Fate and transport of radiocesium (\(^{137}\text{Cs}\)) in terrestrial environment

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ABSTRACT

The major artificial radionuclide present in the environment is \(^{137}\text{Cs}\), which is released as a result of weapon production related to atomic projects, accidental explosions of nuclear power plants and other sources, such as reactors, evaporation ponds, liquid storage tanks, burial grounds, etc. Radiocesium’s high fission yield and ease of detection made it a prime candidate for early radio-ecological investigations. The facility sitting provide diverse background for the better understanding of various factors contributing more towards the fate and transfer of radionuclides in the terrestrial ecosystem. It has been found that \(^{137}\text{Cs}\) can trace the transport of other radionuclide that have a high affinity for binding to soil particles (silts and clays).

1. INTRODUCTION

Cesium-137 (a medium half-life about 30 years) is one of the most important and largely distributed radionuclide in the terrestrial environment due to anthropogenic source such as: nuclear bomb tests, nuclear power plant accidents, leaching from waste disposal radionuclide activities, nuclear weapon race etc. (Zaborska et al., 2014; Steinhauser et al., 2013; Vallés et al., 2009). For last so many years, considerable interest has arisen with regard to the fate and transport of radionuclides in the terrestrial ecosystem after the Chernobyl born fallout in forest ecosystem of Russian federation and Ukarain badly affected by Chernobyl accident (1986) and the Fukushima Daichi Nuclear Power Plant (FNPP) disaster on March 11, 2011. This led to the release of large amounts of radionuclides into terrestrial and aquatic ecosystem. [4-6]. Large amount of radionuclide released into the environment within a month after these accidents estimates hazardous behavior ~100 \(\times\) 2015 Bq of \(^{134}\text{Cs}\) and ~13-50 \(\times\) 1015 Bq of \(^{137}\text{Cs}\). (Hinton et al., 2013; Hou et al., 2003; Zalewska and Lipska, 2004). The \(^{137}\text{Cs}\) distribution and accumulation also occur in the history of Baltic sea-sediments. The Baltic Sea is susceptible to pollution by hazardous substances due to limited water exchange, shallowness, and the large catchment area (Zaborska et al., 2014). These anthropogenic sources increased radionuclide pollution more quickly so there was a need to monitor their dispersion, understand the resulting contamination levels and mitigate the human health and environmental risks. Therefore this research...
was conducted to overview the fate and transfer of radiocesium in terrestrial environment.

$^{137}$Cesium in the environment originates from a variety of sources. Early in 1990, the Institute of Nuclear Energy Research (INER, Taiwan) accidentally discharged radionuclide wastewater into an irrigation ditch and contaminated the agricultural ecosystem. Part of the agricultural soil contaminated with radionuclide was removed by INER in 1994 and 1995 (Chiang et al., 2010). Mobility and bioavailability of radionuclides is determined by ratio of radionuclide chemical forms in fallout and the site-specific environmental characteristics. This determines (a) the rates of leaching, (b) fixation/remobilization, (c) sorption-desorption of mobile fraction (its solid-liquid distribution). The total distribution coefficient for radionuclides can vary in a wide range (4 orders of magnitude for radiostrontium and 5 orders of magnitude for radiocaesium) as a function of fallout characteristics and environmental conditions (Ritchie and McHenry, 1990). The compartment model POSEIDON-R for the northwestern pacific and adjacent seas stimulate the fate and transfer of radioactivity in the period 1945-2010, and to perform a radiological assessment on the release of radiocesium due to Fukushima Daiichi accident for the period 2011-2014. A generics predictive dynamic food chain model was used instead of biological concentration factor (BCF) (Maderich et al., 2013). Spent nuclear fuel re-processing wastes may introduce small amounts of contamination into the environment (Simpson and Law, 2013). Industrial instruments containing $^{137}$Cs are misplaced (Leon et al., 2011) so people handling directly with these may be effected. When this radionuclide enters the environment causes serious pollution (Mihaela et al., 2012). The largest deposition of Cesium is in the forest soil ecosystem due to the Chernobyl born fallout in Russian federation and Ukrarin badly affected by Chernobyl accident (1986). In soil profile the intensity of $^{137}$Cs transport depends largely on the type of ecosystem and soil properties, along with forest litter structure and depth (Shcheglov et al., 2013) solid liquid distribution coefficients ($K_d$) of radiocesium for various soil types is based on texture and organic matter content, derived from geometric means (GM) and also using soil cofactors governing soil–radionuclide interaction (Nakanishi et al., 2014). The geographical trends in $^{137}$Cs fallout from Chernobyl accident and leaching from natural surface soil in Norway were also evaluated (Gjelsvik and Steinnes, 2013). Due to its chemical properties, $^{137}$Cs is readily transported through the environment and food chains. A part from abundance cesium possesses many properties like half-life (~30 y), emits relatively high-energy beta particles and its rather short-lived daughter, 137m Ba, emits strong gamma rays which makes it a good contributor towards terrestrial ecosystem (Chino et al., 2011). The Fukushima Dai-ichi nuclear power plant accident in Japan triggered by a big earthquake and the resulting tsunami on 11 March 2011 were the major source of radiocesium ($^{137}$Cs and $^{134}$Cs) and contamination of soils in terrestrial ecosystem. Vertical distributions of radiocesium and physicochemical properties in soils (to 20 cm depth) at 15 locations under different land-use types (croplands, grasslands, and forests) within a 2 km × 2 km mesh area in Fukushima city were predicted (Nakanishi et al., 2014). Two Italian stations in the north Apennines: Mt. Cimone (Modena) and Montecuccolino (Bologna) used to follow up the Fukushima radioactive plume resulting from 11th March 2011 overwhelming tsunami analysis.
confirms the arrival of radionuclides following atmospheric transport and processing. Fukushima radioactivity data at the two stations were usually comparable, suggesting a good vertical mixing of the plume; discrepancies were occasional and attributed to different occurrence of wet removal, typically characterized by a scattered spatial pattern (Tositti et al., 2012). The total distribution coefficient for radionuclides is a dynamic characteristic and depends on transformation rates of chemical forms. High retention of radiocaesium in soils is caused by two main processes: 1- selective reversible sorption on illitic clay minerals and fixation. 2- Advanced methods have been proposed for determining the capacity of selective sorption sites (Frayed Edge Sites – FES) and exchangeable radiocaesium interception potential (RIPex). Quantitative data were obtained for a wide range of soils and bottom sediments with respect to FES capacities and RIPex (Wampler et al., 2012).

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\text{FES} - M + ^{137}\text{Cs}^+ \leftrightarrow K_c^{\text{FES}(\text{Cs}/M)} \rightarrow \text{FES}^{^{137}\text{Cs}} + M^+ \\
\text{RIP}^{\text{ex}}(M) = K_d^{\text{ex}}(\text{Cs}) \times m_M = K_c^{\text{FES}(\text{Cs}/M)} \times [\text{FES}]
\]

The effect of accompanying anions (\(\text{NO}_3^{-1}\), \(\text{SO}_4^{2-}\), \(\text{CO}_3^{2-}\), and \(\text{I}^-\)) on retention and translocation of cesium performing experiment in a greenhouse by using droplets of stable cesium solution to upper surface of four soybean trifoliate leaves shows that once cesium dissolved, absorbed and penetrate the cuticle very rapidly (Yan et al., 2013). The uptake and translocation of radionuclide in tropical crops and ecosystems depend on the conditions of tropical soil types and the factors influencing radionuclides biogeochemistry (Tagami et al., 2013). The soil to plant transfer of radionuclides and their effects on humans and the environment also depends on the change in global warming (Dowdall et al., 2008). The rate and capacity of transformation and dynamics of Cs and Sr sorption by selected subtropical and tropical soils was also established under highly weathering intensity in Taiwan (Chiang et al., 2010). The effect of soil characteristics and fungal and plant transporters on radiocaesium uptake and
accumulation in plants focus on the radiocesium immobilization and plant accumulation on mycorrhizal fungi. Fungi develop hyphae in the soil, a source to transfer nutrients from soil to plants (Dupré de Boulois et al., 2008). The movement of soil organic carbon (SOC) down the soil profile compared to environmental tracer $^{137}$Cs at two neighboring field sites comprising clay soils with different cracking characteristics (cracking black Vertisol and a red Luvisol) was observed (Wells and Hancock, 2014). Sensitivity of rural and semi natural environments towards radionuclide contamination by $^{137}$Cs, $^{90}$Sr and $^{131}$I, after major nuclear accident also taken into account indicates that environmental sensitivity was highly radionuclide specific and time dependent, with $^{137}$Cs giving highest doses for adults in terrestrial ecosystem and fresh water pathways than in coastal marine environment where $^{131}$I was more noticeable during first year. Sensitivity dependent on social and economic factors like individual living habits, food consumption preferences, and agricultural practices (Tracy et al., 2013) Water drawn from streams for irrigation result in the transfer of contaminated material from the stream to the land. On the other hand, runoff can carry small soil particles containing $^{137}$Cs from the land to waterways (Yasunari et al., 2011). Compared to agricultural land, forests acts as complex ecosystem as they involve miscellaneous plant species associations, several vegetative strata (overstorey, shrubs, herbaceous and other annual plant layer) and multi-layered soil profiles (forest floor, hemi-organic and mineral layers) (Rochette et al., 2000). Large areas of forest in Sweden for the deposition of radiocesium ($^{137}$Cs) after Chernobyl accident in 1986 also show large scale accumulation. 20 woody plants (12 evergreen and 8 deciduous species) grown in Abiko indicates radiocesium deposition after Fukushima power plant accident show average radiocesium activity 7.7 times to that in the leaves of deciduous species (Vinichuk et al., 2010; Okumura et al., 2013).The other factors which show potential to release radiological contaminants to environment include radionuclide activity concentration in Litter (i.e. vegetative debris) and duff (i.e. highly decomposed vegetative debris).Study was carried out in United States Department of Energy's Savannah River Site (SRS), Aiken, South Carolina. Lemon tree (Citrus limon B) indicates the uptake and transfer of soil of $^{137}$Cs to fruits in tropical plants during fruit growth. Maximum values of transfer factors were reached in the initial phase of fruit growth and decreased as the fruit develops (Velasco et al., 2012). Japanese population put emphasis on the estimation of the radiation doses for radionuclide deposition from inhalation of contaminated air, terrestrial and marine food contamination, compared with other sources of anthropogenic (global fallout, Chernobyl accident), natural (radionuclides in food, cosmic radiation) and medical applications (X-ray tests, CT-tests, etc.) of ionizing radiation (Koizumi et al., 2012). Peat deposits in SW Spitsbergen for man-made radionuclide activity. Maximum activities evident in the peats correspond to the 1963/1964 global maximum fallout from atmospheric testing of nuclear weapons; some of the activity profiles have been altered post-deposition by water infiltration (Lozano et al., 2011). The measurement of $^{137}$Cs in Bratisalava stratospheric air put a great impact on the $^{137}$Cs activity concentrations until early 1980s, recognized by typical spring or early summer maxim and winter minima (Povinec et al., 2012). Due to similar chemical behavior, potassium and phosphorous put influence on radiocesium, transported by arbuscular mycorrhizal fungi (AMF). The arbuscular mycorrhizal-plant (AM-P) in vitro culture system associating Medicagotruncatula plantlets with Glomusintraradices
indicates the increasing concentration of both elements on the transport of cesium (Gyuricza, 2010). The activity concentration of radionuclide in edible wild berries and mushrooms collected from Øvre Dividalen national park, Northern Norway. $^{210}$Po, $^{210}$Pb, $^{40}$K and $^{137}$Cs indicates the effective ingestion doses to man based on high consumption rates. Activity concentrations of $^{137}$Cs in edible wild berries and mushrooms reflected the lower levels of fallout of this radionuclide in Northern Norway compared to more central areas following the Chernobyl accident (Ben-Asher, 2011). Seasonal variations in foliar $^{137}$Cs levels were examined in Norway spruce ($Picea abies$ (L.) Karst.), and Scots pine ($Pinus sylvestris$ L.) in western Finland. The analyzing levels of total $^{137}$Cs in the three youngest needle age classes were compared to the levels of potassium and carbon. For needle content and activity concentration of $^{137}$Cs in the time series phases of intensive growth, needle elongation and dormancy were apparent (Paasikallio et al., 1994). The Chernobyl accident in 1986 and the recent Fukushima Daii Chi disaster have produced many new studies in radioecology, primarily in Japan, the USA, the former Soviet Union and other European countries. Earlier work on $^{137}$Cs in the environment was motivated by scientific curiosity and concerns over the health and environmental impacts of global fallout from nuclear testing, and later on the safety of nuclear reactor to produce electricity. Due to rapid distribution of cesium in the environment and global climatic changes, it is going to be more hazardous. Most of the previous work on cesium distribution shows its importance in the environment. This work adds to the significant body of literature from earlier studies.

This review complies and updates our knowledge of transfer parameters in terrestrial ecosystem. Furthermore from the radiocesium study in this review, the fate of other radionuclides in terrestrial ecosystem should also be better documented. Since the knowledge in this review indicates the fate and transfer of radiocesium and share to people so that its impact on terrestrial ecosystem can be minimized and provide answers to questions which arise after its distribution.

2. BEHAVIOR OF CESIUM-137 IN TERRESTRIAL ECOSYSTEMS

The environmental transport of cesium is governed by many factors, most of which vary over space and time. The accumulation of cesium varies by orders of magnitude between different biological components within a single environment and also among different ecosystems. Much of this behavior can be understood from the chemical properties of cesium and its interactions in the soil.

The soil is particularly important because it is the primary reservoir of 137Cs in most ecosystems. The fraction of the 137Cs in soil that is available for biological uptake and transport is determined by the strength of its binding to soil particles. This binding strength is mainly dependent on the clay mineral composition and abundance in the soil. Other chemical factors that modify its transport include the soil CEC, and the pH and potassium concentration of the soil water. For example, acidic conditions tend to enhance the biological availability of 137Cs in soil, while high concentrations of potassium in soil water tend to depress cesium uptake by plants and subsequent transfers to higher trophic levels. Cesium exists in the environment in the +1 oxidation
state. There is little tendency for cesium to form aqueous complexes in the soil/water environment. Thus, the formation of inorganic complexes does not have a major influence on the chemical speciation of cesium, and the dominant aqueous species in most soil and groundwater systems is the uncomplexed Cs+ ion. Unlike many other radionuclides, sorption of cesium to sediments is highly dependent on the mineralogy of the sediment. Unweathered phyllosilicates like micas can be transformed into illites, vermiculites or smectites, depending on the extent to which they have been physically, chemically and biologically weathered. The extent of weathering increases in the order of: mica < illite < vermiculite < smectite < kaolinite. Weathering has a profound effect on a number of physical and chemical properties that in turn have a direct effect on the mineral’s tendency to sorb cesium.

The transport of 137Cs through the environment involves a number of biogeochemical pathways. These include mainly physical processes that move contaminated particles and that are not specifically affected by the chemical nature of the contaminant of interest. For example, the processes of deposition from the atmosphere on to soil or plant surfaces, soil erosion by wind or water, physical percolation of particles into the soil profile, weathering of particulate bound material from plant surfaces, senescence of plant parts, and inhalation or ingestion rates of animals are generally not contaminant-specific. On the other hand, there are processes that depend greatly on the specific contaminant, and on its physical and/or chemical form. Such contaminant-specific processes include foliar absorption, plant uptake from the soil, translocation within the plant, and assimilation, distribution and retention in animals. In the case of cesium, these contaminant-specific processes are affected by the concentrations of specific elements such as potassium, soil pH, and other site-specific conditions. With respect to cesium-specific processes, it has been observed that if in a soluble state, plant leaves will absorb a significant fraction, on the order of 40 to 80 % of a surface deposit, depending on the species of plant. Absorption can approach 100 % in lichens and mosses. Because of the greater absorption of cesium by lichens, and a much longer retention after deposition, these organisms reach steady-state concentrations of 137Cs that can be over an order of magnitude higher than in adjacent herbaceous plant species.

The migration of initially soluble cesium in soil down through the soil profile by aqueous phase transport, or leaching, is normally very small because of its strong binding to clay minerals in the soil, however physical transport of material bound to small soil particles does occur. The transfer of 137Cs from soil to plants occurs by different mechanisms, including uptake through the roots from the soil solution, resuspension from the soil surface, and rain splash of contaminated soil particles. Uptake can be strongly affected by plant species and soil properties, as well as influences from fungi and other microbes.

The transfer of 137Cs from ingested plants and soil to animal tissues is also complex, and depends on a host of conditions. One way of expressing the effectiveness of transfer from food to animal tissues is the simple $Cr$. For example, the animal tissue/food $Cr$ ranges from less than one in herbivores to 20 in predators, reflecting a
general trophic level increase for 137Cs. For animal products such as meat, milk and eggs, a preferred parameter is the feed transfer coefficient ($C_{ft}$), which is the fraction of radiocesium ingested daily that is transferred to 1 L of milk ($d_{L}^{-1}$) or 1 kg of meat or eggs ($d_{kg}^{-1}$) under steady-state conditions. Experimentally-determined values have ranged from 0.004 to 0.012 $d_{L}^{-1}$ for milk, 0.003 to 0.06 $d_{kg}^{-1}$ for beef, and 0.3 to 3 $d_{kg}^{-1}$ for eggs. Animals consume soil, both inadvertently and purposefully. The absorption of 137Cs from ingested soil can be considerably less than absorption from ingested biological material, depending on the degree of binding to clay minerals. The passage of radiocesium up through animal food chains, unlike the vast majority of other radionuclides, often increases from one trophic level to the next higher trophic level. For example, predatory animals tend to concentrate 137Cs in their soft tissues to a higher degree than do the animals upon which they feed. This trophic level increase in cesium concentrations is frequently in the range of two- to fourfold for each step in the animal food chain. The physiological basis of this trophic level effect is the chemical similarity of cesium and its nutrient analogue potassium, and the roughly threefold more rapid excretion rate of potassium. While potassium is homeostatically maintained within a certain range of concentrations in soft animal tissues, the molar concentrations of 137Cs are far too low to be limited by physiological mechanisms. Assimilation fractions across the gut wall for cesium and potassium are generally similar. As a result of these comparative behaviors, the ratios of intake to loss for body compartments (muscle tissue being the dominant compartment) are typically two to four times higher for cesium than for potassium.

Assimilation fractions ($f_a$) from the gut to the blood or body fluids of animals varies with the physical/chemical form of cesium, species, potassium ingestion rate and other factors, but the range of variation is relatively small. Most $f_a$ values fall in the range of ~0.6 to 0.9, but some can fall well outside this range. Radiocesium bound to 2:1 clay minerals is a case where assimilation fractions can be much smaller than the normal range. Considerable variation within a given species can occur depending on body mass and other factors. The body mass effect reflects metabolic rates per unit mass that typically increase as body mass decreases. High metabolic rates are characterized by increased food intake per unit body mass and high nutrient turnovers in muscle and other soft tissues. This is accompanied by rapid excretion rates of various elements, including cesium.

3. RADIATION EXPOSURE OF BIOTA

Major pathways leading to radiation exposure of plants and animals, in aquatic and terrestrial ecosystems, can be usefully considered in several different ways, as follows.

3.1. One of the pathways is Inhalation of (re)suspended contaminated particles or gaseous radionuclides (from air). This is significant for terrestrial animals and aquatic birds, reptiles, amphibians, and mammals. Gaseous exchange of volatile and respired forms of radionuclides at the stomata of plants also contributes to exposure as in Fig. 1.
Fig. 1 Terrestrial exposure pathways. (i) Inhalation of particles or gases. (ii) Contamination of fur/feathers/skin. (iii) Ingestion of animals of lower trophic levels. (v) Drinking contaminated water. (vi) External exposure through (a) air or (b) soil.

3.2 Another path is the Contamination of fur, feathers, skin, and vegetation surfaces comprising both external exposure component (e.g. beta- and gamma-emitting radionuclides on or near the epidermis cause irradiation of the underlying living cells) and internal exposure component (i.e. contaminants are ingested and incorporated into the body of the animal).

3.3 Ingestion of plants and animals. This leads to direct irradiation of the digestive tract, and internal exposure of radionuclide within the animal’s body. For some faunal types, ingestion of detritus and sediment is included.

3.4 Direct uptake from the water column. This may lead to direct irradiation of radionuclide of the gills or respiratory system, and internal exposure if the radionuclide becomes assimilated and distributed within the animal’s body.

3.5 Ingestion from water. This also leads to radiation exposure to animals. For plants, the corresponding pathway relates to root uptake of water.

3.6 Habitat exposure. This occurs mostly from gamma irradiation and, to a much lesser extent by beta irradiation, originating from radionuclides present in the organism’s habitat. The configuration of the source relative to the target clearly depends on the organism’s ecological characteristics and habitat. A benthic-dwelling adult fish will, for example, be exposed to radiation from radionuclides present in the water column and deposited sediments, whereas a pelagic fish may only be exposed to the former, although its eggs may be laid on or in the sediment.
3. EFFECTIVE MEASURES TO REDUCE THE RISK OF $^{137}\text{Cs}$

Cesium-137 is the radionuclide that generally drives human and ecological risks in the ecosystem and thus the radioactive contaminant targeted during remediation operations. Aquatic ecosystem including lakes, rivers, groundwater and seas, all have characteristic and site specific behavior governed by hydrological and morphological parameters of the reservoirs and its drainage area. Remediation of such reservoirs following the contamination of $^{137}\text{Cs}$ is therefore largely dependent on site specific parameters. Also, in general remediation plans for contaminated waters may be expensive and may include large engineering costs. The effects of such remediation efforts therefore normally must be on a cost benefit basis and chosen according to the well known ALARA principles and be compared to risks from other toxic substances present in the water (Brenner, 2003). Remediation of contaminated ecosystem involves physical, chemical and biological procedures which are described as follows;

A number of remediation techniques are available that can be employed to remove or immobilize hazardous substances in environmental systems. Radiological risks associated with radiocesium in soils and sediments are not likely to be eliminated by current remediation technologies, but levels or mobility can sometimes be reduced by application of an appropriate technique. Because of the strong and often irreversible binding of the majority of cesium in soils and sediments, complete elimination of the radionuclide is likely to require the physical removal of the soils or sediments themselves, followed by transport of the material to another site. Because this process is extremely expensive and may entail significant ecological damage, the so-called "no-action" alternative is usually considered (Zheng, 2012). It is generally believed by experienced scientists that the majority of cesium-contaminated areas may not warrant remedial action, based on the actual human health or ecological risk posed by the contamination. However, it can be a significant challenge to convince the public, and sometimes regulators and other decision makers, of this notion.

3. CONCLUSIONS

The evidence discussed in the present review article indicates that, following release into the environment, $^{137}\text{Cs}$ may persist in a biologically available form for many years. The high solubility of cesium, and its almost exclusive existence as $^{137}\text{Cs}$ in the ecosystem, dictate a high degree of mobility and bioavailability. Moreover, $^{137}\text{Cs}$ displays very similar chemical properties to other alkali metal cations with a similar ionic radius and charge, particularly the biologically essential $\text{K}^+$. Thus, $\text{Cs}^+$ may be taken up via transport systems which under normal circumstances catalyze the intracellular accumulation of $\text{K}^+$, and toxic effects of $^{137}\text{Cs}$ may result from perturbation of $\text{K}^+$ mediated processes. Despite these properties, the mobility of $^{137}\text{Cs}$ is strongly influenced by a number of parameters and this appears to be particularly the case in terrestrial ecosystems. One factor that is critical in determining cesium mobility in the environment is its tendency to become strongly complexes with inorganic components of soils. This behavior contrasts markedly with its weak coordination with organic
ligands. Complexation of $^{137}$Cs coincides with a dramatic reduction in its bioavailability. However, binding of $^{137}$Cs to minerals is not necessarily irreversible and this is particularly the case in terrestrial ecosystems, where recycling of bound $^{137}$Cs may result from changes in the redox conditions of the water column, for example. Remediation of aquatic systems following the contamination of $^{137}$Cs is largely dependent on site specific parameters. The effects of such remediation efforts therefore normally must be on a cost benefit basis and chosen according to the environmental conditions. Remediation of contaminated ecosystem involves physical, chemical and biological procedures that have its own advantages and disadvantages.

REFERENCES


