Remediation of Radionuclide-Contaminated Sites Using Plant Litter Decomposition

Jörg Schaller, Mohammad Nassour and Carsten Brackhage

Abstract Radionuclide contamination of ecosystems is a commonly known problem for many sites. A frequently used option in dealing with such contamination is phytoremediation. But when thinking of phytoextraction measures, the process of radionuclide enrichment in plant material is not terminated at the end of the growing season, but may increase during decomposition of the litter afterwards. We show that the process of litter decomposition may be mostly important in remediation of radionuclide-contaminated sites for both aquatic and terrestrial ecosystems. Radionuclide concentrations within organic soil/sediment layers increase strongly during decomposition in terrestrial ecosystems as well as in aquatic systems of temperate zones although there are large differences. This is attributed to emerging fixation sites where differences in aquatic and terrestrial systems are dependent on the particular chemistry (e.g. redox chemistry) of the radionuclides. The potentially high accumulation in developing layers of organic matter on the soils/sediments of aquatic/terrestrial ecosystem can easily be removed from the contaminated sites by removing the organic matter. In summary, beside autochthonous processes (e.g. phytoremediation), especially allochthonous processes (e.g. litter decomposition) are very important for the remediation of radionuclide-contaminated sites.

Keywords Biosorption · Carbon turnover · Decay · Ecosystem engineers · Fixation · Litter processing · Organic matter · Radioactivity

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1 Introduction

Radionuclide contamination of soils and sediments is known for many different ecosystems in many different countries. Apart from radionuclide-rich bedrock (Barth et al. 1998), main sources of radionuclides in the environment are mining activities and industrial waste. Radionuclides may either be drained into hitherto uncontaminated ecosystems or transported even long distances by dust thereby polluting soils and sediments (Entry et al. 1999; Meinrath et al. 1999; Jakubick and Kahnt 2002; Thabayneh and Jazzar 2013).

Phytoremediation would be an option in dealing with such contamination. It comprises techniques that are used especially on sites with comparably low levels of contamination (Salt et al. 1998). As gentle remediation for even larger areas, it became an intensively used technique all over the world (Tripathi et al. 2007) and still is subject to many associated projects on the extension of its operating possibilities/capabilities. For inorganic contaminants like radionuclides, plants in the course of remediation may be used primarily for phytostabilization, rhizofiltration or phytoextraction. The latter result either in a species-dependent accumulation pattern or may even result in phytovolatilization mainly when contaminants like arsenic are methylated (Zhao et al. 2009). Because the accumulation potential is quite different between plants, they have been grouped into species with very high (hyperaccumulators), moderate (accumulators) and no accumulation potential (mainly excluders) (Salt et al. 1998). In any case, this is not only species but also element and site-specific. For radionuclides, the accumulation pattern is not different between different isotopes of the same element. Therefore, the following chapters refer mostly to the element rather than different isotopes. Hyperaccumulating plants of radionuclides have not been described so far. Nonetheless, the accumulation potential of some plant species for uranium led to their use in ore prospection (Cannon 1960). Using the accumulative potential of plants may partially reduce radionuclide concentrations in soils and to a minor extent in sediments (phytoextraction) but without harvest this organically fixed portion cycles back to the soil/sediment via litter fall.

The process of radionuclide enrichment in plant material is not terminated at the end of the growing season, but may increase during decomposition of the litter afterwards (Schaller et al. 2010b, 2011b). More than 80 % of the plant biomass produced in terrestrial ecosystems is directly proceeding to detritus (Gessner et al. 2010). Litter decomposition generally proceeds in three distinct temporal stages of leaching, microbial conditioning and fragmentation (Berg and McClaugherty 2008; Gessner et al. 2010). During the primary decomposition by microorganisms, dissolved organic carbon (DOC) emerges from the litter and microorganisms form together with their exudates a heterotrophic biofilm (Kominkova et al. 2000; Berg and McClaugherty 2008). After the formation of this heterotrophic biofilm (including heterotrophic fungi and bacteria), the litter will be microbially decomposed. In the last step of the decomposition process, the litter will be processed/decomposed/fed on by higher trophic levels (vertebrate and invertebrate animals) (Berg and McClaugherty 2008; Gessner et al. 2010). The fixation and hence the remediation potential for radionuclides therefore are not only given by the accumulation potential of living plants but also by their biomass production and its reformation in the course of decomposition with emerging fixation sites.

This book chapter therefore tries to elucidate the interplay between decomposition of dead plant material (litter) and radionuclide fixation/remobilization potential in aquatic and terrestrial ecosystems and its importance for phytostabilization.

2 Radionuclide Fixation During Litter Decomposition Within Aquatic Ecosystems

On a continental scale not regarding oceans, terrestrial ecosystems may be more important for radionuclide fixation/turnover compared to aquatic ecosystems. But the interplay between organisms and litter in the course of decomposition and hence their importance for radionuclide fixation/turnover may be easily explained for aquatic ecosystems.

The first step during litter decomposition in aquatic ecosystems is the leaching process (Fig. 1). Leaching of DOC as a substantial part herein is mainly occurring during the first 24 h (Gessner et al. 1999). The type of DOC and hence its composition depends on the decomposition stage of the litter and the litter quality. At first, water-soluble fractions (e.g. sugars, amino acids) are released. In a later stage in the course of humification, humic and fulvic acids are leached (Schumacher et al. 2006). These humic and fulvic acids enhance the remobilization of elements from the sediment (Franke et al. 2000). A main part of elements in the so-called dissolved fraction is bound in colloidal form (Baalousha et al. 2006; van Leeuwen and Buffle 2009). In contrast to the above-mentioned findings of DOC impact on element mobilization, other studies found no correlation between an increasing DOC level and the remobilization of radionuclides (Schaller et al. 2008, 2010b). The leaching of radionuclides from the litter (especially litter originating from accumulating plants) is low at the beginning and negligible over the entire time of

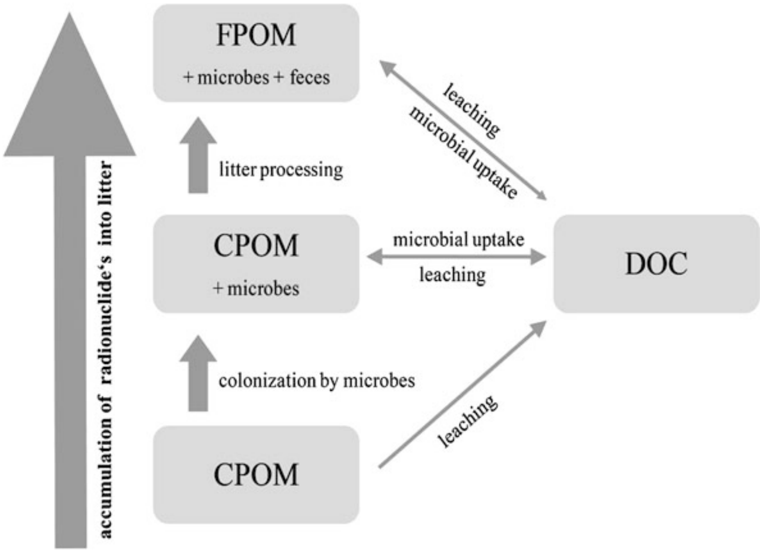

Fig. 1 The process of litter decomposition from coarse particulate organic matter (*CPOM*) to fine particulate organic matter (*FPOM*) and dissolved organic carbon (*DOC*), whereas the resulting changes in litter properties affect the radionuclide fixation, modified after Schaller et al. (2011b)

the decomposition process in relation to the high elemental load of the water passing by during decomposition.

In the second step of decomposition, plant litter will be colonized by microbes (Dang et al. 2007) (Fig. 1). Microbial litter decomposition depends primarily on fungal decomposer and litter diversity (Dang et al. 2005), bacteria and detritivores (Hieber and Gessner 2002), but also on litter quality. Main parameters affecting litter quality are lignin, cellulose, poly-phenol, tannin, nitrogen and phosphorus content (Gessner and Chauvet 1994). But also environmental factors such as pH are important (Lecerf et al. 2007). Decomposition rates of plant litter also depend on water chemistry like acidification (Dangles et al. 2004) and nutrient concentration (Suberkropp and Chauvet 1995). Metals and metalloids themselves affect the microbial decomposer community and consequently affect the decomposition rate (Berg et al. 1991). The amount of fungi growing on the litter decreases in the presence of high radionuclide concentrations (Ferreira et al. 2010). Consequently, the decomposition rate decreases in environments with high radionuclide loads compared to environments with lower radionuclide concentrations, as shown for other elements (Sridhar et al. 2001; Duarte et al. 2008). It was also suggested that microbes produce more exopolysaccharides (EPS) to protect themselves against high concentrations of contaminants (Pirog 1997). These EPS are well known to immobilize elements in high amounts (Huang et al. 2000) by their functional groups and expanded surface area. On the other hand, microbes themselves

Table 1 Activity concentrations shown for ^{238}U, ^{235}U, ^{226}Ra, ^{210}Pb and ^{137}Cs for leaf litter after 8 weeks of microbial decomposition within a stream affected by former uranium mining (Neuensalz, Germany)

Isotope	Activity concentrations (mean ± SD in Bq kg^{-1})
^{238}U	8,300 ± 1,100
^{235}U	390 ± 50
^{226}Ra	650 ± 50
^{210}Pb	<112
^{137}Cs	<5

accumulate high amounts of radionuclides (D'Souza et al. 2006; Purchase et al. 2009). The resulting changes in chemical properties lead to high amounts of radionuclide fixation into plant litter (Flemming et al. 1996; Schaller et al. 2008, 2010b) (see Table 1). The heterotrophic organisms of the biofilm are consumers of oxygen, which results in a redox gradient within the biofilm. There are positive redox conditions at the surface of the biofilm, while negative redox conditions develop towards the centre of the biofilm (thickness 100–1,000 μM) (Paerl and Pinckney 1996). But the negative redox potential formed by heterotrophic biofilms can be inverted in the presence of high concentrations of manganese oxidizing the biofilm (Chinni et al. 2008), which in turn leads to a remobilization of radionuclides with reverse redox chemistry (e.g. uranium) (Schaller et al. 2010b). In contrast, in the absence of high amounts of manganese, it was revealed that high amounts of radionuclides (e.g. lead and uranium) are fixed by these heterotrophic biofilms (with negative redox potential) attached to plant litter (Schaller et al. 2008).

The fragmentation of plant litter depends on the feeding activity and behaviour of animals like invertebrates (Graça 2001). The preferred food of, for instance, invertebrate shredders as key species in aquatic litter decomposition is plant litter colonized by microbes in the primary stages of decay (full established biofilm) (Graça et al. 2001; Franken et al. 2007). These invertebrate shredders cut the litter into smaller particles, which increases the surface area (Schaller et al. 2010a; Schaller and Machill 2012). On this enhanced surface area, more microbial biofilm will grow, which significantly increases the fixation capacity for elements (Schaller et al. 2010a) (Fig. 1). An effect of invertebrate shredders on contaminated leaf litter was first described for uranium (Schaller et al. 2008). It was found that high uranium concentrations in water and litter have no negative effect on the survival rate of invertebrates (Schaller et al. 2009). In contrast, a significant lower decay rate of leaf litter in metal-polluted compared to unpolluted environments was found by experiments with shredding invertebrates (Medeiros et al. 2008), depending on different element speciation and pH values (Schaller et al. 2008). Furthermore, invertebrate shredder shows a lower feeding rate in radionuclide-polluted environments compared to uncontaminated environments (Goncalves et al. 2011). Data from an in situ experiment revealed that invertebrate shredders facilitate radionuclide (e.g. lead) enrichment into smaller particle sizes of POM in

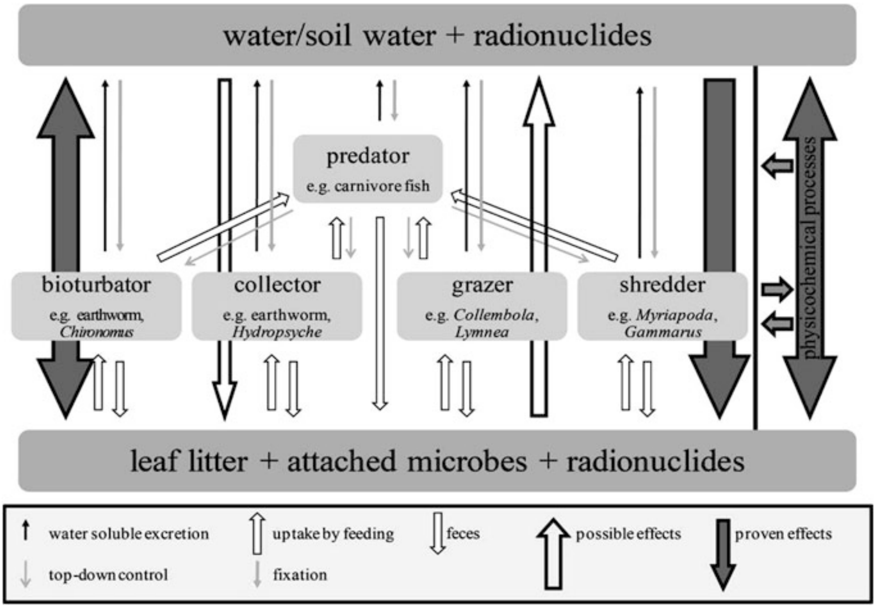

Fig. 2 Overview of proven and possible effects of different types of functional animal groups on metal/metalloid accumulation/remobilization within terrestrial/aquatic ecosystems, modified after Schaller et al. (2011b)

the presence of high concentrations of DOC (Schaller et al. 2010b). These high DOC levels were expected to bind radionuclides (Sachs et al. 2007), but the opposite was shown in different experiments (Schaller et al. 2010b). Current studies reveal a control of radionuclide fixation by silicon availability during plant growth reducing the binding sites at the litter surface for plant material grown under high silicon availability (Schaller 2013), forming a so-called silicon double layer within the epidermis near area (Schaller et al. 2013a). This seems to be quite important because of the enhanced decomposition rate of this plant litter (probably more biofilm) with high silicon content (Schaller and Struyf 2013). The role of other important ecosystem processes has still to be elucidated (see Fig. 2). For some chironomids, it is known that they have a positive effect on radionuclide remobilization (Schaller 2014), but less is known about the effect of other keystone species so far. Hence, a broad area of research opens here. Altogether, the developing layers of organic matter on/in aquatic sediments have a very high potential to trap radionuclides during organic matter decomposition. Hereafter, radionuclides can be easily removed from contaminated sites by removing the organic matter.

3 Fixation Potential of Terrestrial Ecosystems and the Influence of Litter Decomposition

The fixation potential of terrestrial ecosystems such as grasslands or forests is highly dependent on soil organic layers and therefore influenced to a great extent not only by the accumulation potential of abundant plant species resulting in contaminated litter but also by litter decomposition affected in turn by emissions of radionuclides. Terrestrial ecosystems are more complex compared to aquatic systems regarding litter decomposition. The complexity is based on spatial and temporal heterogeneity of the soil texture resulting in a high density of different habitats. This leads to a more or less simultaneous occurrence of leaching, microbial decomposition, fragmentation and decomposition by animals (Berg and McClaugherty 2008). In many cases, fallout radionuclides were investigated for their mobility in different soils and soil layers. This was done with a compartment model using the activity concentrations over time for the different soil layers resulting in residence half-times (e.g. Bunzl et al. 1994). It was shown that long-lived radionuclides such as $^{239+240}Pu$, ^{241}Am and ^{137}Cs are still found within a 30 cm depth of a grassland soil after 30 years of deposition (Bunzl et al. 1994). Residence half-times were found not to be significantly different for all three radionuclides, but derived migration rates taking into account the thickness of each soil layer are increased with depth. Further investigations showed that especially organic matter is involved in the fixation potential and hence low migration rates in upper soil layers (Bunzl and Trautmannsheimer 1999). Rafferty et al. (2000) summarize the migration mechanisms and hence the pools and pathways for ^{137}Cs in a coniferous forest soil. They propose a three-phase model where the phases are discrete but may occur in a given area in parallel. In the first phase, ^{137}Cs is intercepted by the forest canopy and washed onto the soil and a portion of 20–40 % immediately percolates through the organic layers into the underlying mineral soil. Over the next estimated 5 years, the remaining ^{137}Cs moves by leaching and/or decomposition from the Ol- and Of-horizons to the Oh-horizon, where here in the third phase, it is more or less permanently fixed. This is due to a very slow decomposition of organic matter in the Oh-horizon of coniferous forest soils.

A leaching of radionuclides from the plant litter coincidently to the mobilization of DOC may probably take place if the concentration of the fresh fallen plant litter is very high (Sauras et al. 1994). Vice versa plant litter may accumulate radionuclides from soil water/splashing water during litter decomposition as was shown for ^{137}Cs. During plant growth on contaminated soils, the radionuclides will accumulate within the soil organic layer due to plant uptake and even more during litter decomposition afterwards (Choppin 1988; Tikhomirov and Shcheglov 1994; Fesenko et al. 2001). The radionuclide accumulation/fixation by plant litter during decomposition in terrestrial systems takes place as described for aquatic systems (see above). Such an accumulation of radionuclides in litter during decomposition was described for many other ecosystems (Agapkina et al. 1995; Bunzl et al. 1998;

Copplestone et al. 2000; Vaca et al. 2001), where it was indicated that the radionuclides are bound organically (Virchenko and Agapkina 1993). If the soils of terrestrial ecosystems develop such a layer of organic matter and trap the radionuclides within this layer during decomposition, the radionuclides can be easily removed from the contaminated sites by removing the layer of organic matter.

Unfortunately, organic matter is transported from the litter layer into the mineral soils, mostly by soil fauna (Chamberlain et al. 2006; Frelich et al. 2006). This mixing of organic matter with the soil mineral phase is in turn a process diminishing remediation options for radionuclides. In contrast to predicted effects of the soil fauna, such a radionuclide transport into deeper soil horizons was found only in very low amounts (Bunzl et al. 1992). The assemblage of the soil fauna changes during decomposition. For larger detritivores, the litter transformer or shredder such as isopodes, millipedes and epigeic earthworms feed earlier on the organic matter (detritus) than soil-dwelling animals (bioturbation) such as endogenic earthworms, insect larvae and some collembolans (Bastow 2012). Hence, the detritivores feeding first transforming the litter into smaller particles (shredder) lead to an enhanced fixation of radionuclides to organic matter (litter under decomposition), by extending the surface area of the litter, as shown earlier for aquatic systems (see above). In contrast, detritivores following after these first processes affect the radionuclide fixation and distribution by bioturbation (see above).

In conclusion, radionuclide concentrations within organic soil layers may increase even strongly during decomposition in terrestrial ecosystems, as shown earlier for aquatic systems. But this depends on factors such as type of radionuclide, bedrock concentrations, emissions, plant community, climate conditions and hence development of organic layers. In temperate zones, litter decomposition and organic soil formation following phytoremediation by planting accumulating species can be highly complementary for remediation of radionuclides. This combination has the potential to increase the effectiveness of remediation measures of radionuclide-contaminated sites tremendously. Autochthonous processes (e.g. phytoremediation during plant growth) and allochthonous processes (e.g. litter decomposition) together are probably more efficient in element fixation/immobilization as has been indicated already for aquatic ecosystems (Schaller et al. 2013b).

4 Radionuclide Impact on Higher Trophic Levels

There is a general trend that organisms in a contaminated environment show increased concentrations of the contaminants (Rainbow 2002). Hence, concentrations of radionuclides well above background levels may affect multiple levels of biological organization from ecosystem, community, population, individual, cellular, subcellular to molecular levels (Peplow and Edmonds 2005). Effects of stressors on the food web are described to be dampen at higher trophic levels in the course of transfer to the food web (Schindler 1990), but can also be intensified (Breitburg et al. 1999). Still, bioaccumulation differs depending on species (Cain et al. 2004), age (Wallace et al. 2003), sex and fitness of the animals. The uptake of potential toxic trace elements can take place via two different pathways: either directly via surface and/or via ingestion and uptake in the digestive tract (De Schamphelaere et al. 2004). Regulation of potential toxic trace element uptake includes one of the following adaptive strategies: (1) limiting entrance into the body directly, (2) balancing uptake by increasing excretion thereby maintaining a constant total concentration or (3) by detoxifying and storing elements when entering organs or cells (e.g. metallothioneins) (Barka et al. 2010). Some invertebrates show only a low accumulation potential presumably excluding uptake which has been explained by the detoxification of potential toxic elements in gastrointestinal epithelial cells and excretion together with these epithelial cells (Ahearn et al. 1999; Amiard et al. 2006). A higher accumulation of radionuclides into the gut system compared to the remaining tissues of the body was shown for species of the genera *Gammarus* (Sola and Prat 2006; Schaller et al. 2011c). Also potential toxic elements/radionuclides adsorb onto the chitin cuticle of invertebrates (Lenhart et al. 1997) depending on cuticle properties (Lightner et al. 1995). Thereby, a size-dependent bioaccumulation of elements on the surface of invertebrates was observed (Wang and Zauke 2004) resulting in an increased tolerance. Some invertebrates living in environments with high element load may be able to excrete ingested elements to clean their body and avoid high internal concentrations (Tessier et al. 1994; Alves et al. 2009; Schaller et al. 2011c). But many species associated to litter decomposition are described to be highly sensitive to radionuclides (Borgmann et al. 2005; Sola and Prat 2006). Elements can enter organisms via passive transport across the plasmalemma using carrier systems that yield them to higher affinity sites (S- and N-protein binding). Elements can also enter cells down a concentration gradient through specific hydrophilic transmembrane channels. Passive diffusion may also be a way when being in a lipid-soluble (non-polar) form or elements are taken up by endocytosis (Rainbow 1997). It is therefore unlikely that differences in element accumulation between the gut system and other (remaining) tissue(s) of invertebrates are exclusively due to the prevention of entrance into gut epithelial cells. Dietary uptake of elements into invertebrate shredder may cross into gut epithelial cells from the faeces. However, significant differences in radionuclide concentrations between the gut system (and its content) and remaining tissues (Schaller et al. 2011c) show that effective detoxification mechanisms (like sequestration and/or excretion) exist to prevent their entrance into the haemolymph and subsequent dispersal throughout the body. Within hepatopancreatic cells, dietary non-essential elements are isolated by complexing with metallothioneins, glutathione and inorganic anions including sulphur and phosphorus removing them from metabolic activities and hence reducing the haemolymph concentration (Barka et al. 2001; Chavez-Crooker et al. 2003; Amiard et al. 2006). Non-essential elements are precipitated into sulphur and phosphorus containing granules (Nassiri et al. 2000; Sterling et al. 2007).

These elements are sequestered for the life of the cell and then expelled into the gut system at the end of the cell cycle (Sterling et al. 2007; Barka et al. 2010).

Invertebrate shredders are known from toxicity tests to be sensitive to radionuclides such as uranium (Robertson and Liber 2007). It was shown that mortality increases by increasing pollutant concentration. On the other hand, some populations of the European shredders *Gammarus* sp. show no significant differences in survival rate in experiments between polluted and non-polluted water and food (Schaller et al. 2009, 2011a). Furthermore, for *Tubifex* sp. (Oligochaeta) another litter processing invertebrate, no effect of metals/radionuclides on the survival rate was found (Kaonga et al. 2010). In addition, some collectors of the genus *Hydropsyche* (Trichoptera) seem also not affected by high metal/radionuclide concentrations (Clements et al. 2000). For terrestrial systems, less is known about the toxicity of radionuclides on soil animals. For earthworms and soil arthropods, no effects were detected below 1,000 mg U kg^{-1} (earthworm) and 350 mg U kg^{-1} (soil arthropods) (Sheppard and Stephenson 2012). This very high threshold may indicate an adaption to habitats with high radionuclide load. A biomagnifications (e.g. for invertebrates) was proven (Copplestone et al. 1999) but may be rather explained by the amount of radionuclide containing food and faeces within their body than by a real uptake and accumulation of radionuclides within tissues other than the gut system, as it was found earlier for aquatic detritivores (Schaller et al. 2011c). Interestingly, earthworm and collembola seem to be highly tolerant regarding even radiation (Fuma et al. 2011). With an ED50 of about 1,300 Gy for collembola (Nakamori et al. 2008) and about 800 Gy for earthworms (Fuma et al. 2011), an effect of radiation from radionuclides on soil animals on contaminated sites can be excluded. Hence, effects of radionuclides on health and mortality of animals on contaminated sites are more due to their chemical toxicity.

We suggest that organisms living in habitats which tend to high radionuclide accumulation may have been adapted during their evolution, because they had to handle these conditions the whole time. If they had not be adapted to high radionuclide load (probably by minimizing the uptake or maximizing the excretion), they would not still live in such habitats. Hence, a low mortality rate in the presence of high concentrations of metals/radionuclides in the environment can possibly be explained by the evolutionary adaptation to these environments with high bioavailability of radionuclides/metals.

5 Conclusions

In this chapter, we have seen that the process of litter decomposition can be used to enhance the effectiveness of the remediation of radionuclide-contaminated sites. The litter decomposition and radionuclide accumulation is controlled by microbes (bacteria and fungi), their exudates (e.g. EPS), nutritional properties and higher trophic levels (e.g. shredder, grazer and bioturbator). Litter decomposition in aquatic systems proceeds in three distinct temporal stages of leaching, microbial conditioning and fragmentation. Terrestrial ecosystems are more complex compared to aquatic systems regarding litter decomposition, because of the complexity based on spatial and temporal heterogeneity of the soil texture resulting in a high density of different habitats. This leads to a more or less simultaneous occurrence of leaching, microbial decomposition, fragmentation and decomposition by animals in terrestrial systems. The most important process for radionuclide accumulation/fixation is the formation of the heterotrophic biofilm by the microbial decomposer community. These biofilms have a very high capability for radionuclide accumulation/fixation. Within these biofilms growing on plant material/organic matter, the radionuclides can be enriched up to ore level. The impact of higher trophic levels in turn controls the radionuclide accumulation/fixation by influencing the amount of heterotrophic biofilm growing on the litter (e.g. invertebrate shredders) or changing the redox conditions (e.g. bioturbators). Altogether, litter decomposition is highly prone for remediation of radionuclide-contaminated sites, because of the very high capability of radionuclide accumulation/fixation of litter/organic matter under decomposition. We suggest that the remediation of radionuclide-contaminated sites will be much more efficient with the implementation of litter decomposition into the phytoremediation techniques compared to techniques without litter decomposition.

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