, Lloyd JR, Law GTW, Bryan ND, Atherton N, Livens FR, Morris K (2014) The interactions of strontium and technetium with Fe(II) bearing biominerals: implications for bioremediation of radioactively contaminated land. Appl Geochem 40:135–143

UK DEFRA (2003) The Radioactively Contaminated Land Exposure Assessment Methodology (RCLEA). https://www.gov.uk/government/publications/rclea-software-application

Ulanovsky A, Jacob P, Fesenko S, Bogdevitch I, Kashparov V, Sanzharova N (2011) ReSCA: decision support tool for remediation planning after the Chernobyl accident. Radiat Environ Biophys 50:67–83

US DOE (1999) Final site observational work plan for the UMTRA project Old Rifle site GJO-99–88-TAR. Colorado, United States Department of Energy (US DOE)/Grand Junction

US DOE (2004) Monticello mill tailings site, operable unit III–remedial investigation addendum/focused feasibility study. GJO-2003-413-TAC.United States Department of Energy

US DOE (2009) Explanation of significant difference for the Monticello mill tailings (USDOE) site operable unit III, surface water and ground water, Monticello, Utah.Doc. No S0429400. United States Department of Energy

US DOE (2010) 200 west area groundwater pump-and-treat remedial design report, DOE/RL-2010-13, rev. 1.United States Department of Energy, Richland Operations Office. Richland, Washington, DC

US DOE, US EPA, Ecology (2009) Explanation of significant differences for the 300-FF-2 operable unit interim action record of decision Hanford site Benton County, Washington. United States Department of Energy, United States Environmental Protection Agency, Washington State Department of Ecology. Richland, Washington, DC

US EPA (2002) Preliminary Remediation Goal (PRG). https://epa-prgs.ornl.gov/cgi-bin/radionuclides/rprg_search

US EPA (2004) Dose Compliance Concentration (DCC). https://epa-dccs.ornl.gov/cgibin/dose_search

US EPA (2007) The technology reference guide for radioactively contaminated media EPA-402-R-07-004. United States Environmental Protection Agency (US EPA). Washington, DC

Van der Perk M, Burrough PA, Voigt G (1998) GIS based modelling to identify regions of Ukraine, Belarus, and Russia affected by residues of the Chernobyl nuclear power plant accident. J Hazard Mater 61:85–90

Vives i Batlle J, Beresford NA, Beaugelin-Seiller K, Bezhenar R, Brown J, Cheng JJ, Ćujić M, Dragović S, Duffa C, Fiévet B, Hosseini A, Jung KT, Komboj S, Keum DK, Kryshev A, LePoire D, Maderich V, Min BJ, Periáñez R, Sazykina T, Suh KS, Yu C, Wang C, Heling R (2016) Inter-comparison of dynamic models for radionuclide transfer to marine biota in a Fukushima accident scenario. J Environ Radioact 153:31–50

Vovk IF, Blagoyev VV, Lyashenko AN, Kovalev IS (1993) Technical approaches to decontamination of terrestrial environments in the CIS (former USSR). Sci Total Environ 137:49–63

Wada K, Yoshikawa T, Murata M (2012) Decontamination work in the area surrounding Fukushima Dai-ichi Nuclear Power Plant: another occupational health challenge of the nuclear disaster. Arch Environ Occ Health 67:128–132

Wallace SH, Shaw S, Morris K, Small JS, Fuller AJ, Burke IT (2012) Effect of groundwater pH and ionic strength on strontium sorption in aquifer sediments: implications for $^{90}Sr^{2+}$ mobility at contaminated nuclear sites. Appl Geochem 27:482–1491

Wallace SH, Shaw S, Morris K, Small JS, Burke IT (2013) Alteration of sediments by hyperalkaline k-rich cement leachate: implications for strontium adsorption and incorporation. Environ Sci Technol 47:3694–3700

Wellman DM, Glovack JN, Parker K, Richards EL, Pierce EM (2008) Sequestration and retention of uranium(VI) in the presence of hydroxylapatite under dynamic geochemical conditions. Environ Chem 5:40–50

Wilkins MJ, Livens FR, Vaughan DJ, Beadle I, Lloyd JR (2007) The influence of microbial redox cycling on radionuclide mobility in the subsurface at a low-level radioactive waste storage site. Geobiology 5:293–301

Williams KH, Long PE, Davis JA, Wilkins MJ, N'Guessan AL, Steefel CI, Yang L, Newcomer D, Spane FA, Kerkhof LJ, McGuinness L, Dayvault R, Lovley DR (2011) Acetate availability and its influence on sustainable bioremediation of uranium-contaminated groundwater. Geomicrobiol J 28:519–539

Williams KH, Bargar JR, Lloyd JR, Lovley DR (2012) Bioremediation of uranium contaminated groundwater: a systems approach to subsurface biogeochemistry. Curr Opin Biotechnol 24:489–497

Wu WM, Carley J, Luo J, Ginder-Vogel MA, Cardenas E, Leigh MB, Hwang C, Kelly SD, Ruan C, Wu L, Van Nostrand J, Gentry T, Lowe K, Carroll S, Luo W, Fields MW, Gu B, Watson D, Kemner KM, Marsh T, Tiedje J, Zhou J, Fendorf S, Kitanidis PK, Jardine PM, Criddle CS (2007) In situ bioreduction of uranium (VI) to submicromolar levels and reoxidation by dissolved oxygen. Environ Sci Technol 41:5716–5723

Yabusaki SB, Fang Y, Long PE, Resch CT, Peacock AD, Komlos J, Jaffe PR, Morrison SJ, Dayvault RD, White DC, Anderson RT (2007) Uranium removal from groundwater via in situ biostimulation: field-scale modelling of transport and biological processes. J Contam Hydrol 93:216–235

Yang JS, Kwon MJ, Choi J, Baek K, O'Loughlin J (2014) The transport behavior of As, Cu, Pb, and Zn during electrokinetic remediation of a contaminated soil using electrolyte conditioning. Chemosphere 117:79–86

Yeh GT, Tripathi VS (1990) HYDROGEOCHEM: a coupled model of HYDROlogical transport and GEOCHEMical equilibrium of multi component systems, ORNL 6371, Oak Ridge National Laboratory. Oak Ridge, Tennessee

Yeh GT, Gwo JP, Siegel MD, Li M-H, Fang Y, Zhang F, Luo W, Yabusaki SB (2013) Innovative mathematical modeling in environmental remediation. J Environ Radioact 119:26–38

Zachara JM, Long PE, Bargar J, Davis JA, Fox P, Fredrickson JK, Freshley MD, Konopka AE, Liu C, McKinley JP, Rockhold ML, Williams KH, Yabusaki SB (2013) Persistence of uranium groundwater plumes: contrasting mechanisms at two DOE sites in the groundwater-river interaction zone. J Contam Hydrol 147:45–72

Zhang F, Yeh GT, Parker JC, Brooks SC, Pace MN, Kim YJ, Jardine PM, Watson DB (2007) A reaction-based paradigm to model reactive chemical transport in groundwater with general kinetic and equilibrium reactions. J Contam Hydrol 92:10–32

Zhang R, Chen C, Li J, Wang X (2015) Investigation of interaction between U(VI) and carbonaceous nanofibers by batch experiments and modeling study. J Collo Inter Sci 460:237–246

Zyvoloski GA, Robinson BA, Dash ZV, Trease LL (1994) Models and methods summary for the FEHM application, FEHM MMS SC-194, Los Alamos National Laboratory. Los Alamos, New Mexico